



Université du Québec à Chicoutimi

Tribological performance enhancement of AlTiN/AlTiCrN-based physical vapour deposition coatings for wood-processing application

By

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Amélioration des performances tribologiques des revêtements

AlTiN/AlTiCrN déposés par dépôt physique en phase vapeur pour les applications de transformation du bois

Résumé

La nature stochastique de l'usure des outils en usinage du bois, résultant de l'interaction complexe entre les propriétés du matériau bois, les paramètres de coupe et les conditions environnementales, continue de poser des défis majeurs à l'efficacité des opérations d'usinage. Parallèlement, la demande industrielle croissante pour une durabilité accrue et des performances économiques renforcées stimule l'intérêt pour des stratégies avancées d'amélioration tribologique, notamment l'utilisation de revêtements céramiques déposés par dépôt physique en phase vapeur (PVD). Dans ce contexte, cette thèse examine trois facteurs fondamentaux, mais encore peu étudiés, qui gouvernent de manière critique les performances des revêtements AlTiN/AlTiCrN destinés aux applications d'usinage du bois : (i) l'état de surface du substrat avant dépôt, (ii) la conception architecturale des revêtements multicouches, et (iii) la température de dépôt. En isolant et en analysant systématiquement chacun de ces paramètres, ce travail vise à établir un cadre scientifique cohérent pour optimiser le comportement des revêtements et combler des lacunes importantes dans l'amélioration des performances tribologiques des aciers à outils revêtus.

La première étude expérimentale analyse l'effet de la rugosité du substrat ($R_a = 0,0025$ à $1,7 \mu\text{m}$) sur les performances tribologiques d'un acier à outil AISI A8 revêtu d'AlTiN/AlTiCrN. Grâce à des essais de nanoindentation, des analyses d'adhérence, des caractérisations microstructurales et des tests d'usure abrasive, l'étude montre que les substrats plus lisses favorisent une texture cristallographique (111) plus marquée, une dureté supérieure, une meilleure résistance à la fracture ainsi qu'une adhérence accrue. Ces effets synergiques se traduisent par une réduction d'environ 15 % du taux d'usure par rapport aux substrats plus rugueux. Les résultats soulignent l'importance cruciale de la préparation du substrat et démontrent que l'optimisation de la rugosité constitue un levier essentiel pour améliorer le comportement tribologique des outils revêtus utilisés en usinage du bois.

La deuxième étude expérimentale examine la performance tribologique de revêtements multicouches AlTiN/AlTiCrN conçus avec des architectures bilayer et trilayer, déposés par PVD à évaporation cathodique. Trois configurations, désignées AlTiCrN I, II et III, ont été évaluées en termes de microstructure, densité de défauts, texture cristallographique, dureté, adhérence, résistance à la fracture et comportement à l'usure. Les architectures trilayer (AlTiCrN II et III) présentent une densité de macroparticules plus faible, une surface plus lisse, une orientation (111) renforcée et des propriétés mécaniques nettement améliorées par

rapport au revêtement bilayer (AlTiCrN I). De plus, le revêtement AlTiCrN III atteint les meilleures performances, avec des taux d'usure inférieurs de 29 % et 15 % à ceux des revêtements AlTiCrN I et II, respectivement. Cette performance supérieure est attribuée à sa dureté plus élevée, à sa meilleure adhérence, à son architecture optimisée et à sa réduction des défauts de surface. L'étude démontre ainsi le potentiel stratégique de l'ingénierie architecturale pour améliorer la fonctionnalité des revêtements en usinage du bois.

La dernière étude expérimentale porte sur l'influence de la température de dépôt (350 – 450 °C, désignée T350 à T450) sur l'évolution microstructurale, les propriétés mécaniques et le comportement à l'usure des revêtements AlTiN/AlTiCrN déposés sur acier AISI A8. Les résultats montrent que, bien que le revêtement T400 présente la dureté la plus élevée (36 GPa) due à des cristallites plus fines et une microdéformation accrue, sa densité supérieure de macroparticules et sa rugosité plus importante compromettent sa performance tribologique. À l'inverse, le revêtement T350 offre la meilleure résistance globale à l'usure, avec des taux d'usure inférieurs de 36 % et 56 % à ceux de T400 et T450, respectivement. Cette performance optimale est attribuée à une adhérence plus élevée (charge critique ~11 N), une surface plus lisse ($R_a = 0,25 \mu\text{m}$), une épaisseur accrue ($2,8 \mu\text{m}$) et un rapport H/E plus favorable, malgré une dureté intermédiaire. Ces résultats mettent en évidence la nature spécifique du rôle thermique dans les revêtements PVD à arc cathodique et montrent que 350 °C constitue une température optimale pour un compromis idéal entre propriétés microstructurales et performance tribologique en environnement d'usinage du bois.

Dans l'ensemble, cette thèse propose une compréhension intégrée de l'influence de l'ingénierie du substrat, de la conception architecturale et de la température de dépôt sur les performances des revêtements AlTiN/AlTiCrN utilisés en usinage du bois. Les résultats contribuent à l'avancement des connaissances en tribologie des revêtements durs et fournissent une base scientifique pour le développement de systèmes de revêtements de nouvelle génération offrant une durabilité, une adhérence et une résistance à l'usure accrue

Tribological performance enhancement of AlTiN/AlTiCrN-based physical vapour deposition coatings for wood-processing application

Abstract

The stochastic nature of tool wear in wood machining, driven by the complex interaction of wood properties, machining parameters, and environmental conditions, continues to pose major challenges to machining efficiency. At the same time, the growing industrial need for higher durability and cost-effective performance has intensified interest in advanced tribological enhancement strategies, particularly ceramic coatings deposited via physical vapour deposition (PVD). Building on these motivations, this thesis examines three fundamental, yet under-explored factors that critically govern the performance of AlTiN/AlTiCrN coatings for woodworking applications: (i) substrate surface condition prior to deposition, (ii) architectural design of multilayer coatings, and (iii) deposition temperature. By systematically isolating and analyzing each factor, the work aims to establish a coherent scientific framework for optimizing coating behaviour and addressing long-standing gaps in enhancing the tribological performance of coated tool steels.

The first experimental study investigates how substrate roughness ($R_a = 0.0025 - 1.7 \mu\text{m}$) affects the tribological performance of AlTiN/AlTiCrN-coated AISI A8 tool steel. Through nanoindentation, adhesion, microstructural analyses, and abrasive wear testing, the study demonstrates that smoother substrates promote a stronger (111) crystallographic texture, improved hardness, enhanced fracture resistance, and higher adhesion strength. These synergistic effects result in approximately 15 % lower wear rates compared to rougher substrates. The findings underscore the critical importance of substrate preparation and suggest that optimizing roughness is essential for achieving superior tribological behavior in coated wood-machining tools.

The second experimental study examines the tribological performance of engineered multilayer AlTiN/AlTiCrN coatings with distinct bilayer and trilayer architectures deposited via cathodic arc evaporation PVD. Three coating configurations designated as AlTiCrN I, II, and III were evaluated for their microstructure, defect density, crystallographic texture, hardness, adhesion, fracture resistance, and wear behavior. The trilayer designs (i.e. AlTiCrN II and III) exhibited reduced macroparticle density, smoother surfaces, improved (111) orientation, and markedly enhanced mechanical properties relative to the bilayer coating (i.e. AlTiCrN I). Furthermore, the AlTiCrN III coating achieved the highest performance, with wear rates 29 % and 15 % lower than AlTiCrN I and II coatings, respectively. The superior performance of the AlTiCrN III coatings was attributed to its higher hardness, improved adhesion strength, optimized architecture, and reduced surface defects. The study demonstrates how architectural tailoring can strategically enhance coating functionality for woodworking applications.

The final experimental investigation focuses on the influence of deposition temperature (350 – 450 °C, designated as T350 to T450) on the microstructural evolution, mechanical properties, and wear performance of AlTiN/AlTiCrN coatings deposited on AISI A8 tool steel. The results revealed that, although the T400 coating exhibited the highest hardness (36 GPa) due to finer crystallites and elevated microstrain, its higher macroparticle density and increased roughness compromised tribological performance. In contrast, the T350 coating showed the best overall wear resistance, with wear rates 36 % and 56 % lower than T400 and T450, respectively. This superior performance is attributed to its stronger adhesion (critical load ~11 N), smoother surface ($R_a = 0.25 \mu\text{m}$), greater coating thickness (2.8 μm), and higher H/E ratio, despite its intermediate hardness. These findings highlight the system-specific nature of thermal effects in CAE-PVD coatings and reveal that 350 °C presents an optimal deposition temperature for achieving balanced microstructural and tribological performance in woodworking environments.

Overall, this thesis provides an integrated understanding of how substrate engineering, architectural design, and deposition temperature govern the performance of AlTiN/AlTiCrN coatings used in wood machining. The findings contribute to advancing knowledge in the tribology of hard coatings and offer a scientific basis for developing next-generation coating systems with enhanced, adhesion, and wear resistance.

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List of Abbreviations

Abbreviation	Full Form
AISI	American Iron and Steel Institute
CAE	Cathodic Arc Evaporation
CAE-PVD	Cathodic Arc Evaporation Physical Vapour Deposition
COF	Coefficient of Friction
EDX / EDS	Energy-Dispersive X-ray Spectroscopy
E	Elastic Modulus
fcc	Face-Centred Cubic
H	Hardness
H/E	Hardness-to-Elastic Modulus Ratio
H^3/E^2	Plastic Deformation Resistance Index
MP	Macroparticle
PVD	Physical Vapour Deposition
SEM	Scanning Electron Microscopy
XRD	X-Ray Diffraction

List of Symbols

Symbol	Meaning
Al	Aluminium
AlTiCrN	Aluminium Titanium Chromium Nitride
AlTiN	Aluminium Titanium Nitride
Cr	Chromium
CrN	Chromium Nitride
Fe	Iron
mm/min	Millimetre per minute (scratch speed)
mm ³ /Nm	Cubic millimetre per newton-metre (specific wear rate)
N	Nitrogen (element)
N ₂	Nitrogen gas
N/min	Newton per minute (loading rate)
R _a	Arithmetic average roughness
R _z	Maximum height of the roughness profile
sccm	Standard cubic centimetres per minute (gas flow unit)
Si	Silicon
TiN	Titanium Nitride
WC-Co	Tungsten Carbide–Cobalt

Dedication

I dedicate this work to my family, loved ones, and friends, whose unwavering support, encouragement, and belief in me have been a constant source of strength. Your contributions (big and small) have shaped this journey and made the achievement of this milestone possible. This accomplishment is as much yours as it is mine.

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Chapter 1

Introduction

1.1 Background of study

Over the years, the demand for wood and wood-based composites has risen considerably due to global population growth and the increasing need for sustainable construction, furniture, and packaging materials [1]. However, machining wood presents substantial challenges. During the process, cutting tools encounter a highly aggressive environment characterized by abrasive hardwood fibres, high-impact loads from knots and trunk irregularities, and tribochemical degradation caused by embedded silica and organic acids [2]. Additionally, the high temperatures (which could exceed 800 °C) generated by friction during cutting can lead to oxidation and adhesive wear, particularly when processing resinous wood species [3]. These complex interactions frequently result in rapid tool wear, increased energy consumption, and a deteriorated surface finish of the machined components, ultimately compromising both productivity and product quality [4].

To address these challenges, numerous performance enhancement strategies have been developed, beginning with bulk hardening techniques such as heat treatment and cryogenic processing. However, these methods are energy-intensive and provide suboptimal performance relative to their cost [5], [6]. Surface hardening methods, including carburizing and nitriding, have shown some improvement in tribological performance, yet they remain constrained by inflexibility and the strong dependence of the performance on the base material's properties [7]. As a result, researchers have increasingly turned to surface coating

technologies, particularly physical vapour deposition (PVD), due to their ability to improve tribological performance without significant bulk modification. Compared to conventional methods, PVD coatings offer superior adhesion, chemical stability, wear resistance, and compatibility with diverse substrate materials, making them highly attractive for cutting tool applications such as woodworking [8]. Furthermore, advancements in coating architectures from binary (e.g., CrN) to quaternary (e.g., AlTiCrN) systems and layered designs have led to significant improvements in tribological performance [9], [10].

Recently, layered AlTiN/AlTiCrN-based coatings have emerged as promising candidates for woodworking applications. These coatings combine the benefits of a multilayered architecture with the superior properties of quaternary AlTiCrN components to achieve enhanced mechanical strength, oxidation resistance, and chemical stability, ultimately extending cutting tool life [11]. Moreover, the performance of these coatings is strongly influenced by processing parameters, with substrate preparation, deposition temperature, substrate bias, and architectural design identified as critical factors [8]. Therefore, optimizing these parameters is essential for achieving superior tribological properties and ensuring consistent, high-performance outcomes in wood machining operations.

1.2 Problem statement

Since the last four decades, PVD coatings such as AlTiN and AlTiCrN have gained increasing attention for their ability to enhance tool life and performance in demanding applications. However, despite their promising potential, the tribological behaviour of AlTiN/AlTiCrN-based coatings remains relatively underexplored, particularly in the context of wood-processing tools where severe wear and friction are critical challenges. Existing

studies have largely examined AlTiCrN in monolayer [12], or multilayer architectures combined with binary (TiN, CrN) or ternary (TiSiN) coatings [13], while systematic investigations into layered AlTiN/AlTiCrN systems are scarce. Moreover, prior research has primarily focused on compositional variations while maintaining constant deposition parameters [14], overlooking the significant role of process variables such as substrate roughness, bias voltage, and deposition temperature.

The complexity of substrate–coating interactions further amplifies this knowledge gap. While the influence of substrate roughness on PVD coating performance has been studied in limited contexts, the findings are often system-specific and not directly transferable. Notably, no study has addressed the effect of substrate roughness on AlTiN/AlTiCrN-coated AISI A8 tool steel, despite its widespread industrial use. Additionally, limited substrate roughness ranges have been considered, restricting understanding of how tribological performance evolves across broader conditions. Similarly, the effects of deposition parameters, such as substrate bias and nitrogen gas flow, on the tribological response of layered AlTiN/AlTiCrN coatings remain unexplored, even though these factors critically govern coating microstructure and performance.

Therefore, there is a clear need for systematic investigation into the relationship between deposition parameters, substrate condition, and the tribological behaviour of AlTiN/AlTiCrN-based coatings. Addressing this gap will provide fundamental insights for optimizing the coating performance and extending tool life in wood-processing applications.

1.3 Objectives

The primary objective of this study is to enhance the tribological performance of AlTiN/AlTiCrN-based coatings deposited on AISI A8 tool steel for wood-processing applications. The research aims to systematically evaluate how substrate preparation, layered architectural design, and deposition temperature influence the microstructure, mechanical properties, and tribological behavior of these coatings. This study is organized into three parts with the following specific objectives:

1. Effect of substrate preparation on the tribological performance of AlTiN/AlTiCrN coated-A8 tool steel

- I. Investigate the influence of varying substrate roughness on the microstructural evolution of AlTiN/AlTiCrN-coated A8 tool steel.
- II. Evaluate the effect of substrate roughness on hardness, fracture toughness, and adhesion strength.
- III. Examine the relationship between substrate roughness and wear resistance.
- IV. Correlate the microstructure, mechanical properties, and tribological performance at different substrate roughness conditions.

2. Influence of layered architectural design on the tribological performance of AlTiN/AlTiCrN-based coatings

- I. Develop AlTiN/AlTiCrN-based layered coatings by varying substrate bias and reactive gas flow parameters.
- II. Characterize the microstructure of the developed coatings and compare them with conventional AlTiN/AlTiCrN coatings.

- III. Evaluate hardness, fracture toughness, and adhesion strength of the newly designed coatings relative to conventional counterparts.
- IV. Assess the wear resistance of the developed coatings and benchmark against conventional designs.
- V. Correlate microstructural features and mechanical properties with tribological performance, and identify the critical factors governing coating performance.

3. Effect of deposition temperature on the tribological performance of AlTiN/AlTiCrN-coated A8 tool steel

- I. Investigate how deposition temperature influences surface morphology and roughness of AlTiN/AlTiCrN coatings.
- II. Assess changes in hardness, fracture toughness, and adhesion strength under varied deposition temperatures.
- III. Examine the effect of deposition temperature on wear resistance.
- IV. Correlate between microstructural features, mechanical properties, and tribological performance at different deposition temperatures.

1.4 Originality statement

The originality of this research lies in addressing fundamental gaps in the understanding and optimization of AlTiN/AlTiCrN-based coatings for tribological applications. While AlTiN and AlTiCrN coatings have been individually recognized for their superior hardness and wear resistance, systematic investigations into their tribological performance of the bilayer coating on AISI A8 tool steel remain absent. This study represents the first comprehensive effort to investigate how substrate roughness, layered coating

architectures, and deposition temperature influence the microstructure, mechanical properties, and wear behaviour of these coatings in wood-processing applications.

First, the effect of substrate roughness on the tribological performance of AlTiN/AlTiCrN-coated A8 tool steel was systematically examined for the first time. Previous studies have reported inconsistent findings regarding the role of roughness, often limited to specific substrate–coating systems and narrow roughness ranges. By extending the evaluation across broader ranges and directly linking roughness to microstructural features, hardness, adhesion, and wear resistance, this study generates novel insights with direct relevance to coating–substrate engineering.

Second, this research introduces a novel architectural design strategy by developing layered AlTiN/AlTiCrN coatings using controlled variations in bias voltage and reactive gas flow during deposition. Prior works have primarily modified elemental composition under fixed process conditions; here, deposition parameters are strategically altered to produce unique coating architectures. This methodological innovation not only advances understanding of the structure–property relationship but also demonstrates a new pathway for tailoring PVD coatings.

Third, the influence of deposition temperature on the microstructural evolution and tribological behaviour of AlTiN/AlTiCrN coatings on A8 tool steel was systematically investigated for the first time. Although temperature is known to affect PVD coating properties in general, no prior study has specifically addressed its impact in the context of these coatings and substrates.

Finally, by combining wear testing, nanoindentation, adhesion assessment, profilometry, and advanced microstructural characterization, this work establishes robust microstructure–property–performance correlations. Through this integrated approach, the most critical factors governing the tribological performance of AlTiN/AlTiCrN coatings are identified.

In summary, this study makes original contributions by exploring previously unaddressed variables, introducing a novel layered design approach, and providing new insights for optimizing coating performance in tribologically demanding wood-processing applications.

1.5 Thesis Outline

This PhD thesis is organized into seven chapters.

Chapter 1 – Introduction

The first chapter provides the overall background of the project, beginning with an overview of tool steels in wood-processing applications and the challenges posed by severe wear and frictional conditions. It then defines the research problem, states the project objectives, and highlights the originality and significance of the study.

Chapter 2 – Literature review I

Chapter 2 presents a literature review based on the article “*Enhancing the tribological performance of tool steels for wood-processing applications: A comprehensive review*” published in *Metals*. This chapter discusses recent developments in wood machining, emphasizing the role of wood properties and environmental conditions on tool

wear. It further examines different approaches for improving tool performance, including heat treatment, thermochemical treatments, hybrid methods, and coating deposition.

Chapter 3 – Literature review II

Chapter 3 provides a systematic review drawn from the article “*A comprehensive review of cathodic arc evaporated physical vapour deposition coatings for enhanced tribological performance*” published in *Coatings*. Key topics covered include substrate preparation, fundamentals of cathodic arc evaporation (CAE) PVD, coating microstructural evolution, and advances in multilayer architectures. In addition, this chapter reviews characterization techniques for evaluating coating properties and explores how process parameters such as arc current, deposition temperature, and substrate bias influence microstructure and tribological behaviour.

Chapter 4 – Substrate roughness effects

Chapter 4 presents the study “*Effect of substrate roughness on the tribological performance of AlTiN/AlTiCrN-coated A8 tool steel*” published in *Metallurgical and Materials Transactions A*. This chapter systematically investigates the influence of substrate roughness ($R_a = 0.003 - 1.7 \mu\text{m}$) on coating microstructure, morphology, surface roughness, and mechanical properties. The results are correlated with wear performance to identify the most critical microstructural and mechanical factors governing coating behaviour.

Chapter 5 – Layered architectural design

Chapter 5 is based on the article “*Effect of layered architectural design on the tribological behaviour of AlTiN/AlTiCrN-based cathodic arc evaporated physical vapour deposition coatings*” published in *Metallurgical and Materials Transactions A*. This chapter

examines the tribological behaviour of three novel layered AlTiN/AlTiCrN architectures produced by varying the substrate bias (150 – 200 V) and nitrogen gas flow (500 – 700 sccm). Comprehensive microstructural, mechanical, and wear characterizations are presented to establish performance-property relationships and highlight the benefits of layered design strategies.

Chapter 6 – Deposition temperature effects

Chapter 6 is based on the article “*Influence of Deposition Temperature on the Microstructure, Wear and Corrosion Behaviour of AlTiN/AlTiCrN Ceramic Coatings on A8 Tool Steel* ” published in *Ceramics International*. This chapter analyzes the influence of deposition temperatures (350 – 450 °C) on microstructure, mechanical response, and wear performance. The study provides detailed microstructure–property–performance correlations to explain the temperature-dependent tribological behaviour of the coatings.

Chapter 7 – Conclusions and recommendations

The final chapter synthesizes the key findings from all experimental studies, highlighting their implications for improving coating design and performance in wood-processing applications. It concludes with recommendations for future research directions, focusing on further optimization of deposition strategies and extension to other tooling and industrial contexts.

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Chapter 2

Enhancing the tribological performance of tool steels for wood-processing applications: A comprehensive review

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Abstract

The stochastic nature of tool wear during wood machining, owing to the dynamic properties of the biological material and its dependence on various factors, has raised significant industrial and research concerns in recent years. Explicitly, the tool wear is a product of the interaction between wood properties (such as hardness, density, and contamination level), machining parameters (such as cutting speed, feed rate, and rake angle) alongside ambient conditions (such as temperature and humidity). The objective of this review paper is to provide an overview of recent advancements in the field of wood machining. To begin with, it highlights the important role of wood properties and ambient conditions in influencing tool wear. Furthermore, the paper examines the various mechanisms involved in the wood machining process and discusses their cost implications from an industrial perspective. It also covers technological advancements in the characterization of tool wear and explores the relationship between this parameter and other machining variables. It provides critical and analytical discussions on various methods for enhancing tool wear, including heat treatment, cryogenic treatment, thermochemical

treatment, coating deposition, and hybrid treatments. Additionally, the paper incorporates statistical analysis to achieve two objectives: Firstly, it aims to identify the most significant wood property that affects tool wear and establish the correlation between this parameter and wood properties. Secondly, it investigates the effect of heat treatment parameters and carbide characteristics on tool wear, as well as their correlation. Lastly, the review provides recommendations based on relevant literature for prospective researchers and industrial counterparts in the field. These recommendations aim to guide further exploration and practical applications in the subject matter.

Keywords: Wood machining, Tool steels, Tool wear, Wear resistance, Correlation analysis.

2.1 Introduction

Over the past two decades, there has been a significant growth in the demand for wood and wood composites for various applications, driven by the increasing global population [1]–[3]. This can be attributed to their availability [4], high specific strength [5], insulation properties [6], aesthetic appeal [4], and relatively low environmental impact [7]. For instance, wood and its composites find application in building construction [8], energy [9], packaging [4], [10], electronics [9], and furnishing [3], [11]. The transformation of raw wood into finished products involves primary and secondary transformation processes [12], [13]. Primary processes include cutting the tree and branches, debarking, chipping, and cantering [12], [14], while milling [10], drilling [15], shaping, and planing operations [3], fall under the secondary transformation category.

The process of wood transformation is highly intricate, involving the interaction between a cutting tool with a sharp edge and the impact from either the tree trunk or a partially

machined section, such as a precut slice. Additionally, the process involves the presence of other hard materials, such as silica contaminants, as well as organic acids like tree sap [5]. According to forest management inventory data published by Natural Resources Canada, in 2020, approximately 143,061,196 cubic meters of trees were harvested in Canada for both domestic and foreign consumption. This harvesting process results in the generation of approximately 1.75 million metric tonnes of wood waste and contributes to approximately 3.5 megatonnes of CO₂ emissions annually [16], [17]. One significant factor that contributes to wood wastage is the use of low-quality cutting tools. To address this issue, there is a need for technologically advanced cutting tools that can consistently deliver high cutting performance throughout their lifespan. By utilizing such tools, optimal machining can be achieved, resulting in high-quality products and a reduction in wood waste generation and greenhouse gas emissions. This drives the continuous pursuit of high-performance cutting tools by wood processing companies. The cutting performance of a tool plays a crucial role in determining the competitiveness of a cutting tool manufacturing company within the industry. However, it is important to highlight that the property requirement of the cutting tool depends on the wood machining operation. For instance, operations such as debarking, cantering, or continuous sawing are perceived to have a more severe effect on the cutting tool than finishing operations such as planing. Consequently, the cutting tools for the former would necessitate a higher set of property requirements than the latter [18]. Key characteristics used to evaluate cutting performance include hardness, toughness, and wear resistance of the tool. However, the manufacturing of high-performance cutting tools that exhibit prolonged cutting ability with high sharpness, elevated resistance to impact and to

surface deformation, considering the influence of the aforementioned factors, is a highly challenging task [12], [13].

Over time, wood transformation has been achieved using a variety of cutting tools, including tool steels [10], cemented carbides [19], cobalt-base alloys [20], and ultrahard tool materials [21]. Tool steels are iron-carbon alloys with carbon contents ranging from moderate (about 0.5 %) to high amounts (about 2.25 %) [12], [13]. Cemented carbides are composed of a hard carbide material that is bonded together using a soft and ductile binder, such as tungsten, nickel, or cobalt [4], [12]. On the other hand, cobalt-based alloys exhibit properties that lie between those of tool steels and cemented carbides. One popular cobalt-based cutting tool is Stellite [18], [19], [21]. Ultrahard tool materials, on the other hand, exhibit extremely high wear resistance and are primarily used for machining hard woods that cannot be easily transformed using the aforementioned cutting materials. Examples of ultrahard tool materials include Polycrystalline Diamond (PCD) and Polycrystalline Cubic Boron Nitride (PCBN) [12], [22], [23].

The selection of an appropriate cutting tool for wood machining often requires striking a balance between wear resistance, toughness, and cost considerations [12]. For example, while PCD and PCBN exhibit excellent wear resistance, they have lower toughness and higher cost. In contrast, tool steels offer excellent toughness and are more cost-effective, but their wear resistance is comparatively lower [4], [20], [23]. Additionally, achieving a balance between cutting speed and feed rate is crucial. Ultrahard materials can withstand high cutting speeds but can handle relatively lower feed rates, while tool steels exhibit the opposite behaviour [22]. Therefore, the selection of cutting tool materials depends on various factors, including the type of wood being machined [24], desired performance in terms of wear

resistance and toughness [22], and available resources [10]. Additionally, it is important to emphasize the significant role of the wood type on the cutting performance. For instance, a given cutting tool used for machining a very tough wood (e.g., oak wood) [25] or wood species containing a lot of defects (e.g., knotty pine) [26] or even high moisture content (e.g., sugar maple) [27] would likely display a relatively reduced cutting performance when compared to one used for machining one with lower toughness (e.g., black spruce) [28], or one with minimal defects (e.g., african mahogany) [29] or one with low moisture content (e.g., teak) [4], [30]. Again, machining of wood composites characterized by high abrasive properties, such as particle board [31], Medium Density Fibreboard (MDF) [32], Oriented Strand Board (OSB) [33] are also prone to higher tool wear than those used for machining natural wood [11].

In many cases, tool steel is the preferred choice for cutting tools due to several advantages. Firstly, tool steels are readily available and relatively affordable compared to other materials. Secondly, they offer a good balance of wear resistance and toughness, even at high temperatures. Thirdly, tool steels have a low wedge angle, which allows for efficient cutting [10]. Additionally, tool steels can be easily fabricated into various cutting tool shapes, including intricate designs [34]–[36]. Furthermore, tool steels can be modified and tailored to specific applications through heat treatment, providing further versatility and advantage over cobalt-base alloys [12]. Tool steels are commonly classified according to the American Iron and Steel Institute (AISI) and Society for Automotive Engineers (SAE) standards, with the AISI standard gaining wider acceptance in recent years [37]. Under the AISI classification, tool steels are categorized into various types, including water-hardening (W), shock-resisting (S), mold steels (M), high-speed (T and M), hot-work (H), and cold-work

(O, A, and D) tool steels, among others [38]. However, when it comes to wood machining, high-speed and cold-work tool steels are the most commonly used types [5], [12], [39], [40].

Unfortunately, even though tool steel remains prevalent in the wood-cutting industry, manufacturers of wood products frequently face the challenge of low tool life. Tool steels often fall short in sustaining the desired product quality, leading to increased production downtime for tool edge replacement or regeneration. This directly impacts production costs, as the frequent need for tool maintenance and replacement drives up expenses [41], [42]. Consequently, scientists and industry professionals have initially focused on optimizing machining parameters in an attempt to address this issue [43], [44]. Subsequently, attention turned towards technologically enhancing the surface properties of tool steels [12]. Various approaches were explored, including modifying the surface microstructure [45], altering surface chemistry [46], or applying surface coatings [39]. Bendikiene and Pupelis also proposed submerged arc surfacing with alloying as a cost-effective alternative to conventional surface engineering processes [3]. Additionally, the adoption of powder metallurgy as a method for manufacturing tool steels has been recommended by others, citing improvements in microstructure and mechanical properties [23]. As a result, significant advancements in tool life have been observed over the years [4], [41]. These innovative approaches offer promising solutions to enhance the performance and durability of tool steels in wood machining applications.

A literature survey has revealed that studies on wood machining are relatively limited compared to metal machining. However, it should be noted that the technological advancements and innovations in metal machining cannot be wholly applied to wood machining. This is primarily due to the significant differences in the material properties and

machining mechanisms between wood and metal [4]. Wood machining, in fact, is even more complex than metal machining. This is primarily due to the combination of high machining speed [19], intricate machining conditions [47], and the influence of multiple factors [48]. Furthermore, it is worth noting that the number of published review articles on wood machining remains limited, indicating a scarcity of comprehensive literature in this area. In the past three years, Nasir and Cool conducted an extensive analysis on wood sawing, covering various factors that impact the process and exploring methods for optimization [44]. Additionally, Al-Asadi and Al-Tameemi conducted a comprehensive review on different coatings applied to cutting tools, emphasizing the factors that influence their tribological performance [41]. Recently, Warcholinski and Gilewicz provided a comprehensive examination of cutting tools used in wood processing, focusing on enhancing cutting efficiency through coating deposition [4]. However, it is surprising that despite the progress made in research, there has been a lack of effort to quantify the correlation between wood properties and tool wear. This is equally applicable to the properties of the cutting tool and tool wear, particularly the microstructural composition such as the carbide characteristics of tool steels, which have a substantial impact on the tribological performance of the cutting tool. Establishing these relationships can enhance our current understanding of tool wear in wood machining and provide valuable tools for predicting and forecasting related parameters. Therefore, this article begins by addressing the challenges associated with wood machining using cutting tools and presents recent advancements in tool wear characterization. Subsequently, it examines the relationship between tool wear, wood properties, heat treatment parameters of tool steels, and carbide characteristics using correlation and regression analysis. Moreover, it provides critical analytical discussions on

heat treatment, cryogenic treatment, and other surface engineering techniques aimed at enhancing tool life. The objective is to identify research gaps for future researchers and to serve as a valuable resource for industrial professionals in the field.

2.2 Wood machining: challenges, temperature effects, and wear mechanisms

2.2.1 Wood machining

Wood machining involves sectioning the material to achieve a specific product with desired geometry and a high-quality surface finish. The chips generated during this process can be viewed as valuable wood products or waste, depending on the machining operation. Wood industries that carry out operations like milling, drilling, sawing, turning, and planing typically consider the chips as waste. Conversely, industries involved in flat slicing or peeling, such as veneer production, may view the chips differently. However, the majority of wood processing industries fall into the former category. Moreover, wood sawing constitutes the bulk of the wood processing operations, as it is widely used in primary and secondary transformation processes. Band sawing and circular sawing are the most common sawing operations in wood processing [44]. In wood cutting, the cutting tool section responsible for cutting through the wood is typically triangular in shape. Figure 2-1 shows a simplified model of the wood-cutting process [13].

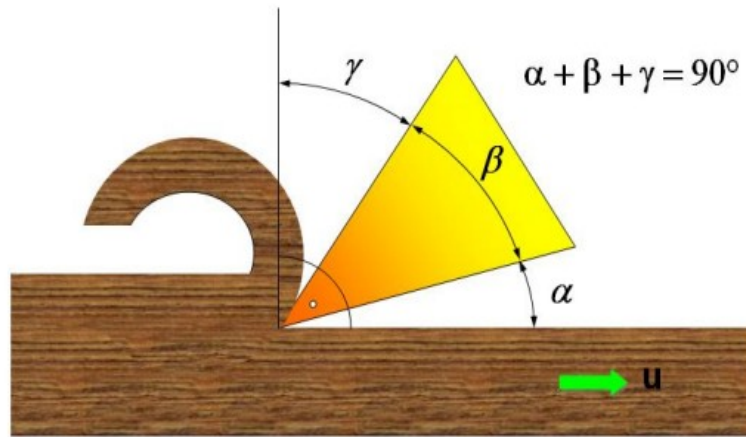


Figure 2-1: A Simple model showing the wood-cutting process (γ – rake angle, β – wedge angle, α - clearance angle and u - cutting velocity) [13].

The edges of the cutting tool engaged during wood machining depend on the type of operation. For instance, in machining operations such as sawing, where the wood thickness is more than the length of the tool edge, both edges are usually engaged. Meanwhile, during wood planing, only one cutting edge is engaged, as the length of the cutting edge is usually more than the wood thickness. However, during wood milling, both aforementioned

conditions are applicable [13]. Figure 2-2 shows some contemporary cutting tools for different wood machining operations.

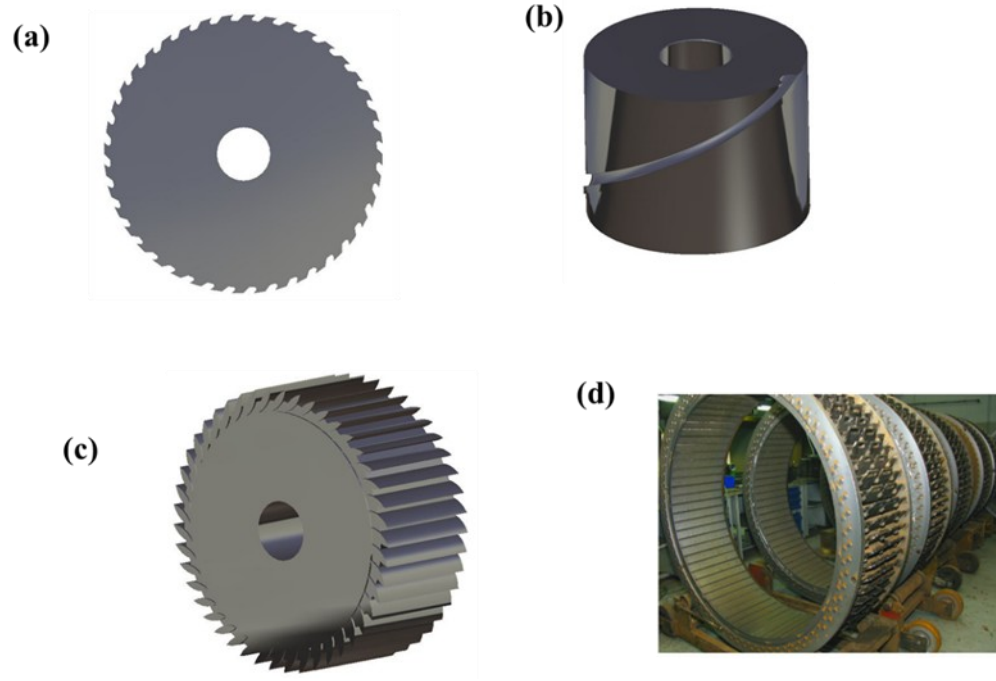


Figure 2-2. Wood machining tools: (a) Saw (for wood sawing); (b) Milling cutters (for wood milling); (c) Planer head (for wood planing); (d) Drum chipper (for chipping of wood composites [6], [49].

2.2.2 Challenges of wood machining: The dynamic properties of wood

Since the inception of wood machining, the wood industry has been faced with the challenge of producing good quality wood products while simultaneously catering for the market demand. However, over time, though the wood machining industry has adopted automation of the machining process to cater for the ever-increasing demand for wood products, the industry is still confronted with the need to produce better quality wood products which requires high-performance cutting tools. Unfortunately, this objective is yet

to be fully actualized, primarily owing to the insufficient tool life of the existing cutting tools [10], [19]. This situation is further aggravated by the contrasting properties (physical, mechanical, and chemical) of the wood and the cutting tools [5], [34].

On one hand, tool steel possesses a considerable combination of hardness and toughness, while wood, on the other hand, is a natural composite material whose property is significantly influenced by its composition [3]. Chemically, it contains cellulose, hemicellulose, lignin, extractives, organic acids, and mineral contaminants. Macroscopically, it contains piths, growth rings, sapwood, heartwood, rays, and resin canals [6], [50], [51]. Firstly, the biological nature of the material accounts for the variability in the properties even within a given specie. Secondly, its properties vary across cross-sections, radial, and transverse sections (anisotropy). Additionally, due to its hygroscopic nature, wood is in constant equilibrium with atmospheric water vapor, allowing it to absorb and release moisture from the surrounding environment. This implies a constantly changing wood moisture composition [6], [7], [52]. Therefore, the anisotropy and hygroscopicity of wood directly influence other properties such as strength, hardness, and workability. The dynamic properties of wood result in constantly changing machining parameters such as the cutting force and resistance [53]. Furthermore, the presence of knots and hard mineral contaminants of various sizes and colours also affect the wear of tool steels [5], [54]–[56]. Moreover, organic acids such as acetic acid (e.g., in red oak) [57], formic acids (e.g., in pine) [58], oxalic acids (e.g., in walnut) [59], and tannic acids (e.g., in chestnut) [60] present in different concentrations also affect tool wear. Their effect is even more severe at higher concentrations as they contribute to tribochemical reactions during wood machining. But, unfortunately, the concentration of these acids also varies within a given wood specie due to

changes in climatic conditions [61]. Consequently, the dynamic properties of wood make wood machining very complex. For instance, a given cutting tool which had an excellent wear performance in the machining of a given wood specie may perform poorly when used for machining another wood of a similar specie [4], [34], [62]. Furthermore, fluctuations in climatic factors, especially temperature, further contribute to the dynamic nature of wood properties, affecting the machining process.

2.2.3 The role of temperature in wood machining

The temperature of the cutting tool and that of the surroundings play a pivotal role in the wood transformation process. Regrettably, despite the dynamic machinability of wood, lubricants or coolants are not used. This is due to the hygroscopic nature of the material. Already, the water and other organic acids contents of wood have a corrosive effect on the cutting tool. The addition of lubricants or coolants would result in unforeseen reactions, which might further increase the corrosivity or compromise the wood quality [10], [19], [52]. Hence, the inability to use lubricants or coolants creates the tendency for temperature rise during machining. The temperature rise varies from small increments (about 50 °C) to large amounts (about 800 °C) depending on the machining conditions [4], [63]. During wood machining, the friction between the tool edges and the wood, alongside the continuous shear and plastic deformation during chip formation, leads to the generation of thermal energy resulting in the temperature rise. The energy is absorbed partly by the cutting tool and the wood, while the remainder is lost to the surroundings. The detrimental effect of the temperature rise is in two folds: on one hand, it compromises the desired accuracy and quality of the wood products. On the other hand, it degrades the cutting tool and affects the

machining parameters. The tool steel degradation is characterized by a reduction in mechanical properties (hardness and toughness) and chemical stability, resulting in a reduced life span. Machining parameters such as cutting force [54], power consumption [10], depth of cut [5], and cutting speed [3] are also negatively affected. In this regard, a study was conducted by Zhu et al. on the relationship between cutting force, machining temperature, and tool wear during the milling of wood plastic composites using cemented carbide cutters. Their findings revealed that an increase in temperature of the cutting edge was accompanied by an increase in the cutting force of the cemented carbide cutters. This is an indication of increased power consumption. Additionally, the temperature rise was also correlated with increased tool wear [64]. In another study, Pei et al. reported a negative correlation between the machining temperature and the feed rate during the milling of wood plastic composite [65].

Furthermore, the ambient machining temperature affects the properties of the wood and also the quality of the machined products. For instance, the strength of wood varies with fluctuations in ambient temperature caused by seasonal changes [6], [12]. Meanwhile, in a study conducted by Hernandez and Boulanger on the effect of ambient temperature on chip dimension for pulp industry application, it was observed that the sizes of the chips produced during summer were quite distinct from those produced in winter. Irrespective of the machining speed, the number of fines and pins produced in winter were twice those produced in summer, while fewer over-thicks were produced during summer than in winter. The fluctuation in the chip dimension was attributed to the changes in the wood properties with ambient temperature [66].

2.2.4 Wear and its mechanisms

Wear is the gradual material loss from the surface of a material due to mechanical or chemical factors during relative motion [41], [54], [67], [68]. The surface properties of the participating materials, such as roughness and hardness, significantly influence the process [67]. For instance, the process has been observed to proceed faster with a lower hardness ratio (substrate/abradant) of the participating materials [69]. The product of the process (wear debris) either sticks with one of the materials or is released into the environment. Its aftermath includes loss of cutting ability [5], reduction in cutting speed [10], [37], increase in power consumption [4] , increase in downtime [41], and reduction in the quality of machined products [53]. Even after grinding (re-sharpening), the functional surface of the knives cannot be regained [53]. As a matter of fact, the wear of cutting tools is of great concern to the industrial counterpart due to its direct impact on the cost of production. Over time, researchers have recognized that the wear of tool steels progresses in three distinct stages. The first stage is usually characterized by a very fast rate of wear followed by a stable linear wear rate, as shown in Figure 2-3. The last stage represents tool failure where it is no longer usable [5], [54], [70], [71].

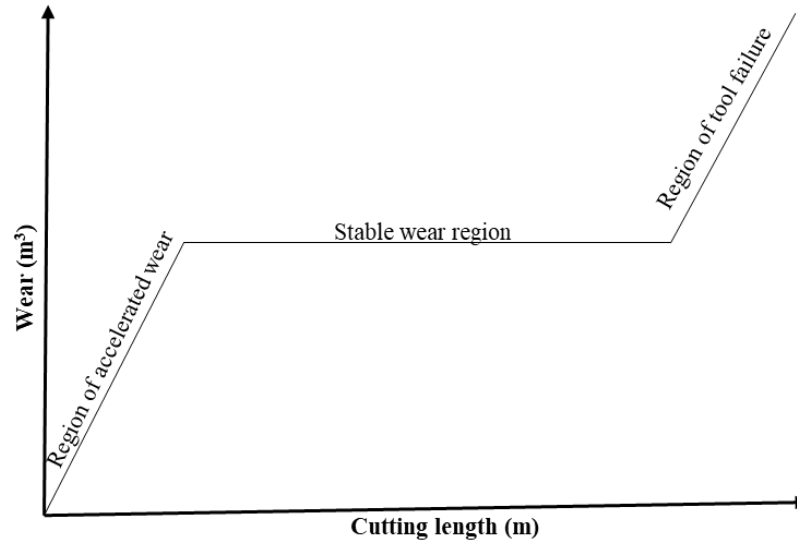


Figure 2-3: Stages of cutting tool wear during wood machining [72].

Tool wear during wood machining is a product of the interaction of the cutting tool with the wood, involving physical, mechanical, and chemical processes that lead to the transfer of materials and energy. The most prevalent wear mechanisms reported in literature are abrasion, adhesion, surface fatigue, and tribochemical reaction [12]. Abrasion is characterized by material removal from a material surface as a result of scratching by an abrasive material. It mainly involves two or three bodies in contact. Microcutting, microploughing, and microcracking are associated with abrasive wear [12], [68], [73]. Meanwhile, in adhesive wear, adhesive bonds are disintegrated due to the surface interaction during relative motion [12], [74]. Surface fatigue failures are either mechanically or thermally activated. It results from the repeated sliding in the presence of hard asperities and is characterized by cracking, flaking, work hardening and plastic deformation. Tribochemical reaction involves intermittent removal of corrosion products facilitated by a combination of relative motion and a corrosive environment [12], [75], [76]. However, in

service, tool wear involves a combination of the different wear mechanisms. Moreover, abrasive wear has been identified as the most prevalent wear mechanism and accounts for the bulk of the cost expended on wear maintenance, as shown in Figure 2-4.

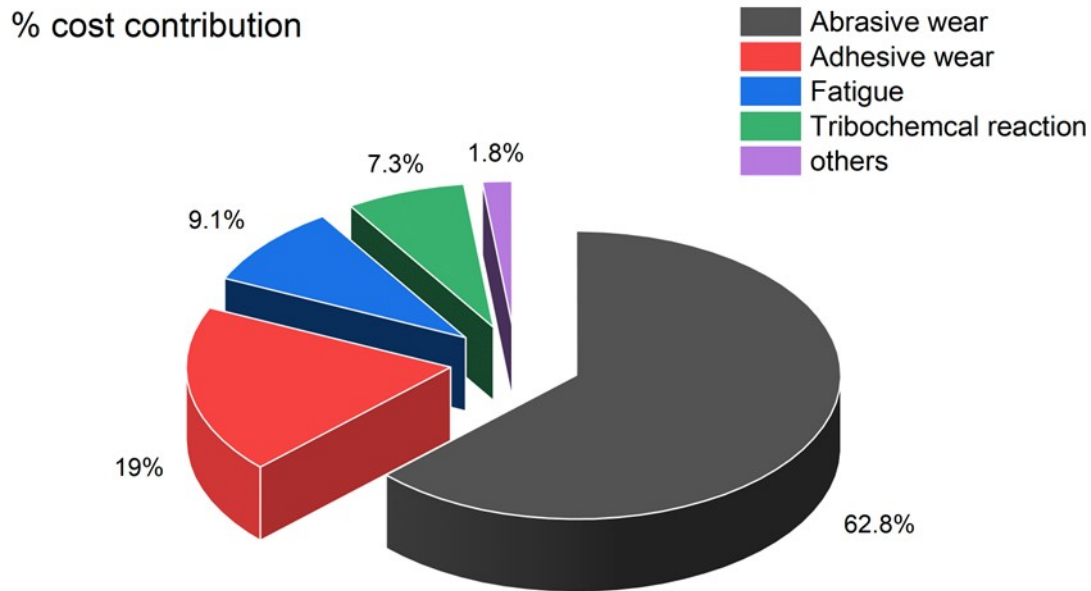


Figure 2-4: Cost of wear maintenance based on the results from an industrial survey conducted by Neale Consulting Engineers Limited in the UK [74].

Despite attempts by scientists to minimize the occurrence of these wear mechanisms, they are still prevalent in different tool steels, irrespective of the surface engineering treatment. Firstly, a study conducted by Heidari et al. [12] to understand the prevailing wear mechanisms in AISI A8 chipping and finishing knives for processing black spruce revealed the occurrence of microploughing, microcutting, cracking, and edge deformation of the cutting tool. The abrasive wear was characterized by uniform grooves whose depth were proportional to the machining force. The observation was attributed to wood contaminations culminating in a three-body abrasion. Meanwhile, the initiation and propagation of the

cracks were ascribed to the high impact, when the cutting tool comes in contact with defects such as knots during wood machining. Though, this form of wear has received less concern, if not properly managed, it results in wood products with poor surface finish and also renders the cutting tools unusable. The consequence of the inability of the cutting tool to withstand further cutting force due to excessive wear was the deformation of the cutting edge [12].

These mechanisms are also not absent from tool steels with surface engineering treatments. In this regard, extensive cracking, protective layer delamination, and microcutting were observed in nitrocarburized Cr-Mo-V tool steels. The findings indicated that the alloy composition of the tool steels significantly affected the resulting hardness and wear resistance of the nitrocarburized tool steels. However, the tool steels displayed superior wear resistance compared to the ones without nitrocarburizing treatment due to the formation of a compound layer comprising ϵ -carbonitride and γ' -nitride. But, with increased loading, the cracking extended beyond the compound layer towards the sub-surface [77]. In another study, Erdogan reported micro-scratching and micro-grooving in borided AISI H13 tool steel [46]. Nevertheless, the application of boriding treatment significantly minimized the micro-abrasive wear. This was due to increased hardness resulting from the formation of FeB-Fe₂B double phase boride layers. As previously observed by Psyllaki et al. [77], the authors also reported that the micro-abrasive wear accelerated with increased loading [46]. Furthermore, abrasive wear was also observed in shot-peened AISI D3 tool steel with CrN/CrAlN nanolayer coating [40]. This occurrence can contribute to coating failure in the form of cracking and delamination [78]. Nonetheless, the hybrid treatment led to about 47 % increment in wear resistance compared to the untreated tool steel. This was attributed to the

increased surface hardness, reduced surface roughness, and improved adhesion strength [40].

Summarily, the wear of tool steels is one of the major constraints preventing the full exploitation of the production capacity of the wood processing industries. The situation is further aggravated by the constantly changing wood properties coupled with the inability to use coolants or lubricants during wood processing. Consequently, the temperature rise is encountered during the machining process, which has a detrimental effect on the cutting tool and the quality of the machined products. Additionally, the ambient temperature also significantly influences tool wear and the quality of machined products. Though scientists have identified several wear mechanisms, most of the cost expended in industrial wear maintenance has been attributed to abrasive wear. More so, despite the success recorded in minimizing the occurrence of these mechanisms, they remain prevalent in the industrial daily production cycle.

2.3 Recent advances in tool wear improvement during wood machining

Before now, significant progress has been made in trying to curb the expenditure accrued to annual wear maintenance. Firstly, researchers developed techniques to characterize the mechanism so as to grasp a thorough understanding of the process. Thereafter, relationships were established between machining parameters, wood properties, and tool wear. Afterwards, several engineering treatments, including heat treatment,

cryogenic treatment, thermochemical treatments, coating deposition, and hybrid treatments, were applied to the cutting tools. These subjects are discussed in the succeeding sections.

2.3.1 Wear characterization

The direct impact of tool wear on production cost has attracted significant research concerns. One of the outcomes is wear characterization. Both direct and indirect methods have been developed [14], [79]. The direct techniques are based on the measurement of dimensional changes in the cutting tool, particularly changes in the cutting-edge region. Mass loss, volume loss, edge recession and changes in width track have been reported [5], [80], [81]. Szwajka and Trzepiecinski [11], Porankiewicz et al. [55] and Torkghashghaei et al. [82] characterized the edge recession of different tool steels using microscopy techniques. Szwajka and Trzepiecinski [11] in their study characterized the maximum edge recession in high-speed steel and cemented carbide cutting tools for the milling of particle board at different cutting speed. It was observed that the edge recession increased with the cutting speed for both cutting tool types. Porankiewicz et al. [55] characterized the edge recession of high-speed cutting tools used for milling different wood species with varying density, silica contamination, and corrosivity. They reported that the recession of the cutting edge was more correlated with the corrosivity of the wood species than the density and silica contamination. Meanwhile, Torkghashghaei et al. [82] characterized the recession in the cutting edge of cemented carbide circular saws with different edge geometries (waterfall and up-sharp geometries) used in the sawing of spruce-jack pine-basalm fir. Their findings revealed that circular saws with waterfall geometry showed less recession than up-sharp geometry. Furthermore, Heidari et al. [12] characterized the effect of tempering temperature

(500 – 540 °C) on the wear resistance of AISI A8 tool steels by measuring the volume loss. They noted that samples tempered at high temperatures displayed minimal volume reduction. Kumar et al. [83] characterized the wear resistance of high silicon alloy steel with different surface engineering treatments using weight loss. They resolved that the minimum weight loss was recorded for samples with the highest hardness. The depth of wear track was adopted by Zeghni and Hashmi to characterize the effect of TiC coating and nitriding hybrid treatment on the wear resistance of AISI D3 and Vanadis 4 tool steels using stylus profilometry [81]. They reported that tool steels with hybrid treatment exhibited lower wear track depth than those with only TiC coating. Advances have also been made in measuring real-time tool wear using simulation techniques and microscopic analysis [5]. Accordingly, Lins et al. [84] successfully developed a cyber-physical system based on tool wear vision monitoring for real-time measuring of tool wear. In another study, Qiao and co-workers [85] developed models based on deep learning and fog computing for real-time measurement and prediction of the wear of a twist drill. They reported a reasonable correlation between the predicted and actual tool wear.

Meanwhile, the indirect measurement involves the estimation of the tool wear using other machining parameters based on an already established relationship between the tool wear and the parameter [14]. Parameters such as power consumed, cutting torque, cutting force, acoustic emission, quality of machined product, and tool temperature have been used to indirectly estimate the wear of cutting tools during wood machining [54], [86]–[90]. Like the direct techniques, real-time assessment of tool wear has also been attempted for the indirect methods. In this regard, Aknouche et al. investigated the real-time tool wear during the machining of Aleppo pine wood with the cutting force as the parameter. Significant

variations in cutting force were observed in some sections along the cutting length, which was attributed to the presence of knots with high density [54]. Cuppini et al. [86] indirectly measured the wear of cutting tools through cutting power measurement. Lemaster et al. conducted indirect wear characterization of tungsten carbide inserts for machining medium density particle board by measuring power consumption, machining noise, machine vibration and acoustic emission. They observed that measurement of the machine vibration and noise were more correlated to the tool wear than the other parameters [87], [88]. Comparing both methods, the direct approach, though mostly characterized by intermittent measurements, is more reliable. On the other hand, the indirect method allows continuous measurement. However, Cuppini et al. [86] and De Lacalle et al. [52] opined that the efficiency of the measuring device strongly influences the reliability of the indirect method. Hence, they recommended setting a threshold for the parameter during real-time measurement to avert catastrophic failures [52], [86]. Generally, for both methods, in order to ascertain the full extent of tool wear, it is better to use more than one parameter, such as cutting force and power consumption or machining noise and cutting force.

Studies have also been conducted to ascertain methods for correctly measuring the recession of the cutting tool edge. Tavodova et al. used three approaches. The first involved comparing the distance between the tip of a used cutting edge to that of an un-used one. The second approach calculated the difference in the radius of the circle inscribed on the cutting-edge profile of the used and un-used one, while the last approach focused on the alteration in wedge angle. They concluded that the second approach was more accurate as it showed the least variance [79].

Additionally, some experimental methods have also been developed to characterize tool wear mechanisms. Some of these techniques include the popular Pin On the Disk (POD) [91], Dry Sand Rubber Wheel (DSRW) test [92], and Taber abrasive wear test [93], among others. Wear characterization in these methods is based on weight loss after abrading the material for a period of time. The characteristic appearance of abrasive wear mechanism is the presence of grooves, scratches, ploughed ridges (microcutting and microploughing) or even microcracks whose intensity correlates with the extent of wear. The rate of abrasive wear increases as the mechanism translates from microcutting through microploughing to cracking [68], [73]. Figure 2-5a-c shows the characteristic appearance of abrasive wear. Adhesive wear, on the other hand, is typically characterized by inter-surface material transfer, consequently, resulting in surface rupture, as shown in Figure 2-5d. Meanwhile, the effect of surface fatigue, due to impact from knots, sand, and rocks during wood machining, could manifest in the form of microcracks (Figure 2-5) or even result in surface rupture (Figure 2-5d). Tool wear resulting from tribochemical reaction are less prevalent during wood machining [12].

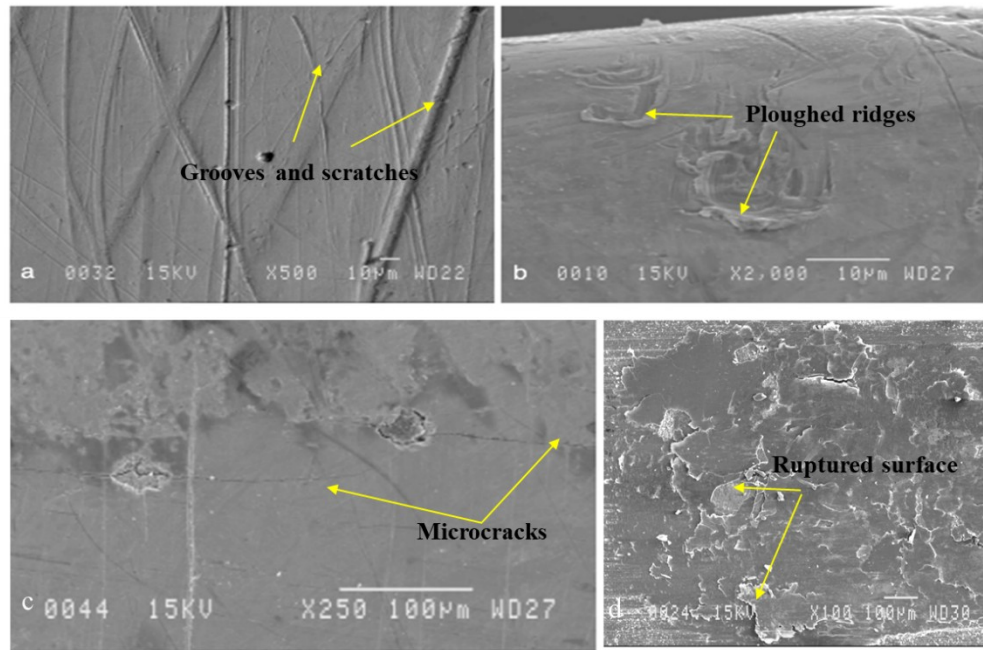


Figure 2-5: Wear mechanisms in AISI A8 chipper-canter knives: (a) microcutting; (b) microploughing; (c) microcracking; (d) surface rupture

2.3.2 Effect of machining parameters and wood properties on tool wear

Wood machining parameters such as cutting speed, rake angle, cutting force, cutting length, machining time, and feed rate significantly affect tool wear [12], [14], [77], [94]. Machining at high cutting speed and cutting forces accelerate tool wear [11]. At high machining speeds, there is an increase in the amount of heat generated as a result of shearing, leading to tool degradation [52]. In most cases, wood machining within ranges that ensure a prolonged cutting tool life, has been found to be inefficient and uneconomic. In this regard, Szwajka and Trzepieciniski reported that increasing the machining speed of high speed tool steel and cemented carbide cutting tools above 18000 rev/min (efficient speed range) during the machining of melamine-faced particle board resulted in reductions in tool life by 91 % and 95 %, respectively [11]. Meanwhile, investigations on the relationship between the

cutting speed and the cutting forces during the machining of high-density fibreboard was reported by Zhu et al. [95]. Interestingly, they observed that the cutting forces on the tool edge were negatively correlated with the cutting speed, as increasing the cutting speed (5000 – 10,000 rev/min) resulted in a reduction in the cutting forces on the edge of the cutting tool. This connotes improvement in wood processing characterized by a reduction in energy consumption. The observation was attributed to the reduction in friction coefficient owing to the temperature rise associated with the increased cutting speed. However, it is important to highlight that the optimum machining parameters depend on the cutting tool material, wood type, and desired quality, among other factors. Nonetheless, in the light of these, wood product manufacturers try to achieve a compromise between tool life, cutting speed, and minimizing production cost.

As expected, the feed rate, machining time, and cutting length are positively correlated with the tool wear [10], [14], [19]. Bendikiene et al. [19] reported a positive correlation between the feed rate and cutting power during the machining of oak wood. Consequently, wood machining at relatively high feed rates and high machining length and time would increase the energy consumption, leading to tool life reduction as compared to lower values of the parameters. The increase in tool wear with increasing feed rate is connected with the increase in chip thickness which is in accordance with the cutting theory [10]. Furthermore, analytical studies conducted by Ghosh et al. [14] revealed that the machining time had a more significant effect on tool wear than the type of knife (chipper or canter), irrespective of the point of measurement of tool wear along the cutting edge. Wood machining is usually carried out at low rake angles because of the desired surface finish. However, this results in increased tool wear [20]. Studies conducted by Zhu et al. [95]

revealed a negative correlation between the rake angle and cutting forces, irrespective of the cutting speed. This is because increasing the rake angle implies a reduction in wedge angle (see Figure 2-1), thus a sharper cutting tool. The authors concluded that for cost-effective wood machining and prolonged tool life, cutting tools with lower rake angles should be operated at higher speed, as vice versa.

Further investigation using regression and correlation analysis was conducted to ascertain the effect of wood properties, including density, silica contamination, and its corrosive effect at elevated temperature (high temperature corrosivity) on tool wear using IBM® SPSS® Statistics version 26 [96]–[98]. Table 2-1 shows the density, silica contamination, high temperature corrosivity, and tool wear for different wood species. The wear of the cutting tools (μm) was obtained by measuring the recession of the cutting edge (clearance face) from the optical images of the respective cutting-edge profile [99].

Table 2-1: Wood properties and tool wear for different wood species [99].

Wood sample	Origin	Density (kg/m^3)	Silica contamination (mg/kg)	High temperature corrosivity (R^w_{mw})	Wear (μm)
Nogal	Columbia	395	8.8	1.7	73
Passang	Japan	550	8.1	21.8	103
Red meranti	Indonesia	669	1.7	3.2	77
Teak	Indonesia	847	2573.8	1.0	77
Coconut	Indonesia	911	1540.6	2.1	69
Oil palm	Indonesia	482	5911.4	246.3	142

The effect of the wood properties on the tool wear can be evaluated using the significance levels of the t-values obtained from the regression analysis. Conventionally, parameters with significant values less than 5 % are considered significant [11]. The result of the statistical analysis of the data in Table 2-1 is presented in Table 2-2 and Figure 2-6. Table 2-2 shows that the effect of the wood density, silica contamination, and high

temperature corrosivity are statistically not considerable as their respective significance values are greater than 5 %.

Table 2-2. Significance values of the effect of wood density, silica contamination, and high temperature corrosivity on tool wear.

Parameter	t-values	Significant values (%)
Density (kg/m ³)	0.113	92.0
Silica contamination (mg/kg)	-0.557	63.4
High temperature corrosivity (R_{mw}^w)	1.639	24.3

However, of all the three factors, a change in the high temperature corrosivity is most likely to affect the wear of the tool steel at a confidence level of 75 % compared to the 36 % and 8 % of the silica contamination and density, respectively, as shown in Figure 2-6a.

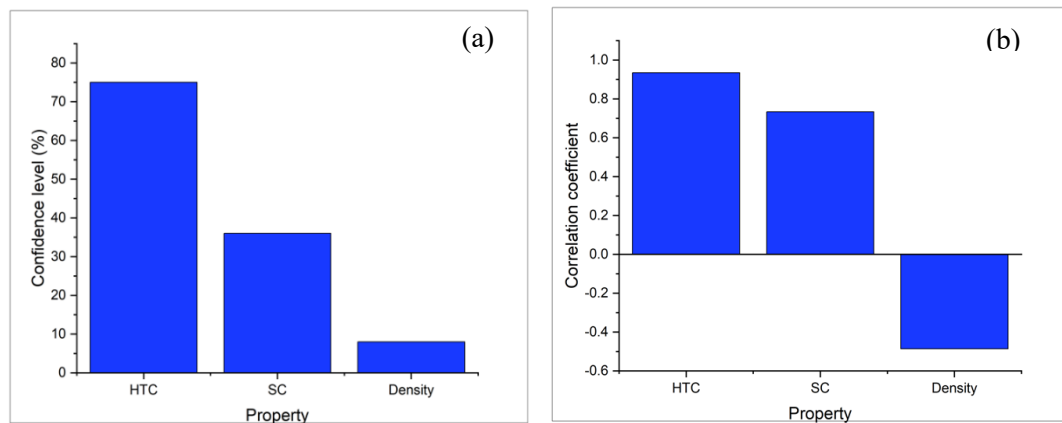


Figure 2-6: (a) Confidence levels for the effect of wood properties on tool wear; (b) Correlation coefficient between wood properties and tool wear (HTC- High Temperature Corrosivity, SC-Silica Contamination).

Figure 2-6b shows the correlation coefficient between tool wear and wood properties. The analysis revealed a very high positive correlation coefficient of 0.935 and 0.734 for high temperature corrosivity and silica contamination, respectively. The observed positive correlations imply that there is a likelihood that an increase in these parameters will increase the tool wear, i.e., reduce tool life based on their respective confidence levels. Besides, the considerable oxygen content of these contaminants may also contribute to their corrosivity [99]. Nevertheless, the findings of Amos revealed that high silica contamination might not be the cause of tool wear in some wood species [100]. Meanwhile, a weak negative correlation was observed for the density. However, the likelihood of an increase in tool wear rate with lower wood density is only 8 %. This is also coupled with the constantly changing wood density with environmental conditions. A similar finding was reported by L. Cristóvão et al. [101]. Moreover, the hardness of wood has been reported to increase with increasing density [53]. Hence, a denser wood is more likely to accelerate the tool wear process.

From the foregoing, machining parameters such as cutting force, speed, power, feed rate, cutting length, and machining time have been reported to be positively correlated with tool wear. On the other hand, a reverse trend is observed for the wedge angle. Furthermore, wood with high temperature corrosivity and silica contaminants is more likely to accelerate the wear of tool steels during machining.

2.3.3 Tool wear enhancement via application of heat treatment

The need to develop cutting tools with high wear resistance birthed the tremendous effort made by scientists to develop technologies capable of predicting material properties and phase compositions [102]. This was based on the underlying understanding that the service performance of materials is largely dependent on their phase composition and the

resulting properties. Fortunately, five decades ago witnessed the arrival of CALculation of PHase Diagrams (CALPHAD) technologies. The technology uses a computational approach involving thermodynamic and kinetic modeling to design materials with specific properties [103]. Some CALPHAD technologies reported in literature include Thermo-Calc [104], MatCalc [105] and JMatPro [106]. These technologies create the possibility of determining the optimum heat treatment conditions which will provide optimum wear performance.

In order to enhance the wear performance of tool steels during wood machining, they are often used in a hardened condition. The process of hardening involves austenitization followed by quenching and tempering. The austenitization process of tool steels involves heating to the austenitic temperature range (usually between 850 – 1200 °C). The extent of austenitization is a function of the temperature [107], [108]. On one hand, while austenitization at higher temperatures results in increased carbide dissolution, it is also accompanied by the tendency of having an increased level of retained austenite, delta ferrite, and coarse austenite grains, which impair the mechanical properties of the tool steels. On the other hand, low temperature austenitization does not result in optimum wear properties due to inadequate carbide dissolution [107]–[109]. So most often, both high and low temperature austenitization are applied to the tool steel to confer the benefits of both processes characterized by optimum combination of hardness and toughness required for increased tool life during wood machining [109], [110]. Accordingly, Salunkhe et al. [110] recommended a double austenitization process: a high temperature austenitization (to dissolve carbides) is followed by a low temperature austenitization (to prevent grain

coarsening and to reduce the retained austenite) to optimize the microstructure and final mechanical properties.

Upon quenching, martensite is formed alongside certain amounts of untransformed austenite (retained austenite), in some cases. The martensitic structure exists in lath, plate, or mixed morphologies depending on the carbon composition of the tool steel [38], [111]. The strength and hardness of the phase are attributed to the different mechanisms hindering dislocation movement, including solid solution strengthening, grain refinement, and strain hardening. Unfortunately, the martensitic structure has limited applications owing to its insufficient toughness [112]. Hence, the tempering process is required.

The tempering of tool steels is usually in the temperature range of 200 – 550 °C [12], [110]. During the process, there is a release of carbon from the saturated martensite matrix, followed by the transformation of retained austenite to martensite leading to the release of residual stresses. The resulting tempered martensite has a slightly lower hardness but improved toughness suitable for wood machining applications. In some cases, when there are still high levels of un-tempered martensite or retained austenite, the need for another tempering process may arise [107], [113]. For instance, Bochnowski et al. reported that three times tempering led to a 90 % reduction of retained austenite in high speed tool steel [114]. Additionally, due to the composition of most tool steels, secondary phases, predominantly carbides, also exist in the microstructure [111], [115]. These carbides provide secondary hardening by further impeding dislocation movement, inhibiting grain growth, and reinforcing the matrix, thus enhancing the wear properties of the tool steels [107], [116], [117]. Therefore, the microstructure of most tool steels comprises predominantly tempered martensite and carbides alongside retained austenite in some cases [53], [79], [118], [119].

Figure 2-7 shows a typical microstructure of AISI D6 tool steel austenitized at 1050 °C and 940 °C, oil-quenched, and tempered at 500 °C. It contains tempered martensite, carbides and retained austenite.

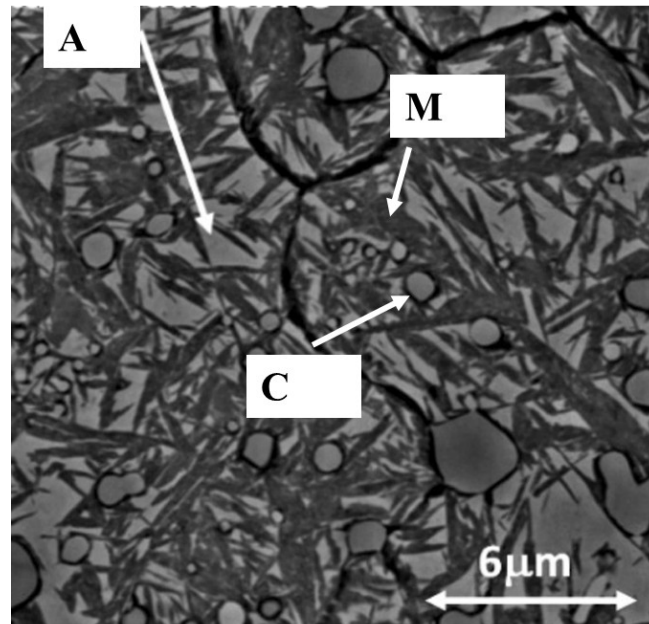


Figure 2-7: Micrograph showing carbides (C), martensite (M), and retained austenite (A) in AISI D6 cold work tool steel austenitized at 1050 °C, and air quenched [120].

The level of retained austenite in the tool steel also affects the cutting tools' tribological performance during wood machining. The austenite phase having a relatively lower hardness than martensite, has a negative effect on the overall hardness of the hardened tool steels [121]. Consequently, some reports have been documented on the detrimental effect of this phase on the tribological performance of tool steels. The study conducted by Colaco and Vilar revealed that as the level of retained austenite increased from 0 – 100 % in AISI 420 steel, a 35 % reduction was observed in the hardness. This resulted in a reduction

in the wear resistance of the tool steel [122]. A similar investigation reported by Moradiani et al. revealed that as the level of retained austenite increased from 42 to 90 % in AISI D2 tool steel, about 8 % reduction was observed in the hardness, which was accompanied by a reduction in wear resistance of the tool steels [123].

Unsurprisingly, the characteristics of the carbides, including carbide-type, their distribution, volume fraction, and sizes, significantly affect the wear properties of the tool steels. Accordingly, extensive reports have been published on tool steels with similar hardness but completely different wear properties, due to variation in the carbide characteristics [92], [116]. In this regard, Bourithis et al. [92] studied the tribological performance of AISI O1 and D2 tool steels having the same hardness. Their findings revealed that the latter displayed superior wear resistance in both two-body and three-body abrasive wear tests. The observation was attributed to the presence of plate-like carbide morphology in the latter, which was absent in the former. In a study conducted by Bochnowski et al. on the effect of tempering on the carbide morphology of tool steel, it was observed that the presence of M_6C , MC , and M_2C primary carbides facilitated the formation of secondary carbides. Furthermore, increasing the tempering temperature beyond the optimum range resulted in a reduction in hardness due to the coarsening of the secondary carbides at higher temperatures. Interestingly, increased volume fraction and finer primary carbides hindered the transformation of residual stresses from tensile to compressive during the tempering process [114]. Meanwhile, Heidari et al. observed that increasing the volume fraction of fine carbides in AISI A8, H13, and S1 significantly improved their wear performance [12].

Expectedly, the austenitization and tempering parameters play significant roles in the wear behaviour of tool steels. Attaullah reported that increasing the austenitization temperature in AISI D2 and H13 tool steels resulted in increased hardness owing to the increase in carbon concentration in the martensite matrix [37]. Similar observations have also been reported by other authors [116], [124], [125]. Meanwhile, poor wear properties were observed in AISI H11 tool steel with low austenitization temperature due to the reduced hardness. However, upon increasing the austenitization temperature, despite the increment in hardness, it was accompanied by a toughness reduction. This was attributed to the decrease in the volume fraction of carbides and the unrestricted austenite growth. Hence, optimum wear properties were obtained at intermediate austenitization temperature (between 1000 - 1200 °C) with the best combination of microstructural composition [116].

Studies have also been conducted on the effect of multiple austenitization on the hardness of tool steel. It was revealed that an increase in single and double austenitization temperatures increased the tool steel hardness. This was attributed to the inhibition of austenite grain growth and increased carbon composition in the martensite matrix following carbide dissolution. However, higher hardness was observed with double austenitization as compared to single austenitization because the former offered greater dissolution of carbides and inhibition to grain growth than the latter [110].

Further statistical analysis was also conducted to ascertain the effect of carbide volume fraction, sizes, and tool steel hardness on the tool wear. Table 2-3 shows the wear of AISI A8 tool steel with single austenitization and tempering treatment temperatures alongside the hardness and carbide characteristics. The wear of the tool steels was characterized using the volume loss (mm^3) obtained from the DSRW test [12].

Table 2-3: Wear, hardness, and carbide characteristics of AISI A8 tool steel at different austenitizing and tempering temperatures [12].

Wear (mm³) ± 0.2	Hardness (HRC) ± 0.5	Carbide volume fraction (%) ± 0.2	Average carbide size (μm) ± 0.01	Austenitizing temperature (°C)	Tempering temperature (°C)
88.9	53	20.15	1.25	980	500
85.3	53	21.3	1.28	980	520
78.0	51	24.79	1.33	980	540
95.5	54	17.3	1.22	1010	500
88.8	56	20.53	1.27	1010	520
83.7	52	24.77	1.30	1010	540
91.1	59	13.22	1.15	1040	500
90.8	58	14.42	1.21	1040	520
82.3	57	19.54	1.27	1040	540

Table 2-4 shows the significance values of the effect of heat treatment parameters, carbide characteristics, and hardness on the wear of AISI A8 tool steels based on the analysis of the results in Table 2-3.

Table 2-4: Significance values of the effect of heat treatment, hardness, and carbide characteristics on tool wear.

Parameter	t-values	Significant values (%)
Hardness (HRC)	-0.990	39.5
Carbide volume fraction (%)	-0.651	56.2
Average carbide size (μm)	1.322	27.8
Austenitizing temperature (°C)	2.081	12.9
Tempering temperature (°C)	-2.678	7.5

From Table 2-4, though the parameters have significant values greater than 5 %, there is a likelihood that changes in the parameter would affect tool wear based on their respective confidence levels presented in Figure 2-8a. Accordingly, the tempering temperature is the most significant parameter influencing the tool wear, with the least significant parameter being the carbide volume fraction. The respective confidence levels for the tempering temperature, austenitizing temperature, average carbide size, hardness, and carbide volume fraction are 92.5 %, 87.1 %, 72.2 %, 60.5 %, and 43.8 %. More so, the correlation analysis revealed that positive correlations were obtained for austenitization temperature (0.325) and hardness (0.479), while negative correlations were obtained for tempering temperature (- 0.854), average carbide size (-0.795) and volume fraction of carbides (-0.749) as shown in Figure 2-8b.

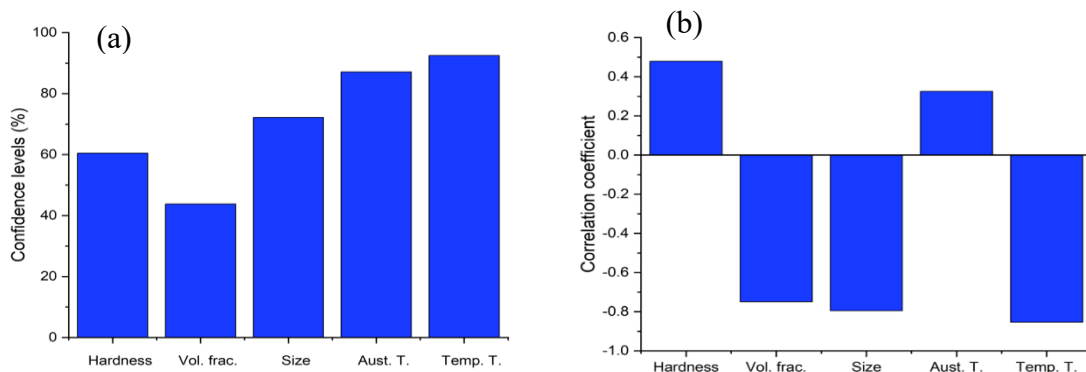


Figure 2-8: (a) Confidence levels for the effect of heat treatment, hardness, and carbide characteristics on tool wear; (b) Correlation coefficient between heat treatment, hardness and carbide characteristics and tool wear (Vol. frac. – carbide volume fraction, Size – Average carbide size, Aust. T. – austenitizing temperature, Temp. T. – tempering temperature).

Based on the correlation results, increasing the tempering temperature would be likely accompanied by an improvement in tool wear resistance (reduction in wear amount) at the respective confidence level. This might be attributed to the increase in carbide volume fraction [12]. Also, the weak positive correlation obtained for the austenitizing temperature implies that increasing the austenitizing temperature is likely accompanied by an increase in tool wear at the respective confidence level. Despite the corresponding increment in hardness (positive correlation) and reduction in average carbide sizes (negative correlation) with increasing austenitization temperature, the increase in tool wear may be attributed to the reduction in the carbide volume fraction (negative correlation). A similar observation was reported by Heidari et al. [12].

In summary, austenitization and tempering processes can potentially improve tool wear resistance during wood machining. If single austenitization and tempering are to be carried out, the temperatures should be intermediate in order to achieve optimum microstructural composition with improved wear performance. Meanwhile, double austenitization and multiple tempering operations confer properties with superior wear performance. Furthermore, the characteristics of the carbides influence the wear of tool steels. The resulting wear properties are a product of the interaction between the strengthening offered by tempered martensite, the volume fraction and sizes of the carbides, and the carbon concentration in the martensite matrix. Meanwhile, the statistical analysis in Figure 2-8 and Table 2-4 revealed that the effect of the heat treatment temperatures, carbide characteristics, and mechanical properties on tool wear was in the order of tempering temperature > austenitizing temperature > average carbide size > hardness > volume fraction of carbides.

2.3.4 Tool wear enhancement via cryogenic treatment

The application of cryogenic treatment (low temperature treatment) is another technique of improving the wear performance of tool steels for wood machining [111]. It is mostly carried out at temperatures in the range of dry ice temperature (about $-80\text{ }^{\circ}\text{C}$, known as shallow treatment) or liquid nitrogen temperatures (about $-196\text{ }^{\circ}\text{C}$, known as deep treatment) [126]–[128]. Like the conventional heat treatment, the process parameters such as the holding time and temperature influence the resulting properties [129]. Some reported effects of cryogenic treatment include increase in hardness, wear resistance, corrosion resistance and reduction in residual stresses and toughness [130]–[133]. Consequently, this can lead to a reduction in the cost of production and production downtime of wood processing [134].

However, over time, some controversial arguments have evolved regarding the application of cryogenic treatment to tool steels. Firstly, while a majority of researchers have reported that the process improves wear resistance [126], [135], [136], some have reported otherwise [91]. Secondly, a group of researchers have opined that wear improvement during the process is due to the transformation of retained austenite to martensite [136], [137] while others claim that the improvement is attributed to the precipitation of fine secondary carbides [127], [138]. The precipitation of secondary carbides during cryogenic treatment has been attributed to the contraction of the martensite matrix leading to the segregation of the released carbon atoms near defects, which eventually transform into carbides [126]. As a matter of fact, some authors have reported a significant reduction in retained austenite composition in tool steel due to cryogenic treatment which did not have any notable impact

on their wear performance [91]. Another controversial issue is the position of the cryogenic treatment in the heat treatment cycle. Some researchers have reported that the cryogenic treatment should be applied prior to the tempering process [95], [109], while others have applied it after the tempering process [134]. Nonetheless, improved wear resistance has been observed for both configurations.

A study was conducted by Molinari et al. [134] to ascertain the effect of the position of the cryogenic treatment in the heat treatment schedule and multiple tempering on the mechanical and wear properties of AISI M2 and H13 tool steels. Their findings revealed that both configurations resulted in improved wear resistance, but the samples with cryogenic treatment applied after tempering showed higher wear performance by about 30 %. More so, double tempering after cryogenic treatment did not significantly improve the wear performance of the tool steels [134]. In another study conducted by Huang et al., it was observed that the low temperature treatment resulted in an increase in the volume fraction of carbides and produced a more homogeneous microstructure, which improved wear performance [127]. Researchers have also attempted multistage cryogenic treatment highlighting its cost benefit. The process was observed to increase the volume fraction of carbides and produced a more homogeneous microstructure relative to the single stage treatment [126]. Additionally, analytical studies conducted by Oppenkowski et al. revealed that the heat treatment parameters have a more significant effect on the mechanical and wear properties of AISI D2 tool steel compared to the cryogenic parameters [139].

2.3.5 Tool wear enhancement via thermochemical surface treatment

Thermochemical treatment is often applied to tool steels for wood machining to improve wear behaviour and load capacity [39]. It includes processes applied to modify the surface composition of the tool steels without major significant changes in the material dimension [12], [140], [141]. This is achieved by heating the tool steels to elevated temperatures in the presence of a medium comprising carbon, nitrogen, or boron, among others [12], [140], [142]. In this process, temperature, time, composition of the tool steels, and the composition of the medium are significant parameters that determine the resulting wear properties [143]–[145]. Wear improvement is achieved due to the increased surface hardness resulting from the formation of intermetallics, coupled with the generated strain field between the tool steel matrix and the intermetallics [146]. More so, the process does not result in a drastic hardness discontinuity between the bulk material and the surface compared to other surface engineering treatments like coatings [147]. Some thermochemical treatments reported in literature are carburizing [120], nitriding [30], carbonitriding [48], boriding [121], and cyaniding [112].

Faga and Settineri presented a study on the effect of high (500 °C) and low (300 °C) temperature nitriding on the wear properties of HSS18 high speed tool steel and 90MCV8 alloy steel. Their findings revealed that low temperature nitriding did not significantly improve the wear behaviour of both tool steels. In fact, a slight deterioration was reported for the alloy steel. However, upon increasing the nitriding temperature, significant improvement was observed for both tool steels, which was characterized by a reduction in friction coefficient and wear track depth [5]. In another study conducted by Psyllaki et al. to ascertain the effect of microstructural composition and liquid nitrocarburizing on the wear

of AISI H13, D6, and Cr-Mo-V tool steels, they reported that the AISI H13 showed superior wear performance due to the formation of finely dispersed carbides, as compared to the elongated carbonitrides and lamellar precipitates found in the AISI D6 and Cr-Mo-V grades, respectively. Additionally, the state of the hardening medium has been reported to significantly impact the wear of tool steels. For instance, liquid carbonitrided tool steels have been observed to exhibit superior wear performance and toughness than those subjected to gas carbonitriding owing to the formation of an ϵ -carbonitride compound in the former [77].

Researchers have also highlighted the benefits of plasma nitriding over the conventional nitriding process. These include reduction in the environmental footprint, lower treatment temperature and greater versatility [147], [148]. Accordingly, an investigation on the effect of plasma nitriding temperature (460 – 520 °C) and time (6 – 12 hours) on the mechanical and corrosive properties of 90CrMoV8 tool steel for the machining of oak wood was conducted by Nouveau et al. They reported that optimum hardness was achieved at intermediate plasma nitriding parameters. However, the plasma nitriding process did not improve the resistance of the tool steel to corrosive wear in the wood electrolyte [147].

The effect of boriding temperature on the wear properties of AISI H13 tool steel was studied by Erdogan [46]. It was reported that increasing the temperature resulted in a corresponding increase in wear resistance. This was attributed to the reduction in surface roughness and the formation of double ferrite phases leading to an increased hardness. The hardness was observed to further increase with the thickness of the intermetallic layers. Other authors have reported that such double intermetallic phases are sites of crack initiation owing to the difference in properties between the phases and the matrix [149]. More so, the

reduction in tool wear was partly attributed to the adherence of abrasive particles to the surface of the tool steels. A similar occurrence was reported by Krelling et al. [150]. The size of the abrasive particle also played a significant role in the wear process, as coarse-sized particles resulted in accelerated tool wear [46]. A similar study on the effect of boriding temperature and time on the mechanical properties of tool steel was reported by Sen and Sen [145]. They reported a positive correlation between the treatment parameters, hardness, and thickness of the intermetallic layers. However, this was accompanied by a reduction in toughness owing to the interaction between the different brittle intermetallic boride phases [151].

From the foregoing, it is evident that tool wear enhancement by thermochemical diffusion treatment is a function of the process parameters. While relatively low temperatures are likely to have an insignificant impact on tool wear performance, improved wear performance would be likely obtained at higher temperatures due to increased hardness resulting from the formation of the hard intermetallic layer. More so, the surface roughness and the composition of the tool steels play significant roles in the wear behaviour. However, the presence of excessive intermetallic phases due to increased process parameters might have a detrimental effect on the toughness of the tool steels resulting in premature failure in the form of cracking during wood machining.

2.3.6 Tool wear enhancement via coating application

Coating deposition is another promising method of improving the wear behaviour of tool steels for wood machining applications. It has previously been adopted for metal machining purposes, but recently, it attracted the interest of the wood machining industry

due to its cost-effectiveness compared to other wear enhancement techniques [41], [52], [53]. Tool wear enhancement is achieved through tool edge temperature reduction by heat dissipation, reduction in frictional force and stresses at the cutting edge, increase in hardness alongside improvement in corrosion resistance to the corrosive effect of the wood fluids [53], [152], [153]. In coating selection, some of the properties sought after are hardness, toughness, adherence to the substrate, and chemical stability, especially at elevated temperatures [41], [97], [154]–[158]. Generally, the coating hardness is considered the most significant parameter affecting the wear resistance of tool steels [12], [159]. It is a function of several factors, including the method of deposition, coating composition, and the size of the crystallites, among others [41], [160], [161]. For uncoated substrates, increased hardness correlates to improved wear resistance in most cases [41]. However, the same was believed for coated substrates until failures were observed in some hard coatings due to poor adhesion and reduced resistance to elastic and plastic deformation [2]. Hence, in addition to increased coating hardness, parameters such as resistance to elastic strain (H/E) and plastic deformation (H^3/E^2) are often used for the assessment of the quality of coatings [70], [162]–[164]. Like the hardness, a positive correlation has been observed between H/E , H^3/E^2 and wear resistance of tool steels [157], [165].

Prior coating deposition, adequate substrate preparation is necessary so as to make the substrate more receptive of the coating and ensure proper adhesion. The importance of surface preparation during coating deposition has not received due attention from researchers in the past years despite its significant role in the overall performance of the coatings [4], [42], [118], [166]. Accordingly, a considerable amount of coating failure has been attributed to insufficient bond strength owing to inadequate surface preparation [165].

The process has been reported to account for at least 45 % and up to 90 % of the labour cost during the entire coating process. The surface preparation of tool steels involves the removal of surface contaminants followed by profiling [157]. While the effect of surface contaminants has been well documented, insufficient literatures are available regarding the latter [41], [167]. Again, another issue that is less discussed is the bond coating; an adhesive layer deposited on the substrate prior coating deposition. In addition to enhancing coating adhesion, it also protects cracks initiated at the coating from easily propagating to the substrate [118], [165].

2.3.6.1 Method of coating deposition

The method of coating deposition and the deposition parameters significantly affect the wear properties of the tool steel [5],[53]. Several coating deposition methods involving electrochemical treatments (electroplating and anodizing), chemical treatment (physical and chemical vapour deposition), and thermal spraying have been reported in literature [12], [41], [141]. However, chemical deposition treatments have received a wider range of acceptance due to their large application spectrum, as little or no metallurgical compatibility between the substrate and coating is required. For this reason, their application has been extended to ceramic coating depositions [159].

Coating deposition by Physical Vapour Deposition (PVD) technique is achieved either by vaporization or atomization of the coating material from the target thermally or kinetically followed by deposition on the substrate. On the other hand, chemical vapour deposition is achieved by chemical reactions between the coating materials at much higher temperatures (800 °C – 1500 °C) [141], [159], [168]. Meanwhile, despite the high bond strength previously obtained from chemical vapour deposition coatings, the physical vapour

techniques are preferred because of the lower temperature of deposition, increased hardness, reduced surface roughness, and finer crystallites [39], [40]. The deposition temperature is of prime importance as higher values have a detrimental effect on the hardened substrate [79]. Some physical vapour deposition techniques reported in the literature are cathodic arc evaporation, electron beam physical vapour deposition, magnetic sputtering deposition, and ion beam-assisted deposition, among others [41]. PVD techniques involving arc deposition are preferred to sputtering techniques because of the high deposition rate and bond strength of the former [40]. The reduced bond strength in the latter is due to the high residual compressive stresses in the coated substrate resulting from its ionic bombardment during deposition [165], [169].

2.3.6.2 Coating types

Previously, coatings were classified according to their resistance to deformation (hard or soft coatings) and chemical bonds (metallic, covalent, or ionic coatings). However, the recent trend involves the development of coatings with composite compositions and structures having scales ranging from micro to nano [39], [159], [168]. In the past years, different coating types, including nitride-based, carbide-based, and diamond-based coatings, have been deposited on tool steels, resulting in significant wear improvement [39]. Most significantly, advances have been made in developing coatings with multiple layers [170]. This offers benefits such as enhancing corrosion resistance, surface residual stress of the substrate, and resistance to plastic deformation due to the possibility of combining layers with different properties to suit the desired application [118], [157], [171]. In a recent study, conducted by Naghashzadeh et al., the wear behaviour of a ternary, binary TiN based, and

uncoated AISI D3 tool steel, was investigated. It was observed that the improvement in wear performance was in the order of ternary multilayer > binary multilayer > uncoated substrate [157]. Similar findings were recently reported by Çelik et al. [172]. However, limited studies are available regarding the number of layers for optimized wear performance. Though the coating composition plays a more significant role in the tool wear performance compared to the number of layers.

A few authors have studied suitable coatings for optimum wear performance during wood machining. Firstly, Heidari et al. investigated the wear performance of CrN, CrAlN, TiAlN, TiAlCrN, TiAlCrN-CrC, and AlCrTiN coatings deposited on AISI A8, H13, and S1 tool steels. The AlCrTiN and TiAlCrN-CrC coating showed the optimum wear performance for the A8 and H13 tool steels, respectively, while TiAlCrN showed the least wear performance for both grades. Contrastingly, TiAlCrN and AlTiCrN exhibited the highest and lowest wear performance for the S1 tool steel. They attributed the observation primarily to the surface stiffness of the substrates and their residual stress support capacity [12]. In another study, Faga and Settineri investigated the wear performance of CrN and diamond-like carbon single and multilayer coatings deposited on HSS18 and 90CMV8 tool steels. Their investigations revealed that the multilayer coatings generally exhibited superior wear performance. This improvement was ascribed to the reduction in residual stresses on the surface of the substrate [5]. Kazlauskas et al. also investigated the wear performance of CrN, TiCN, TiAlN, AlTiN, and AlCrN coatings deposited on tungsten carbide for the machining of oak wood. The wear performance was in the order of CrN > AlCrN > TiCN > TiAlN > AlTiN [2]. One notable finding from their study is that the TiCN coating with the highest hardness did not exhibit the optimum wear performance. This might be attributed to the

reduced resistance of the coatings to elastic strain and plastic deformation which the authors did not take into consideration.

In another report, Liu et al. conducted a comparative study on the tribological performance of TiN, TiAlN, AlTiN, and CrAlN coatings deposited on cemented carbide cutting tools. Their results revealed that the tribological performance of the coatings was in the order $\text{TiAlN} > \text{TiN} > \text{CrAlN} > \text{AlTiN}$. The observed trend in tribological performance was attributed to the hardness and friction coefficient of the respective coatings. This study emphasized the detrimental role of high aluminium composition (in CrAlN and AlTiN), leading to their reduced mechanical and tribological properties. Property deterioration was attributed to the high chemical reactivity of the metal resulting in the formation of interfacial bonds with the abrading surface leading to adhesive wear [173].

The thickness of the coatings also plays a significant role in the wear performance of tool steels. Many authors have reported a negative correlation between the coating thickness and the tool wear. However, a threshold of coating thickness was observed, above which the wear performance begins to deteriorate [4], [39], [41]. This is because an increase in the thickness of the coatings is accompanied by increased residual stress, which is detrimental to the growth of the coating films. When the stress exceeds the bond strength of the coatings, it results in the delamination of the coating from the substrate [39]. Meanwhile, as the thickness of the coating reduces, the effect of the substrate on the tribological properties increases [168]. However, irrespective of the thickness of the coating, the substrate should have sufficient strength to support the load applied to the coating. Otherwise, even with a high wear-resistant coating with the appropriate thickness, premature coating failure would likely occur [41], [174].

2.3.6.3 Effect of coating doping

The doping of coatings is another attempt by researchers to further enhance the wear resistance of tool steels. Coatings doped with carbon, nickel, and silicon, among others, have been reported by different authors [41]. Some of the benefits of doping documented in literature are an increase in hardness, resistance to plastic deformation, corrosion, and oxidation [165], [175]–[177].

Studies were conducted by Kong et al. [70] on the effect of carbon doping on the wear performance of CrN coating deposited on AISI M2 tool steel. They reported that carbon doping improved the wear performance of the tool steel, which was characterized by the increment in hardness, resistance to elastic strain, and plastic deformation. The improvement was attributed to the formation of carbides. However, increasing the carbon composition above a certain threshold had a detrimental effect on the wear performance despite the increment in coating thickness. This was due to the presence of excess amorphous carbon, which led to the formation of a porous microstructure. Interestingly, the doped multilayer coating also showed a superior wear performance compared to the single layer coating [70]. Similar observations were reported for TiAlCrN coatings doped with carbon [71]. Recently, Akhter et al. investigated the effect of Ni doping on CrN coatings deposited on AISI M2 tool steel. They recorded optimum wear performance at lower compositions of the dopant. Wear enhancement was attributed to the increase in hardness resulting from grain reformation and solid solution hardening [165]. As observed by Kong et al. [70], they also reported deterioration in tool wear performance at higher dopant compositions.

To add up, wood machining with an uncoated substrate is already complicated. The incorporation of the coating further complicates the process due to the increase in the factors upon which tool wear is dependent. However, from an economic standpoint, the achieved wear improvement correlates with the cost implication. Furthermore, in the decision-making for coating selection for wood machining, resistance to elastic strain and plastic deformation should also be considered coupled with the hardness. More so, the substrate should possess sufficient strength and toughness to support the load applied to the coatings. The difference in surface chemistry between the substrate and coating accounts for the difference in wear performance when a given coating is deposited on different substrates. Nonetheless, the properties of the coatings can be further enhanced by doping with optimum elemental compositions.

2.3.7 Wear enhancement via hybrid treatment

In an attempt to further enhance the wear of tool steels, researchers have also explored the possibility of hybrid treatments. This involves a combination of two or more surface modification processes, such as nitriding and coating or shot peening and coating, among others [159], [178], [179].

A significant number of studies have been documented on the effect of combined nitriding and coating on the wear performance of different tool steels. The nitriding process prior coating deposition has been reported to improve the adhesion of the coating to the substrate [118], [180]. Firstly, Zeghni et al. investigated the wear behaviour of AISI D3 and Vanadis 4 tool steels, which were nitrided prior to the coating process. For most of the investigated coatings, the authors reported an improvement in wear performance

accompanied by an increment in hardness and bond strength. Meanwhile, drastic reductions in wear resistance were recorded for other coated tool steels [81]. In another study, Faga and Settineri investigated the effect of nitriding temperature on diamond-like carbon and CrN coatings deposited on tool steels for the machining of black spruce. While the nitriding temperature had no significant effect on the tool wear with diamond-based coating, low temperature nitriding improved the wear performance of the substrates with CrN-based monolayer coatings. However, inconsistent wear performances were recorded for tool steels with CrN-based multilayer coatings. Nonetheless, compared to the overall wear performances of the coated tool steel without nitriding, they exhibited inferior wear performances [5]. In a related study, Panjan observed that plasma nitriding at lower temperatures and shorter time improved the coating adhesion to the substrate due to the reduction in the formation of nitride layers. However, no significant improvement in tool wear was recorded [146]. Some authors have opined that the nitride layers have high compressive stresses and affect the growth of the coating films [5], [146]. Others have attributed the observation to the reduction in toughness upon nitriding [181]. However, current reports by Çelik and co-workers revealed that nitriding of Cr-Mo-V tool steel at 585 °C for 6 hours, followed by the deposition of CrN, AlTiN, and CrN/AlTiN coatings resulted in tribological performance with the multilayer coating exhibiting the optimum performance. This was due to the increased hardness resulting from the combined effect of the hard ceramic coatings and the diffusion layer produced by nitriding [182].

Researchers have also explored hybrid treatments combining shot peening treatments and coating on tool steels [40], [183]. Shot peening involves altering the surface microstructure by plastic deformation resulting in the inducement of compressive residual

stresses [140], [184], [185]. It confers benefits such as increase in hardness and reduction in surface roughness [40]. Accordingly, a study conducted by Pak et al. on the hybrid coating and ultrasonic peening treatment revealed an improvement in wear performance by 47 %. More so, the coating did not significantly influence the surface roughness but contributed substantially to the surface hardness, which accounted for the improved wear performance. However, high compressive stresses resulted in a reduced wear performance due to the reduction in bond strength [40].

Similarly, hybrid treatments involving burnishing, high-energy beam treatments, and nitriding of tool steels have been reported [23], [186], [187]. In this regard, Ormanova et al. studied the effect of electron beam treatment and nitriding on the hardness of AISI W320 tool steel. They observed an increase in hardness with the hybrid treatment. But an additional electron beam treatment after nitriding resulted in hardness reduction primarily due to grain coarsening [187]. In another study, Tobola et al. investigated the effect of nitriding and burnishing on the wear performance of AISI D2 and Vanadis tool steels. Prominently, they found that nitriding and burnishing enhanced the wear of the tool steels owing to the increase in compound layer accompanied by increased surface hardness. However, nitriding offered a more significant improvement in tool wear relative to the latter. Consequently, they recommended that the burnishing should precede the nitriding treatment for optimum wear performance [23].

In summary, hybrid treatment has the potential to provide tool wear enhancement if appropriately applied. In the case of the combination of nitriding and coating, despite exhibiting superior wear performance compared to the untreated substrate, based on the preponderance reports, it is not recommended if optimum wear performance is desired. This

is because the coated substrate without nitriding exhibits far superior wear performance in most cases. However, if nitriding is supposed to be applied prior coating, in that case, it should be carried out at lower temperatures and shorter times in order to maximize the adhesive benefits of the nitride layers while minimizing the thickness due to their detrimental effect on the wear performance.

2.4 Summary and research gaps

2.4.1 Summary

Tool wear, characterized by various mechanisms, significantly limits the overall productivity of the wood machining industry by reducing the lifespan of cutting tools. This issue is further exacerbated by the dynamic properties of wood and dry machining conditions. To address this challenge, several techniques for characterizing tool wear have been developed, alongside promising methods for enhancing tool lifespan. Multiple austenitization and tempering operations have demonstrated optimal tool wear performance, while cryogenic treatment has also shown improvements. Thermochemical treatments have shown positive outcomes when applied at intermediate process parameters. When it comes to coating selection for wood machining application, factors such as hardness, resistance to elastic strain (H/E), and plastic deformation (H^3/E^2) should be taken into consideration to make informed decisions. Furthermore, the utilization of hybrid treatments can ultimately result in significant enhancements in tool wear performance.

2.4.2 Research gaps

In light of the review, the following recommendations are outlined for researchers and industrial practitioners involved in enhancing tool wear for wood processing applications:

- Researchers should explore the development of innovative wood machining lubricants or coolants, similar to those used in metal machining, that do not compromise wood quality or negatively affect cutting tools. If achievable, the use of hardened tool steel without additional surface modification treatments could be considered, leading to potential cost savings in wood machining.
- In addition to the experimental tests reported in existing literature, more field tests conducted under practical industrial scenarios with optimized machining parameters are needed. Subsequently, further studies should be undertaken to investigate any disparities in tool wear behavior.
- Given the absence of an all-inclusive coating, researchers should focus on developing coatings specifically tailored for particular wood species to achieve optimal wear performance. This approach can then be expanded gradually to cover a wider range of wood species.
- More studies should be conducted to understand the bonding mechanism and surface chemistry at the interface between the substrate and coating. This understanding could shed light on the variations in wear behavior exhibited by the same coating deposited on different substrates.

- Most coatings examined in the review employed Ti or Cr bond coatings/interlayers. Further studies should investigate the effect of bond coating/interlayer thickness and explore alternative options to enhance bond adhesion to the substrate.

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Chapter 3

A comprehensive review of cathodic arc evaporation physical vapour deposition (CAE-PVD) coatings for enhanced tribological performance

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Abstract

In the realm of industries focused on tribology, such as the machining industry, among others, the primary objective has been tribological performance enhancement, given its substantial impact on production cost. Amid the variety of tribological enhancement techniques, cathodic arc evaporation physical vapour deposition (CAE-PVD) coatings have emerged as a promising solution offering both tribological performance enhancement and cost-effectiveness. This review article aims to systematically present the subject of CAE-PVD coatings in light of the tribological performance enhancement. It commences with a comprehensive discussion on substrate preparation, emphasizing the significant effect of substrate roughness on the coating properties and the ensuing tribological performance. The literature analysis conducted revealed that optimum tribological performance could be achieved with an average roughness (R_a) of 0.1 μm . Subsequently, the article explores the CAE-PVD process and the coating's microstructural evolution with emphasis on advances in macroparticles (MPs) formation and reduction. Further discussions are provided on the characterization of the coatings' microstructural, mechanical, electrochemical and

tribological properties. Most importantly, crucial analytical discussions highlighting the impact of deposition parameters namely: arc current, temperature and substrate bias on the coating properties are also provided. The examination of the analyzed literature revealed that the optimum tribological performance can be attained with a 70 to 100 A arc current, a substrate bias ranging from -100 to -200 V and a deposition temperature exceeding 300 °C. The article further explores advancements in coating doping, monolayer and multilayer coating architectures of CAE-PVD coatings. Finally, invaluable recommendations for future exploration by prospective researchers to further enrich the field of study are also provided.

Keywords: physical vapour deposition; cathodic arc evaporation; tribological performance; tool steel

3.1 Introduction

Over the years, the concept of tribological performance has received significant attention from industrial practitioners, particularly in the machining [1], mining [2], and agricultural sectors [3]. This is driven by the need to improve the production efficiency and minimize production cost [4,5]. In the decision-making process for selecting materials for tribological applications, tool steel is the first resort of most industrial practitioners due to its hardness, toughness, wear resistance and cost-effectiveness [6]. However, over the course of time, the tribological performance offered by these tool steels cannot sustain the desired production output and product quality [7,8]. Consequently, this has prompted researchers to explore different tribological performance enhancement techniques [9,10].

The earliest tribological performance enhancement techniques were aimed at the bulk hardening of the tool steel. These include processes such as heat treatment (multiple

austenitization and tempering) [11,12] and cryogenic treatment [13,14]. However, these processes are energy and time-consuming [15,16]. Additionally, the resulting tribological performances were still insufficient and did not correlate with the expended cost [17]. Moving forward, the next tribological performance enhancement techniques were designed to harden the surface of the tool steels by modifying the surface microstructure and composition. While processes such as carburizing [18], nitriding [19], and nitrocarburizing [20] altered the surface composition, processes such as flame hardening [21], induction hardening [22], high energy beam hardening [23], and shot peening [24] resulted in surface microstructure modification. Nonetheless, the processes in the former category were not versatile, required postprocessing of the hardened tool, and were also time-consuming [12,25]. Meanwhile, in the latter category, the maximum hardenability and the resulting tribological performance enhancement were constrained by the tool steel's composition [26]. In addition, these processes, generally, did not offer remarkable tribological performance improvement [10]. Consequently, the desire to remain economically competitive has motivated researchers and industrial professionals to explore further alternatives [27].

Recently, tribological performance enhancement via coating deposition has attracted the attention of industrial practitioners, especially in the machining industry, primarily due to its cost benefits coupled with the excellent improvement in mechanical and tribological performance compared to the previous methods [28,29]. Despite the existence of several coating deposition methods, coating deposited via physical vapour deposition (PVD) and chemical vapour deposition (CVD) have been the subject of contemporary research [30]. This is due to their broad substrate compatibility spectrum, as a minimal metallurgical compatibility between the coating and substrate is required [31]. The improvement in

tribological performance is attributed to the increase in hardness and fracture toughness [32], as well as the increase in chemical stability which enhances the resistance of the coated material to the detrimental effect of heat generated during tribological interactions [33]. Additionally, the coatings also offer improved electrochemical resistance to corrosive agents, such as organic acid or contaminants typical for wood machining applications [34].

PVD involves the atomization or vapourization of coating atoms from a target, transportation through a vacuum or plasma, followed by condensation on the substrate [35]. Meanwhile, CVD employs the reactions among thermally activated gaseous species to produce chemical compounds under vacuum, anhydrous or anaerobic conditions and, subsequently, deposited on the substrate [30,36]. However, in recent times, the former has received wider acceptance in the industry. This is due to the energy and cost savings [36], environmental friendliness [37], lower deposition temperature ($<500\text{ }^{\circ}\text{C}$) [38], higher deposition rate (up to 10 nm/s) [35], greater flexibility in coating architecture [36], and the remarkable coating quality [39]. Generally, PVD is classified based on the method of ejecting the coating atoms from the target into evaporative and sputtering processes [31]. However, evaporative PVD coatings dominate the industrial terrain owing to their relatively higher ionization rate, higher deposition rate, more uniform deposition, and less stringent vacuum requirements [35,36]. The evaporative-based PVD process can be achieved using a variety of heat sources, including resistance, induction, electron beam and arc sources. The several limitations associated with the resistance, induction, and electron beam evaporative–PVD systems make the arc evaporative PVD-based system more attractive than industrial counterparts. For instance, there is a melting point constraint associated with resistive-based systems [31], electron gun deterioration and substrate geometry restriction [40] associated

with electron-beam-based systems, and induction-based systems are quite expensive [38,41]. Additionally, arc evaporation PVD systems provide greater versatility, higher ionization, a flexible target configuration, and a high deposition rate [4,31,35].

Following the commercialization of the arc evaporation PVD technique in the early 1980s, significant technological advances aimed at improving the process efficiency and the quality of the coatings have been attempted by scientists [37]. The first attempts were targeted at optimizing the quality and ionization of the coating materials. This entailed improving the quality of targets, using high-purity reactive gases, and incorporating additional anode and plasma sources to enhance the ionization process [36,37]. Thereafter, hybrid deposition systems utilizing evaporative and sputtering techniques were developed with the objective of maximizing the benefits of both processes [42]. However, there is still a lot of ongoing research on these systems to improve their stability and reliability to befit large-scale industrial applications [30]. Furthermore, because of the market demand, there has been an evolution of several coating architectures, including binary (e.g., CrN [10]), ternary (e.g., AlTiN [27]), quaternary (e.g., TiAlCrN [33]), and even quinary nitride-based coatings (e.g., TiAlCrCN, [43]), through appropriate elemental doping. Consequently, following these innovative advances, a significant improvement in the coating's tribological performance has been achieved over the years [44].

Unfortunately, despite the widespread adoption of CAE-PVD coatings for tribological performance enhancement, the deposition process still suffers some setbacks. Initially, controlling the location of the generated arcs on the target was a chief concern. This created a lot of instability in the deposition process, making it difficult to control the properties of the deposited coatings [45]. Although, in recent times, modifications

incorporating a magnetic field have been made to the traditional systems, making it easy to control the arcs during coating deposition, only a few are commercially available [46]. Nonetheless, the main contemporary drawback associated with CAE-PVD coatings is the formation of a significant amount of macroparticles (MPs) and craters during the deposition process [47,48]. These defects have been reported to have a detrimental effect on the coating quality and service performance [49]. To address this issue, some scientists opted for the modification of the deposition chamber configuration by incorporating filter systems [46] and the modification of the cathode geometry [37,50]. Furthermore, reports have it that the top manufacturers of CAE-PVD PVD systems, such as PLATIT, Swiss-PVD, and Hauzer, have developed advanced coating systems that produce coatings with minimal defects. However, these technologies are considered trade secrets [37]. Nevertheless, these modifications complicated the deposition system, increased their acquisition cost, and, by extension, the cost of the coated material [51]. Apart from that, some of these filter systems have insufficient filtration efficiency, thus impeding the coating deposition rate [52]. Considering these setbacks, such technologically modified systems are less attractive to the industrial counterparts. Nonetheless, addressing the shortcomings associated with these technologically advanced systems could unlock greater efficiency and durability in CAE-PVD coatings.

Moving forward, researchers decided to explore other efficient and cost-effective alternatives. This time, attention was directed towards the optimization of the deposition parameters [53,54]. Over the last decade, a considerable number of research articles have been published on this subject. The main parameters reported to have the most significant influence on these defects are the substrate bias [55], arc current [56] and the pressure of the

reactive gas [51]. The findings from these studies revealed that the optimization of these parameters is a very promising, cost-effective and practical alternative for mitigating defect formation in the coatings.

A review of the literature revealed that a significant number of articles on PVD coatings have been published, particularly over the last three decades. This also applies to the number of review articles. Approximately three decades ago, the review published by Rother [57] focused on the construction of the CAE setup, the fundamentals of the generation and acceleration of plasma and the comparison between CAE and other PVD techniques. The author also collected relevant data on arc-related material properties for different target materials. A decade later, Sanders and Anders [46] reviewed the principles of plasma production, recent advances in filter systems, growth of PVD films and industrial applications of the deposition process. About a decade ago, Bouzakis and co-workers' [30] review addressed the differences between CVD and PVD techniques, general methods of coating characterization, substrate mechanical treatments, and postdeposition treatments. Later, in 2018, Baptista et al. [36] published a detailed review of the research advances on the sputtering PVD process. This included the principle of the deposition technique, the effects of the deposition parameters on coating properties, as well as recent advances in improving the coating deposition process. Two years later, Deng et al. [37] published an interesting review with extensive discussions on the principles of CAE-PVD, cathode configuration, power supply for CAE-PVD systems, magnetron sputtering, hybrid systems, and plasma etching, as well as their industrial applications for general cutting tool applications. Recently, Warcholinski and Gilewicz [10], in their review, discussed the

research advances in binary and multilayer coatings, as well as the effect of doping on the properties of PVD coatings.

From the foregoing, it was discovered that despite the vast number of research articles on PVD coatings, very few articles are available on the effect of substrate morphology, particularly the effect of the substrate profile on the properties of PVD coatings, despite its significant influence on the coating's tribological performance [7,35]. As a matter of fact, based on the knowledge of the authors, no review article has been published on this subject. Surprisingly, this is also true for the effect of deposition parameters (especially the arc current, deposition temperature, and substrate bias) on the properties of CAE-PVD coatings. A comprehensive understanding of the effect of these parameters on the properties of the coatings not only creates the pathway to obtaining coatings with a superior tribological performance but also has the potential to result in energy savings during the deposition process. This is of prime importance, especially in large-scale industrial production where profit maximization and production cost minimization are the primary objectives. Hence, part of the objective of this review is to present relevant discussions on these crucial subjects. However, the review is presented systematically, beginning with discussions on substrate preparation and its effects on the properties of CAE-PVD coatings. Thereafter, insightful discussions are presented on the process and principles, microstructural evolution during coating formation, MPs, and characterization of CAE-PVD coatings. Afterward, an intuitive literature analysis is presented on the effect of arc current, deposition temperature, and substrate bias on the properties of the coatings. Later, research advances in coating architecture and the corrosion properties of CAE-PVD coatings are discussed. The idea behind this systematic approach is to bridge the gap in the literature by integrating previous

works into contemporary studies so as to clearly spot the research gaps and also keep industrial counterparts abreast of the most relevant findings to facilitate the making of informed decisions.

3.2 Substrate preparation

The primary objective of substrate preparation is to make the substrate more receptive to the coating layer, ensuring effective coating–substrate interfacial bonding [40]. Interestingly, this process accounts for the bulk of the cost expended in the entire deposition process [36]. But, unfortunately, despite its significant effect on the coating’s tribological performance and cost contribution, it has not received due attention from researchers over the past years [38]. The insufficient number of studies on this subject matter could be the result of the influence of the historical emphasis on other areas (such as composition, architecture, process parameters, etc. [7]), the constraints of the substrate finish’s requirements for some applications (such as medical implants and optical components [58,59]) and complexities in the substrate profile variation in some substrate materials (such as composite materials [60]). The entire substrate preparation process involves operations targeted at eliminating contaminants emanating from substrate preprocessing and the annihilation of surface defects through adequate profiling [40]. These processes are illustrated in Figure 3-1.

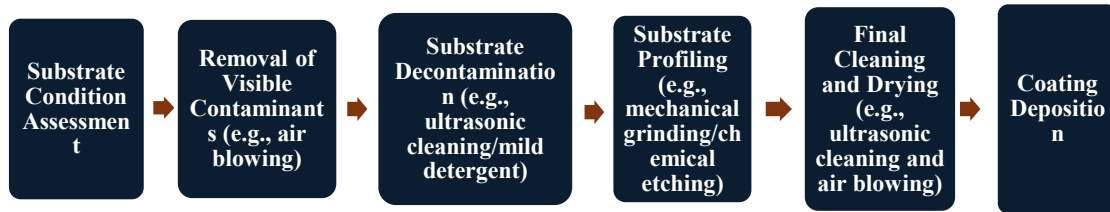


Figure 3-1: The processes involved in PVD substrate preparation.

Firstly, the surface condition of the substrate should be assessed for the presence of visible loose oxide layers (such as metallic rusts in the case of tool steels), dust particles, or organic dirt [37]. If present, they could be removed by air blowing. Thereafter, the substrate is subjected to decontamination. The decontamination process is aimed at eliminating contaminants, such as grease, oils, chlorides, oxides, and hydrocarbons, from the surface of the substrate. Sometimes, these contaminants are microscopic and may require analytical techniques to ascertain their presence [35]. The presence of these contaminants, even in minute concentrations, can compromise the coating adhesion and the resulting properties [61]. Substrate decontamination can be achieved through a variety of methods. Some of the decontamination techniques reported in the literature are ultrasonic cleaning with alcohol [62,63] or hot alkaline [64]. However, in an industrial setting, decontamination is mostly achieved using mild solvent detergent owing to the hazards posed by the former [49]. It is important to mention that even with high-purity decontaminants, the substrate cannot be entirely contaminant free, mostly due to the presence of oxygen-containing species in the atmosphere. These oxide contaminants have a high surface energy and tend to absorb other contaminants with lower energies (particularly hydrocarbons) as a means of lowering their surface energy. However, these contaminants are removed subsequently by etching prior to

coating deposition [65]. Figure 3-2 shows a typical surface of a substrate before and after the decontamination process.

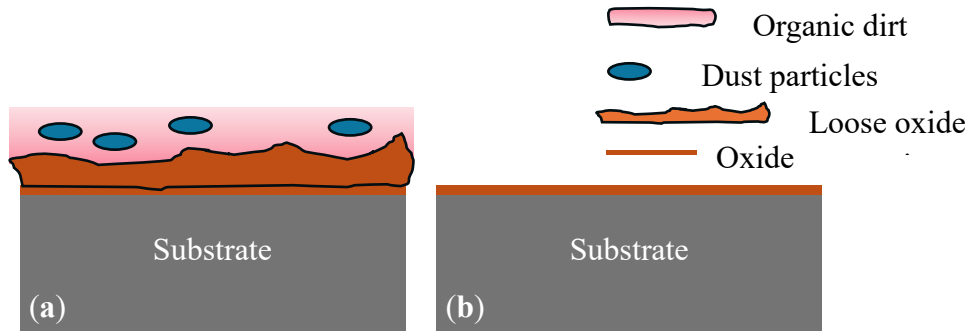


Figure 3-2: Surface of the substrate: (a) before decontamination; (b) after decontamination.

The next step is the profiling of the decontaminated substrate. The significance of this stage cannot be overemphasized, as it determines the extent of the mechanical interlocking and chemical bonding of the coating to the substrate [66,67]. Substrate profiling can be achieved through mechanical grinding/polishing, microblasting, or chemical etching, among others [35]. Mechanical substrate profiling is performed using an abrasive wheel or SiC-based grinding papers followed by polishing with an appropriate medium, such as diamond suspension. The induced substrate profile is a function of the grit size of the SiC-based papers, the grain size of the abrasive wheel, or the suspension size of the polishing media [8,40]. Microblasting, however, involves focusing abrasive particles (such as sand particles) at a high pressure (up to 4 bar) on the substrate. Here, the resulting substrate profile depends on the pressure and abrasive properties of the abrasive medium [62]. Meanwhile, chemical substrate profiling is achieved by etching the substrate using appropriate etchants, such as Murakami's reagent or Caro's solution [68]. Nevertheless, the profiling method to be adopted depends on the type of substrate. For instance, while mechanical treatments are

mostly used for tool steels [69], chemical etching is mostly adopted for cemented carbides and cobalt-based substrates [5]. However, irrespective of the profiling method, the emphasis is laid on producing a homogenous surface profile with minimal surface flaws such as microcracks [35]. More so, care should be taken to avoid inducing additional residual stresses on the substrate, as it has a detrimental effect on the coating adhesion [68]. While microblasting techniques induce more residual stresses on the substrate than other profiling techniques, mechanical profiling is more prone to generating surface flaws on the substrate [7,70]. However, the optimum microblasting treatment has been reported to result in improved tribological performance, characterized by increased hardness compared to the other surface profiling techniques [30,62]. Following substrate profiling, it is necessary to further clean the substrates to eliminate contaminations emanating from the profiling process. This could be achieved by ultrasonic cleaning with detergent and subsequent rinsing with deionized water [63]. Drying the substrate after profiling before coating deposition is of the utmost importance to minimize pinhole formation [35].

3.2.1 Effect of substrate profiling on the properties of CAE-PVD coatings

The preceding section delineated that achieving an optimum substrate profile is very crucial for producing coatings with superior mechanical properties and tribological performances. Hypothetically, the rougher the substrate, the higher the coating–substrate bond strength due to the enhancement in the mechanical coating–substrate interlocking [63]. More so, it also increases the number of nucleation sites, facilitating the coating’s growth [66]. However, an excessively rough substrate profile (up to an average roughness (R_a) >10 μm) could result in coatings with a nonuniform thickness [68]. This is usually accompanied by an increase in the R_a of the coatings, which has been reported to negatively impact their

tribological performance [71]. The deterioration in the tribological performance is attributed to the relatively small contact area on the rougher substrate, producing higher pressures on the coating during abrasive contact [72]. Although the effect of a rough substrate can be minimized by increasing the coating thickness [73], but then, thicker coatings are accompanied by high residual stresses, which also negatively affect coating adhesion [5]. Moreover, the substrate roughness has been reported to correlate with pinhole formation in the deposited films. Hence, coatings deposited on a rougher substrate would have a greater pin hole density, making them more susceptible to tribochemical reactions [34].

Meanwhile, a very smooth substrate (e.g., $R_a < 0.01 \mu\text{m}$) has limited nucleation sites and provides insufficient coating–substrate interlocking, thus resulting in poor adhesion [70]. Coatings deposited on such substrates are prone to spalling [68]. Apart from coating–substrate bonding, the substrate profile also has a significant influence on the morphology of the deposited coatings [74]. Accordingly, coatings deposited on rougher substrates have a more columnar morphology compared to those deposited on smoother substrates [7,34]. At this juncture, several puzzling questions may arise, such as the following, among others: What is the optimal substrate roughness? Does it vary for different substrates with similar coatings? Does it vary for different coatings with similar substrates? Table 3-1 presents significant findings on the effect of the substrate profile on the properties of CAE-PVD coatings and other relevant information related to the profiling methods, substrates, and coatings.

Table 3-1: Summary of the effect of the substrate profiling on the properties of CAE-PVD coatings.

Profiling Method	Profiling Details	Average Roughness, Substrate R_a (μm)	Coating	Thickness (μm)	Significant Findings	Ref.
Mechanical treatment	Mechanical grinding discs (600 and 1200 mesh sizes) and polishing with diamond powder.	0.016–0.038	Tungsten carbide	TiAlN	3.5	1—Increase in coating's R_a and COF with the substrate's R_a . 2—No significant effect on the crystal structure, residual stresses, and coating thickness. [66] 3—Increase in the adhesion strength and wear resistance at lower R_a .
Mechanical treatment	Mechanical grinding with emery paper (800 and 1000 grit sizes) and 1 μm diamond polishing.	0.01–0.08	AISI M2 HSS	TiN and nc-TiAlSiN	2	1—Minimal wear rates at high substrate R_a . 2—Increase in the adhesion strength of TiN and a decrease for the nc-TiAlSiN coating with an increasing substrate R_a . [70]
Chemical treatment	Etching with Murakami's reagent and Caro's solution.	0.05–0.11	Tungsten carbide	AlCrN	1.24–2.78	Increase in the coating thickness, homogeneity, and adhesion strength at higher substrate R_a . [68]
Mechanical treatment	Mechanical grinding with emery paper	0.05 and 0.25	AISI 100 steel	TiN	0.432 and 2.83	1—Increase in the MP density, COF, H/E , H^3/E^2 , adhesion strength, [63]

	(600–2500 mesh sizes)					and wear resistance with increased substrate R_a . 2—No significant changes in the coating thickness.
Mechanical treatment	Not provided	0.1–0.31	AISI M2 HSS	CrN	2.36	Reduced adhesion strength, corrosion, and high-temperature oxidation resistance at higher substrate R_a . [34]
Not provided	Not provided	0.037– 1.348	Cr ₁₇ Ni ₂ stainless steel	Ti-TiN-Zr- ZrN	11–15	Reduction in the adhesion strength and coating homogeneity at higher substrate R_a . [73]

COF—coefficient of friction; H—hardness; E—elasticity; MP—macroparticle; R_a —average roughness.

The succeeding literature analysis presents some insight into the optimum surface roughness for an enhanced tribological performance of CAE-PVD coatings. The findings of Huang et al. [66] revealed a reduction in the adhesion strength and wear resistance of TiAlN-coated carbide with a substrate roughness up to an R_a of approximately 0.04 μm . This observation resulted from the cumulative increase in the R_a and coefficient of friction of the coatings with an increasing substrate roughness [66]. Contrastingly, for a higher substrate R_a (reaching 0.08 μm), Ravi and co-workers [70] reported a positive correlation between the wear resistance and R_a for a tool steel substrate coated with TiN and TiAlSiN. Similar findings were also reported by Uddin et al. for TiN-coated tool steel [63]. The improvement in the wear resistance was attributed to the prevalence of three-body abrasion owing to the formation of more wear debris facilitated by the rougher coatings. However, the adhesion of

both coatings varied differently with the surface roughness. While TiN exhibited superior adhesion strength at lower substrate roughness, the reverse trend was observed in the latter [70]. The behaviour of the TiAlSiN nanocomposite coating was expected because the rougher substrate enhanced the interlocking mechanism of the coating–substrate system [75]. On the other hand, the behaviour of the TiN was attributed to the reduction in the surface ridges, which act as stress raisers and, subsequently, result in coating failure at lower loads [76]. Further investigations by Ling et al. for a similar substrate R_a for AlCrN-coated carbide revealed that coatings deposited on the rougher substrate showed superior adhesion strength owing to the improvement in the interlocking mechanism [68]. This finding conflicts with previous reports by Huang et al. [66] considering that tungsten carbide was the substrate in both studies. This discrepancy could stem from the difference in the coating composition (see Table 3-1). Additionally, investigations by Lee et al. [34] for substrate R_a reaching 0.3 μm for CrN-coated tool steel revealed the optimum tribological performance at a 0.1 μm R_a owing to the lower crater density and coefficient of friction. Lastly, studies by Lin et al. showed that when the substrate R_a exceeds 1 μm , there is a reduction in the coating's adhesion strength [73].

From the foregoing, the general trend observed for most of the coating–substrate system is increased adhesion strength with increasing surface roughness, up to about an R_a of 0.1 μm . The increase in the adhesion strength is primarily attributed to the increase in the nucleation density facilitating the interlocking mechanism, as the asperities in the rougher substrate facilitate the penetration of the coating material, resulting in stronger bonding. But at a higher substrate R_a , especially when $R_a > 0.2 \mu\text{m}$, there is an observed general decline in the adhesion strength. This is due to the stress concentration on the ridges of the rough

substrate, resulting in coating failure at much lower loads. Likewise, the wear resistance of most of the coated substrates followed a similar trend. Some contributing factors to the increased wear resistance are increased adhesion strength, hardness, and H^3/E^2 . Again, it is important to mention that while this optimal range of substrate roughness is achievable for machining applications, such as woodworking and metal machining, it might not be applicable for applications requiring a high surface finish, such as deep drawing. Nonetheless, as it currently stands, it appears that the effect of the substrate profile on the properties and the tribological performances of CAE-PVD coatings have not been well explored. Many research questions, such as the influence of the surface profile on the interfacial chemistry, still need to be answered. Again, more systematic studies on the correlation between the substrate profile, coating morphology, mechanical properties, and tribological performance need to be conducted.

3.3 Process and principles of CAE-PVD

The series of operations involved in the CAE-PVD is shown in Figure 3-3. An understanding of the sequence of processes and the principles of the deposition process are essential to facilitate improvement in the coating quality and the resulting tribological performance.

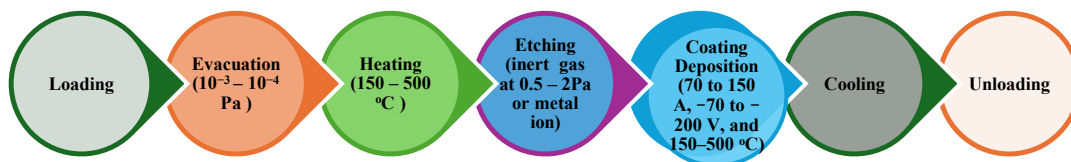


Figure 3-3. Processes involved in CAE-PVD coating

The process commences with loading the adequately prepared substrates in the deposition chamber. Generally, the target-to-substrate distances is maintained between 18 and 25 cm [4,48] to minimize the effect of radiant heating during the evaporation process [35]. Thereafter, chamber evacuation to create a vacuum in the range of 10^{-3} – 10^{-4} Pa [32,74] is required to prevent gaseous contamination during coating deposition [35,40]. This is followed by heating to temperatures between 150 and 500 °C as a precursor to the etching process [77].

During the etching process and the subsequent coating deposition, substrate rotation at a 1.5 to 2 rpm [32,48] or higher is required to ensure uniformity in these processes [40]. The purpose of the etching process is to get rid of the residue contaminants from the decontamination process [36]. Most often, etching is achieved using an inert gas or metal ion plasma. During the process, a high-speed plasma containing an inert gas, such as argon at pressures in the range of 0.5–2 Pa [34], or metal ions, such as Ti, Cr, and Al [37], is directed to a highly negatively biased substrate (200–1000 V) for a period of time (typically 10–45 min) [32,74,78]. This leads to the sputtering of surface contaminants and the activation of a superficial layer on the substrate's surface [37]. The purpose of the heating during this process is to facilitate the transportation of the etching species while the high substrate biasing promotes the sputtering of the surface contaminants and also prevents the incorporation of the etching species into the substrate [7,79]. Apart from decontamination, the process also results in roughening of the substrate's surface [37,80], facilitating further coating–substrate interlocking. Comparing metal ion and inert gas etching, Sproul and co-workers reported that the former is less time-consuming and offers superior coating adhesion

[81]. Nonetheless, a combination of both etching technologies could also be adopted [48]. The etching process is succeeded by evacuation of gaseous contaminants and re-establishment of the vacuum condition in preparation for coating deposition. Figure 3-4 shows the layout of a typical CAE-PVD system.

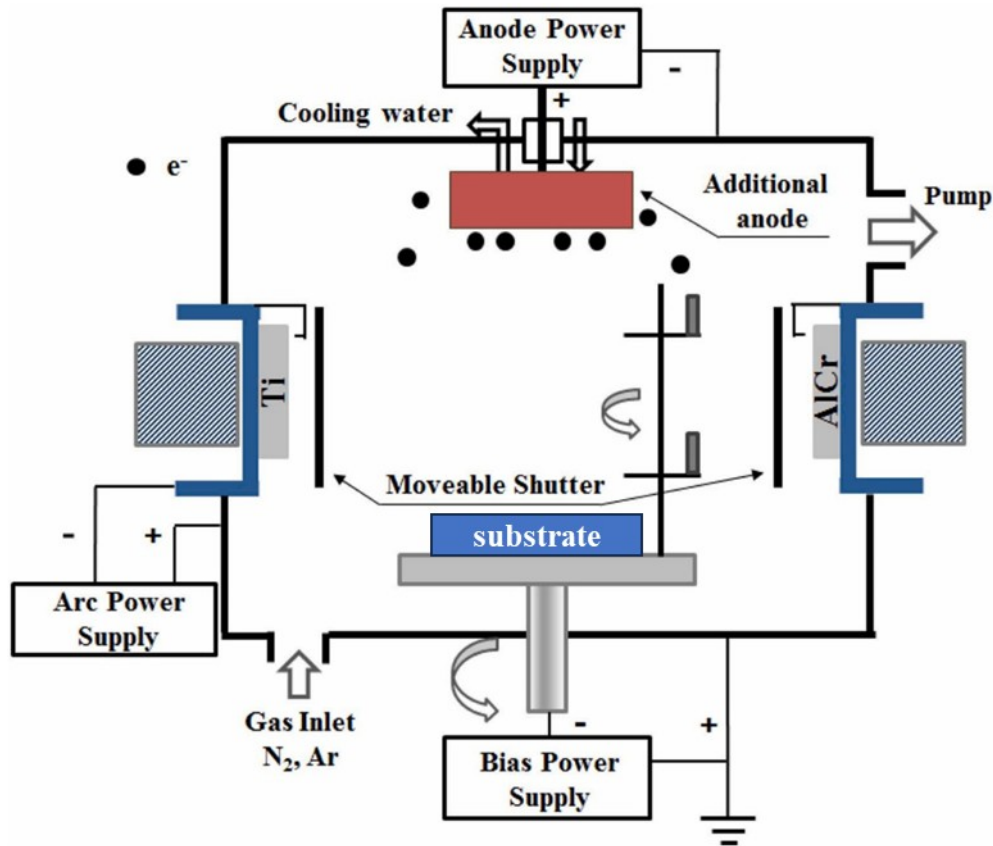


Figure 3-4: Schematic of a typical CAE-PVD system [82].

The coating deposition process commences with the evaporation of the coating atoms from the target by initiating an arc on the cathode [37]. Depending on the system configuration, arc initiation can be achieved through a trigger arc obtained from a high voltage on an auxiliary electrode usually located near the cathode surface or applying a low-voltage electric current between two slightly separated electrodes in a vacuum or through

laser ionization [35,83]. Ideally, the generated arc moves randomly on the cathode surface, but in some system designs, the arc can be pulsed using a direct current power supply, steered around a particular path, or filtered using a magnetic field [83]. The behaviour of the arc is also influenced by the properties of the cathode, including the cathode geometry, cathode material, and purity level [35]. The generated arc, which is concentrated on the surface of the cathode, results in the formation of nonstationary microspots (known as cathode spots) on the target. These cathode spots are usually characterized by a high current density (10^4 – 10^{11} A/cm²), short life span (in the range of microseconds), and a characteristic size reaching 1 μ m. However, the specific properties of the cathode spot depend on the cathode material [46,84]. The number of cathode spots generated is largely dependent on the arc current [83]. Because of the short residence time of these spots, they are quickly extinguished. However, when this happens, they are re-ignited by a voltage spike provided by the inductance in the arc current [35]. Figure 3-5 illustrates the principles of the CAE-PVD.

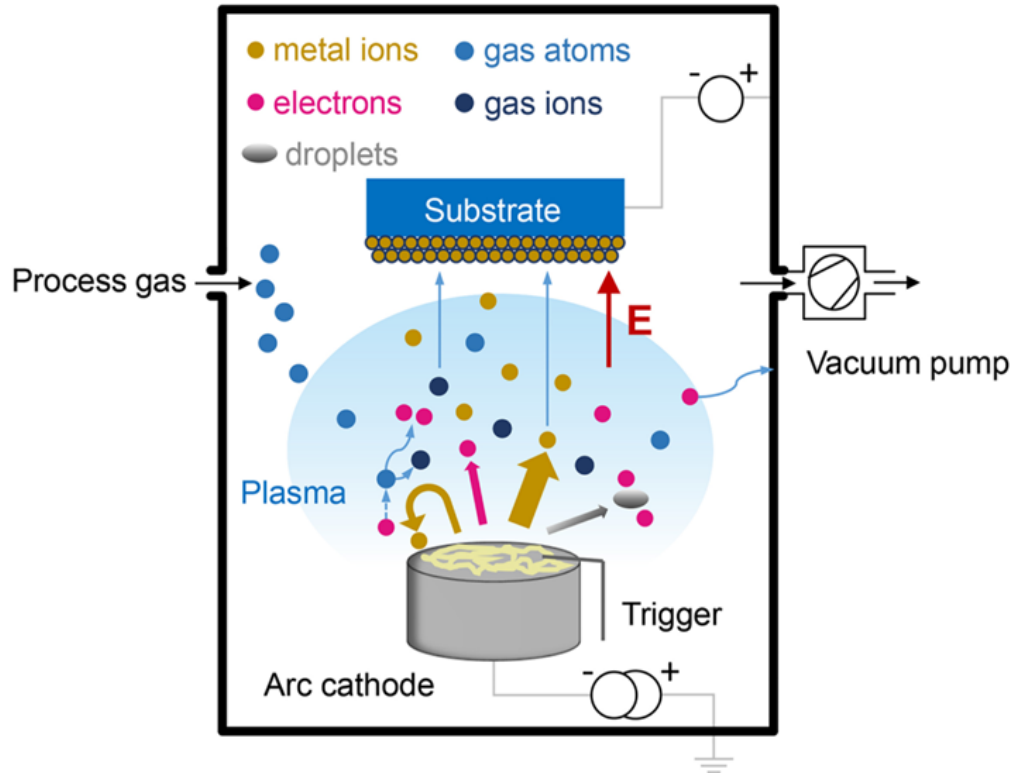


Figure 3-5: An illustration of the principles of the CAE-PVD [37].

The high current density of the cathode spot erodes the cathode, leading to the localized melting, ionization and evaporation of the cathode material alongside a significant number of MPs [46]. The extent of the cathode ionization and the number of MPs depend on the melting temperature of the cathode material [37,85]. In some system designs, an additional anode is incorporated to improve the ionization efficiency and the quality of the deposited coatings [32,37]. The generated gaseous system comprising electrons, MPs, and ions constitute the plasma. In reactive CAE-PVD, a reactive gas (usually nitrogen) introduced into the deposition chamber is activated by the plasma, leading to its dissociation to produce ions and more electrons. Afterwards, the plasma is transported to the substrate. The particles in the plasma travel at different velocities, typically between 10 and 10,000

m/s, depending on their atomic mass and energy [46,83,85]. Upon arriving at the substrate's surface, the plasma volume near the substrate suffers a loss of plasma species to the substrate. Since electrons have greater mobility, they are lost at a relatively faster rate, thereby creating a potential between the substrate and the plasma [35,46]. This justifies the requirement of a negative substrate bias to accelerate the positive coating ions to the substrate, resulting in a faster deposition rate [86]. Subsequently, the plasma species, including the MPs, condenses on the surface of the substrate [31,35]. This is followed by the cooling of the deposition chamber achieved mostly by water cooling [35,83]. Eventually, the coated substrates are unloaded from the deposition chamber.

3.4 Microstructural evolution of CAE-PVD coating

An understanding of the processes involved in the microstructural evolution of CAE-PVD coatings is essential for developing coatings with improved tribological performance. The process involves the condensation of mobile coating atom species (known as adatoms) on the surface of the substrate, followed by nucleation, growth of the nuclei, formation of the coating–substrate interface, and continuous film growth [35]. These processes are illustrated in Figure 3-6.

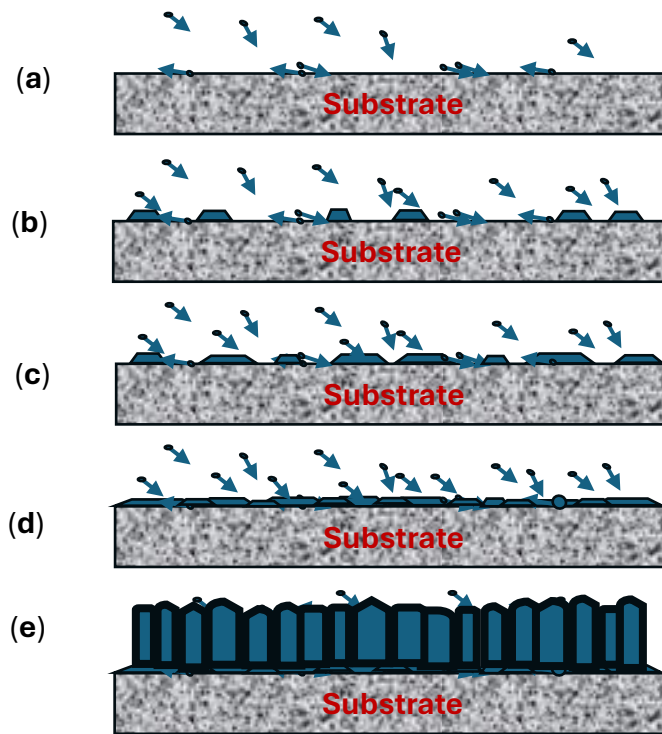


Figure 3-6: Schematic diagram illustrating the microstructural evolution during the CAE-PVD coating's formation: (a) condensation of adatoms; (b) nucleation; (c) growth of nuclei; (d) interface formation; (e) continuous film growth.

The coating evolution process commences with the condensation of the adatoms on the surface of the substrate as shown in Figure 3-6a. Subsequently, the condensing adatoms experience energy loss due to the collision with other adatoms, chemical bonding with the substrate surface atoms, or collision with other adatoms already bonded to the substrate [35]. Most often, the preferential sites for the impinging adatoms are surface defects or impurities [66]. At this stage, the mobility of the adatoms is primarily influenced by the bond strength [48,86]. The condensation of the adatoms is followed by nucleation as shown in Figure 3-6b. Every respective condensing adatoms constitute a nucleation site for coating growth. The nucleation of coating atoms is largely influenced by the crystal structure of the substrate, the

binding energy of the adatoms, contaminations, and the surface profile of the substrate, among others [61,87]. Following adatoms nucleation, the nuclei growth involves the bonding of the formed nuclei with other incoming adatoms or with existing condensed adatoms through surface diffusion [35]. An outcome of the nuclei growth is the formation of randomly oriented islands (as shown in Figure 3-6c). This is then followed by the coalescence of the islands to form the interfacial region (Figure 3-6d).

Following the interface formation is the stage of continuous film growth. This stage is characterized by the continuous nucleation of the condensing adatoms on the interface accompanied by continuous growth, where previously deposited coating species are continually buried by the incoming ones [35]. During this process, there is grain coarsening, which is evident in the coalescence of the islands (as shown in Figure 3-6e). The driving force for this migration of island boundaries is the need to minimize the edge and surface energies, whereby the islands with lower energy densities consume others with higher densities. This process is very competitive, as crystallographic planes with higher densities are the preferred orientation, causing them to grow faster than less dense planes [61]. This difference in the growth rates of different crystallographic planes produces a nonuniform coating surface morphology, thus contributing to the increased surface roughness of the deposited coating. This effect is more evident when the flux of the incoming adatoms is off-normal or when the adatoms have low energy. Under these two conditions, the effect of geometric shadowing is very dominant, whereby the peaks of the faster-growing planes receive all of the incoming adatoms, thus shadowing the valleys of the slower-growing planes. The resulting coating microstructure, characterized by a columnar morphology, appears like aligned plates piled together, as shown in Figure 3-7 [35].

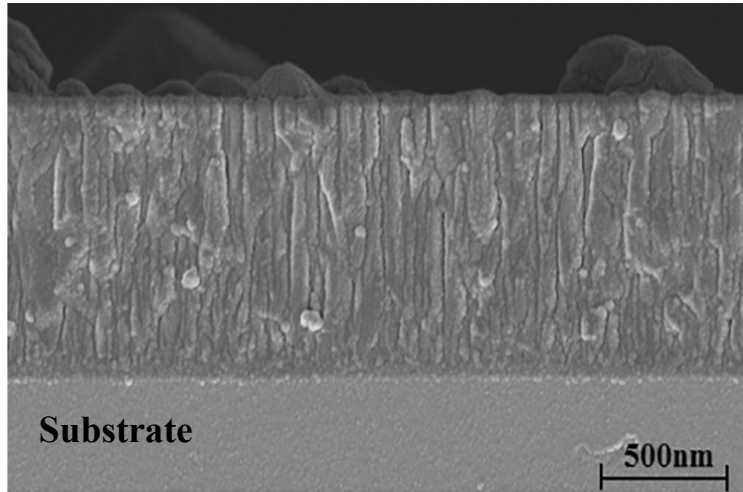


Figure 3-7: Columnar morphology of the CrN/CrCN CAE-PVD coating deposited on AISI M2 high-speed steel [32].

3.4.1. Structural zone model (SZM)

In an attempt to engineer the microstructural evolution during coating formation for different technological applications, significant efforts have been put forth by researchers to model the microstructural evolution in terms of the coating growth parameters. One of the earliest successful attempts was the collective efforts of Movchan and Demchishin in 1969 [88] and, later on, Thornton in 1974 [89], which birthed the Structural Zone Model (SZM). According to this model, the resulting coating microstructure is a function of the competitive interplay between the rate of condensation of the adatoms and the rate at which these adatoms are able to re-arrange themselves before the arrival of new ones. On the basis of this model's concept, dense and compact coatings can be obtained at high deposition temperatures and slower deposition rates. Under these conditions, there is sufficient energy and time for the arrangement of the condensed adatoms as the rate of arrival of new adatoms is reduced [46]. Figure 3-8 shows a schematic of a typical SZM.

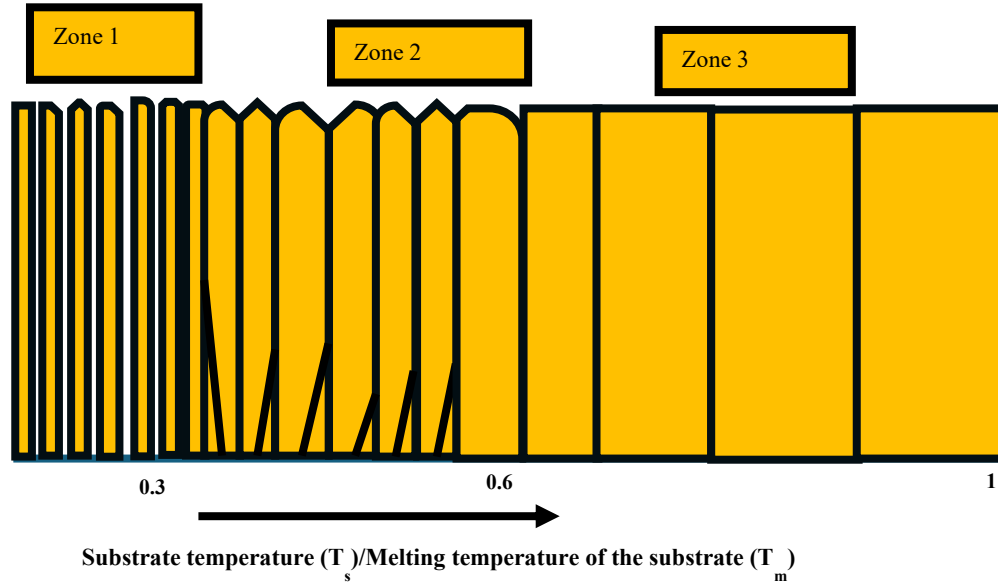


Figure 3-8: A schematic diagram of the structural zone model.

The model shows the microstructural evolution of the coating as a function of the ratio of the substrate temperature (T_s) to its melting temperature (T_m). Movchan and Demchishin [88] were the first to classify the model into three distinct zones. Zone 1 represents a region of low substrate temperature where the mobility of the adatoms is relatively low. Consequently, they are not able to overcome the geometric shadowing effect of the substrate profile, leading to the formation of an under-dense film structure characterized by open boundaries between the columns and increased surface roughness morphology. The typical T_s/T_m temperature ratio for this zone is usually in the range of 0.1 to 0.3. In zone 2, the diffusion of the adatoms is significant (higher than that in zone 1) because of the higher substrate temperature leading to the coalescence of the columnar boundaries and the formation of denser coatings with faceted tops. The temperature rise is facilitated by substrate biasing leading to an increase in energetic substrate bombardment [90]. The faceting of the column is a product of the competition among the different crystallographic

planes having different growth rates. The typical T_s/T_m ratio for this zone is between 0.3 and 0.5. Later attempts by Thornton [89] to modify the initial model developed by Movchan and Demchishin [88] suggested the existence of a transition zone ($T_s/T_m = 0.4$ to 0.5) between zones I and II characterized by poorly defined columnar grains. In zone 3, the diffusion rate is much higher than in the other zones, occurring on the surface and bulk of the coating. This promotes recrystallization and grain growth, producing dense columnar coatings with a flat surface, as the growth orientation is more pronounced. This zone's typical T_s/T_m ratio is between 0.5 and 1 [35,61]. Furthermore, Anders also recently proposed a modified version of the model taking into consideration parameters related to the coating growth process, such as the film thickness and the heating effect resulting from energetic substrate bombardment [91]. Moreover, the understanding derived from this model on the effect of substrate temperature on the coating morphology could be adapted to improve the tribological performance of CAE-PVD coatings.

Further critical analysis of the model to ascertain the region of likely optimum coating properties for tribological applications revealed the following. In zone 1, the reduction in coating properties is mostly attributed to the porous columnar structure. However, in zones 2 and 3, the increased surface roughness and grain coarsening account for the reduced service performance of the coatings, respectively. Therefore, it appears that the optimum coating properties would likely be obtained at regions between the end of zone 2 and the beginning of zone 3 (i.e., about $T_s/T_m = 0.5$ – 0.7). This is because, in this region, the effect of surface roughness and grain coarsening are minimal. However, in the actual CAE-PVD process, especially with tool steel substrates, such a temperature range corresponding to about 790 – 1100 °C is not feasible. This is because, at such temperatures,

there is a likelihood of substrate softening, particularly due to carbide coarsening, resulting in reduced tribological performance [1]. Consequently, most CAE-PVD is carried out at much lower temperatures corresponding to a T_s/T_m of 0.1–0.3 (zone 1). However, the literature reports revealed that these coatings exhibited good service performance [4]. Despite the low deposition temperature, the observed service performance of the coatings could be attributed to the intense bombardment of the energetic ions resulting in dense compact coatings with good mechanical and tribological properties [92].

3.4.2 Macroparticles (MPs) in CAE-PVD coatings

From the time CAE-PVD coatings were introduced to the coating industry until the present day, the inclusion of micro-sized MPs embedded in the coating structure or situated on the coating surface alongside the associated pitting defects (called craters) have been a major challenge. In fact, these MPs present a significant threat to the widespread adoption of these coatings for tribological applications, particularly on an industrial scale [46,51]. The mechanism of formation of these MPs is complex and is still the subject of contemporary research [54]. Over time, several explanations have been offered regarding their formation. According to Anders [93], MPs are formed as a result of the combination of thermal shock and hydrodynamic effects on the cathode spot, leading to the ablation of molten particles, which eventually solidify on the substrate. Lee et al. [34] attributed their formation to the occurrence of micro-explosions on the cathode caused by the generated plasma or the product of the intense backflow of the generated ions on the cathode spot during ionization. Meanwhile, the most popular opinion regarding the formation of the craters is that they are the outcome of the geometric shadowing by the MPs and become apparent after the flaking of the loosely bound MPs [52]. However, Naghashzadeh and co-workers attributed their

formation to the incomplete reaction between the evaporated MPs and the reactive gas during the deposition process [74]. Figure 3-9 shows the surface of a typical CAE-PVD coating with MPs and craters.

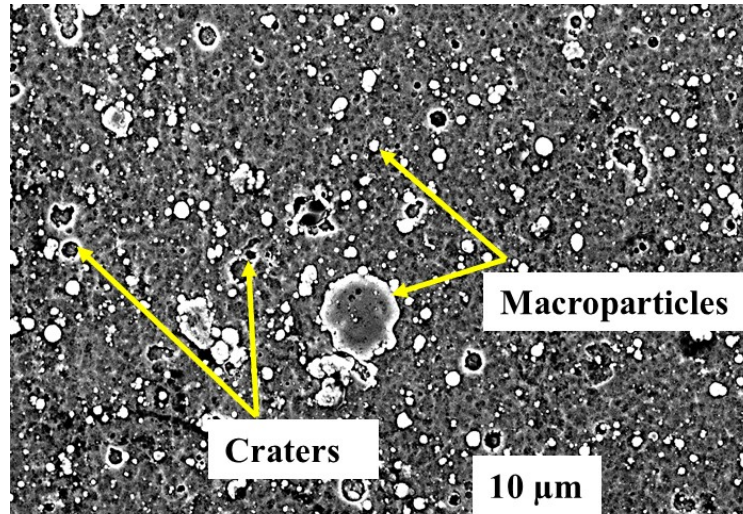


Figure 3-9: Micrograph of AlTiN/AlTiCrN coating surface showing macroparticles and craters.

The generated MPs have different characteristic shapes, including spherical, oval, elongated, and lens-like, depending on the size, condition, and velocity at which they arrive at the substrate [54,71,94]. The sizes of the MPs typically vary between 0.1 and 10 μm depending on the extent of MPs' agglomeration during the coating deposition [4]. However, the MP density is largely dependent on several factors, such as the melting temperature of the cathode, the substrate–cathode distance, and the deposition parameters [35]. Researchers have reported that the MP density is positively correlated with the arc current and negatively correlated with the melting temperature of the cathode and substrate–cathode distance [35,52,95]. The MP density is also largely dependent on the velocity and trajectory of the

MPs [47]. For instance, an MP travelling at a high velocity (>5 m/s) with a trajectory grazing the substrate MP could exhibit an elongated morphology after impact with the substrate and, in some cases, may become almost invisible. The composition of these MPs is also complex and is a function of the position of the MPs within the coating structure rather than their sizes. For instance, MPs embedded in the central part of the coating structure are more likely to have a lower amount of reactive gas than those located on the coating surface or just below the coating surface [71,93]. According to Vereschaka et al. [47] and Xu et al. [96], the compositions of MPs may include pure metal (corresponding to the cathode material, e.g., Ti), metal nitrides (e.g., TiN), or multicomponent nitrides (such as those comprising a metallic core with multiple outer nitride layers), which are formed as a result of the diffusion of the metallic ions from the target to the surface of the MPs (e.g., Ti-TiN-TiAlN).

The reason why MPs have received significant attention from researchers in recent years is due to their detrimental effect on the coating quality and the ensuing tribological performance. Firstly, their varying compositions and sizes across the coating thickness result in heterogeneity in the coating composition and properties [35]. Secondly, their presence on the surface of the deposited coatings increases the surface roughness, thus accelerating degradation in tribological performance [4,50]. Thirdly, they have also been reported to negatively impact the adhesion strength and coating density [7,97]. Furthermore, researchers have also hypothesized that the impingement of the coatings by MPs induces compressive deformation, which can facilitate crack initiation [98]. In fact, recent studies conducted by Vereschaka et al. revealed that MP impingement results in significant compressive coating deformation, making the region of impingement distinct from the rest of the coating structure [47]. This occurrence is shown in Figure 3-10a. Also, in connection with this, studies

conducted by Tkadletz et al. on the effect of MPs on the tribological performance of a TiAlTaN coating revealed that during abrasive wear, the MPs are embedded in the coating matrix, thus serving as nucleation sites for shear crack initiation, as shown in Figure 3-10b [71]. Additionally, Warcholinski and Gilewicz expressed that the difference in the thermal properties between these MPs and the coating matrix could initiate localized thermal stresses, eventually resulting in crack initiation, especially at the coating–MP boundary [4]. Furthermore, the craters associated with the MPs facilitate tribochemical reactions, as the corrosive agent could readily be transported to the substrate [52].

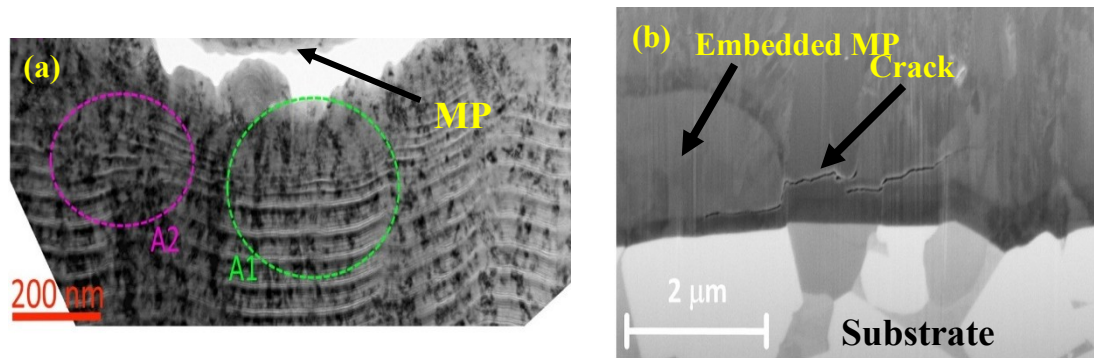


Figure 3-10: (a) TEM micrograph showing compressive deformation induced by a Ti microdroplet on a TiN-(Ti, Mo, Al)N coating (A1—region of compressive deformation; A2—undeformed region) [47]; (b) SEM micrograph showing crack initiation and propagation from an MP embedded in a TiAlTaN coating [71].

Despite the negative impacts of MPs on the quality of the deposited coatings, some potential benefits have also been highlighted by some researchers. In this regard, Vereschaka et al. likened the bombarding effect of MPs on the coating to shot peening and, thus, hypothesized that they could strengthen the deposited coatings [47]. Aside from the hypothesis not being supported by experimental validation, it appears that the tendency of

the MPs to act as crack nucleation sites outweighs any strengthening they could possibly confer. This is justified by the reduction in the tribological performance of CAE-PVD coatings with an increasing MP density [47,86]. Researchers have also hypothesized that MPs could offer a form of lubrication and potentially enhance the thermal stability of the deposited coatings. The lubricative property is attributed to the possibility of MPs forming an intermediate layer between the coated material and the abradant. The emphasis was laid on aluminium containing multicomponent nitride MPs such as Al-AlN. In this case, the high temperatures (typically up to 600 °C or higher [1]) encountered during tribological interactions, particularly during woodworking and metal machining, could melt the aluminum component, thus forming a highly ductile layer in the cutting zone. Furthermore, the high melting temperature of the AlN phase (2200 °C) could possibly increase the thermal stability of the coatings [47]. However, further experimental studies are required to validate these hypotheses.

3.4.3. Research advances in MP reduction in CAE-PVD coatings

Over the years, significant efforts have been put forth by researchers to minimize MPs in CAE-PVD coatings. One of the outcomes of such efforts is filtering the MPs from the plasma stream prior to condensation on the substrate [37]. The concept was first introduced by Aksenov and co-workers in the late 1970s [99]. Plasma filtering can be achieved mechanically by shielding the cathode or applying an external magnetic field. However, magnetic filtering has received greater attention due to its superior filtering efficiency [37]. Magnetic filtering of the plasma is made possible because of its directional line-of-sight trajectory coupled with the difference in charge-to-mass ratio and velocity of the MPs, ions, and electrons contained in the plasma stream [51]. In the presence of an

applied magnetic field, ions and electrons with a relatively lower mass-to-charge ratio are bound to the magnetic field lines and, subsequently, guided outside the line of sight, while the MPs continue along the line-of-sight trajectory, resulting in their separation from the plasma stream. Over the past years, significant efforts by researchers to improve the filtering efficiencies of the magnetic filters have birthed several magnetic filter types with different geometries (e.g., linear duct and S-filter) and architectures (closed or open) currently available on a commercial scale [46,51]. However, apart from the complexity and increased cost of these deposition systems, the disruption of the motion of the plasma during filtering results in a drastically reduced deposition rate [51]. In this regard, a study conducted by Steffens et al. revealed that the application of a magnetic field (10 mT) during the deposition of TiN and CrN coatings on AISI D2 tool steel resulted in a 50% reduction in coating thickness despite the 25% reduction in surface roughness [50].

One of the cost-effective methods for MP reduction reported by researchers is increasing the substrate bias during coating deposition [100]. Two major mechanisms have been put forward to explain the observed reduction in MP density at a higher substrate bias. The first is the mechanism of ion bombardment. On the basis of this mechanism, the higher substrate biases increase the energy of the bombarding ions on the substrate, consequently leading to the re-sputtering of loosely bound MPs from the coating [101,102]. However, a more popular opinion is the electrical repulsive mechanism [55,103]. Accordingly, scientists have explained that increasing the substrate bias increases the collision frequency of the ions, MPs, and electrons in the plasma. Since electrons have much greater velocity than ions, the MPs take up more electrons than ions at any given time, therefore resulting in negatively charged MPs [104]. Upon arriving at the negatively biased substrate, they are subsequently

repelled. The repulsive effect is proportional to the applied substrate bias [102,105]. Nonetheless, a common challenge with this technique is the likelihood of increased coating residual stresses and re-sputtering of coating atoms due to energetic substrate bombardment [55]. Figure 3-11 depicts the reduction in MPs with an increasing substrate bias.

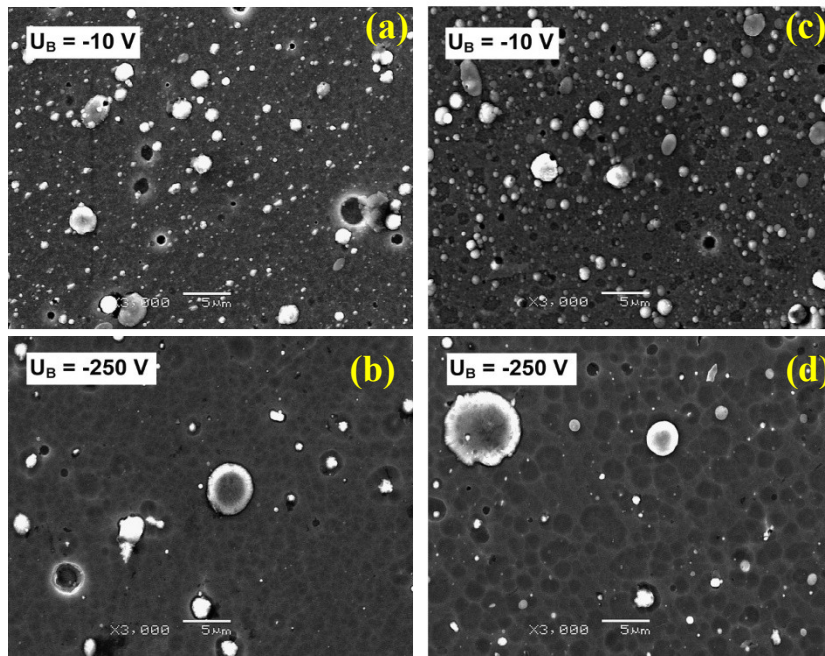


Figure 3-11: Effect of substrate bias (U_B) on MPs in HS-6-5-2 tool steel coated with: (a,b) CrN; (c,d) CrCN [55].

Also, researchers have highlighted the reduction in the arc current as another cost-efficient alternative for MP reduction [106]. This is because, at a lower arc current, there is a reduction in the temperature and energy at the cathode spot, leading to a reduction in the number of MPs emitted from the cathode. However, the concern with arc current reduction is its negative effect on the plasma density and the resulting coating properties. In this regard, a recent investigation conducted by Warcholinski et al. revealed that reducing the arc current

from 120 to 50 A during the deposition of AlCrN coatings led to a reduction in coating adhesion strength despite the reduction in MPs [56]. Figure 3-12 shows the reduction in MPs with a decrease in the arc current. Furthermore, researchers have proposed using a pulsed arc to achieve a greater reduction in MPs compared to the continuous DC arc supply. During pulsing, the high arc current intermittently supplied to the cathode is sufficient to initiate the ionization of coating species, but the short pulsing time prevents the formation of MPs [37]. Additionally, arc pulsing also results in a lower power consumption, more efficient consumption of target material, and a reduction in the cooling requirement [46,107].

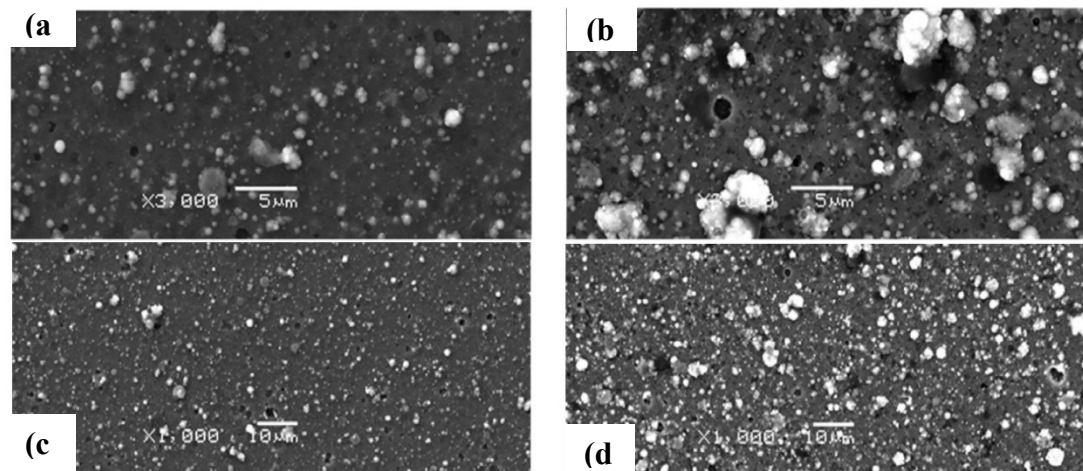


Figure 3-12: Effect of arc current on MPs in AlCrN coating deposited at an arc current of: (a,b) 50 A; (c,d) 120 A [106].

Additionally, increasing the flow rate or partial pressure of the reactive gas (usually nitrogen) has also been reported as an economically viable and feasible method for MP reduction [48,108]. In fact, it is perceived by some researchers to offer more significant MP reduction than filtering, arc current reduction, or increase in substrate bias [50,109]. In this method, a reduction in the number of MPs is achieved through a mechanism known as “target

poisoning”. This occurs if sufficient reactive gas is present during the deposition process, leading to the formation of a compound layer (usually the nitride of the cathode material) on the cathode surface, thus resulting in the poisoning of the cathode. In some cases, these compound layers have significantly higher melting points than the cathode material (e.g., melting temperature of Ti = 1668 °C and TiN = 2930 °C [47]), thus leading to a reduction in the evaporation of the cathode material and, consequently, the emitted MPs [51]. Wang et al. further explained that optimizing the cathode poisoning by feeding the reactive gases directly to the surface of the cathode provides additional reduction of the MPs emitted [110]. The major drawback associated with MP reduction by target poisoning is the accompanying reduction in the deposition rate. In reference to this, an investigation conducted by Harris et al. revealed that increasing the partial pressure of the reactive gas (nitrogen) from 0.1 to 1.2 Pa led to a 23% reduction in the coating thickness despite the 283% increment in the tool life [51]. Figure 3-13 shows the reduction in MPs with an increasing partial pressure of the reactive gas.

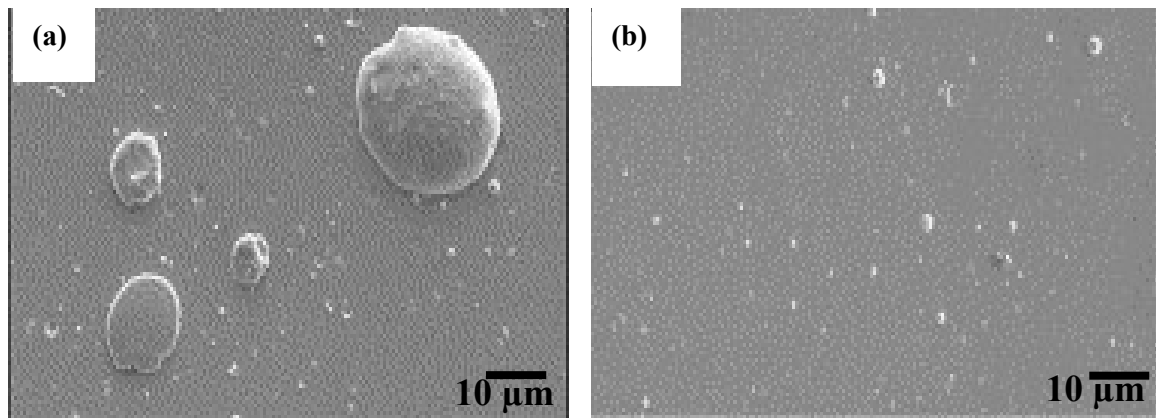


Figure 3-13: Effect of partial pressure of nitrogen on MPs of TiN coatings deposited: (a) 0.1 Pa; (b) 1.2 Pa [51].

3.4.4. Research advances in CAE-PVD microstructural characterization

Over the past years, extensive studies have documented the microstructural characterization of the properties of CAE-PVD coatings, which have a significant effect on their tribological performance [111]. Some of the properties reported by researchers include the MP statistics [102,105,112], coating composition [113–115], thickness of the coating [51,105,112], crystallite size [78,106,116], residual stresses [97,117], and coating density [52]. However, limited studies have documented the substrate–coating interfacial characterization despite the significant effect of the interfacial chemistry and the resulting composition on the tribological performance of the coatings [118,119]. A few of the studies reported are discussed in the succeeding paragraph.

Dejun and co-workers characterized the AlTiCrN-YT14 tungsten carbide interface using EDX analysis, which revealed coating–substrate elemental diffusion and an interfacial composition comprising coating and substrate elements [120]. In another study, Tian et al. investigated the effect of interfacial carbon composition on the adhesion strength of CrN coating deposited on AISI 52100 and SAE 1045 steels using a GDOES line scan. They observed that increasing the carbon content at the interface had a negative effect on the bond strength [121]. Further interfacial characterizations can be performed using transmission emission microscopic (TEM) techniques [122]. However, the main challenge with this technique is the sample preparation, as the sample must be thinned without introducing any deformation [35]. Investigations on the interfacial composition and microstructure of Cr films deposited on Zr were conducted by Ribis et al. using energy-filtered TEM (EFTEM) and electron backscatter diffraction (EBSD). They observed the formation of a 50–100 nm Zr (Fe, Cr)_2 intermetallic phase with equiaxed grains with complex hexagonal and cubic

structures. However, the equiaxed interface did not affect the columnar growth and crystallinity of the deposited coatings [123]. Similar observations were reported by Brachet et al. [124]. Figure 3-14a depicts a typical coating–substrate interface, while Figure 3-14b shows the equiaxed grains at the interface. Nonetheless, the interfacial composition could also enhance the properties of the deposited coatings. The outcome of the investigations conducted by Pang et al. on Cr_2O_3 -coated low-carbon steel interface using high-resolution TEM (HRTEM) revealed that the amorphous Fe-Cr intermetallic phases improved the corrosion resistance and conferred additional strengthening by inhibiting columnar grain growth and preventing dislocation movement [125]. From the foregoing, it appears that the evolution of the interfacial composition and the morphology, particularly with CAE-PVD process parameters and the corresponding effect on the tribological performance is still not very clear. This warrants further studies.

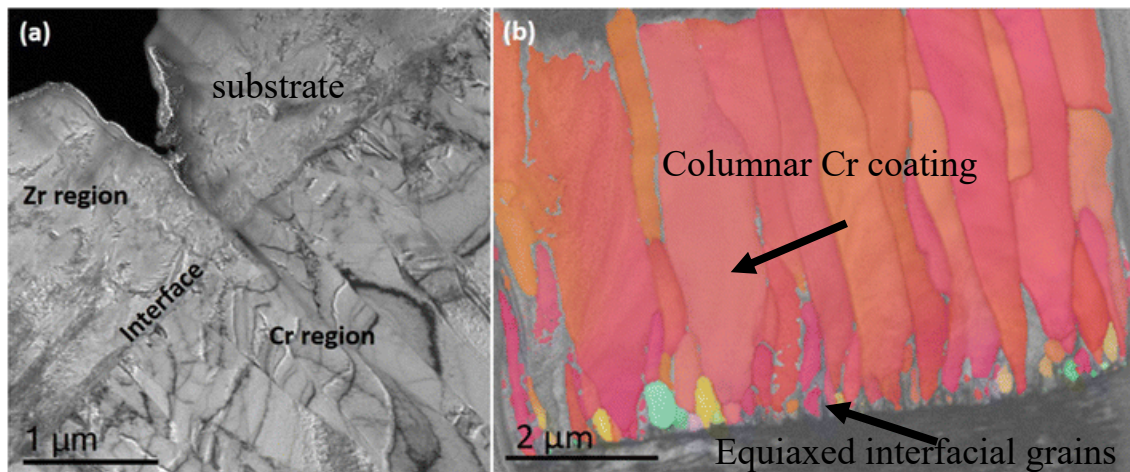


Figure 3-14: (a) TEM micrograph showing the Cr-Zr interface; (b) EBSD micrograph showing equiaxed interfacial grains and the columnar coating morphology [123].

In summary, it is apparent that the detrimental effect of MPs and the accompanying craters in the CAE-PVD coatings pose a significant threat to their wide-spread commercialization for tribological application. Among the outcomes are inhomogeneity in the coating composition, increased surface roughness, facilitating crack nucleation, and promoting tribochemical reactions. Although researchers have also highlighted the potential benefits of MPs, such as strengthening, lubricating properties, and increasing thermal stability, these positive impacts require experimental validation. Furthermore, attempts made by researchers to minimize MPs include filtering and optimizing the deposition parameters, such as the substrate bias, the arc current, and the partial pressure of the reactive gas. However, despite the cost-intensive nature of the filter systems, they are often preferred in practice compared to the other techniques due to the greater MP reduction efficiency. Additionally, further studies need to be conducted to unravel the relationship between interfacial composition, process parameters, and the tribological performance of the coatings.

3.5 Characterization of mechanical and tribological-related properties of CAE-PVD coating

This section discusses the research advances in the characterization of the mechanical properties, surface roughness, wear, and adhesion of CAE-PVD coatings.

3.5.1. Hardness (H), plasticity index (H/E), and resistance to plastic deformation (H^3/E^2) and roughness

Conventionally, the H of CAE-PVD coatings is the primary indicator of their tribological performance [1,63]. Two main techniques for CAE-PVD coating hardness

measurement reported in the literature are Vickers microhardness testing and nanoindentation [117,126]. The major challenge with Vickers microhardness measurement is the likelihood of the indentation depth exceeding the standard 10% of the coating thickness [127]. Consequently, the obtained hardness measurement might not be the accurate hardness of the deposited coatings. On the other hand, apart from the more accurate hardness measurement obtained from nanoindentation, it provides additional information about the coating's reduced elastic modulus (E). Nonetheless, despite the use of nanoindentation, the common challenge encountered is the difficulty of obtaining repeatable results due to the large amounts of MPs and craters inherently present on the surface of the coatings. In the past, some authors have reported that polishing the surface of the coatings using diamond suspension (1–3 μm) significantly improved the repeatability of the measurements [71]. However, most studies do not provide information regarding the polishing time, thus making it a qualitative approach. Moreover, there are two main questions that require further clarification. First, is the effect of the polishing on the coating thickness, and second is the effect of the thickness reduction on the resulting hardness considering the hardness variation with the coating thickness [128,129].

In recent times, it has become common knowledge that the hardness of CAE-PVD coatings is not sufficient for assessing their tribological performance [1]. Studies conducted by several researchers revealed that the coating's tribological performance can be better characterized using a combination of H and E [130,131]. Two major parameters were developed: H/E and H^3/E^2 . The former is known as the plasticity index and provides a measure of the elastic behaviour of the coatings, while the latter is a measure of the coating's resistance to plastic deformation [106]. Like the hardness, these ratios positively correlate

with the coating's tribological performance [128]. However, some researchers have expressed a contrasting opinion as they observed improved tribological performances for coatings with lower H/E and H^3/E^2 [132]. Accordingly, they explained that the tribological behaviour of the coatings is very complex and uncertain. Consequently, they concluded that the existing indicators (H , H/E , and H^3/E^2) are not sufficient for a complete assessment of the coatings' tribological performance. In addition, they recommended the analysis of the wear track and the debris alongside the above parameters so as to have a comprehensive understanding of the coating's performance [56,63]. More so, the coating's composition could also be another contributing factor to the observed discrepancy in the coating's tribological performance.

The roughness of the deposited coatings also affects the coating's tribological performance. Coatings with a higher roughness typically exhibit a reduced tribological performance, as the rougher surfaces have higher contact pressures due to the lower contact areas [70]. Ideally, the deposited coatings are expected to exhibit a similar roughness as the profiled substrate. However, because of the presence of MPs, the accompanying craters, and the variation in the coating morphology with the process parameters, the resulting roughness is mostly higher than that of the profiled substrate [34,39]. The coating roughness can be characterized using profilometers [55], surface texture meters [63], or atomic force microscopy (AFM) [8]. The two most widely used parameters for surface roughness assessment are the average surface roughness (R_a) and the peak-to-valley height (R_z), as illustrated in Figure 3-15. In spite of the wide adoption of these parameters, some researchers have argued that they fall short in portraying the surface defects present in the coatings [133].

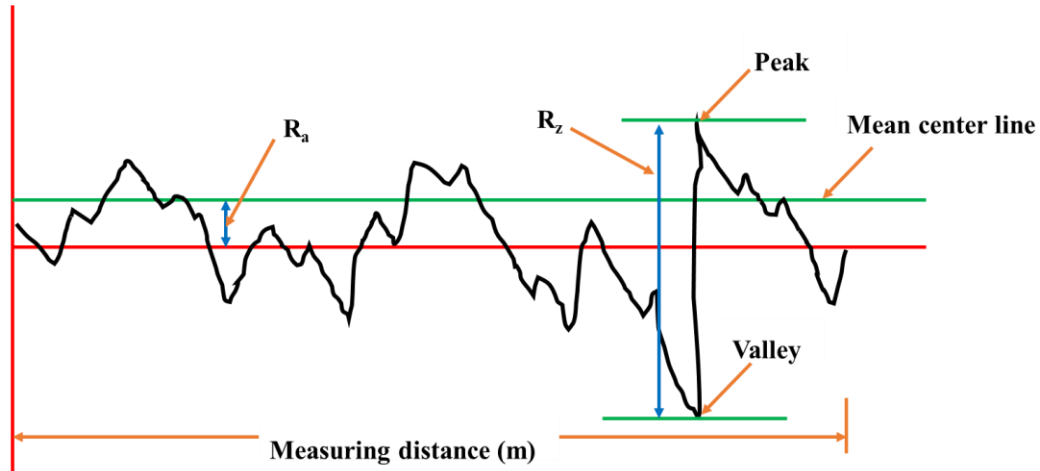


Figure 3-15: A surface profile showing the average surface roughness (R_a) and the peak-to-valley height (R_z).

3.5.2 Wear characterization

Wear characterization is considered the most important parameter for tribological performance assessment [1]. Different methods exist for characterizing this parameter. Nonetheless, the method adopted hinges on the desired tribological application. The objective is to select the wear testing method that best replicates the actual service conditions of the coatings. However, most wear assessment tests developed over the years are based on abrasion and sliding. One of the most popular sliding tests is the pin-on-disk (POD) [134], as depicted in Figure 3-16a. Meanwhile, two widely used abrasion tests are the Taber [135,136] and dry sand rubber wheel (DSRW) [137] tests, as illustrated in Figure 3-16b, c. Nevertheless, irrespective of the wear testing method adopted, better insight into the evolution of the coating degradation could be obtained by examining the coated samples at regular intervals during the test. This approach is exemplified in the studies conducted by

Scholl to understand the evolution of the wear behaviour of TiN-coated high-speed steel using the DSRW test. In their studies, the samples were removed at intervals of 4 to 10 revolutions and, subsequently, re-weighed and re-examined, thereby providing a deeper understanding of the progression of the coating deterioration [138].

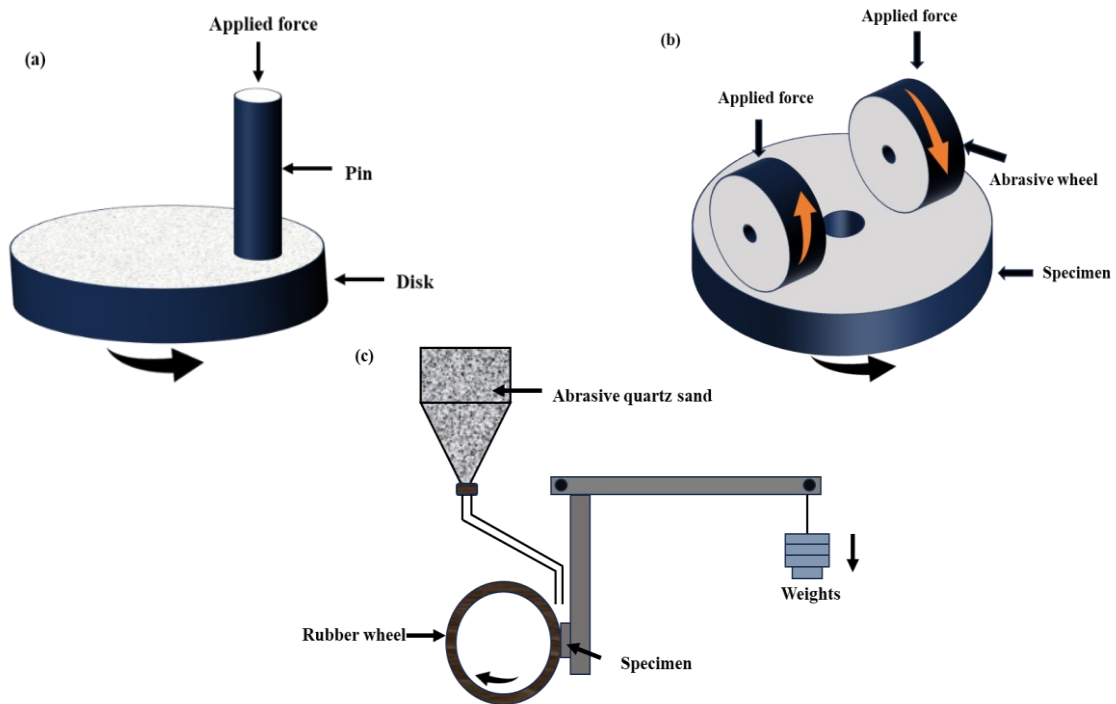


Figure 3-16: Illustrations: (a) pin-on-disk (POD) test; (b) Taber abrasive test; (c) dry sand rubber wheel (DSRW) test.

Furthermore, in both the abrasive and sliding tests, the wear track and wear debris characterization also provide additional information to further understand the prevalent wear mechanisms. Information relating to wear track dimensions, such as the wear track width, can be obtained using image processing software such as image J (Version 1.54h) [74]. Profilometry could be used to evaluate the depth of the wear track [139], while wear debris can be analyzed using scanning electron microscopy and energy-dispersive X-ray analysis

[43]. Over time, some researchers have shown a preference for sliding tests, because they also provide a measure of the coating's COF in addition to the wear rate [63]. However, on many occasions, researchers observed that coatings with a low COF exhibit a contrasting inferior wear performance. This led to the conclusion that the COF might not be a good indicator of the tribological performance of the coatings [19,32,140].

3.5.3 Coating adhesion

The adhesion strength of the coating also influences the coating's tribological performance as it significantly affects the coating functionality. Explicitly, a low coating adhesion strength can result in premature coating failure, thus preventing the effective utilization of the mechanical and tribological properties of the coatings [30,31]. The coating adhesion strength can be characterized using qualitative and quantitative techniques. The most popular qualitative adhesion test is the Daimler–Benz indentation test (also known as the VDI 3198 test) [39,141]. In this test, the adhesion is assessed based on the coating damage compared with the VDI 3198 standard when indented by a Rockwell indenter. According to the standard, coatings exhibiting a failure mode ranked HF1 to HF4 are considered acceptable failures, while HF5 and HF6 failure modes are characterized by poor adhesion strength, as shown in Figure 3-17a [142]. Several authors have reported the failure modes of HF1 to HF3 for different CAE-PVD coatings [142].

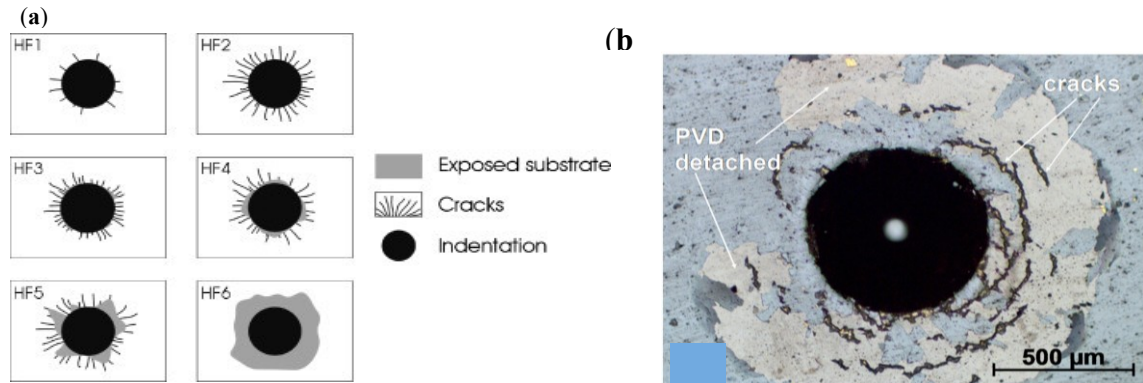


Figure 3-17: (a) VDI 3198 standard for classification of coating adhesion strength [143]; (b) circumferential cracks in TiAlN-coated waspaloy [144].

However, investigations by some researchers have shown the occurrence of circumferential cracks contrary to the conventional radial cracks proposed by the VDI 3198 standard, as shown in Figure 3-17b. According to Biava et al., such an occurrence may not result from poor adhesion but rather because of the higher plasticity of the substrate relative to the coating [144]. Meanwhile, Warcholinski et al. attributed the occurrence to be a characteristic of hard and brittle coatings [56,142]. The limitation of this test is that it relies solely on crack propagation to the surface for assessing the coating adhesion strength. Sometimes, coating debonding can occur without the emergence of surface cracks. Hence, such failures cannot be revealed by this test. Again, the result of the assessment is relative depending on the assessor; thus, this might result in inconsistencies in the reported results. Nonetheless, this test has widespread acceptance in the industry because it is an easy, rapid and convenient qualitative indication of coating bond strength [31].

Quantitative techniques for assessing coating bond strength include the pull-off test and scratch test. The former relies on the formation of a bond between the coated surface and a loading fixture using an epoxy resin or other adhesives. Subsequently, the adhesion

strength is indicated by the force required to pull-off the fixture from the coated surface [145]. However, this test is sparsely reported for the characterization of PVD coatings, as the maximum adhesion strength that can be measured is limited by the bond strength between the fixture and the adhesive [31]. In fact, investigations conducted by Ang et al. on the assessment of the bond strength of TiN and CrN coatings deposited on SiC using epoxy adhesives revealed inconclusive results due to the adhesive failure at the fixture–adhesive interface [146]. Therefore, the pull-off test might not be suitable for characterizing the adhesion strength of CAE-PVD coatings. However, if adhesive–fixtures with super bond strength exceeding the coating bond strength are developed, this test may be considered.

On the other hand, in the scratch test (sometimes called the stylus method), the adhesion strength is evaluated by the load required to results in coating failure when an indenter is pulled across the coating at a constant speed. The load at which coating failure occurs (called the critical load (L_c)) provides the adhesion strength of the coating [31,69]. The standard provides the allowance for adopting multiple critical loads associated with reproducible, specific and defined coating damage [147]. However, two main types of critical loads (L_{c1} and L_{c2}) have been adopted by researchers. L_{c1} represents the critical load at which cohesive coating failure occurs, and it is usually characterized by the appearance of the first crack, while L_{c2} represents the critical load at adhesive failure, i.e., complete delamination of the coating from the substrate. These failures are shown in Figure 3-18 [142]. Authors have reported L_{c1} and L_{c2} of 15–40 N and 70–110 N, respectively, for different CAE-PVD coatings [56,148]. Furthermore, approximately a decade ago, Zhang et al. [149] proposed the crack propagation resistance ($CPR = L_{c1} (L_{c2} - L_{c1})$) as a parameter for the quick

qualitative assessment of coating toughness. Since then, it has been widely adopted in the characterization of the fracture toughness of PVD coatings [56].

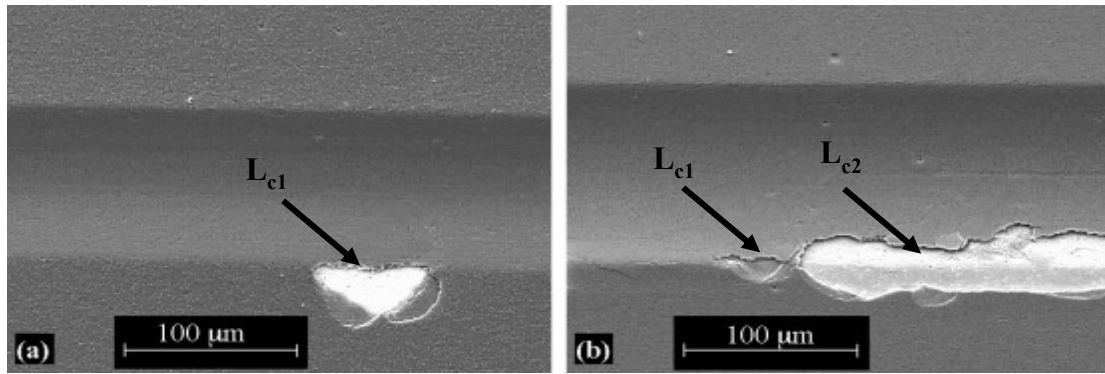


Figure 3-18: Micrographs showing TiN-coated high-speed tool steel coating damage corresponding to (a) L_{c1} and (b) L_{c2} [143].

To sum up, characterizing the mechanical and tribological properties of CAE-PVD coatings is essential for optimizing their tribological performance, as they provide valuable insight for developing more wear-resistant coatings. Significant advances have been made in the characterization of the mechanical properties, surface roughness, wear and adhesion of CAE-PVD coatings. Regarding hardness measurements, nanoindentation is preferred to Vickers microhardness, as it provides a more accurate measurement and E of the coatings. Surface roughness can be characterized using profilometers, AFM or surface texture meters. Furthermore, some popular wear characterization techniques include the POD, DSRW and Taber abrasive tests. The preference for POD over the others is because it provides additional information about the coating's COF. Additionally, the adhesion strength of the coatings can be characterized qualitatively using the VDI 3198 test and quantitatively using scratch tests. In general, because of the complexity of coating's tribological behaviour, it is very important

to conduct a wholistic assessment of the aforementioned characterizations alongside the wear track analysis so as to have a comprehensive understanding of the phenomenon.

3.6 Electrochemical properties of CAE-PVD coatings

The electrochemical properties of CAE-PVD coatings also play a significant role in their tribological performance, especially in applications involving presence of corrosive environment such as in green wood machining. During this process, the presence of organic acids (e.g., tannic acids) resulting in acidic pH levels (usually between 4 and 5.5) coupled with the silica contaminants cumulatively culminating in tribochemical reactions, thus facilitating coating degradation [1,150]. The coating electrochemical degradation process is also accelerated by high temperature [144,151]. The electrochemical properties of CAE-PVD coatings are mostly characterized using potentiodynamic polarization [152]. Predictably, the coating composition and morphology play a significant role in their corrosion resistance. Most importantly, researchers have observed that columnar morphology coating and craters are undesirable for corrosion resistance [73,153] This is because these microstructural features facilitate the penetration of the corrosive wood electrolyte to the substrate, resulting in pitting [154,155]. The following paragraphs present significant findings regarding the electrochemical behaviour of CAE-PVD coatings under different conditions.

Bilgin et al. [116] investigated the corrosion behaviour of monolayered TiN, TiAlCN, AlCrN, AlTiN and multilayered AlN-TiN deposited on tungsten carbide in neutral (sodium chloride solution, pH 7), acidic (sulfuric acid solution, pH 2) and (ammonium hydroxide solution, pH 10) electrolytes. Their findings revealed that in the neutral electrolyte, the corrosion resistance of the coatings was in the order of $\text{TiAlCN} > \text{AlN/TiN} > \text{AlTiN} > \text{AlCrN}$

> TiN. Meanwhile, under acidic and alkaline conditions, the corrosion resistance was in the order of AlCrN > AlN/TiN > TiN > AlTiN > TiAlCN and AlCrN > AlN/TiN > AlTiN > TiAlCN > TiN, respectively. The authors further highlighted the superior stability of the chromium oxide layers over other passive oxide layers, which contributed to the excellent performance of the coatings under acidic and alkaline conditions. A notable observation from this study is the consistent performance of the AlN-TiN multilayer coatings under all conditions. This is noteworthy because the pH of wood species fluctuates over a wide range from acidic, neutral to alkaline depending on the effect of climatic conditions [150]. Therefore, to ensure consistent electrochemical performance, the multilayer architecture might be a more suitable selection.

In another study, Fazel et al. [141] compared the electrochemical behaviour of CrN/TiN multilayer and TiN monolayer coatings deposited on Ti-6Al-4 V alloy in Hank's solution. As expected, they reported that the multilayer CrN/TiN coating exhibited superior performance. Three factors accounted for the observation. Firstly, the monolayer coating was characterized by higher crater density, thus facilitating the electrolyte transfer to the substrate, resulting in coating degradation. Secondly, the presence of more coating layers in the CrN/TiN inhibited the transfer of the corrosive electrolyte, increased the time taken for the electrolyte to reach the substrate and, consequently, resulted in improved corrosion resistance. Thirdly, the formation of stable chromium oxides on the coating surface also inhibited the electrochemical degradation of the coating [141]. In a separate study, Biava et al. [144] investigated the high-temperature (700 °C) electrochemical performance of CrN, TiAlN and AlCrN coatings deposited on waspaloy in an aqueous solution of sodium sulfate and sodium chloride. Their findings revealed that the corrosion resistance of the coatings

was in the order of $\text{AlCrN} > \text{TiAlN} > \text{CrN}$. The superior performance of the TiAlN coating to CrN was attributed to the formation of passive Al_2O_3 and TiO_2 , while the more stable Cr_2O_3 coupled with the Al_2O_3 resulted in the superior performance of the AlCrN relative to the TiAlN. The findings of this study further emphasized the important role of Cr in the electrochemical performance of CAE-PVD coatings, even at high temperatures typical of those encountered in wood machining, as previously pointed out by Bilgin et al. [116].

Researchers have also reported the effect of dopants and nitrogen reactive gas on the electrochemical behaviour of CAE-PVD coatings. In this regard, Wu et al. [43] investigated the effect of carbon (10–50 sccm of C_2H_2) doping on the corrosion behaviour of TiAlCrN coatings in sulfuric acid solution electrolyte. Their findings revealed that increasing the dopant composition reduced the coatings' corrosion resistance. However, compared to the undoped TiAlCrN coating, the improvement in the corrosion resistance was not significant. They attributed the observation to the dominant effect of the existing stable metallic nitride phases (TiN, AlN and CrN) in the coating and that the introduction of C did not destabilize these phases coupled with the insignificant corrosion resistance of the newly formed CN phase. In another study, Warcholinski et al. [142] investigated the effect of silicon doping (0 at. %–4.4 at. %) on the properties of AlCrN coating in sodium chloride electrolyte. They observed a positive correlation between the dopant composition and the corrosion resistance of the coatings. The authors attributed this observation to the resulting dense coating structure preventing electrolyte transport to the substrate coupled with the formation of a passive SiO_2 on the coating surface. Meanwhile, another study conducted by Warcholinski et al. [48] on the effect of nitrogen pressure (3–5 Pa) on the electrochemical behaviour of

AlCrN coatings in sodium chloride solution electrolyte revealed that increasing the pressure of nitrogen did not have any significant effect on the corrosion behaviour of the coatings.

Nonetheless, for some machining applications characterized by the occurrence of wear and tribochemical reactions, improved perspectives of the electrochemical performance of the coatings could be obtained through tribocorrosion testing [156]. By conducting such test, it would be possible to study the interplay between mechanical and chemical mechanisms responsible for coating degradation and could provide better insight into the expected service life and performance of the coating compared to assessments using wear or corrosion tests separately. However, such characterizations are sparsely reported in the literature for CAE-PVD coatings.

From the foregoing, it is evident that a coating's defects, composition and morphology play a significant role in its electrochemical performance. Increasing the crater density and a less dense coating morphology correlate with poor corrosion resistance. Some authors have also highlighted the vital role of passive chromium oxides in corrosion resistance improvement, especially in monolayer-coated substrates. The multilayer configuration exhibited the most consistent electrochemical behaviour under all conditions. Additionally, the effect of coating doping on the electrochemical resistance might differ depending on the coating composition, dopant concentration and the type of dopant.

3.7 Effect of deposition parameters on the properties of CAE-PVD coatings

The deposition parameters, mainly arc current, temperature and substrate bias, play a significant role in the deposited coatings' properties and resulting performance [46]. These

parameters affect the morphology, adhesion strength and mechanical properties of the coatings [68,123]. The precise and optimal control of these parameters not only ensures a high coating quality but also results in an improvement in the energy efficiency during coating deposition, especially for industrial-scale production [36]. The succeeding sections highlight the effects of the arc current, deposition temperature and substrate bias on the properties of CAE-PVD coatings.

3.7.1 Effect of arc current

The arc current primarily determines the rate of evaporation of the coating species from the cathode [27]. Table 3-2 presents significant findings on the effect of arc current on the properties of CAE-PVD coatings and other relevant information related to the substrates, other deposition parameters and the coatings.

Table 3-2. Summary of the effect of the arc current on the properties of CAE-PVD coatings.

Arc Current (A)	Temperature, Substrate Bias	Substrate	Coating	Thickness (μm)	Significant Findings	Ref.
50–90	400 °C, –70 V	Stainless steel	Cr	1.8	1—No effect on coating texture. 2—Increase in the deposition rate with the arc current.	[100]
0–100	500 °C, –100 V	High-speed steel	CrN and AlN	3.1–6.6	1—A positive correlation exists between the arc current and the coating's metallic composition.	[157]
75–175	300 °C, –150 V	M2 high-speed steel	TiAlN	1	1—No significant effect on the coating's hardness and residual stress.	[97]

					2—A reduction in the adhesion strength with an increasing arc current. 3—An increase in the deposition rate and aluminium concentration with the arc current.
50–100	350 °C, –100 V	Not provided	AlCrN	3–4	1—No significant effect on the coating texture, crystallite size, composition, H, H/E and H^3/E^2. 2—An increase in the coating defects, surface roughness and deposition rate with the arc current. [106] 3—A reduction in the COF with an increasing arc current. 4—A fluctuation in the adhesion strength and wear rate with the arc current, reaching an optimum at 80 A.
50–120	350 °C, –100 V	4H13 and HS6-5-2 steel	AlCrN	Not provided	1—An increase in the crystallite size, deposition rate, surface roughness and residual stresses with the arc current. 2—The H, H/E and H^3/E^2 decreased with an increasing arc current. [56] 3—The adhesion strength and wear rate followed a similar trend, reaching an optimum at 80 A.

Further critical analysis of the literature presented in Table 3-2 revealed the following: There are two main divergent opinions on the effect of the arc current on the hardness and residual stresses of the coatings. The first point of view is that increasing the arc current does not significantly affect the hardness and residual stresses [97]. In support of this argument, the authors explained that increasing the arc current does not necessarily connote an increase in the energy of the evaporated species [158]. In fact, Harris and Lafferty [159] reported that the ion current is only 8%–12% of the applied arc current. Hence, the increase in energy of the ion resulting from the increase in arc current is not sufficient to result in significant changes in coating compressive residual stresses. The static behaviour of the hardness is connected to the insignificant changes in crystallite size, which is an outcome of the negligible variation in plasma energy with increasing arc current [97]. Gilewicz et al. [106] attributed this observation to the dominating effect of increased coating defects (producing less dense and compact coatings) over the reduction of the crystallite size due to the coating bombardment. The second point of view is the increase in residual stresses and decrease in hardness with an increasing arc current. Contrary to the previously presented arguments, they attributed the increase in the residual stresses with the arc current to the increase in the energetic bombardment of the coating by the evaporated species. More so, despite the increment in the residual stresses, the reduction in the hardness was attributed to the dominating effect of the temperature rise leading to crystallite growth (an aftermath of coating bombardment) coupled with the reduction in coating density due to the increased craters and MPs [56].

Similarly, three divergent views on the effect of the arc current on the adhesion strength and wear resistance were deduced. From the first standpoint, there is a reduction in

the adhesion strength and wear resistance with an increased arc current. The authors attributed this observation to the increase in the thermal component of the residual stresses with an increasing arc current, leading to accelerated coating degradation [97]. The second viewpoint is the increase in adhesion strength and wear resistance with an increasing arc current. The authors attributed this observation to the shallow implantation of the coating species on the substrate, leading to the formation of adherent layers coupled with a reduced COF [106]. The last point of view is the fluctuations (increasing and later decreasing) in the adhesion strength and wear resistance with an increasing arc current. In this regard, the authors linked this observation to the fluctuation in the H/E and H^3/E^2 with an increasing arc current [56]. An attempt to resolve the conflicting observations on the effect of the arc current on the properties and performances of the coatings revealed that the substrate bias, target composition and domain of arc current investigated in the studies were different. This could justify the discrepancy in the observations [56,97,106].

In summary, it appears that very limited studies have been conducted on the effect of the arc current on the properties of CAE-PVD coatings. Generally, increasing the arc current is mostly accompanied by an increase in the deposition rate, coating defects, surface roughness, and no significant changes in the structure and texture of the coatings. Regarding the composition, some authors observed an increase in the coating's metallic composition with the arc current, while others considered it insignificant. The conflicting reports of the effect of the arc current on the residual stresses, hardness, adhesion strength and wear resistance might be attributed to the differences in the coating elemental composition, cathode material and substrate bias used by the different authors. Furthermore, an overall

assessment of the literature in this study hints that an arc current in the range of 75–100 A is sufficient for an optimal bond strength and tribological performance.

3.7.2. Effect of deposition temperature

The deposition temperature has a significant effect on the kinetic energy of the evaporated species (i.e., diffusion rate) and the activation energy of reactions leading to coating formation (phase transformation) [68]. Consequently, variations in this parameter could have a significant effect on the coating properties and tribological performance [112]. Table 3-3 presents significant findings on the effect of deposition temperature on the properties of CAE-PVD coatings and other relevant information related to the substrates, other deposition parameters and the coatings.

Table 3-3: Summary of the effect of the deposition temperature on the properties of CAE-PVD coatings.

Temperature (°C)	Arc Current, Substrate Bias	Substrate	Coating	Thickness (μm)	Significant Findings	Ref.
250–450	80 A, –100 V	WC-Co	AlCrN	1.24–2.78	1—Reductions in the crystallite size, compressive residual stresses and wear rate with an increasing temperature. 2—An increase in the hardness with the deposition temperature.	[68]
250–500	80 A, –70 V	Stainless steel	Cr	3	A change in the coating texture from (110) to (200).	[100]
350–450	80 A, –100 V	HS6-5-2 steel	AlCrN	3.9–6	1—Reductions in the hardness, deposition rate and adhesion	[48]

					strength with an increasing temperature.
					2—An increase in the crystallite size with the temperature.
150–450	100 A, bias not provided	HSS	TiN	6.3	Increases in the COF and surface roughness with the deposition temperature. [133]
250–500	80 A, –100 V	Stainless steel	Cr	2.35–4.35	1—The crystallite size increased with temperatures up to 400 °C and remained constant afterward. [98] 2—The hardness, wear resistance, and lattice perfection increased with the temperature.

The following important points are noteworthy following a detailed analysis of the literature presented in Table 3-3. Firstly, there is an unexpected change in the texture of the coatings to a plane of lower reticular density at higher temperatures [100]. Ideally, the growth of planes with higher reticular densities are promoted at high temperatures [160]. However, the authors attributed the occurrence to chamber impurities, which were activated at high temperatures, incorporated into the film and hindered the growth of the preferred planes [100]. Another critical issue is the effect of increasing deposition temperature on the hardness, crystallite size and tribological performance of the coatings. Here, three main divergent opinions were deduced. Firstly, is the expected increase in crystallite size accompanied by an unexpected increase in hardness and wear resistance. In support of this, the authors explained that increasing the mobility of the adatoms at high temperatures

facilitated the growth of the crystallite. Despite the crystallite growth, the increase in coating hardness was attributed to the enhanced texture. Meanwhile, the cumulative increase in the hardness and enhanced texture led to improved wear resistance [98]. The second viewpoint is the unexpected reduction in crystallite size accompanied by a reduction in the hardness and wear rate. Contrary to the previous explanations, the authors attributed the reduction in crystallite size at higher temperatures to the increased coating bombardment by the energetic coating species. Meanwhile, the observed increment in hardness was attributed to the reduced crystallite sizes and reduction in detrimental intermetallic phases. Cumulatively, the increased hardness and adhesion strength resulted in improved wear resistance [68]. The third viewpoint is the reduction in crystallite size, hardness, and wear resistance with increasing deposition temperature. Contrary to previous accounts, the authors attributed the crystallite growth at higher deposition temperature to the reduction in the deposition rate, resulting in a reduction in coating bombardment and the subsequent migration of the crystallite boundaries. The authors further explained the surprising reduction in deposition rate to target poisoning and re-sputtering. Target poisoning results when reactions leading to the formation of nitrides on the target surface are promoted, thus hindering the evaporation rate of coating species. Again, at high temperatures, the incident adatoms have greater mobility, thus increasing their probability of re-sputtering. The reductions in hardness and adhesion strength contributed to the reduction in wear resistance [48].

In summary, like the arc current, the effect of deposition temperature on the properties of CAE-PVD coatings is sparsely reported in the literature. Generally, coating deposition at higher temperatures is likely characterized by enhanced preferred texture and reduced incomplete phase transformation. However, there seem to be conflicting views on

the effect of the parameter on the crystallite size, hardness, and the resulting tribological properties. Consequently, further studies need to be conducted to better understand the effect of the parameter on the mechanical and tribological performance of CAE-PVD coatings.

3.7.3 Effect of substrate bias

The substrate bias influences the impact of the adatoms on the substrate. This parameter plays a vital role in the resulting morphology and properties of the coatings [46,92]. Table 3-4 presents significant findings on the effect of substrate bias on the properties of CAE-PVD coatings and other relevant information related to the substrates, other deposition parameters and the coatings.

Table 3-4: Summary of the effect of the substrate bias on the properties of CAE-PVD coatings.

Negative Bias (V)	Arc Current, Temperature	Substrate	Coating	Thickness (μm)	Significant Findings	Ref.
0–100	80 A, 400 °C	Stainless steel	Cr	1.4	1—Increase in the crystallite size and enhancement of (110) texture with the increasing bias. 2—Reduction in MPs with increasing bias.	[100]
0–300	80 A, 350 °C	HS6-5-2 steel	AlCrN	4.5	1—Increases in the crystallite size and hardness with bias. 2—Reductions in the deposition rate and surface roughness with an increasing bias.	[48]

50–400	80 A, 400–420 °C	Hardened steel	CrAlN	1.9–3.6	1—Reductions in the wear rate, crystallite size, residual stresses and deposition rate with an increasing bias.	[92]
					2—Increases in the Cr/Al ratio and hardness with bias.	
					3—A change in texture from (100) to (220) and a constant N composition with increasing bias.	
10–300	80 A, 300 °C	HS6-5-2 steel	CrN and CrCN	2.2	1—Fluctuations in the hardness and residual stresses with bias (an increase up to 150 V followed by a decrease).	[86]
					2—A reduction in substrate roughness and no effect on the Cr/N ratio with an increasing bias.	
					3—The adhesion strength of CrCN was unaffected while that of CrN reduced with an increasing bias.	
					4—Transformation of coating morphology from fine-grained to a polycrystalline dense structure with increasing bias.	
50–250	75 A, 300 °C	M2 High-speed steel	TiAlN	1	1—Increase in residual stresses, hardness, and enhanced coating texture (111) with increasing bias.	[97]
					2—Increase in adhesion strength with	

						bias up to 150 V followed by a decrease. 3—Reduction in Al content with an increasing bias.
20–400	135 A, 200–420 °C	High-speed steel	CrN	8.6–9.7		1—A positive correlation between the substrate temperature and bias. 2—Fluctuation in residual stresses with the bias (an increase up to 100 V, followed by a decrease) with an increasing bias. [161] 3—No changes in the coating's composition. 4—Hexagonal Cr₂N phases were formed at 400 V.
70–300	80 A, 300 °C	HS6-5-2 steel	CrN	9		1—Hardness, surface roughness, and defects were reduced with an increasing bias. 2—Deposition rate and coating composition remained fairly constant. [52] 3—Crystallite size and density were positively correlated with the bias. 4—Hexagonal Cr₂N phase was formed at a higher bias.
20–180	280–320 °C, Arc current not provided.	SK H9 steel	AlTiN/CrTiSiN	1.6–1.7		1—Hardness decreased with an increasing bias. [162]

	2—Residual stresses increased with the bias.
	3—A less columnar morphology was observed with an increasing bias.
	4—The coating thickness and composition were not affected by changes in the bias.

A critical analysis of the literature presented in the table revealed the following. There are discrepancies in the effect of the substrate bias on the deposition rate, hardness, surface roughness, residual stresses, and the overall tribological performance of the coatings. Firstly, the deposition rate could either decrease or remain constant with an increasing substrate bias. The former could be attributed to the re-sputtering of weakly bound adatoms due to the energetic ion bombardment at high bias [48]. Meanwhile, the constant deposition rate could be attributed to the lower tendency of re-sputtering due to the relatively lower substrate bias or a relatively high arc current coupled with the difference in coating composition contributing to an increase in cation density, which counteracts the effect of target poisoning and re-sputtering [48,50,162]. Concerning the hardness, it was observed to reduce, increase or fluctuate with an increasing bias. The reduction in the hardness with an increasing substrate bias could be attributed to the crystallite growth facilitated by the rise in the coating temperature due to the increase in the energetic ion bombardment [48,100,162]. Alternatively, it could be attributed to the formation of intermetallic phases possessing relatively lower hardness [52]. Meanwhile, the increase in the hardness was correlated with the change in the coating composition and texture as an aftermath of the re-

sputtering at high bias [92]. The following explanations were offered regarding hardness fluctuation (increase followed by a decrease) with bias. The increase in hardness with the bias was attributed to the less columnar and dense coatings, while the subsequent reduction was attributed to the formation of unstable intermetallic phases coupled with stress relaxation following saturation due to excessive bombardment [86,162,163].

Concerning residual stresses, two dominant views were reported. Firstly, is the increase in residual stress with bias, which was linked to the increase in the rate of impinging coating atoms [100,162]. Secondly, is the fluctuation (an increase followed by a decrease) in the residual stresses with the bias. The increase in the residual stresses with the bias and the subsequent reduction was due to the energetic coating bombardment followed by stress relaxation, respectively [86,97,163]. Regarding the surface roughness of the coatings, it was observed to either reduce or fluctuate with an increasing bias. The former was linked to coating densification and MPs' reduction [48,100]. The fluctuation in the surface roughness with the substrate bias characterized by a reduction and a subsequent increment could be attributed to the distortion in coating morphology by excessive ion bombardment following coating densification [86]. Lastly, two main divergent views were reported for the parameter's effect on the coatings' tribological performance. The first view is the reduction in tribological performance with increasing bias due to a reduction in hardness, H/E , and H^3/E^2 [162]. The second view is the fluctuation in the tribological performance owing to fluctuations in properties like the residual stresses, surface roughness, and hardness [97,163].

Furthermore, it appears that the effect of the substrate bias on the coating properties seems to be complicated. However, the effect can be categorized based on the bias intensity as low, optimum, and high. At a low negative bias (usually < 100 V), the resulting coating is

mostly characterized by defects, columnar morphology, and low hardness, which are undesired for tribological applications. Meanwhile, for optimum negative bias (usually between 100 and 200 V), the resulting coatings were dense and less columnar with reduced defects and increased residual stresses and hardness. Meanwhile, at high substrate bias (typically > 250 V), there is a high likelihood of re-sputtering coupled with residual stress release, leading to a reduction in hardness. However, it is important to mention that the optimum substrate bias largely depends on the coating composition and other deposition parameters.

In summary, it appears that more studies have been documented on the effect of substrate bias as compared to the arc current or temperature. Nonetheless, the literature analysis in this study hints that the substrate bias and the temperature have the most significant effect on the coating properties and the resulting performance. Gautier and Machet [100] previously proposed a similar argument. However, further studies need to be conducted to ascertain which parameter has the most significant effect. Again, it was discovered that only separate studies of the effect of the individual parameters have been mostly documented. Further studies can be conducted to optimize these parameters and also understand the interactions among these parameters, as well as the resulting effects on the coating properties and service performance.

3.8 CAE-PVD coating architecture

Over the years, the desire to improve the service performance of the existing CAE-PVD coatings birthed the significant efforts by researchers in developing different coating architectures. This section presents recent advances in the evolution of monolayer CAE-PVD coatings, coating doping and multilayer architectures.

3.8.1. Evolution of monolayer CAE-PVD coatings

Most monolayer coatings for tribological applications usually comprise Al, Ti or Cr, with N as the base element [12]. However, the earliest industrial PVD coatings developed were CrN and TiN [30,164]. While CrN showed superior thermal stability (approximately 100 °C higher) and corrosion resistance [157,165], TiN possesses higher hardness (approximately 3 GPa higher) [4,32]. Nonetheless, the insufficient tribological performances of these coatings led to the introduction of Al-based ternary PVD coatings, leading to AlCrN and AlTiN coatings [10,128]. This resulted in an increase in the hardness (up to 32 GPa), thermal stability (up to 800 °C), and tribological performance [114,142,166]. The improvement in the performance of these coatings was attributed to the formation of a passive aluminum oxide layer and solid solution strengthening by the cubic structure AlN phase [27,106]. However, it has been reported that when the concentration of Al exceeds 60%–70% (i.e., $\text{Al}_x(\text{Cr or Ti})_{1-x}\text{N}$, $x > 60\%–70\%$), there is a reduction in the tribological performance of these coatings [92,167]. This is due to the formation of the detrimental hexagonal AlN phase [48,168]. In fact, studies conducted by Benlatreche et al. [169] on the effect of 0 at. %–51 at. % Al on the wear resistance of AlCrN coatings revealed that the optimum tribological performance was obtained at 5 at. % Al. Additionally, an insufficient supply of the base nitrogen reactive gas during coating deposition could also result in the formation of the AlN phase. Nitrogen pressures in the range of 3–5 Pa have been reported by different researchers to result in optimal tribological performance of CAE-PVD coatings [48,106]. Nonetheless, in recent years, quaternary AlTiCrN coatings have been hinted as promising candidates for tribological applications and are the subject of contemporary research [33]. These coatings typically have relatively higher hardness (up to 36 GPa) and

thermal stability (up to 1100 °C) [4,170]. The improvement in the properties of the quaternary coating is attributed to the combined solid solution strengthening offered by TiN, AlN, and CrN phases coupled with the formation of dense and stable oxide layers of Al, Cr, and Ti [171]. Additional investigations revealed that 3 at. % Cr is required to optimize the thermal stability of AlTiCrN coatings [27]. However, further studies need to be conducted to optimize the coating composition, particularly for improved tribological performance.

3.8.2 Doping of coatings

In the past years, the need to further improve the quality of the existing coatings led to the introduction of other metallic (e.g., Ta, Y and V) and non-metallic (e.g., C, Si and B) dopants to the coating composition [172–174]. When the dopant is introduced to the coating matrix, it substitutes atoms from the matrix or occupies matrix interstices (depending on the dopant size relative to the size of the coating crystallite) to form solid solutions. This is usually accompanied by distortion of the coating lattice structure and increased residual stresses [32,170]. Meanwhile, the increase in nucleation sites by the incoming dopants results in the finer coating crystallite, thus providing additional strengthening per Hall–Petch principle [128,142]. However, excessive doping is characterized by superfluous nucleation, leading to the formation of amorphous phases, which are detrimental to the coating properties and the resulting service performance. The optimal dopant composition is governed by the maximum solubility of the dopant in the coating matrix, thus varies from coating to coating for a given dopant [142]. For instance, Kong et al. [32] reported 6.9 at. % of C as the optimal dopant for CrN/CrCN multilayer coating for maximum mechanical and tribological performance, while Wu et al. [43] reported 22.35 at. % of C for TiAlCrCN coating.

Lately, the use of carbon-doped CAE-PVD coatings for tribological application has received significant attention from researchers due to the strengthening properties of the dopant [32]. However, unlike other dopants, which are typically evaporated from the cathode, the carbon is mostly introduced into the deposition chamber as a reactive gas in the form of a hydrocarbon (mainly CH_4 and C_2H_2) [74,165]. This is because of the poor electrical conductivity and the high melting point of the nonmetal, leading to the evaporation of a significant portion of the metal as clusters (MPs). Furthermore, carbon, unlike other dopants, which are usually atomized, has a very high tendency to undergo sublimation coupled with its high melting point. This leads to the formation of carbon-containing species which act as nucleation sites for other incoming species thereby, facilitating their clustering to form MPs. Again, it is more energy-conservative and easier to control the coating composition by introducing the dopant as a reactive gas than cathodic evaporation [37,83]. Like other reactive gases, when the hydrocarbon gas is introduced into the chamber, it is ionized by the plasma, producing hydrogen and carbon ions. The relatively low momentum of the hydrogen ions due to their atomic mass makes them susceptible to easy displacement by other coating elements with higher momentum; hence, they mostly remain as gaseous residue in the deposition chamber and are subsequently evacuated via the exhaust system [35].

Furthermore, researchers have also recently attempted co-doping as a promising alternative for further improving the tribological properties of CAE-PVD coatings. In this process, two dopants are simultaneously introduced to the coating matrix [172]. This creates the possibility of combining the properties of different dopants, thus enhancing the performance of the deposited coatings. In this regard, a recent study by Mei et al. revealed that optimal 0.5 at. % C and 0.2 at. % B co-doping of AlTiN coatings improved the

mechanical and tribological properties compared to doping with C or B alone [175]. Also, in an effort to combine the strengthening properties of Ta and the anti-oxidative properties of Y, Aninat et al. reported that the co-doping of TiAlN with 2 at. % Ta and 1 at. % Y resulted in improved mechanical properties and thermal stability of the coatings [132]. Additionally, V and Si co-doping has also been explored by researchers to amalgamate the lubricative properties of V and the strengthening properties of Si. Studies reported by Chen et al. revealed that optimal 0.04 at. % V and 0.07 at. % Si doping of AlCrN coatings resulted in improved hardness and wear performance of the coatings [176].

3.8.3 Multilayer architecture

The multilayer architecture is a product of the attempts by researchers to fully explore the properties of different CAE-PVD coatings and tailor the coating morphology and properties to suit the desired application. For instance, a layer with good adhesive properties (e.g., CrN [86]) and lubricative properties (e.g., VN [176]) could be combined with one with good mechanical properties (e.g., AlCrN [177]), resulting in improved tribological performance. This might be very difficult to achieve with monolayer configurations [5]. Moreover, the superior performance of these coatings compared to the monolayer counterpart is mainly attributed to their improved resistance to crack propagation, as the multilayer boundaries inhibit dislocation movement [162]. Over the past years, this coating architecture has been found to be very beneficial, particularly in the machining applications [148,178]. As this coating architecture has received great attention from researchers, different multilayer architectures have been developed. Some of these architectures are illustrated in Figure 3-19.

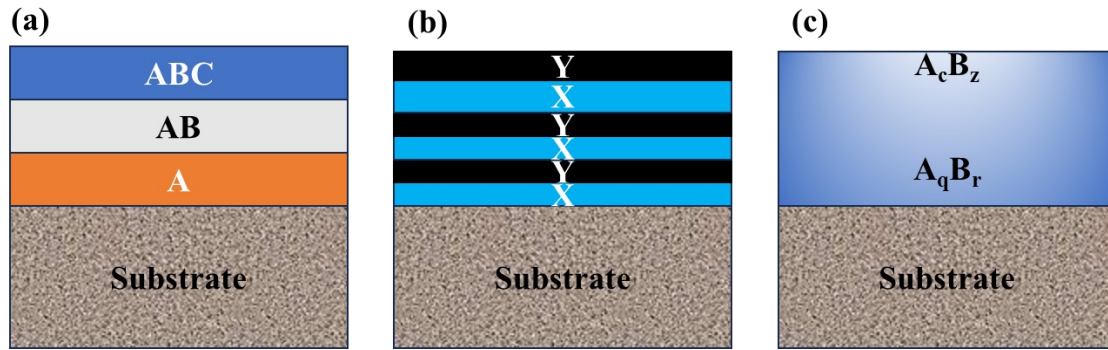


Figure 3-19: An illustration of different multilayer architectures: (a) trilayer architecture; (b) nanolayer architecture; (c) gradient architecture (A, B, C, X, and Y represent coating composition, while q, r, c, and z represent the % atomic compositions).

The first multilayer configuration (Figure 3-19a) is one in which each distinct layer has at least one additional element in excess with respect to the other layers. Most often, it comprises two (bilayers) or three (trilayers) distinct layers. This multilayer configuration is exemplified in the study conducted by Naghashzadeh et al. on the comparative assessment of the tribological performance of TiN-TiCN-TiN/TiAlN binary and TiN-TiCN-TiN/TiAlN/TiN-TiCN-TiN ternary multilayer coatings. Their findings revealed that the ternary multilayer coating exhibited superior tribological performance [74]. In this layer configuration, the order of layer arrangement significantly affects the resulting coating performance. Ideally, it is generally preferred that the first layer deposited on the substrate should have better adhesive properties. For instance, for a bilayer configuration comprising CrN and CrCN, it is recommended that the former be the first deposited layer due to its superior adhesive properties [86,179].

The second multilayer configuration is a variant of the first configuration. It comprises two major layers repeated throughout the coating thickness, as shown in Figure

3-19b. It is sometimes referred to as a nanolayer configuration, as the layer thickness is usually in the nanometric scale [180]. An example of this configuration can be found in the study conducted by Kong et al. on the tribological performance of three bilayers CrN/CrN multilayer coating. They observed superior performance with the multilayer coating compared to the respective monolayer configurations [32]. In another study, Warcholinski and Gilewicz conducted a comparative study between seven bilayers of CrN/CrCN and TiN/TiAlN coatings. They reported that both configurations resulted in improved performance compared to their respective monolayer configurations, with the former exhibiting superior tribological performance [4]. Additionally, in this configuration, the thickness of the layers plays a significant role in the resulting coating properties [128]. Accordingly, Warcholinski and Gilewicz investigated the effect of 1:5, 1:2, and 1:1 bilayer thickness ratios on the properties of CrN/CrCN multilayer coatings. Their findings revealed that optimum adhesion strength and wear resistance are obtained when the layers have the same thickness (i.e., thickness ratio of 1:1). At higher bilayer thickness ratios, the reduction in coating quality was due to the accompanying residual stresses and reduction in H/E. It is also important to mention that layer thickness in the nanometric range (<200 nm) have been observed to exhibit superior tribological performance than thicker layers [180]. The improved performance could be explained in terms of Hall–Petch effect (i.e., reduction in crystallite size and increased crystallite boundaries following layer thickness reduction) and Koehler effect (i.e., increase in interfacial density with reduction in layer thickness) leading to improved resistance to plastic deformation. Furthermore, authors have also reported a positive correlation between the tribological performance and the number of repetitive layer elements in a given bilayer thickness [165,180]. In other words, given two nanolayer

coatings with the same thickness, the one with more repetitive layers would likely exhibit superior performance. This is justifiable as increasing the number of layers increases the number of layer interfaces, thus providing additional impedance to dislocation movement. The last multilayer configuration is best described as a gradient-type architecture. In this configuration, the coating composition is varied across the coating thickness, as illustrated in Figure 3-19c. A typical example of this configuration can be found in the study conducted by Dobrzanski et al. [181] on the tribological performance of a gradient TiAlN multilayer coating deposited on different tool materials including cemented carbides, oxide tool ceramics among others. They observed an improvement in the tribological performance of the tool materials as a result of the significant increase in the hardness offered by the gradient composition.

In summary, over time, there has been an evolution from binary monolayer coatings to coating doping and the development of different multilayer architectures. However, irrespective of the coating architecture, the elemental composition of the coating is critical in achieving optimal tribological performance. Meanwhile, while optimal coating doping, particularly with carbon, is effective for tribological performance enhancement, exploring optimal co-doping holds greater prospects due to the possibility of combining desirable properties of different dopants. Furthermore, the multilayer architecture has been suggested by several researchers to provide superior improvement in tribological performance as compared to the monolayer counterpart.

3.9 Summary and future directions

3.9.1 Summary

In summary, the preparation of the substrate before coating deposition, particularly the substrate profile, significantly affects the coating morphology and the resulting tribological performance. Despite the promising prospects of CAE-PVD coatings for tribological applications, the contemporary challenge militating against the wide spread of these coatings in the wood-cutting industry is the inherent defects. However, researchers have identified deposition parameter optimization (particularly the partial pressure of the nitrogen reactive gas) as the most cost-conservative and effective method of minimizing these defects. Furthermore, because of the complex tribological behaviour of the coatings, a holistic approach involving the characterization of the mechanical, morphological, electrochemical, and tribological properties should be adopted during coating performance assessment. Additionally, the deposition parameters influence the properties and the service performance of the coatings. On the basis of the literature analysis, the substrate bias and the temperature are most likely to cause more significant changes in the coating properties than the arc current. Finally, several coating architectures have been developed over the years which exhibit different tribological performance. However, the nanometric multilayer configuration seems to hold the most potential for tribological applications as they offer superior improvement in the overall service performance of the coatings.

3.9.2 Future directions

Following the review of the recent advances and the comprehensive literature analysis of CAE-PVD coatings for enhanced tribological performance conducted, the ensuing recommendations are provided for researchers in the field for further exploration.

- (1) More systematic studies on the optimization and the effect of substrate profile with specific emphasis on the correlation with the resulting morphology, mechanical properties, the evolution of the coating–substrate interface, and the tribological performance should be conducted. Achieving this would reinforce the existing understanding of the parameter’s effect on the coatings’ service performance.
- (2) Researchers should explore the effect of the property mismatch between the coating and the substrate on the adhesion and tribological performances of CAE-PVD coatings. In this discourse, the focus can be directed towards the effect of the coating–substrate chemistry, hardness and elastic modulus differentials on the resulting residual stresses and bond strength. The results of such investigation can provide some valuable insight for making informed decisions during the coating–substrate selection for optimized tribological performance.
- (3) More systematic studies should be conducted on the effect of the deposition parameters on the coating properties, with more emphasis on process parameter optimization. Regarding the effect of process parameters, attention should be directed towards the evolution of the coating–substrate interface and the resulting impact on coating quality. In order to achieve the goal of parameter optimization, adopting a design of experiment (DOE), such as the response surface methodology (RSM) or Taguchi method, might

come in very handy. The objective of such an optimization process should be targeted towards improving energy efficiency during coating deposition and maximizing the coating quality. This holds great potential benefits, particularly for industrial practitioners.

- (4) Researchers should also investigate the effects of substrate heating resulting from high deposition temperature, arc current heating, and substrate temperature rise due to energetic bombardment, on the properties of the substrate, particularly tool steels. This is of utmost importance as the substrate heating might influence the carbide characteristics, retained austenite composition, and the resulting mechanical and tribological performance of the coated substrate. To achieve this objective, an in situ temperature measurement device such as a thermocouple can be used to obtain information about the thermal cycle of the coated substrate during coating deposition. Such a cycle can then be replicated in an uncoated substrate in a furnace, followed by subsequent mechanical and tribological characterizations. The result of such an investigation would facilitate informed decision making in the selection of deposition parameters for optimal tribological performance of the coated substrate.
- (5) Further comparative studies on the tribological performance of the different optimized coating architectures should be conducted to ascertain which would provide superior tribological enhancement. If possible, such an investigation should be supported by field tests to further validate the suitability of the optimal architecture for the intended application.

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Chapter 4

Effect of substrate roughness on the tribological performance of AlTiN/AlTiCrN-coated A8 tool steel

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Abstract

Substrate preparation is known to influence the tribological performance of coated tool steels. However, despite extensive research on the subject, accurately predicting the impact of substrate roughness on the tribological performance of a given coated tool steel remains a prevalent challenge due to its significant variation with the coating-substrate combination. Therefore, this study investigates the effect of the substrate roughness ($R_a = 0.0025 - 1.7 \mu\text{m}$) on the tribological behaviour of AlTiN/AlTiCrN-coated AISI A8 tool steel widely used in the woodcutting industry. The coated tool steel with varied roughness is evaluated through a comprehensive series of tests, including wear, nanoindentation, adhesion, profilometry, and microstructural characterizations. The results reveal that smoother substrates exhibit superior performance, characterized by about 15 % lower wear rates. This improvement is attributed to the enhancement of the (111) plane orientation, which contributes to improved adhesion, increased hardness, and greater fracture resistance, alongside a higher density of fine macroparticles and reduced surface roughness. Based on these findings, optimizing the substrate roughness to achieve a smoother surface for

AlTiN/AlTiCrN-coated AISI A8 tool steel appears promising for enhancing tribological performance.

Keywords: AISI A8 tool steel; Substrate roughness; AlTiN/AlTiCrN; Cathodic arc evaporation; Tribological performance.

4.1 Introduction

In recent years, AISI A8 tool steel has received attention from the woodworking industry due to its availability, cost-effectiveness, and wear resistance [1]. However, even in hardened conditions, the material still falls short of providing the desired tribological performance [2]. To address this limitation, tribological performance enhancement techniques, including cathodic arc-evaporated physical vapour deposition (CAE-PVD) coatings, have been explored [3]. Lately, layered coatings have become widespread in the coating industry, as they combine the properties of different coating-types, thus outperforming their monolayer counterparts [4]. In the last 20 years, layered architectures such as Cr/CrN [5], CrN/CrCN [6], and TiN/TiAlN [7] have been explored. Additionally, quaternary-based architectures, including TiN/AlTiCrN [8], TiSiN/AlTiCrN [9], and AlTiN/AlTiCrN [10], have emerged as promising candidates for enhanced tribological performance in woodworking applications. These advanced architectural designs, for instance, the AlTiN/AlTiCrN, leverage the benefit of solid solution strengthening offered by the metallic nitrides, the thermal stability of the passive metallic oxides, and the interfacial strengthening offered by the layered structure. However, despite the promising properties of these layered coatings, their performance shows considerable variability, with the substrate preparation identified as a critical influencing factor [4], [11], [12]. Adequate substrate

preparation is therefore required prior to coating deposition due to its significant impact on the coating performance and costs, especially on commercial-scale production [1].

Substrate preparation serves two major objectives: decontamination and surface profiling [4]. Decontamination aims to eliminate contaminants and surface defects, typically achieved through ultrasonic cleaning with alcohol, hot alkaline [13], or mild solvent-based detergent [14]. Surface profiling, on the other hand, is intended to modify the substrate topography to enhance the substrate-coating bonding and improve tribological performance [4]. Common profiling techniques include mechanical grinding/polishing [15], chemical etching [16], or microblasting [17]. However, despite the critical role of surface profiling in coating performance, its effect and optimization strategies remain relatively underexplored in literature. Furthermore, it is important to highlight the complex nature of substrate profiling, requiring a balance between coating adhesion and surface roughness, which are crucial for enhanced performance [4]. On one hand, increasing substrate roughness is expected to improve coating adhesion due to the enhancement of mechanical interlocking [18], [19]. But at the same time, it may be accompanied by higher friction and wear rates [16]. Meanwhile, while smoother substrates lower the coating roughness, they may weaken the adhesion, potentially compromising the tribological performance [4]. Thus, this raises an important question: How do different substrate roughness levels influence the tribological performance of CAE-PVD coatings? The following literature survey explores this issue.

Huang et al. [15] found that TiAlN-coated tungsten carbide with an average Roughness (R_a) between 0.02 and 0.04 μm showed better adhesion and wear resistance at lower R_a . The authors cited the reduction in the coating's R_a and Coefficient of Friction (COF) as the reason for the observation. However, contradicting findings were recently

reported by Ling et al. [16] for a similar substrate, but with an AlCrN coating. The authors observed an improvement in coating adhesion strength and wear resistance with increasing R_a . The observation was attributed to the enhancement of the mechanical interlocking mechanism between the coating and substrate. These contrasting findings suggest that for a given substrate, the coating composition plays a crucial role in determining how the substrate roughness influences tribological performance. This argument is further corroborated by the reports of Ravi et al. [20] on TiN and nc-TiAlSiN-coated AISI M2 tool steel. Their results showed that the adhesion strength of the coatings varied differently with the substrate R_a , depending on the coating-type. Explicitly, the expected increase in adhesion strength observed for the nc-TiAlSiN coating with R_a was connected to the improved coating-substrate mechanical interlocking. However, the contrasting behaviour exhibited by the TiN coating was explained by the increase in surface asperities at higher R_a , acting as stress concentration sites, thus leading to premature coating failure.

The foregoing discussion highlights the complexity of the effect of the substrate roughness on the coating's tribological performance, exhibiting significant variation with the coating-substrate type. This implies that, while existing studies may provide valuable insights into the effect of substrate roughness on the tribological performance of PVD coatings, the applicability of the findings may be limited to specific coating-substrate systems. Surprisingly, to the best knowledge of the authors, no studies have specifically addressed the effect of the substrate roughness on the tribological performance of AlTiN/AlTiCrN-coated AISI A8 tool steel, despite its widespread usage for tribological applications [4]. Moreover, previous studies have largely focused on narrow substrate R_a ranges (such as 0.02 and 0.04 μm [15] and 0.05 – 0.11 μm [16]), thus limiting the

understanding of how the performance and properties of the coatings evolve with the parameter. Therefore, these gaps highlight the need for further investigation to provide a more in-depth understanding of the relationship between the substrate roughness and the tribological performance of AlTiN/AlTiCrN-coated AISI A8 tool steel.

This study investigates the effect of substrate roughness on the tribological performance of AlTiN/AlTiCrN coatings deposited on AISI A8 tool steel. A broader R_a range (0.002 – 1.7 μm) was adopted to gain a better understanding of how the AlTiN/AlTiCrN coating properties and performance evolve with extreme variations in the substrate roughness. This was achieved through a series of tests, including wear, nanoindentation, adhesion, profilometry, and microstructural characterizations. These were complemented by microstructure–property–performance correlations and attempts to identify the most significant factors influencing the AlTiN/AlTiCrN coating’s performance. The findings offer valuable insight for optimizing the substrate roughness of AlTiN/AlTiCrN-coated AISI A8 tool steel for enhanced tribological performance.

4.2 Experimental details

4.2.1 Substrate preparation and profilometry

The chemical composition (% wt.) of the AISI A8 tool steel used for the investigation comprises: C – 0.48 %, Cr – 7.9 %, Si – 0.87 %, V – 0.36 %, Mn – 0.46, Ni – 0.14 %, Cu – 0.17 %, Mo – 1.41 %, W – < 0.01 % and Fe – balance. The hardened samples (~ 610 HV) with dimensions of 75 × 25 × 10 mm were mechanically ground to achieve three surface finishes, denoted as Very smooth, Smooth, and Rough, as summarized in Table 4-1. To obtain the Rough surface finish, the substrates were ground using SiC-based emery papers with grit

sizes of 80 and 120, mounted on a grinding disc rotating at 250 rpm for 3 minutes, resulting in an average R_a of about 1.7 μm . For the Smooth condition, grinding was carried out sequentially with 120 and 240 grit emery papers for a total of about 5 minutes at a constant rotational speed, producing an average R_a of about 0.09 μm . Meanwhile, to achieve the Very smooth surface finish, substrates were ground with emery papers up to 600 grit, followed by polishing with 3 μm diamond suspension for a total of 10 minutes, yielding an average R_a of 0.003 μm . Thereafter, all the samples were decontaminated using a mild solvent-based detergent, rinsed with water, and vacuum-dried. The surface roughness of the samples before and after coating deposition was measured using a stylus profilometer (Mahr MarSurf PS 10, Germany). In recent studies, 3D profilometry techniques have also been utilized to achieve higher precision and comprehensive roughness characterization [21]. The resulting R_a and peak-to-valley height (R_z) for each condition represent an average of at least nine measurements.

Table 4-1: Substrate pretreatment procedures and the roughness parameters.

Sample condition	Processing procedure	Total grinding/polishing time (mins)	Average R_a (μm)	Average R_z (μm)
Very smooth	240 + 400 + 600 grits grinding + 3 μm polishing	10	0.0025 ± 0.0004	0.21 ± 0.08
Smooth	120 + 240 grits grinding	5	0.089 ± 0.006	0.68 ± 0.1
Rough	80 + 120 grits grinding	3	1.69 ± 0.2	11.69 ± 1.5

4.2.2 Coating deposition

The AlTiN/AlTiCrN bilayer coating was deposited on the prepared samples using PLATIT (PL1011, Switzerland) CAE-PVD equipment. The apparatus is equipped with an Al₅₀Ti₅₀, two Al₆₇Ti₃₃, and a Cr high-purity planar targets. The chamber was then evacuated to achieve a vacuum of $1 - 2 \times 10^{-5}$ mbar, not only to minimize coating contamination [4], but also to facilitate deposition at lower temperatures (< 500 °C), thereby enhancing energy efficiency during the deposition process [22]. Thereafter, etching of the samples to eliminate possible surface residue contaminants was performed at $380 - 400$ °C using Al₆₇Ti₃₃ metal ion etching followed by Ar inert gas. During coating deposition, the samples were rotated continuously ($\sim 3 - 5$ rpm) around the vertical central axis to ensure uniform deposition. The bilayer AlTiN/AlTiCrN coating deposition began with the AlTiN base layer using Al₆₇Ti₃₃ and Al₅₀Ti₅₀ targets. The arc current, negative bias, temperature, and N₂ gas flow were kept at $250 - 280$ A, $150 - 180$ V, $350 - 380$ °C, and $640 - 660$ sccm, respectively. This was then followed by the deposition of the top AlTiCrN layer using two Al₆₇Ti₃₃ and Cr targets. However, a higher N₂ gas supply (about $680 - 700$ sccm) was employed while the other parameters remained unchanged as in the deposition of the AlTiN base layer. The coating was deposited over 1.8 hours.

4.2.3 Microstructure and mechanical properties

Surface examination of the coated samples, to identify deposition defects, such as macroparticles (MPs) and craters, was achieved using a scanning electron microscope (SEM, JSM-6480LV, Japan) equipped with an energy-dispersive X-ray (EDX, INCA energy 250). Subsequently, post-image processing using Image J software (version 1.54d) was performed to obtain the MP statistics [23]. For this analysis, ten images having $\times 1000$ magnification

and similar contrast properties were used for each condition. The phase composition was analyzed using X-ray diffraction (XRD, D8 Discover, Bruker AXS, Germany) technique with Co $k\alpha$ radiation, step size of 0.05° , step time of 0.5 s/step, and a 2θ diffraction angle ranging from $20 - 115^\circ$. Phase identification was achieved using PANalytical X'pert HighScore plus software with Crystallography Open Database (COD) [24]. The coating thickness was also measured from the SEM cross-sectional micrographs using Image J [23], taking at least nine measurements for each condition. Subsequently, SEM-EDX analysis was used to determine the elemental composition of the coating. The coating hardness (H) and elastic modulus (E) were measured by nanoindentation (Anton Paar MCT³), with an indenting load of 20 mN. The resulting hardness for each condition represents an average of at least five measurements. The coating adhesion was evaluated via scratch testing using the Anton Paar (MCT³) apparatus. During the test, a Rockwell-type diamond indenter (tip radius = 50 μm) under a progressive loading from 0 to 30 N is applied normal to the coating surface. The loading rate, indenter speed, and scratch length were 20.98 N/min, 3.5 mm/min, and 5 mm, respectively. The test was repeated three times for each condition.

4.2.4 Wear test

The wear performance of the coated samples was assessed using the Dry Sand Rubber Wheel (DSRW) test, as per procedure B of the ASTM G65 standard [25]. Most conventional wear testing methods rely on two-body abrasion [4], which fails to accurately mimic the conditions encountered during wood machining. In contrast, the DSRW test employs three-body abrasion, comprising the coated sample, abrasive silica sand, and a rubber wheel, which simulates the cutting tool, wood contaminants, and workpiece, respectively. During the test, the sample undergoes continuous abrasion from 200 – 300 μm

silica sand particles as it is pressed against a 228 mm Chlorobutyl rubber wheel, as illustrated in Figure 4-1.

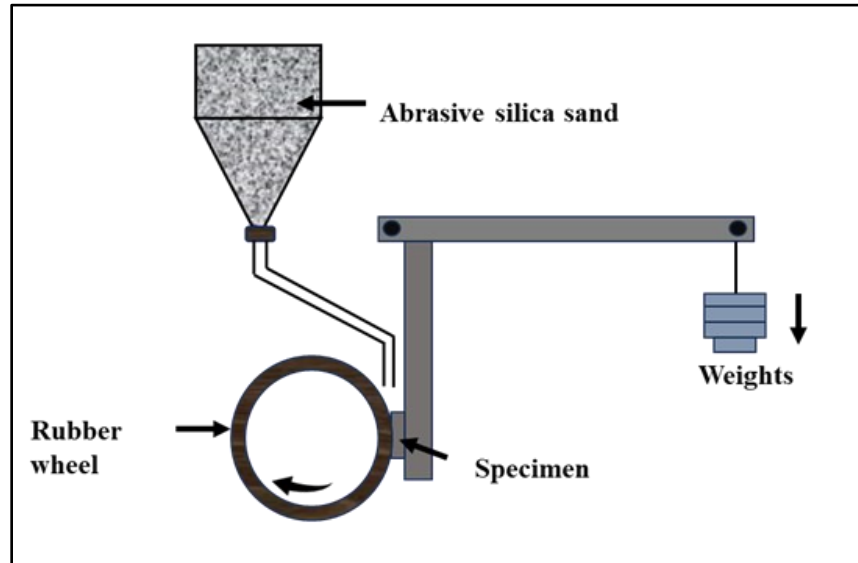


Figure 4-1: An illustration of the DSRW apparatus.

The wear resistance of the samples was evaluated as per the mass loss method using a 1 mg precision balance, i.e., obtaining the difference in the mass of the samples before and after the test. While this balance precision is relatively lower than that employed in micro-abrasion tests, previous studies indicate that it could be suitable for PVD-coated tool steels characterized by the ASTM G65 procedure [10], [26], which is a high stress test involving a high contact force of 130 N. Unlike micro-abrasion tests (contact force < 10 N) [27], where material loss is minimal and requires a higher precision balance to detect, the ASTM G65 test produces greater material loss, which could be measured with a 1 mg resolution balance. The mass loss obtained for each condition represents the average of three measurements and was subsequently used in the computation of the wear volume [10]. In order to standardize

resulting wear volumes, it was necessary to account for the wear of the rubber wheel. This was done by measuring the wheel diameter after each test and then applying Equation 1 to obtain the adjusted wear volumes [25].

$$\begin{aligned} & \text{Adjusted wear volume (mm}^3\text{)} \\ &= \text{wear volume (mm}^3\text{)} \\ &\times \frac{228.6 \text{ mm}}{\text{Wheel diameter after test (mm)}} \end{aligned} \quad (1)$$

The wear rate expressed in mm³/Nm, also known as the specific wear rate [28], [29], was then obtained using the adjusted wear volume derived from Equation 1. Post-wear characterization of the wear track to ascertain the prevalent wear mechanism was conducted using SEM-EDX analysis. Further analysis to quantify the area of delaminated coating ($\bar{A}$) was conducted using Image J software [23], while wear track depth was measured via profilometry. The value reported for each condition represents the average of three measurements.

4.3 Results and discussion

4.3.1 Coating microstructural analysis

4.3.1.1 Surface morphology, MP statistics, and surface roughness

Figure 4-2 presents the surface morphology of the coated tool steels. From the figure, grooves from substrate profiling are evident, particularly in the Smooth and Rough conditions (Figure 4-2b and c). The groove visibility appears to increase with the substrate roughness, with the Rough condition (Figure 4-2c) exhibiting the most pronounced grooves. This is anticipated, as the coating replicates the substrate morphology [20]. Additionally, the

surfaces exhibit MPs and craters; common CAE-PVD coating defects that can degrade the coating performance [4]. The MPs could result from microexplosions and hydrodynamic effects at the target, ejecting molten droplets that eventually solidify on the substrate [4], [30]. Meanwhile, craters may result from the geometric shadowing effect by the MPs [31], or an incomplete reaction between the coating species and the N_2 gas [10], [32], often becoming apparent after the flaking of loosely bound MPs.

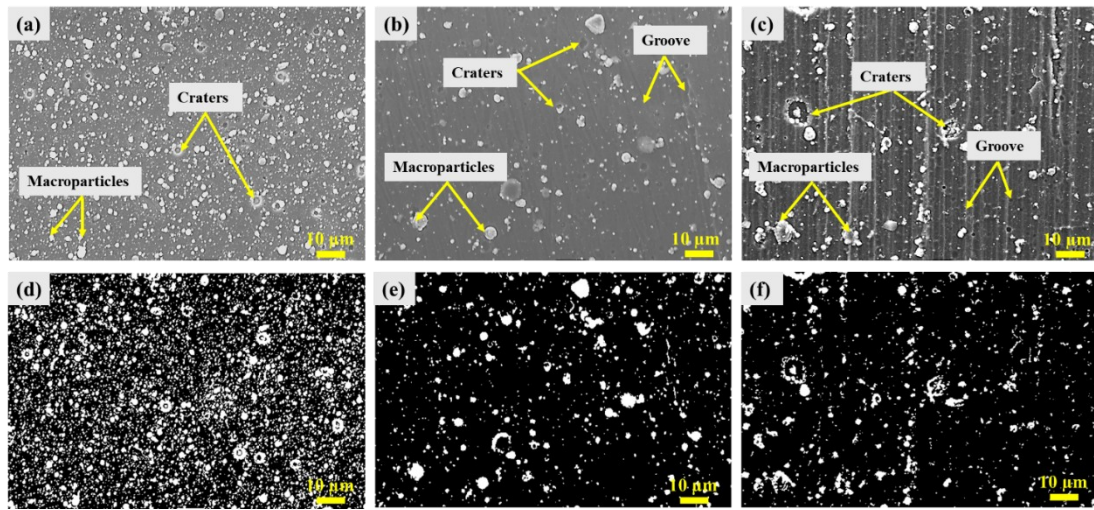


Figure 4-2: SEM micrographs and corresponding ImageJ-processed images showing the surface morphology of AlTiN/AlTiCrN-coated AISI A8 tool steel under three substrate conditions: (a, d) Very smooth; (b, e) Smooth; and (c, f) Rough.

Figure 4-2d-f present the ImageJ-processed micrographs corresponding to Figure 4-2a-c, respectively, which were used for the macroparticle statistics analysis. As shown in the transformed micrographs (Figure 4-2d-f), the processing minimizes the influence of surface grooves, particularly in the Rough condition (Figure 4-2c and f), ensuring a more reliable quantification. The results of the MP statistics coated samples are presented in Figure 4-3. This aspect has not been previously explored in previous studies [15], [16], [20], [33]. Figure 4-3a shows the MP density and average size for the different substrate roughness conditions.

From the figure, the Very smooth condition presented the highest MP density of about 0.27 particle/ μm^2 , decreasing by about 68 and 47 % in the Smooth and Rough conditions, respectively. The volume fraction of the MPs exhibited a similar trend, reaching about 27 % in the Very smooth condition, and decreasing to about 7.9 and 7.3 % for the Smooth and Rough conditions, respectively. Meanwhile, the average MP size followed a contrasting trend, with the Very smooth condition presenting the lowest MP size ($\sim 0.65 \mu\text{m}^2$). This is followed by about 17 and 14 % increase in the Smooth and Rough conditions, respectively.

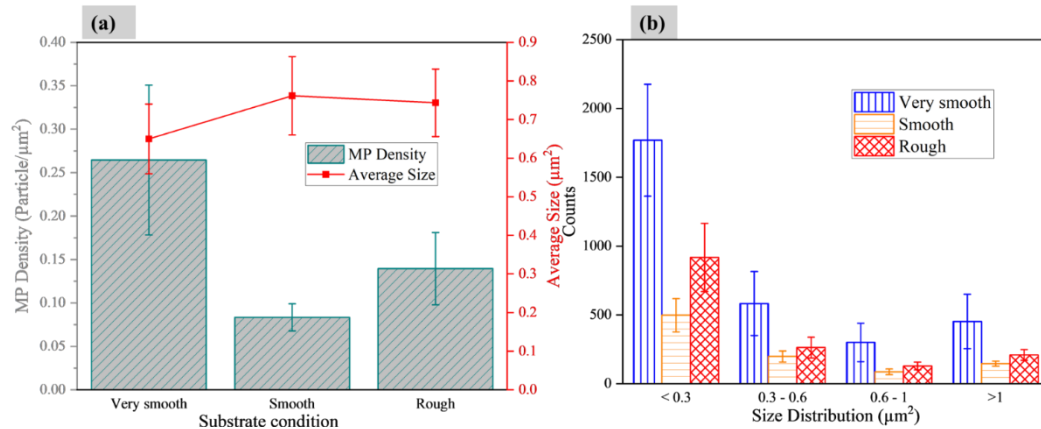


Figure 4-3. MP statistics of the different substrate conditions: (a) MP density and average size, and (b) MP size distribution.

Figure 4-3b shows the MP size distribution. The figure reveals that about 75 – 77 % of the MPs have sizes $< 0.6 \mu\text{m}^2$. Furthermore, the Very smooth condition consistently showed the highest MP counts across all size ranges, explaining its greater MP density (Figure 4-3a), since higher MP counts directly increase the density [10]. Additionally, the least average MP size exhibited by the Very smooth condition, despite presenting the highest MP density, could be attributed to its greater counts of fine MPs, particularly in the range 0 – $0.6 \mu\text{m}^2$ (70 and 50 % less in the Smooth and Rough conditions, respectively). Meanwhile, for the Smooth and Rough conditions, 16 % of the large MPs with sizes $> 1 \mu\text{m}^2$ in the

former might have contributed to the slightly ($\sim 2\%$) greater average size as compared to the 14% recorded for the latter.

Overall, the MP statistics reveal a general trend: the MP density decreases while the average size increases with increasing substrate roughness. This behaviour could be explained in terms of MP preferential nucleation and agglomeration mechanisms. In this regard, the high surface energy peaks on the rougher substrate act as preferential nucleation sites, promoting the agglomeration of incoming MPs and eventually resulting in their growth [22], [34]. Hence, the larger MP size observed on the rougher substrate. In contrast, these surface inhomogeneities are less prevalent in the smoother substrate, facilitating more uniform MPs nucleation with limited agglomeration [15]. Hence, the finer MPs observed in the Very smooth condition. Furthermore, the preferential nucleation of the incoming MPs at the peaks increases the susceptibility of MP re-sputtering due to energetic bombardment by incoming coating species, ultimately leading to a reduction in MP density observed with increasing substrate roughness [4].

Figure 4-4 shows the results of the R_a and R_z of the coated and uncoated samples. From the figure, the parameters are expectedly higher after coating deposition, following the trend: Very smooth < Smooth < Rough. For instance, in comparison with the uncoated conditions, the R_a is about 8.4, 1.8, and 1.2 times higher in the Very smooth, Smooth, and Rough conditions, respectively. Similar roughness changes after coating deposition have been reported by Huang et al. [15] and Pak et al. [35]. Notably, while the roughness parameters for the Very smooth and Smooth conditions after coating deposition are relatively comparable, the Rough condition exhibits distinctively higher values. Moreover, the change in the roughness after the coating deposition could be linked to the MPs [4]. Accordingly,

higher MP density and larger average MP sizes have been reported to contribute to an increase in the surface asperities, thereby increasing the roughness of the coated sample [26].

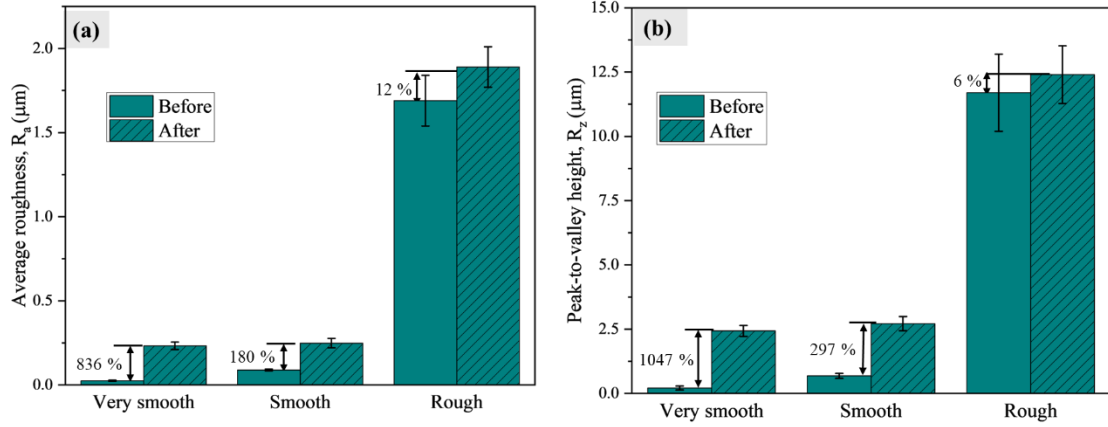


Figure 4-4: Surface roughness parameters of the different substrate conditions before and after coating deposition: (a) R_a and (b) R_z .

Figure 4-5 shows the relationship between the changes in R_a , R_z , and MP statistics. Figure 4-5a reveals an increase in ΔR_a with an increasing MP density (highest for Very smooth conditions), but decreases with the average MP size. The increase in ΔR_a despite the reduction in the average MP size might be attributed to the likely greater influence of the accompanying 70 % increase in MP density compared to the less than 20 % reduction observed for the average sizes. Meanwhile, Figure 4-5b reveals no clear correlation between ΔR_z and the MP statistics. However, the Very smooth condition still presented the highest ΔR_z .

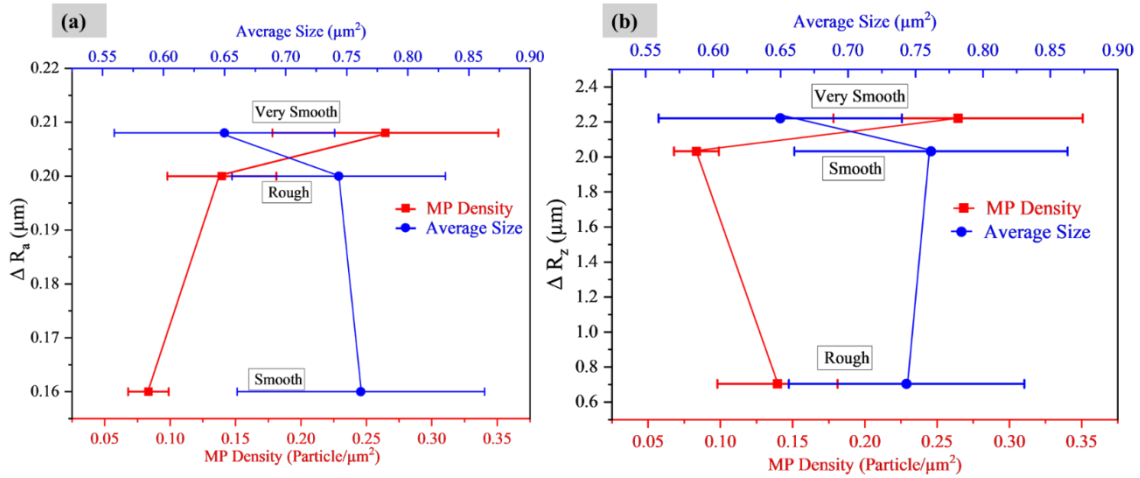


Figure 4-5: Relationship between MP density, average MP size, and: (a) ΔR_a and (b) ΔR_z

Another noteworthy observation is the diminishing influence of coating-induced roughness changes compared to the uncoated counterparts with increasing substrate roughness. For instance, ΔR_a decreased sharply from 836 % (Very smooth) to 12 % (Rough) while ΔR_z reduced from 1047 to 6 % (Figure 4-5a and b). This trend suggests that for rougher substrates ($R_a > 0.1 \mu\text{m}$), the substrate roughness largely dictates the final surface roughness of the coatings, whereas for smoother substrates ($R_a < 0.1 \mu\text{m}$), the surface features introduced by the coating (such as the MPs) have a more pronounced effect. In summary, while the roughness of the coated samples correlated with the substrate roughness, the greatest changes in the roughness parameters after coating deposition were observed at lower substrate roughness, with the ΔR_a showing a significant correlation with the MP statistics.

4.3.1.2 Cross-sectional and compositional analysis

Figure 4-6 presents the cross-sectional micrographs of the coated samples. The coatings present a dense morphology and appear to be well-adhered to the substrate. The coating morphology typically replicates the substrate topography, particularly evident in the Rough condition (Figure 4-6c), characterized by a wavy coating-substrate interface and the subsequent growth. However, a more uniform morphology is obtained for the smoother substrate (Figure 4-6a), consistent with previous reports [36].

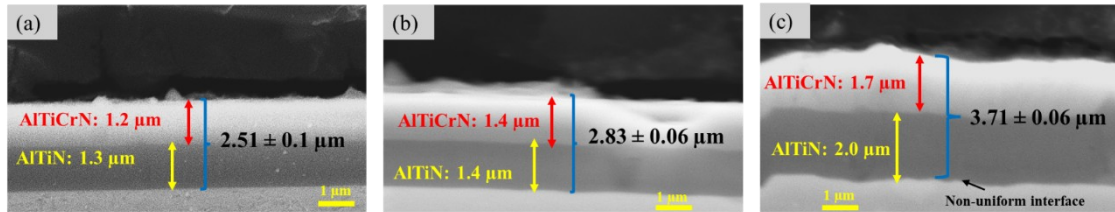


Figure 4-6: SEM micrographs showing the AlTiN/AlTiCrN coating cross-section:

(a) Very smooth; (b) Smooth; (c) Rough.

The cross-sectional micrographs reveal a bilayer structure: a dark AlTiN base layer and a bright AlTiCrN top layer, as identified by EDX analysis (Table 4-2). The thickness of the AlTiCrN top layer was generally lower than the AlTiN base layer due to the shorter deposition time. The total coating thickness increased by about 50 % with an increase in substrate R_a from 0.025 to 1.69 μm (i.e., Very smooth to Rough). Previously, Ling et al. [16] reported up to a 90 % increase in coating thickness for substrate R_a between 0.05 – 0.1 μm. The thicker coatings observed for the rougher substrate could be attributed to the increase in nucleation sites [4], [16].

Table 4-2. EDX elemental analysis of the AlTiN/AlTiCrN coatings for the different substrate conditions.

Layer	Condition	Composition (at. %)				$\frac{[Al]}{[Al] + [Ti]}$	$\frac{[Al]}{[Al] + [Ti] + [Cr]}$	$\frac{[N]}{[Al] + [Ti] + [Cr]}$
		Al	Ti	Cr	N			
AlTiN	Very smooth	23.80	18.20	2.80	55.10	0.57	0.53	1.23
	Smooth	26.10	21.10	2.20	50.70	0.55	0.53	1.03
	Rough	27.40	20.70	1.20	50.70	0.57	0.56	1.03
AlTiCrN	Very smooth	18.50	10.60	16.70	54.20	0.64	0.40	1.18
	Smooth	16.90	10.10	17.80	55.20	0.63	0.38	1.23
	Rough	16.70	8.60	18.00	56.70	0.66	0.39	1.30

Notably, as summarized in Table 6-3, the coating thickness increased by only 13 % when substrate R_a increased by about 3460 % from Very smooth and Smooth conditions. However, a much greater increment of about 31 % was observed upon increasing the substrate R_a by about 1799 % from the Smooth to the Rough condition. It appears that the increase in coating thickness becomes more pronounced at higher substrate R_a , consistent with the findings of Ling et al [16]. In summary, with increasing substrate roughness, the AlTiN/AlTiCrN coating morphology tends to be less uniform, accompanied by an increase in thickness.

Table 4-3: Changes in the R_a and coating thickness for the different substrate conditions.

Condition	R_a (μm)	Change in R_a (%)	Coating thickness (μm)	Change in Coating thickness (%)
Very Smooth	0.0025 ± 0.0004	–	2.51 ± 0.1	–
Smooth	0.089 ± 0.006	3460 ± 603	2.83 ± 0.06	12.7 ± 2.4
Rough	1.69 ± 0.2	1799 ± 255	3.71 ± 0.06	31.1 ± 2.2

Note: The change in R_a and thickness for the Smooth is relative to the Very smooth, while the Rough is relative to the Smooth.

Furthermore, the EDX elemental analysis (Table 4-2) revealed that, for all the substrate conditions, the AlTiN base layer contained Al, Ti, and N alongside trace amounts of Cr. This is further evidenced by the EDX mapping in Figure 4-7a. The detected traces of Cr likely originated either from diffusion from the Cr-rich AlTiCrN top layer or the Cr-containing substrate, as supported by the EDX line scan profile (Figure 4-7b). Similarly, the analysis of the AlTiCrN top layer confirms the presence of Al, Ti, Cr, and N for all the conditions. Expectedly, the incorporation of Cr in this layer resulted in a reduction in the composition of other metallic components (i.e., Al and Ti) compared to the base AlTiN layer, for all the coated samples. Notably, variations in the substrate surface roughness prior to deposition did not appear to significantly influence the elemental incorporation efficiency of Cr in the top AlTiCrN layer, resulting in a generally fairly constant Cr content across all the coated samples. This may be attributed to the relatively higher atomic mass of Cr atoms [26], which makes them less susceptible to re-sputtering effects caused by local inhomogeneities in the plasma-substrate interaction induced by substrate surface profile variations.

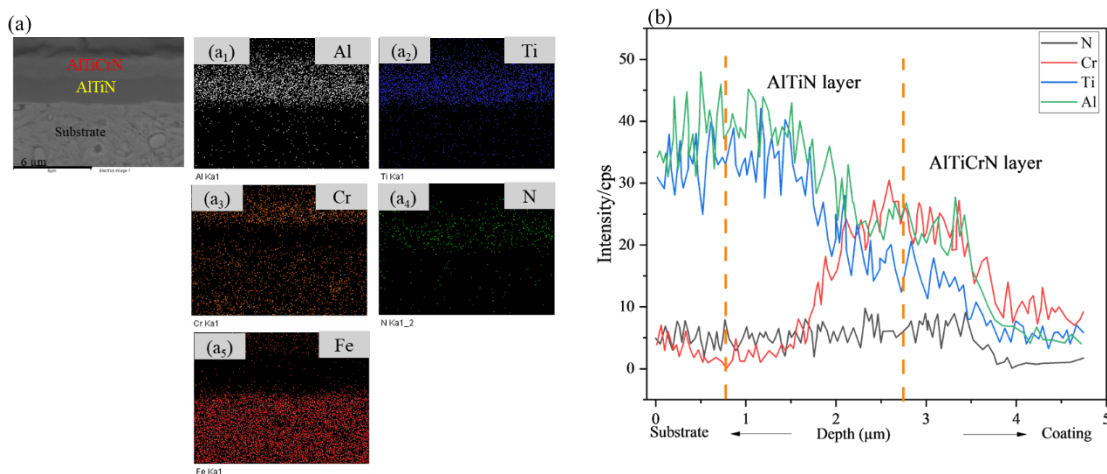


Figure 4-7: (a) EDX map of the AlTiN/AlTiCrN coating and (b) line scan profile of the Rough condition.

Furthermore, the stoichiometric analysis indicated by the $N / (Al + Ti + Cr)$ ratio revealed that both coating layers exhibited ratios > 1 for all substrate conditions. This signifies a sufficient N_2 gas supply facilitating the formation of different metallic nitrides (i.e., AlN, TiN, and CrN) [6], [16]. Further EDX examination revealed that, with increasing substrate roughness, the AlTiN base layer appears to be less stoichiometric, evident in a reduction in the ratio (from 1.23 to 1.03) and N content (from 55.1 – 50.7 at. %). This suggests the preferential re-sputtering of N atoms. This is likely because the lighter N atoms (~ 14 g/mol) are more easily displaced from the rough surfaces during energetic collision compared to heavier Al and Ti atoms (> 25 g/mol) [4]. Meanwhile, a reverse trend was observed for the top AlTiCrN layer. Rougher substrates exhibited higher N content (increasing from 54.2 to 56.7 at. %), following Cr addition. More so, the increase in N content correlated with higher Cr content, accompanied by lower Al and Ti concentrations. It appears that higher Cr content

on the rougher substrates may have contributed to the stabilization of N by promoting the formation of metallic nitrides, thereby reducing N re-sputtering [37].

Additionally, the Al/(Al+Ti) ratio for the AlTiN layer across all substrate conditions is observed to be fairly constant ($\sim 0.55 - 0.57$), but generally higher for the AlTiCrN layer ($\sim 0.63 - 0.66$). The higher ratios observed for the AlTiCrN layer are connected to the higher Al/Ti ratio of the deposition target material [32]. Furthermore, higher Al/(Al+Ti+Cr) ratios, particularly below 0.7, have been linked with improved performance [38]. In this study, the AlTiN layer showed a slightly higher ratio (~ 0.56) for the Rough condition and fairly constant (~ 0.53) for the other conditions. Meanwhile, for the AlTiCrN layer, the Very smooth condition presented a higher ratio (~ 0.4) while the ratios for the others were fairly constant (~ 0.38). The general notable reduction in the Al/(Al+Ti+Cr) ratio observed for the AlTiCrN layer as compared to the AlTiN layer could be due to Cr atoms substituting Al atoms [29]. Nonetheless, considering both layers collectively, the effect of the substrate roughness on the Al/(Al+Ti+Cr) ratio could be considered negligible ($\sim 0.45 - 0.47$). In summary, the coatings generally presented stoichiometric compositions, with the substrate roughness showing no significant impact on the Al/(Al+Ti+Cr) ratio.

Figure 4-8 shows the XRD patterns of the AlTiCrN/AlTiN coatings with different substrate roughness. The lines represent standard peak positions based on the Crystallography Open Database (COD24) [39]. The peaks are a combination of those from the substrates and the AlTiN/AlTiCrN coatings [10]. Three substrate peaks ($2\theta = 52.2^\circ$, 77° , and 99.4°) were identified according to the reference code 96-901-3474. Notably, with increasing roughness (Very smooth to Rough), the substrate peaks intensities decrease, suggesting an increase in the coating thickness [32], [40]. This is consistent with the cross-

sectional micrographs in Figure 4-6. This likely occurs because thicker coatings absorb more X-rays, thereby reducing penetration to the substrate [41]. Meanwhile, the coating-related peaks appear to be positioned between the reference c-TiN (96-101-1103), c-CrN (96-101-0975), and c-AlN (00-046-1200). However, compared to the reference peaks, the peaks appear to be shifted to higher or lower angles, suggesting the formation of a solid solution of the metallic nitrides (i.e., TiN, CrN, and AlN) [29].

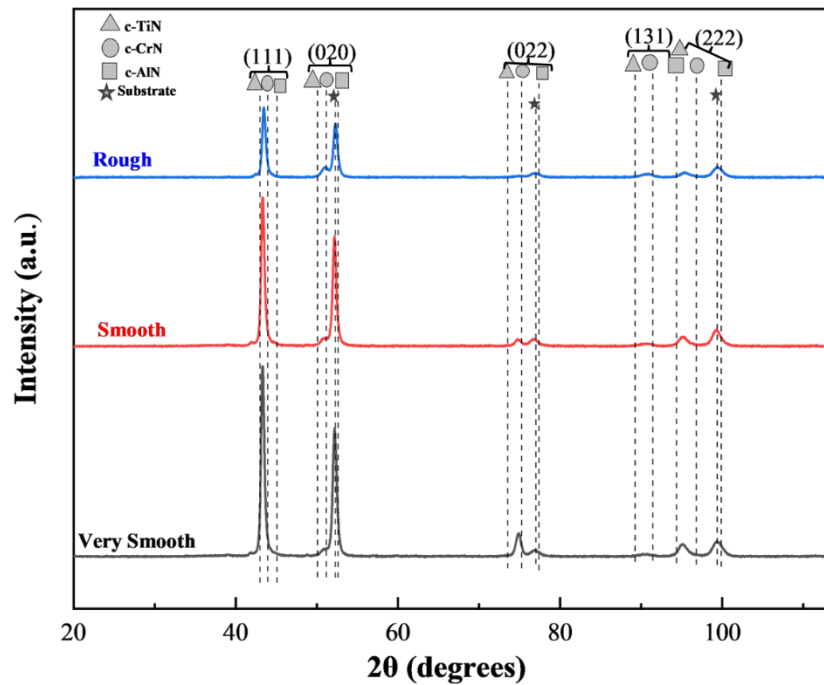


Figure 4-8: The XRD profiles of the AlTiN/AlTiCrN coating with different substrate conditions.

The AlTiCrN/AlTiN coatings generally presented a (111) preferred orientation and minors of (020), (022), (131), and (222). The high planar density of the (111) orientation likely promoted the preferential arrangement of the adatoms in the closed-pack plane [22]. Figure 4-9 quantifies the plane distribution of the coating peaks with 2θ angles

summarized in Table 4-4. The percentage plane distribution values were calculated by expressing the intensity of a given coating diffraction peak as a proportion of the sum of all the coating peaks' intensities. The figure shows that as the substrate roughness increased (from Very smooth to Rough), the (111) orientation decreased from 59 to 48 %. This is consistent with the corresponding reduced peak intensities in Figure 4-8. This might be attributed to the disruption in adatom nucleation by the surface irregularities of the rougher substrates, which promotes the competitive growth of various crystallographic planes and results in more random plane orientations [22]. This reduction in the percentage of dense (111) plane with increasing substrate roughness suggests a reduction in the coating densification [42]. This phenomenon can be explained by the geometric shadowing effect, which is more prominent on rougher substrates. In such cases, incoming coating adatoms tend to preferentially nucleate on the elevated regions or peaks of the substrate surface. Meanwhile, the valleys receive fewer adatoms, facilitated by the limited line-of-sight exposure, leading to slower coating growth in those areas. This uneven growth results in a

less dense coating structure [22]. In summary, higher substrate roughness led to a reduction in the preferred closed-packed (111) plane composition.

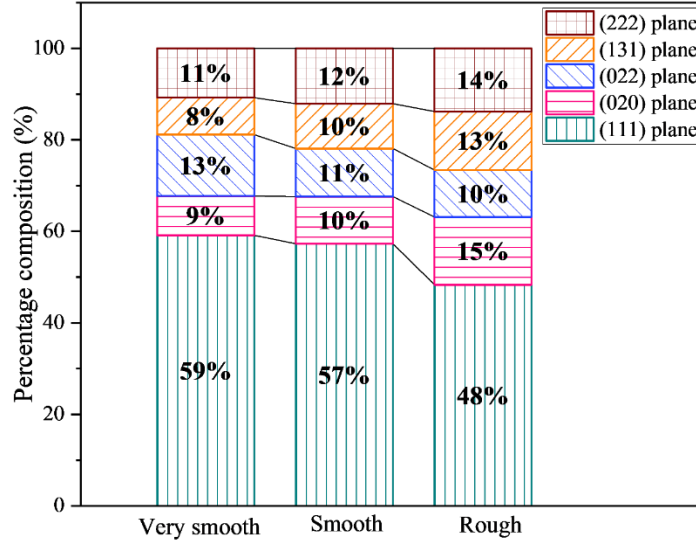


Figure 4-9: Plane distribution of the AlTiN/AlTiCrN coatings deposited on different substrate profiles.

Notably, no peaks corresponding to the detrimental h-AlN phase were detected. This could be explained by the fact that all the coatings had Al/(Al+Ti+Cr) ratios well below the 0.7 formation threshold (Table 4-2). Also, no peaks associated with incomplete phase transformation products (such as Cr₂N) were observed. This could also be related to the stoichiometric compositions of the coatings with N/(Al+Ti+Cr) ratios > 1 (Table 4-2). Further examination of the full-width at half-maximum (FWHM) values (Table 4-4) revealed the following, with increasing substrate roughness: (1) peak broadening for the (111), (022), and (222) planes indicated by the general increase in their FWHM values (2) peak narrowing for (020) and (131) planes evident in the reduction in their FWHM values.

Table 4-4: Peak positions and FWHM of the AlTiCrN/AlTiN coatings with different substrate roughness.

Condition	(111) plane		(020) plane		(022) plane		(131) plane)		(222) plane	
	2 θ (°)	FWHM M (°)	2 θ (°)	FWHM M (°)	2 θ (°)	FWHM M (°)	2 θ (°)	FWHM M (°)	2 θ (°)	FWHM M (°)
Very Smooth	43.3 0	0.51	50.84	2.97	74.88	0.71	90.5 9	2.32	95.09	1.19
Smooth	43.3 2	0.51	50.76	1.55	74.74	0.82	90.6 3	2.00	95.11	1.22
Rough	43.4 7	0.53	50.98	1.15	74.92	1.95	90.8 2	1.80	95.34	1.21

Generally, FWHM changes suggest changes in crystallite size and microstraining. While peak broadening is associated with finer crystallites and increased microstrain, peak narrowing indicates crystallite growth and reduction in microstrain [10]. The crystallite size and microstrain were computed using Equations 2 and 3 [9].

$$D = \frac{K \cdot \lambda}{\beta \cdot \cos \theta} \quad (2)$$

$$\varepsilon = \frac{\beta \cdot \cos \theta}{4 \cdot \sin \theta} \quad (3)$$

Where D – crystallite size, K – Sherrer constant, λ – X-ray wavelength, β – FWHM, θ – peak position, and ε – microstrain.

Figure 4-10 shows the average crystallite size and average microstrain for the different substrate conditions. The substantial increment in the substrate roughness appears not to have a significant effect on the average crystallite size, with values between 10.5 – 11.3 nm. Meanwhile, the average microstrain generally decreased with increasing substrate

roughness, suggesting that roughness affected the microstrain more significantly than the crystallite size. This differs from Ling et al.'s [16] observation of $> 100\%$ crystallite size reduction when the substrate roughness was increased by 120% . The discrepancy is likely due to the difference in the coating-substrate system and deposition conditions. Specifically, Ling et al. [16] investigated AlCrN-coated WC-Co substrates, with substrate roughness induced by chemical etching, and coatings deposited at a negative bias of 100 V and arc current of 80 A . These parameters differ significantly from those employed in this present study. Nonetheless, the higher microstrain observed in the Very smooth condition suggests greater coating lattice distortion within the coating, which may be attributed to the enhanced (111) crystallographic ordering [10]. Gui et al. [43] previously reported that in sputtered AlTiCrN coatings, variations in Ti contents led to the formation of substitutional and interstitial solid solutions of transition metallic nitrides, which in turn induced lattice distortions and microstrain. Their interpretation aligns with our current observation, where increased microstrain recorded in the Very smooth condition could be linked to the formation of solid solution, potentially promoted by a stronger (111) preferred orientation (Figure 4-8 and 9). This crystallographic texture, which is denser and more energetically favorable [22], may intensify lattice distortion effects in the smoother substrate, thereby supporting the observed microstrain behaviour. In summary, while the increase in substrate roughness led to a reduction in the microstrain, the crystallite size appears to be fairly constant.

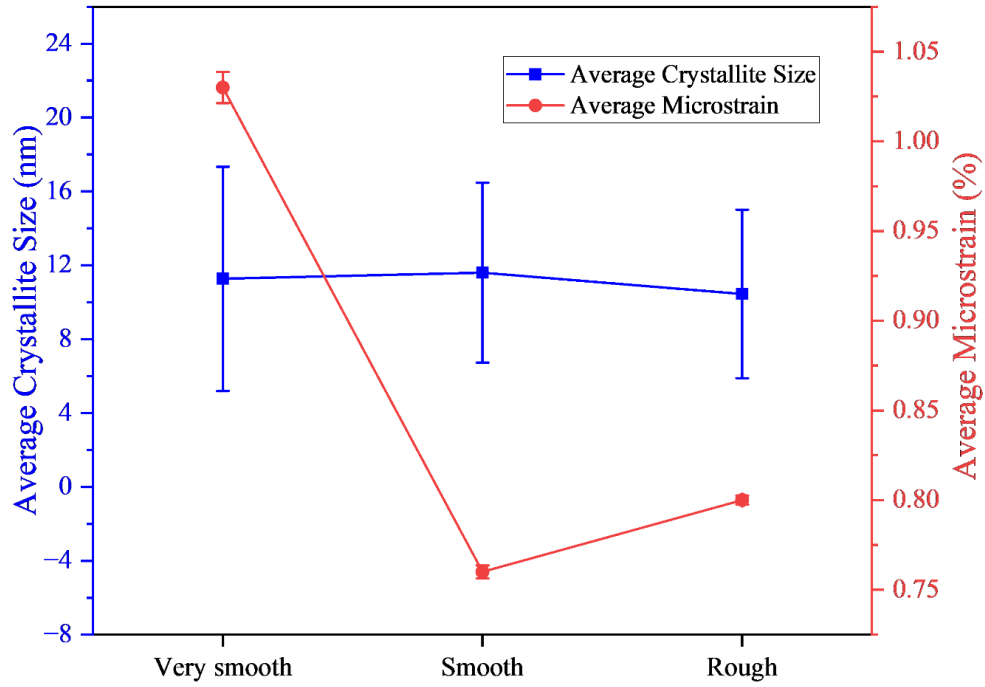


Figure 4-10: Average crystallite size and microstrain for AlTiN/AlTiCrN coatings with different substrate roughness.

4.3.2 Hardness, H/E and H^3/E^2 ratios

Figure 4-11 shows the results obtained from the nanoindentation test. Figure 4-11a shows the indenter displacement versus the load curves. For all the substrate conditions, two regions corresponding to the loading and unloading stages are evident [44]. This is characterized by increasing displacement with load (loading stage), followed by decreasing displacement with load (unloading stage). It is also worth mentioning that the maximum indenter displacement is less than the stipulated 10 % thickness for all conditions [45]. Hence, the substrate contribution is minimized in the resulting measurements. Furthermore, with increasing substrate roughness, the maximum indenter displacement also increases, as evidenced in the shift of the curves to the right. This occurrence signifies a reduced resistance of the AlTiN/AlTiCrN coatings to plastic deformation [32].

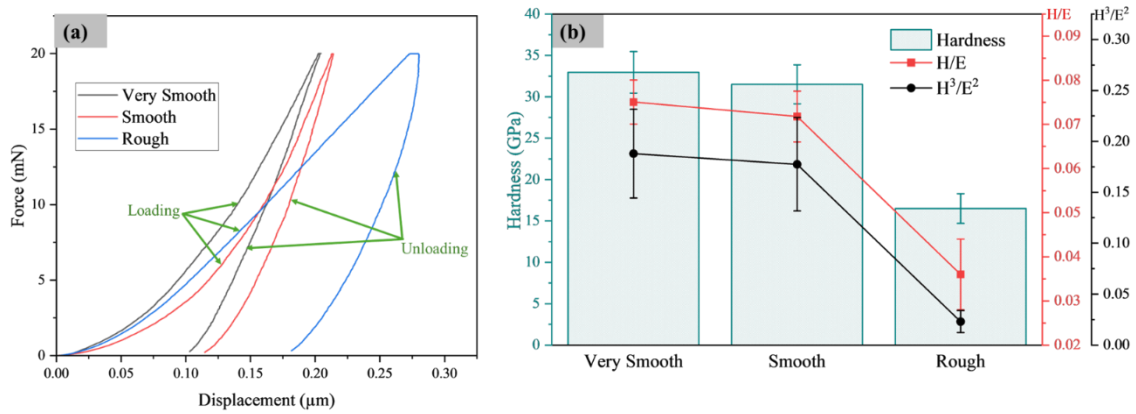


Figure 4-11: Nanoindentation test results for the AlTiN/AlTiCrN coatings with different substrate roughness (a) Load-indentation displacement curves and (b) Hardness, H/E, and H^3/E^2 ratios.

Figure 4-11b shows the hardness, H/E, and H^3/E^2 ratios of the samples. From the figure, the Very smooth condition exhibited the highest hardness (~ 33 GPa), which reduced by 4 % (Smooth) and 50 % (Rough) with increasing substrate roughness. The improvement in the hardness of the AlTiN/AlTiCrN coatings at lower substrate roughness could be attributed to the synergistic effect of several factors [6]. Firstly, higher MP statistics (density and sizes) could negatively impact the coating densification or compactness, culminating in lower hardness [4]. Figure 4-12 reveals no distinct correlation between the hardness and the MP statistics. Nonetheless, the highest hardness was obtained at the smallest average MP size (i.e., Very smooth), despite having the highest MP density. Quantitative analysis revealed that a 1 % change in the MP size resulted in a 6.97 % change in the hardness, compared to the 1.1 % observed for the MP density. This signifies greater sensitivity of the coating hardness to the average MP sizes rather than the density. Therefore, it appears that

the enhanced coating compactness offered by the smaller MPs in the Very smooth condition outweighed the negative impact of its higher MP density, resulting in improved hardness.

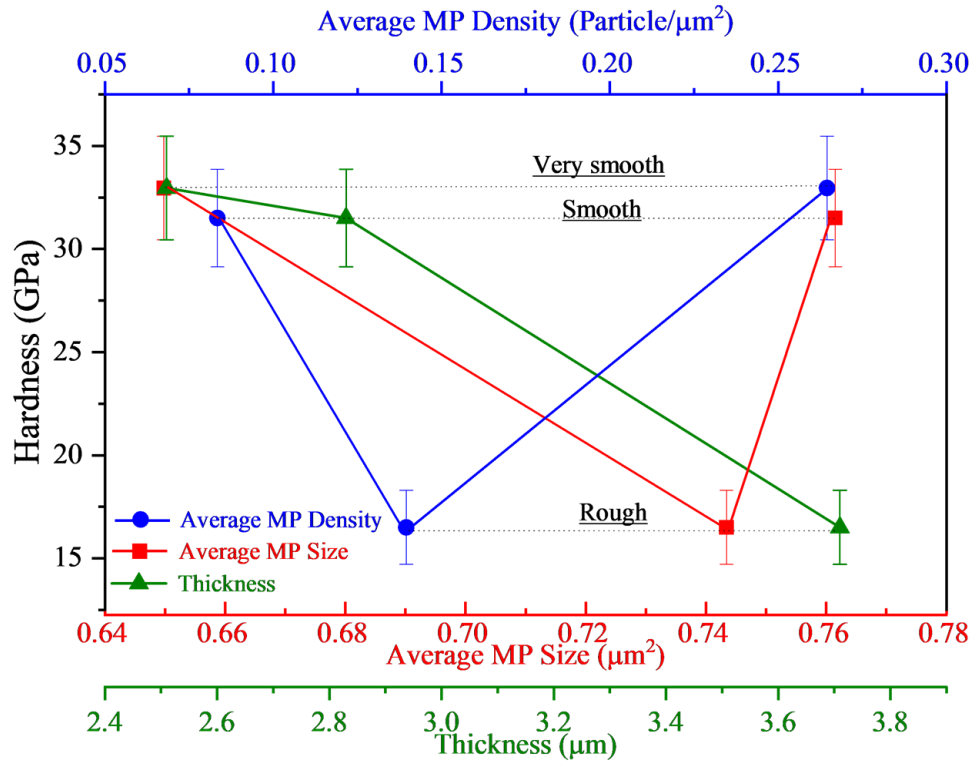


Figure 4-12: Relationship between MP statistics, thickness, and hardness of the AlTiN/AlTiCrN coatings.

Furthermore, the coating thickness could also affect the hardness [16]. From Figure 4-12, a reduction in hardness at higher coating thickness is observed (Very smooth to Rough). Previously, studies by Ling et al. [16] and Uddin et al. [18] revealed an increase in coating hardness with increasing thickness, citing the reduction in crystallite size. However, in this study, the average crystallite size was fairly constant (Figure 4-10), despite the significant changes in coating hardness, suggesting the influence of other factors. Figure 4-13 reveals that coating hardness increases with (111) plane orientation (Rough to Very

smooth). This improvement could be attributed to the increased solid solution strengthening offered by the dense (111) plane, impeding dislocation movement. This is consistent with previous reports by Zhou et al. [29], who observed a positive correlation between the (111) plane peak intensity and the hardness of AlTiCrN coatings.

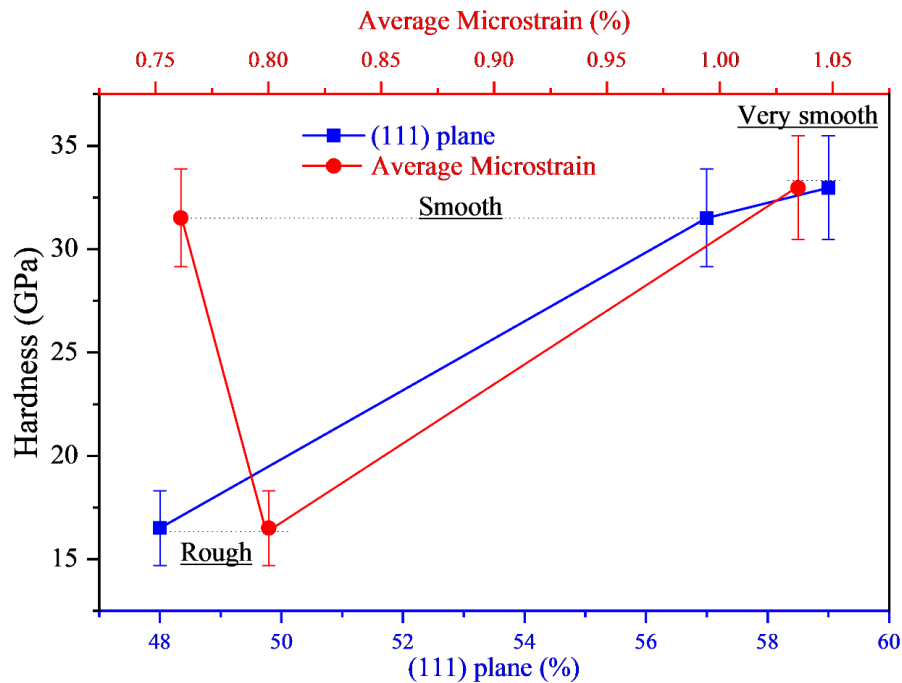


Figure 4-13: Relationship between (111) plane, average microstrain, and hardness of the AlTiN/AlTiCrN coatings.

Furthermore, microstraining resulting from the formation of a solid solution of metallic nitrides could further enhance hardness. The stress field resulting from lattice distortion serves to impede dislocation movement [46]. While Figure 4-13 shows no distinct trend between average microstrain and hardness, higher microstrain generally resulted in improved hardness (Very smooth). In the case of the Rough and Smooth conditions, the

higher hardness of the latter, despite exhibiting lower microstrain, could be due to the influence of the effect of other factors, such as the enhancement of the (111) plane in the Smooth condition. Further analysis to ascertain the sensitivity of the hardness to changes in the average microstrain and (111) plane composition revealed that a 1 % increase in the percentage of (111) plane and average microstrain resulted in 4.4 % and 2.8 % increments in the coating hardness, respectively. This suggests a greater impact of the (111) plane composition on the coating hardness than the microstrain.

Additionally, the coatings' fracture resistance, assessed using the H/E and H^3/E^2 ratios [4], was superior for the Very smooth condition, as shown in Figure 4-11. These ratios (H/E and H^3/E^2) reduced by 52 % and 88 % in the Rough condition, respectively. Similar variations in these ratios with substrate roughness have been previously reported by Meng et al. [47] and Uddin et al. [18]. The superior fracture resistance at lower substrate roughness could be attributed to the cumulative impact of enhanced densely packed (111) plane (Figure 4-8) and higher microstrain (Figure 4-10) serving as hindrances to crack initiation and propagation.

Overall, the hardness of the AlTiN/AlTiCrN coatings showed a significant correlation with (111) plane composition and coating thickness. Explicitly, the highest hardness occurred in the Very smooth condition, which combined the highest (111) plane composition and the lowest coating thickness. Furthermore, while the coating hardness is dependent on several factors, it appears to be more significantly impacted by the (111) plane orientation and MP size. This explains the superior hardness observed at lower substrate roughness, where the (111) plane composition is highest and MPs are finer. Meanwhile, the

fracture resistance metrics (H/E and H^3/E^2 ratios) followed an identical trend as the hardness, with the highest values at lower substrate roughness.

4.3.3 Adhesion strength

Figure 4-14a–c presents the scratch test results showing frictional force, acoustic emission, and normal load against the scratch length for the different substrate conditions.

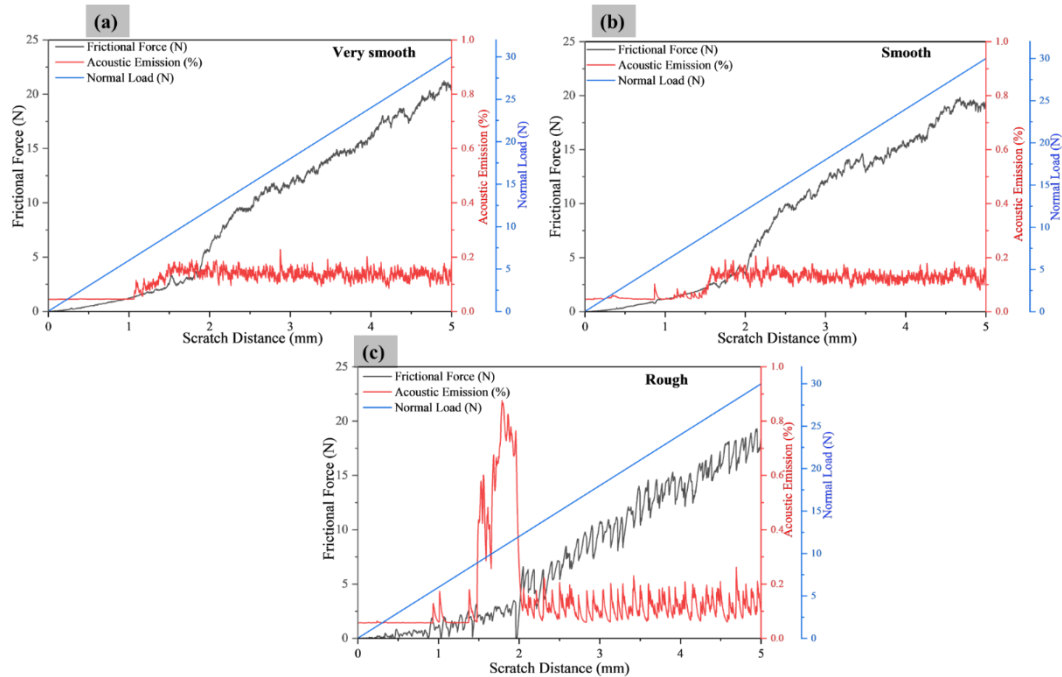


Figure 4-14: A plot of the frictional force, acoustic emission, and normal force against scratch length of the AlTiN/AlTiCrN coatings: (a) Very smooth; (b) Smooth; (c) Rough.

The acoustic emission signals, generated by indenter-coating interactions, have been used to determine critical coating failure points [48]. From the figure, the Very smooth condition showed negligible emissions in the first 1 mm, indicating the preservation of initial coating integrity. However, Smooth and Rough conditions exhibited peak acoustic emissions

reaching ~ 0.1 and 0.18 %, respectively, during the first 1 mm. Moreover, the maximum acoustic emissions generally increased with substrate roughness, reaching ~ 0.23 % at 3 mm (Very smooth), 0.21 % at 2 mm (Smooth), and 0.9 % at 1.7 mm (Rough). The higher maximum acoustic emissions and shorter distance of occurrence with increasing substrate roughness suggest a higher severity of coating failure and a shorter time to failure, in agreement with previous findings [49]. Furthermore, the fluctuations in the frictional force are also highest in the Rough condition, suggesting an increase in the COF of the AlTiN/AlTiCrN coatings [50]. This finding correlates with the results of the roughness (Figure 4-4), as rougher coatings are expected to have higher COF [51]. Cumulatively, the increase in acoustic emissions and frictional force with increasing substrate roughness suggests degradation in the coating adhesion strength. The coating adhesion strength was quantified using the critical load (normal load) corresponding to adhesive coating failure [48]. As shown in Figure 4-15 the Very smooth condition exhibited the highest critical load (~ 12 N), suggesting superior adhesion strength. The critical load reduced by 7 and 29 % in the smooth and Rough conditions, respectively.

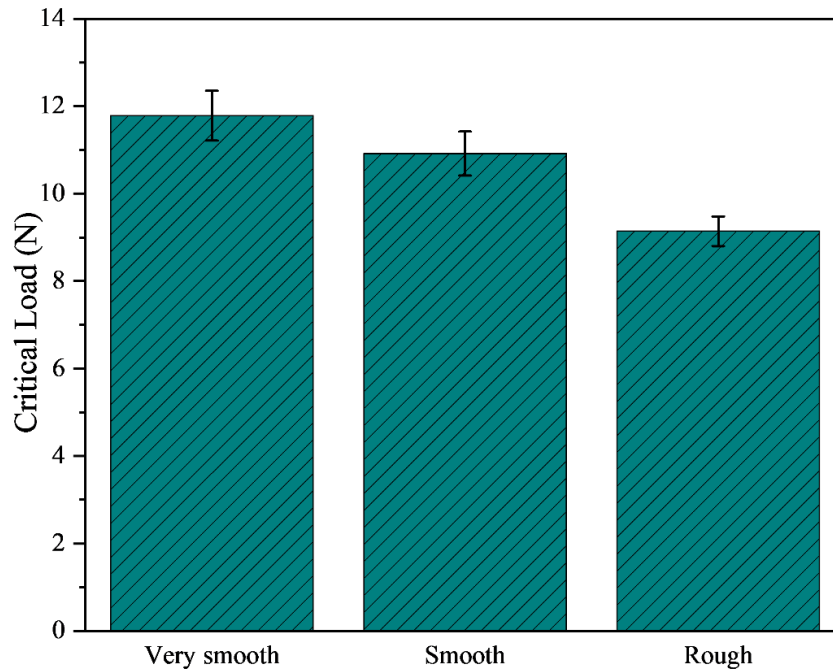


Figure 4-15: Critical load of the AlTiN/AlTiCrN coatings obtained from the scratch test for the different substrate conditions.

4.3.4 Wear performance

Figure 4-16 shows the wear performance of the AlTiN/AlTiCrN coatings, with the Very smooth condition exhibiting the least wear rate ($\sim 1.8 \times 10^{-4} \text{ mm}^3/\text{Nm}$), suggesting superior wear resistance. The wear rate increases with substrate roughness by 7 % (Smooth) and 17 % (Rough).

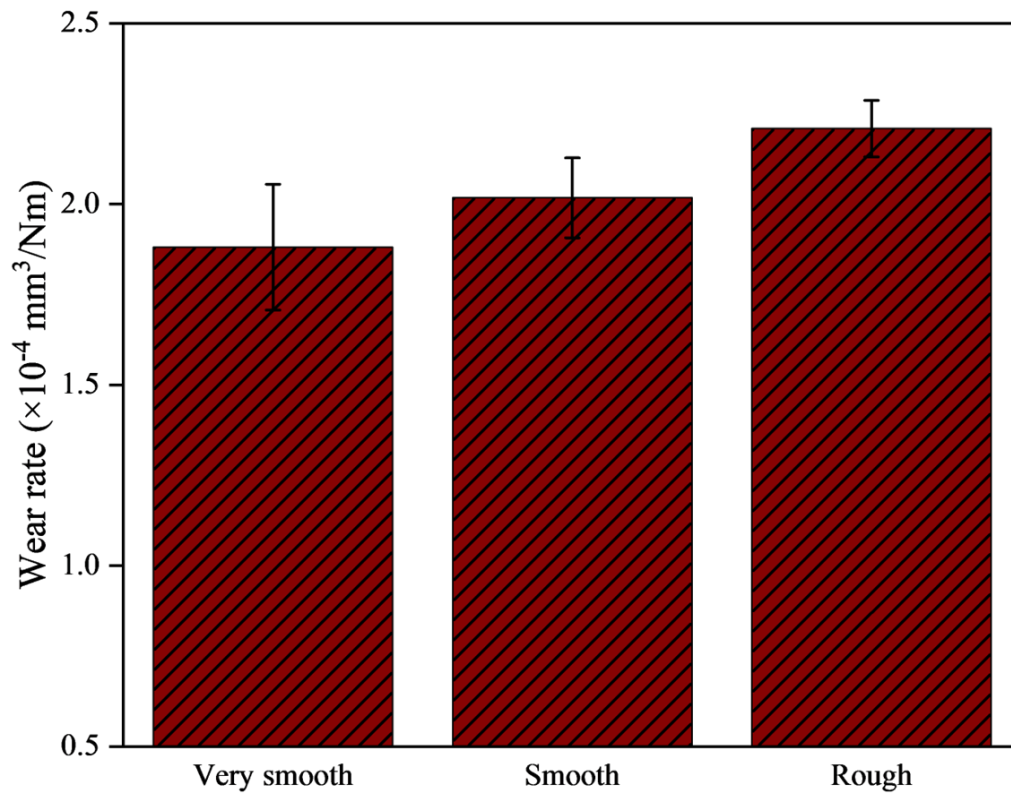


Figure 4-16: Wear rates of the AlTiN/AlTiCrN coatings for the different substrate conditions.

Figure 4-17 shows the SEM micrographs of the wear track, revealing three distinct zones (i.e., A, B, and C, as shown in Figure 4-17S). These zones have been discussed extensively in our prior work [26].

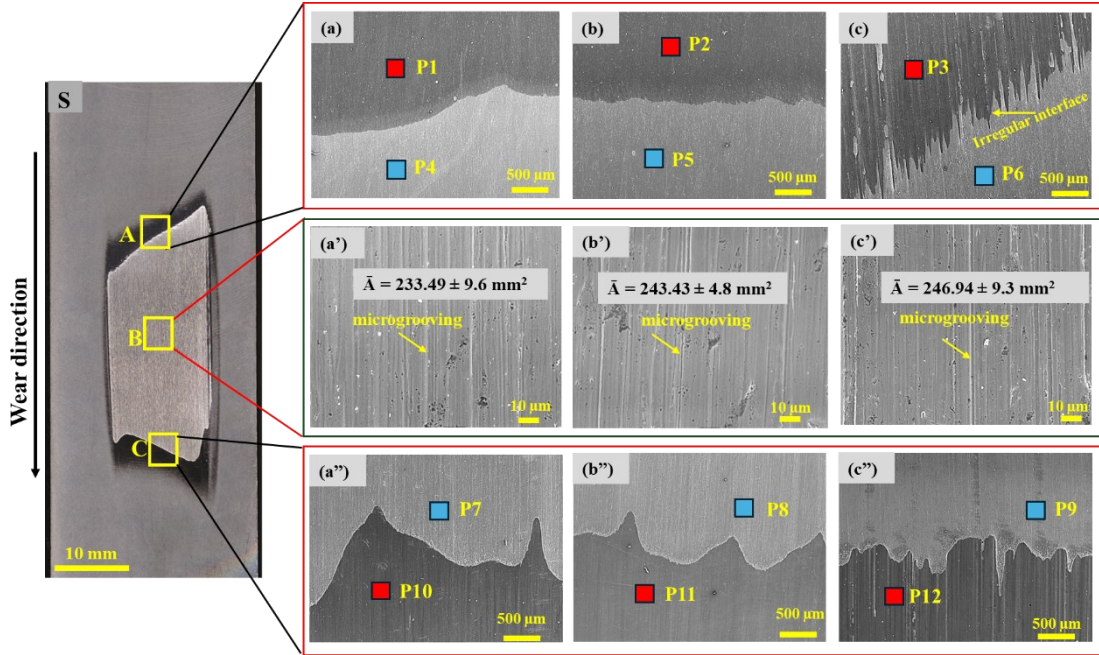


Figure 4-17: Micrographs of the wear tracks: (S) Overview of the wear track; (a), (a') & (a'') Very smooth; (b), (b') & (b'') Smooth; (c), (c') & (c'') Rough.

In zone A (Figure 4-17a–c), all the samples exhibited bright and dark regions. The interface morphology transitions from river patterns at lower substrate roughness (Figure 4-17a and b) to irregular asperities in the Rough condition (Figure 4-17c), likely connected to the higher substrate roughness. The dark regions identified by EDX (P1–3, Table 4-5) contain coating elements (Al, Ti, Cr, and N) and reduced Fe content. Notably, the relatively higher Cr and lower Fe content in the Rough condition (P3) suggests the persistence of the AlTiCrN top layer, unlike the other conditions (P1 and 2). This might be connected to the greater coating thickness in the Rough condition (Figure 4-6c). Meanwhile, bright regions represent substrate exposure (P4 – 6), indicated by high Fe content. Notably, the general increment in Fe content accompanied by depletion in Al and Ti with increasing substrate roughness (P4 – 6) suggests that the coating atoms are more adherent to the smoother substrate, consistent with the adhesion test results (Figure 4-15).

Table 4-5: EDX analysis of the wear track (Figure 4-17).

Spectrums	Composition (at. %)						
	Al	Ti	Cr	N	O	Si	Fe
P1	20.0	11.7	1.8	42.5	12.6	1.0	10.4
P2	23.0	14.9	0.6	46.9	9.0	0.3	5.3
P3	17.8	13.4	14.0	45.8	8.5	-	0.5
P4	9.0	3.0	5.6	16.1	11.3	1.7	53.3
P5	4.6	1.9	7.6	1.0	11.5	2.8	70.5
P6	0.6	0.2	9.7	-	16.3	4.1	69.1
P7	1.0	-	9.1	-	3.1	2.3	84.4
P8	0.9	0.1	6.8	-	18.5	2.4	71.3
P9	0.1	0.3	9.9	-	6.9	2.4	80.4
P10	16.4	11.4	13.9	45.4	11.9	<0.1	0.9
P11	18.2	12.7	13.2	37.9	17.2	0.1	0.8
P12	18.6	14.1	11.0	49.3	6.7	0.1	0.3

Expectedly, zone B (Figure 4-17a'-c') reveals the occurrence of abrasive wear in the form of microgrooving leading to substrate exposure (P7-9) while zone C (Figure 4-17a''-c'') exhibited distinct regions (P7-12) like zone A, for all conditions. Further analysis revealed that the maximum wear track depth occurred in the region of substrate exposure (Zone B), reaching about 0.2 mm for all conditions, which significantly exceeds the coating thickness (Figure 4-6), thereby further corroborating the occurrence of complete coating abrasion in this region. Furthermore, Si contamination (from the abrasive), known to facilitate tribochemical reactions was detected [1]. As shown in Table 4-5, higher Si levels

were detected in regions of substrate exposure (P4 – 9), underscoring the protective role of the AlTiN/AlTiCrN coatings. Meanwhile, the O detected likely originates from atmospheric exposure [52]. Further analysis of the exposed substrate area ($\bar{A}$) revealed that the Rough condition exhibited the highest exposed area (247 mm²), decreasing by ~ 2 % (Smooth) and 5 % (Very smooth). This correlates with the wear resistance results (Figure 4-16), as the minimal substrate exposure (Very smooth) aligns with its superior wear resistance.

4.3.5 Impact of substrate roughness on coating properties and performance

From the foregoing, increasing substrate R_a by 675×10^2 % (Very smooth to Rough) led to ~ 700 % rise in the coating's R_a , along with a 50 % increase in thickness. However, key mechanical properties, including hardness, H/E , H^3/E^2 and critical load, decreased by 54, 52, 88, and 29 %, respectively. Additionally, microstructural and morphological features such as the (111) orientation, microstrain, and MP density decreased by 19, 23, and 47 % while the MP size increased by 14 %. These were accompanied by a 17 % rise in the wear rate. These findings indicate that the coating's roughness is most impacted by substrate R_a variation, and that the substrate roughness exerts a ripple influence on the other coating properties and the resulting performance.

4.3.5.1 Influence of substrate roughness, coating thickness, and fracture resistance on adhesion

The coating adhesion strength was superior at lower substrate roughness (Figure 4-15). Previous reports indicated improvements in coating adhesion strength for rougher substrates, citing enhancement of mechanical interlocking [18], [20], [47]. Contrarily, the

improvement in the adhesion strength for the smoother substrate observed in this study could be explained in terms of the frictional forces and the surface area variation. Firstly, higher frictional forces in rough coatings (Figure 4-14) generate greater compressive stresses during tribological contact, thus accelerating delamination [15], [51]. Secondly, roughness-induced asperities create localized stresses which are usually concentrated on the peaks, leading to premature coating failure [20], [53].

Additionally, beyond the substrate roughness, the AlTiN/AlTiCrN coating adhesion strength could be influenced by the coating thickness and fracture resistance. From Figure 4-18, the critical load decreases with increasing thickness (Very smooth to Rough). This is consistent with Bull and Rickerby's model [33], whereby thicker coatings are expected to accumulate more elastic energy and may develop residual stresses during tribological interactions. This ultimately reduces the work of adhesion, thereby promoting crack formation [54]. Meanwhile, an increase in the critical load with the H/E and H^3/E^2 ratios is also observed (Rough to Very smooth). The enhanced fracture resistance of the AlTiN/AlTiCrN coatings deposited on the smoother substrate indicates that it can absorb more energy during tribological interactions without cracking. Consequently, the coating exhibits increased resistance to crack propagation, thereby lowering the chances of delamination when subjected to external loading [10]. Quantitatively, a 1 % decrease in the substrate R_a and coating thickness, as well as a 1 % increase in H/E and H^3/E^2 ratios, resulted in 0.04, 0.58, 0.56, and 0.33 % increments in the critical load. This implies that the coating adhesion strength is more impacted by the H/E ratio and thickness than by the other factors. In summary, the adhesion strength of the AlTiN/AlTiCrN coatings generally deteriorated

with increasing substrate roughness and was more significantly influenced by the fracture resistance indices and coating thickness.

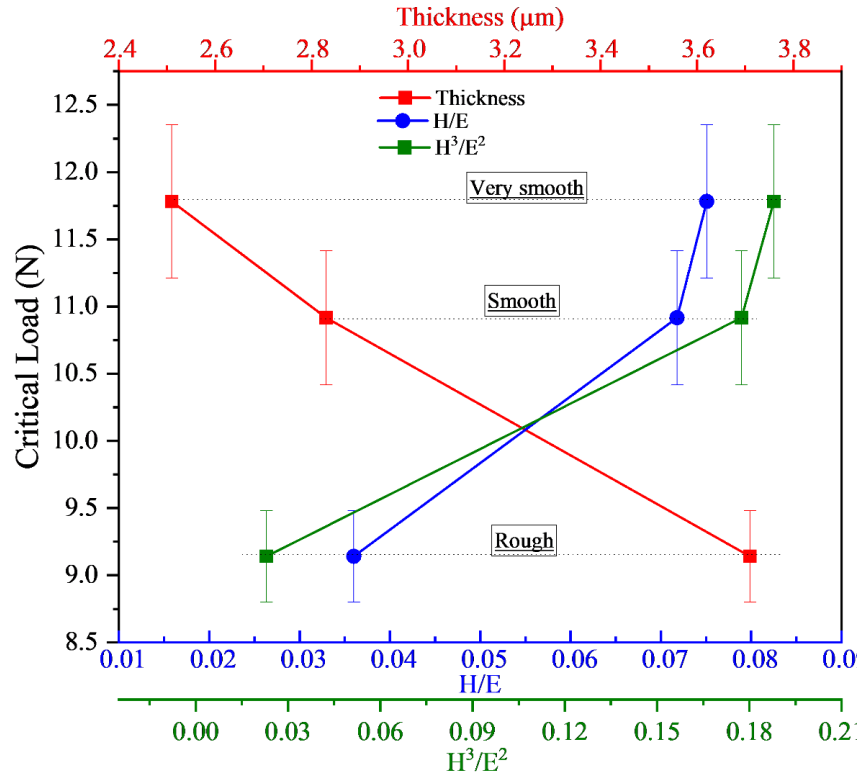


Figure 4-18: Relationship between coating thickness, indices of fracture resistance, and critical load of the AlTiN/AlTiCrN coatings.

4.3.5.2 Influence of coating mechanical and microstructural properties on wear resistance

The superior wear performance exhibited by the AlTiN/AlTiCrN coating deposited on the smoother substrate (Figure 4-16) could be explained in terms of several factors. Figure 4-19 shows decreasing wear rates of the AlTiN/AlTiCrN coatings with increasing hardness, H/E , and H^3/E^2 ratios. The lower wear on the smoother substrate could be attributed to the improved resistance to plastic deformation indicated by higher hardness [10]. Additionally, the enhancement in the coating's fracture resistance, evident in the higher H/E and H^3/E^2

ratios, contributes to lower wear rates by enhancing the coating's resistance to crack initiation and propagation [6]. Quantitatively, a 1 % increase in hardness, H/E , and H^3/E^2 ratios led to 0.35, 0.34, and 0.2 % reductions in the wear rates, respectively, indicating the comparable influence of hardness and the H/E ratio on the wear rates.

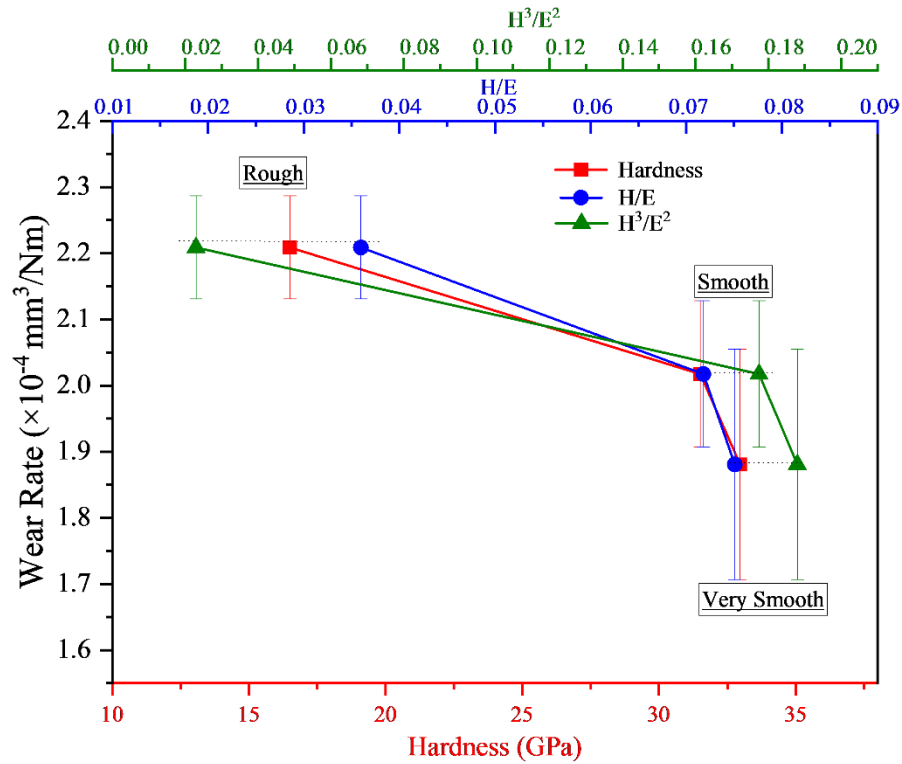


Figure 4-19: Relationship between hardness, H/E , H^3/E^2 ratios, and wear rates of the AlTiN/AlTiCrN coatings.

Figure 4-20 shows increased wear rates at higher coating thickness and R_a . It has been reported that thicker coatings often culminate in lower wear rates by distributing the external load over a greater volume, leading to lower shear stresses [55]. Contrarily, the lower wear rates of the thinner AlTiN/AlTiCrN coatings (Very smooth) suggest mitigating

effects of other factors such as hardness. Nonetheless, this result is consistent with Takadoun and Houmid's findings, who also reported higher wear rates in thicker coatings [33]. Meanwhile, the increase in wear rates with increasing R_a is likely due to reduced contact area and stress concentration at asperities [4]. Additionally, the lower wear rates at higher critical load (Very smooth) could be attributed to the enhancement of the coating bond strength, thereby reducing the chances of premature delamination during external loading [55]. Quantitatively, a 1 % increase in R_a and thickness, as well as a 1 % decrease in critical load, led to 0.02, 0.36, and 0.78 % increments in the wear rates, implying that adhesion has the strongest influence, followed by thickness, with R_a having the least impact.

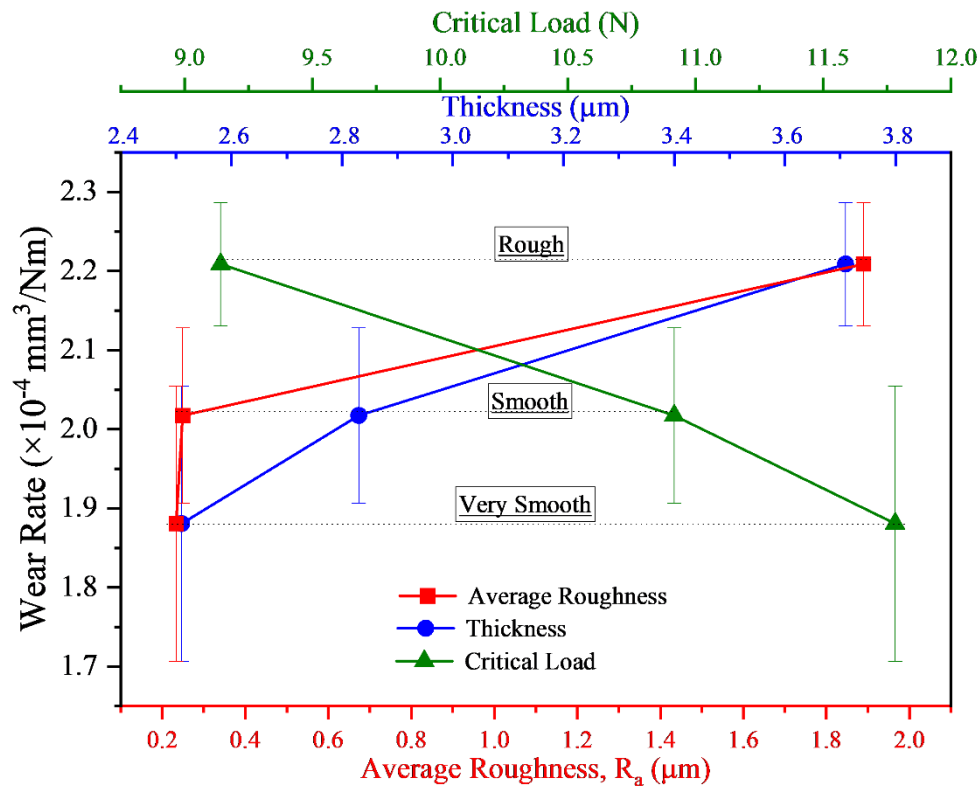


Figure 4-20: Relationship between the R_a , coating thickness, critical load, and wear rates of the AlTiN/AlTiCrN coatings.

Apart from the indirect impact of MPs on wear performance through roughness and hardness variation, MPs may directly affect the wear of the coatings by acting as a 4th body in the tribological system [4], in addition to the rubber wheel, abrasive, and the coated sample. Figure 4-21 shows no clear correlation between the MP density or size and wear rates. Nonetheless, a lower wear rate appears to be associated with higher MP density and smaller MP sizes (Very smooth). Higher MP density implies a likely increase in rolling interactions alongside the abrasive, thereby reducing direct contact between the abrasives and the coating, leading to lower material loss. This is consistent with the reports by Rabinowicz et al. [56], who noted that loose abrasive particles spend about 90 % of their time rolling, rather than abrasion. Moreover, smaller MP sizes may be easily displaced, promoting their rolling [57]. This might serve to further minimize direct contact, culminating in lower wear rates. Quantitatively, a 1 % decrease in the MP density and a 1 % increase in the MP size were accompanied by 0.37 and 1.2 % increments in the wear rates, respectively. This suggests that the MP size has a more significant impact on the coating wear rate than the density.

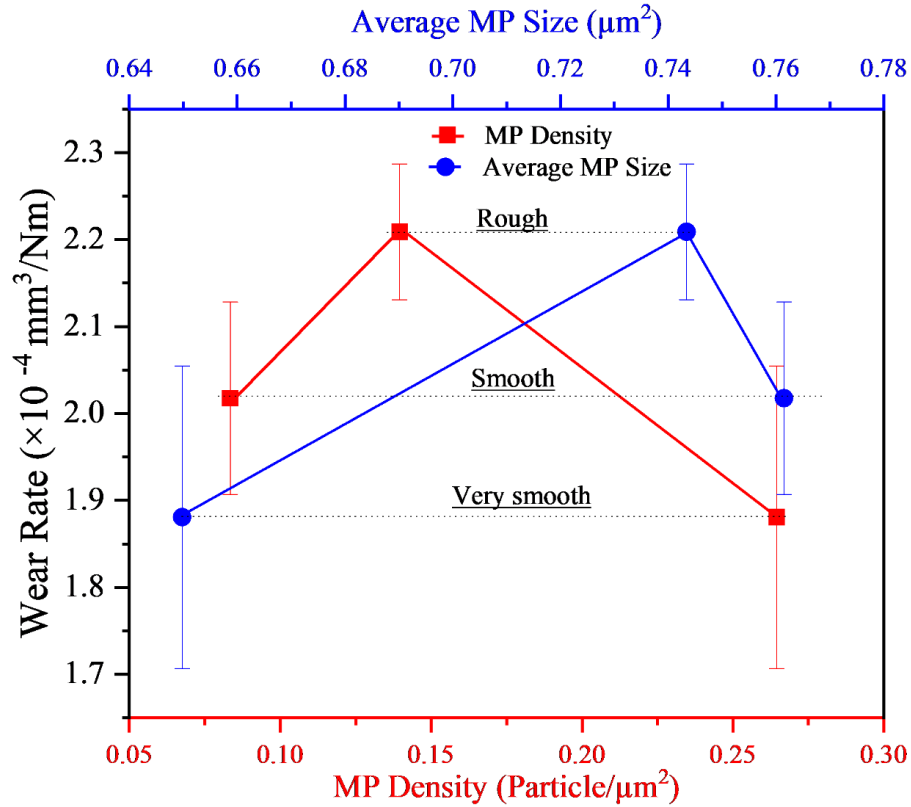


Figure 4-21: Relationship between the MP density, size, and wear rates of the AlTiN/AlTiCrN coatings.

Furthermore, a direct correlation was found between the (111) orientation and the wear resistance of the AlTiN/AlTiCrN coatings. A higher fraction of (111) planes was associated with reduced wear rates, observed in the Very smooth condition (Figure 4-9 and 18). Quantitative analysis showed that a 1 % decrease in the (111) plane composition (from Very smooth to Rough) led to about a 0.94 % increase in the wear rates. This is consistent with the findings from our recent study [10], where AlTiN/AlTiCrN-based coatings with stronger (111) orientation consistently demonstrated superior mechanical properties and tribological performance. Evidently, the improvement in the wear resistance with the

enhancement of the close-packed (111) plane can be attributed to the corresponding improvements in hardness, fracture resistance (Figure 4-11), and adhesion (Figure 4-15).

Overall, the wear performance of the AlTiN/AlTiCrN coatings depends on the coating properties, with the smoother substrates yielding the best results. The wear rate decreased with increasing hardness, H/E , H^3/E^2 , adhesion strength, and (111) orientation. Meanwhile, thickness and roughness increased with the wear rate, translating to poor performance (i.e., Rough). Furthermore, although the coating roughness was most influenced by the substrate roughness, it had the least impact on the wear rates. Instead, factors such as the (111) plane, MP size, adhesion strength, and hardness played more significant roles.

4.5 Conclusions

The key findings from the study on the effect of substrate roughness ($R_a = 0.0025$ to $1.7 \mu\text{m}$) on the tribological performance of AlTiN/AlTiCrN-coated AISI A8 tool steel are as follows:

1. Higher AISI A8 substrate roughness led to an increase in the coating roughness, degrading the tribological performance. However, the most significant changes in the coating roughness compared to the uncoated counterpart occurred at lower substrate roughness, correlated with the MP statistics.
2. The coating thickness increased with the substrate roughness, with all coatings exhibiting stoichiometric compositions. Additionally, as the substrate roughness increased, the preferred (111) orientation and microstrain reduced, while the crystallite size remained fairly constant.

3. The reductions in hardness, H/E , and H^3/E^2 with increasing substrate roughness were linked to the reduction in the (111) orientation, decrease in microstrain, and larger MP sizes, all contributing to higher wear rates. Notably, the (111) plane composition and the MP size had the most significant influence on the mechanical properties.
4. Smoother substrates yielded better adhesion strength. This was linked to reduced substrate roughness, thinner coatings, and improved fracture resistance, reflected in the higher H/E and H^3/E^2 ratios. Among these, the coating thickness and H/E ratio had the greatest impact on the adhesion strength.
5. AlTiN/AlTiCrN coatings deposited on the rougher substrates exhibited inferior wear resistance. This resulted from a reduction in (111) plane orientation, leading to reduced hardness, fracture resistance, and adhesion strength, as well as increased roughness and larger MPs. Notably, (111) plane orientation, adhesion strength, hardness, and MP size were the most critical factors.

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Chapter 5

Effect of layered architectural design on the tribological behaviour of AlTiN/AlTiCrN-based cathodic arc evaporated physical vapour deposition coatings

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Abstract

AlTiN/AlTiCrN coatings have received significant attention for their ability to enhance tribological performance, particularly in the woodworking industry. However, their tribological behaviour remains relatively under-explored. Therefore, this study aims to investigate the tribological behaviour of cathodic arc-evaporated coatings deposited on AISI A8 tool steel, including a bilayer AlTiN (I)/AlTiCrN coating (designated as AlTiCrN I) and two trilayer coatings (AlTiCrN II and III) with an AlTiN(I)/AlTiN (II)/AlTiCrN architecture, differing in composition and thickness. The intent is to forecast the performance of the coatings under woodworking conditions through microstructural, nanoindentation, adhesion, and wear characterizations. The results revealed that AlTiCrN II and III coatings exhibited fewer surface defects and lower surface roughness compared to AlTiCrN I. Compositional analysis indicated the enhancement of the (111) orientation from AlTiCrN I to III. Furthermore, while the AlTiCrN I exhibited a stoichiometric AlTiN (I) base layer, AlTiCrN II and III were characterized by non-stoichiometric AlTiN (II) base layers, which significantly improved the adhesion strength. The hardness, adhesion strength, and fracture

resistance indices consistently increased from AlTiCrN I to III. Notably, the AlTiCrN III coating presented a wear rate that was 15 % and 29 % lower than those of AlTiCrN II and I, respectively. This improvement is attributed to the synergistic effects of higher hardness, enhanced fracture resistance, better adhesion strength, and reduced surface roughness. These findings underscore the potential of engineered layered AlTiN/AlTiCrN-based coatings for enhanced tribological performance in woodworking applications.

Keywords: Coating architecture; AlTiN/AlTiCrN; Cathodic arc evaporation, Tribological performance

5.1 Introduction

In recent years, hard ceramic coatings produced via the cathodic arc evaporation physical vapour deposition (CAE-PVD) technique have become trendy in the machining industries, especially for woodworking and metal machining applications [1], [2]. This growing popularity stems from their numerous advantages, including cost-effectiveness, eco-friendliness, compatibility with a wide range of substrates, and their ability to significantly enhance tribological performance [3]. Some of the earliest commercially available CAE-PVD ceramic coatings, such as binary CrN and TiN, marked a breakthrough in the coating market. However, these coatings exhibited limitations, including relatively low hardness (< 25 GPa), limited thermal stability (< 700 °C), and moderate wear resistance (wear rates: $10^{-6} - 10^{-7}$ mm³/N.m) [3], [4]. These challenges paved the way for further innovations and the development of advanced ceramic coating with superior performance.

The quest for coatings with enhanced tribological performance has driven significant advancements in the development of multicomponent coatings. Notable advancements from

these efforts include the development of AlCrN and AlTiN ternary coatings [5], [6]. These coatings leverage the benefits of Al alloying, such as the solid solution strengthening and passive oxide formation, to achieve remarkable improvements, including increased hardness (up to 30 GPa), enhanced thermal stability (up to 750 °C), and up to 30 % increase in tribological performance [4], [7]. However, several researchers have reported deterioration in the tribological performance of these coatings when the Al concentration exceeds 70 at.%, owing to the formation of the detrimental hexagonal AlN phase [8], [9], [10]. During the last 20 years, multicomponent quaternary AlTiCrN coatings have emerged as promising candidates for various machining applications, with significant research devoted to metalworking applications [11], [12]. However, despite their success in metal machining applications, their potential for the woodworking industry remains relatively underexplored. This offers the opportunity for further research into their advantages under such unique tribological conditions.

Nevertheless, it is important to mention that the properties and performance of the CAE-PVD coatings are highly influenced by the deposition parameters. Two parameters that have been researched extensively are the negative substrate bias and supply of the reactive N₂ gas. For instance, Romero et al. [13] observed that increasing the negative bias from 50 to 400 V resulted in an increase in hardness and improved tribological performance of AlCrN coatings. This occurrence was attributed to coating densification and a reduction in surface roughness as the bias increased. However, findings by Vlasveld et al. revealed that higher negative bias (50 to 250 V) led to a reduction in the coating's adhesion strength, characterized by increased compressive residual stresses caused by the continuous energetic coating bombardment by incoming adatoms at elevated bias [14]. Similarly, the supply of

reactive N₂ gas plays a pivotal role. Gilewicz et al. reported that increasing the pressure of the reactive N₂ gas from 1 to 5 Pa resulted in coatings with fewer defects, lower roughness, and increased hardness. The increase in coating hardness was correlated with the elimination of the detrimental hexagonal AlN phase with increased N₂ supply. However, the adhesion strength, fracture resistance, and wear rate did not improve beyond 3 Pa [5]. These findings collectively emphasize the critical importance of precise and optimal control of the deposition parameters to maximize the tribological performance of CAE-PVD coatings.

Recently, layered architectural CAE-PVD coating designs have received significant attention, particularly from practitioners in the machining industry [15], [16]. These multilayered coatings offer the possibility to combine properties of different coating-types, thus, creating the potential of achieving performance characteristics that might be challenging to achieve in the monolayer designs [3]. The most common layered architectural design is one in which all the individual layers have different elemental compositions. This could be achieved by varying the target composition during the deposition process. A noteworthy example of this type of architectural design is exemplified in a recent investigation conducted by Chang et al. [17], where the properties of Ti/AlTiCrN, TiN/AlTiCrN, Cr/AlTiCrN, and CrN/AlTiCrN deposited on steel substrates were investigated. Their results demonstrated that such architectural variations could lead to up to a 15 % difference in residual stresses and a 20 % difference in hardness. Another popular layered design involves repeating two distinct layers with different elemental compositions throughout the coating thickness. This is exemplified in the reports of Kong et al. [16], who investigated the properties of three bilayers of CrN/CrCN coating deposited on a steel substrate. They found that the multilayer design presented superior performance compared

to the monolayer counterparts. In another study, Warcholinski and Gilewicz [18] investigated the tribological behaviour of seven bilayers of TiN/TiAlN and CrN/CrCN coatings deposited on a steel substrate. They observed that the CrN/CrCN presented superior tribological performance. Consequently, from these findings, it could be deduced that the layered architectural designs hold promising potential for enhanced tribological performance in wood machining applications.

The complexity and demanding operating conditions encountered during wood machining pose significant challenges that degrade cutting performance. During the operation, cutting tools are subjected to severe abrasion from hardwood fibers, impact load from tree trunks and knots, tribochemical degradation caused by contaminants (e.g. silica) and organic acids (e.g. acetic acid), high-temperature oxidation (up to 800 °C) due to frictional forces, as well as adhesive wear from resinous wood species. These factors collectively contribute to accelerated tool wear, increased power consumption, and poor surface finish quality of the machined products [3], [19]. Although conventional coatings have demonstrated improvements in cutting tool performance, they still fall short in addressing these challenges. Fahrussiam [20] reported that TiAlN, TiSiN, and TiAlN/TiBON coatings reduced wear of K-10 cutting tool by at least 30 % during the machining of wood species including particleboard and fiberboard, attributing the improvement to increased hardness, reduction in friction coefficient and enhanced oxidation resistance. Building on this, layered AlTiN/AlTiCrN-based coatings offer promising tribological performance by leveraging their layered architecture and quaternary AlTiCrN component, resulting in improved mechanical properties, oxidation resistance, and chemical stability, thus, leading to prolonged cutting tool life. Some studies have investigated the properties and performance

of AlTiN/AlTiCrN coatings. Santana [21] reported that the AlTiN/AlTiCrN layered architecture exhibited finer grains and columnar structure with a preferred orientation, leading to enhanced coating hardness. Additionally, the coatings also enhance corrosion performance. Additionally, recent field trials by Vanella et al. [22] to ascertain the suitability of AlTiN/AlTiCrN coatings on AISI A8 and W360 tool steels for wood-cutting applications revealed about 17 – 57 % reduction in wear volume compared to the uncoated counterparts, confirming their effectiveness in extending cutting tool life.

A literature survey revealed a significant gap in the available studies examining the tribological behaviour of AlTiN/AlTiCrN coatings, despite their increasing recognition for enhancing tribological performance in woodworking applications. The existing research has focused predominantly on the quaternary multicomponent monolayered AlTiCrN coating [23] or multilayered AlTiCrN coatings involving other binary (such as TiN and CrN) or ternary (e.g. TiSiN) components [17]. In fact, to the best knowledge of the authors, no prior study has systematically investigated the tribological performance of the layered AlTiN/AlTiCrN, particularly in relation to the effects of deposition parameters. Previous AlTiN/AlTiCrN layered coating designs were achieved by varying the elemental composition while maintaining constant deposition parameters. This is exemplified in the studies conducted by Santana [21] where different coating designs with varying Cr content were developed. This presents a critical knowledge gap as the tribological properties of the coatings are significantly impacted by the process parameters. In this study, the tribological behaviour of layered AlTiN/AlTiCrN-based coatings deposited on AISI A8 tool steel substrates, widely used in the wood-cutting industry, was investigated. The study aims to assess the combined tribological advantages of the layered AlTiN/AlTiCrN architectural

design alongside the quaternary multicomponent AlTiCrN coating. These layered coating architectures were uniquely developed by varying the flow of the N₂ reactive gas supply and the bias during the deposition of the individual layers, offering a novel approach to tailoring the coating properties. Furthermore, the review of literature revealed a lack of studies examining the effect of substrate bias and N₂ reactive gas supply on the properties of AlTiN/AlTiCrN layered coatings, despite their potential to significantly impact the coatings' tribological performance. Therefore, this study directly addresses this knowledge gap by simultaneously evaluating these parameters and their effects on the tribological behavior of AlTiN/AlTiCrN-based coatings. The findings provide valuable insights into optimizing these parameters for improved performance in woodworking applications.

5.2 Experimental details

5.2.1 Substrate material and preparation

AISI A8 cold-work tool steel was used as the substrate material for the investigation. The chemical composition is presented in Table 5-1. The material finds application in the manufacture of cutting tools for machining applications such as woodworking [19]. Before the coating operation, the samples, with dimensions of 75 × 25 × 10 mm, were hardened to achieve a bulk hardness of 56 – 57 HRC. Thereafter, the surfaces of the hardened samples were profiled by mechanical wheel grinding and polishing to achieve an average roughness (R_a) of about 1.6 μm. The samples were subsequently decontaminated by cleaning with mild solvent-free detergent, followed by rinsing with water and vacuum drying in preparation for the coating deposition process.

Table 5-1: Chemical composition of the AISI A8 tool steel (% wt.).

C	Cr	Si	V	Mn	Ni	Cu	Mo	W	Fe
0.48	7.90	0.87	0.36	0.46	0.14	0.17	1.41	<0.01	Balance

5.2.2 Coating deposition

Coating deposition was achieved using a cathodic arc evaporator PVD system (PLATIT PL1011, Switzerland). The deposition chamber comprised high-purity planar targets, including two $\text{Al}_{67}\text{Ti}_{33}$, $\text{Al}_{50}\text{Ti}_{50}$, and pure Cr. The prepared samples were loaded into the deposition chamber by mounting on a sample holder continuously rotated at 3 – 5 rpm around the central vertical axis. Rotating the samples ensures uniformity in the deposition process [3]. In order to prevent contamination during coating deposition, the chamber was evacuated to a vacuum of $1 - 2 \times 10^{-5}$ mbar [24]. Subsequently, the samples were heated to 380 – 400 °C for about 45 minutes as a precursor to the etching process. Etching the samples was necessary to eliminate possible residue surface contaminants, particularly oxides and absorbed gases [3]. The etching was achieved using an inert gas (Ar) and $\text{Al}_{67}\text{Ti}_{33}$ ion etching. The subsequent coating deposition process was designed to achieve three different AlTiN/AlTiCrN-based architectures denoted as AlTiCrN I, II, and III, as shown in Figure 5-1.

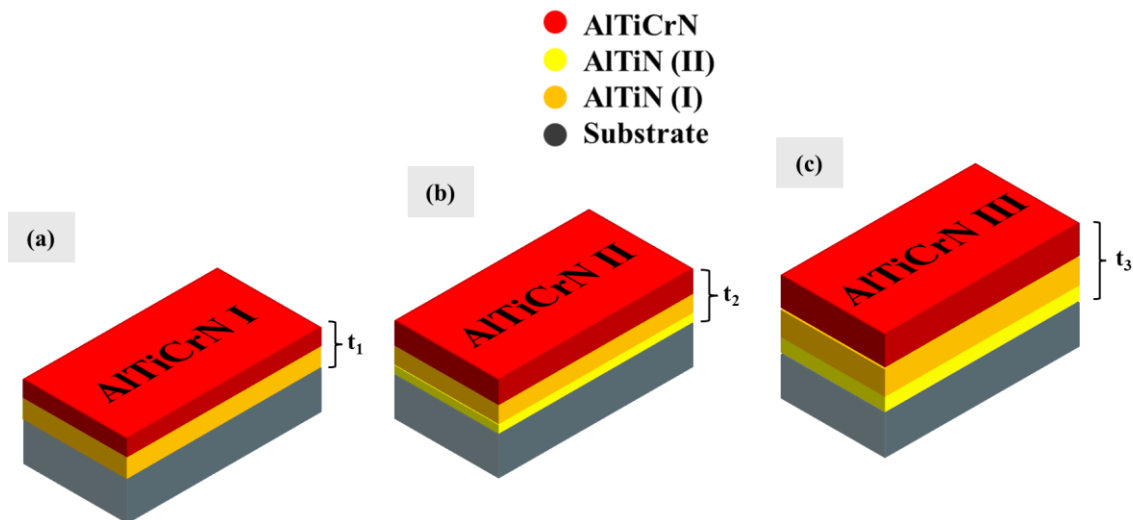


Figure 5-1: Schematic of the different AlTiN/AlTiCrN-based coating architectures: (a) AlTiCrN I; (b) AlTiCrN II; (c) AlTiCrN III (t_1 , t_2 , and t_3 represent the thickness of the respective architectures with $t_3 > t_2 > t_1$).

From the figure, AlTiCrN I architecture features a bilayer design comprising an AlTiN (I) base layer and an AlTiCrN top layer. The AlTiN (I) base layer was deposited using $\text{Al}_{67}\text{Ti}_{33}$ and $\text{Al}_{50}\text{Ti}_{50}$ targets at a temperature of $350 - 370\text{ }^{\circ}\text{C}$, an arc current of $250 - 270\text{ A}$, and a negative bias (U_b) of $130 - 150\text{ V}$, in the presence of pure reactive N_2 gas flowing at $640 - 660\text{ sccm}$ (f_N). The top AlTiCrN layer was deposited using $\text{Al}_{67}\text{Ti}_{33}$, and Cr targets while maintaining the same parameters as in the AlTiN (I) base layer, but with a higher N_2 gas supply ($\sim 1.06f_N$). This bilayer AlTiCrN I architecture achieved a total thickness of approximately $3.7\text{ }\mu\text{m}$. In AlTiCrN II and III architectures, a third AlTiN (II) base layer was introduced. This additional layer was deposited under similar conditions as the AlTiN (I) base layer in AlTiCrN I but with a reduced N_2 gas supply ($\sim 0.83f_N$ for AlTiCrN II and $\sim 0.77f_N$ for AlTiCrN III). The lower N_2 gas supply during the deposition of the AlTiN (II) base layer, peculiar to the AlTiCrN II and III architectures, is consistent with the findings from previous studies showing that reduced N_2 gas flow results in a non-stoichiometric

composition. This might in turn enhance coating adhesion by minimizing the hardness differential between the coating and substrate [25], [26]. Meanwhile, the intermediate AlTiN (I) layers in the AlTiCrN II and III architectures were deposited under similar conditions as the AlTiN (I) base layer in AlTiCrN I architecture, but with an increased negative bias ($\sim 1.4U_b$) for both architectures and, a slightly higher N_2 gas supply ($\sim 1.03f_N$) for the AlTiCrN III architecture. The choice of a higher substrate bias for the AlTiCrN II and III architectures aligns with findings from previous literature, which indicate that increasing substrate bias promotes the formation of denser and more compact structures with fewer defects, leading to improved properties and enhanced tribological performance [13], [14], [25]. Finally, the top AlTiCrN layers in the AlTiCrN II and III architectures were deposited under similar conditions as the top AlTiCrN layer in AlTiCrN I, but with a higher bias ($\sim 1.4U_b$) and reduced N_2 gas supply ($\sim 1.03f_N$ for AlTiCrN II and f_N for AlTiCrN III). This resulted in a total thickness of approximately 4.7 and 4.9 μm for the AlTiCrN II and III architectures, respectively. The key differences between the coating architectures are:

- AlTiCrN I is a bilayer architecture while AlTiCrN II and III are trilayer architectures deposited using relatively higher substrate bias compared to AlTiCrN I.
- The coating thickness increases from AlTiCrN I to III.
- The third AlTiN (II) base layer introduced for the AlTiCrN II and III architectures were deposited under a relatively low supply of N_2 gas compared to the AlTiN (I) base layer in the AlTiCrN I architecture. The supply is lowest for AlTiCrN III architecture.

- The N₂ gas supply during the deposition of the AlTiN (I) layer generally increased from AlTiN I to III.
- The N₂ gas supply during the deposition of the AlTiCrN top layers generally reduced from AlTiCrN I to III.

Table 5-2 provides a detailed summary of the deposition parameters for the coating architectures.

Table 5-2: Deposition parameters (i.e. temperature, bias, nitrogen flow rate, and arc current) for the different AlTiN/AlTiCrN-based coating architectures.

Layer	AlTiCrN I	AlTiCrN II	AlTiCrN III
AlTiN (I)	T = 350 – 370 °C	T and AC same as AlTiCrN I	T and AC same as AlTiCrN I
	AC= 250 – 270 A	SB=1.4 U _b	SB=1.4 U _b
	SB= 130 – 150 V = U _b	N = f _N	N = 1.03f _N
	N=640 – 660 sccm=f _N		
AlTiN (II)	–	T = 350 – 370 °C	T, AC, and SB= U _b same as AlTiCrN II
		AC= 250 – 270 A	N=0.77f _N
		SB= U _b	
		N=0.83f _N	
AlTiCrN	T = 350 – 370 °C	T and AC same as AlTiCrN I	T and AC same as AlTiCrN I
	AC= 250 – 270 A	SB=1.4 U _b	SB=1.4 U _b
	SB= 130 – 150 V = U _b	N = 1.03f _N	N = f _N
	N=1.06f _N		

*T–Temperature; AC–Arc current; SB–Substrate bias; N–Nitrogen flow rate.

5.2.3 Microstructure and profilometry

The surfaces of the coated samples were examined for the presence of deposition defects, including Macroparticles (MPs) and craters, using a scanning electron microscope (SEM, JSM-6480LV, Japan) equipped with an energy-dispersive X-ray (EDX, INCA energy 250). Post-image analysis of the micrographs, to obtain MPs statistics, was performed using Image J software (ImageJ, version:1.54d) [27]. The methodology used for this analysis has been described elsewhere [28]. For the analysis, ten images with a magnification of $\times 1000$ were used for each coating architecture. The surface roughness of the samples before and after coating deposition was evaluated using a stylus-based profilometer (Mahr MarSurf PS 10, Germany). The evaluation yielded the R_a and the peak-to-valley height (R_z). The resulting R_a and R_z represent the average of ten measurements taken for each coating architecture. X-ray diffraction technique (XRD, D8 Discover, Bruker AXS, Germany) with Cu α radiation source was used for phase composition analysis. A step size of 0.05° , step time of 0.5 s/step, and a 2θ diffraction angle ranging from $30 - 50^\circ$ were used for the analysis. Sample preparation for cross-sectional examination to reveal coating thickness was carried out using a technique described in our previous work [28]. The resulting coating thickness also obtained using Image J software represents the average of nine measurements taken from three images for each coating architecture. Following this, EDX analysis was employed to evaluate the coating composition on an elemental basis.

5.2.4 Mechanical properties

The hardness (H) and elastic modulus (E) of the samples were evaluated using a nanoindenter (Anton Paar MCT³). An indenting load of 20 mN was used while maintaining a penetration depth below 10 % of the coating thickness [29]. This was done to minimize the

influence of the softer substrate and improve the measurement accuracy [3]. The resulting coating hardness is the average of at least five measurements taken for each coating architecture. The adhesion strength of the coatings was assessed using qualitative and quantitative techniques. The qualitative assessment was performed per the Daimler-Benz test by indenting the coatings using a Rockwell C indenter (QualiRock-RS, QUALITEST, USA) with a force of 150 Kgf for 25 seconds [30]. During the analysis, three indentations were made for each coating architecture. The deformed coating was subsequently examined using an optical microscope (Nikon Eclipse ME600, Japan) and compared with the VDI 3198 standard [31]. For the quantitative assessment of coating adhesion strength, scratch testing was performed (Anton Paar MCT³). For this analysis, the test was repeated thrice for each coating architecture. During the test, a Rockwell-type indenter having a tip radius of 20 μm under progressive loading from 0 to 30 N was used. The respective indenter speed, loading rate, and scratch length were 3.5 mm/min, 20.98 N/min, and 5 mm.

5.2.5 Wear test

There are different techniques for evaluating the wear resistance of CAE-PVD coatings. However, most of the existing methods are based on two-body abrasion which does not replicate the actual conditions encountered during woodworking, especially green wood machining. Moreover, the findings from our previous work indicated that the method of wear testing can significantly impact the performance of AlTiN/AlTiCrN-coated tool steels [28]. Consequently, the wear performance of the samples was evaluated using the dry sand rubber wheel (DSRW) apparatus, following the ASTM G65 standard under ambient conditions [32]. The test is based on three-body abrasion where the coated sample, abrasive silica sand, and the rubber wheel represent the cutting tool, wood contaminants, and the work piece (i.e.

wood). During the test, the stationary sample undergoes continuous abrasion by silica sand particles (approximately 200 – 300 μm) while being pressed against a 228 mm chlorobutyl rubber wheel. The test parameters are summarized in Table 5-3.

Table 5-3. Test parameters for the dry sand rubber wheel test for wear evaluation.

Parameters	Details
Test type	Procedure B
Silica sand	AFS 50/70
Force against specimen	130 N
Number of revolutions	2000 rev
Linear abrasion	1436 m
Sand flow rate	300 g/min
Rotational speed	200 rpm (2.4 m/s peripheral wheel speed)

After the testing, the wear of the coated samples was evaluated using the mass loss method, with a 1 mg precision balance. This included computing the mass difference before and after the test. The resulting mass loss represents the average of three measurements for each coating architecture. The wear volume and wear rate were subsequently determined using Equations 1 and 2 respectively [12]:

$$\text{Wear volume (mm}^3\text{)} = \frac{\text{Mass loss (g)}}{\text{Density (}\frac{\text{g}}{\text{mm}^3}\text{)}} \quad (1)$$

$$\begin{aligned} \text{Wear rate (}\frac{\text{mm}^3}{\text{N.m}}\text{)} \\ = \frac{\text{Wear volume (mm}^3\text{)}}{\text{Normal force (N)} \times \text{sliding distance (m)}} \end{aligned} \quad (2)$$

In order to ensure consistency in the wear volumes of the coated tool steels, it was pertinent to account for the reduction in the rubber wheel diameter. This was achieved by

measuring the diameter of the rubber wheel after each test and subsequently computing the adjusted wear volumes using Equation 3 [32].

$$\begin{aligned} & \text{Adjusted wear volume (mm}^3\text{)} \\ &= \text{wear volume} \times \frac{228.6 \text{ mm}}{\text{Wheel diameter after test (mm)}} \end{aligned} \quad (3)$$

Upon completing the wear test, SEM-EDX analysis of the wear track was conducted to identify the predominant wear mechanisms. Additionally, the area of the exposed substrate was quantified using Image J software while the wear volume of the worn sample was quantified using a 3D confocal profilometer (Keyence VR-5200, Canada). The resulting exposed area is the average of three measurements.

5.3 Results and discussion

5.3.1 Microstructure analysis

5.3.1.1 Surface morphology, macroparticle statistics and surface roughness

Figure 5-2 shows the micrographs of the surface morphology of the different AlTiN/AlTiCrN-based coating architectures. Grooves emanating from substrate profiling are evident in all architectures, highlighting the role of the substrate profile on the morphology of the deposited coatings [33]. The surfaces are characterized by deposition defects, namely, MPs and craters. From the figure, the MPs present several morphologies, including irregular, elongated, and spherical. This is typical for CAE-PVD coatings [3]. These defects pose a significant setback for the coatings owing to their negative impact on their tribological performance [25], [34], [35]. The following explanations have been offered

regarding MP formation: (1) They could be formed as a product of combined thermal shock and hydrodynamic effects occurring on the target, leading to molten particles ablation, which eventually solidify on the substrate [3]. (2) They could also be formed as a result of the generated plasma causing microexplosions on the target, leading to the ejection of molten particles, which ultimately solidify on the substrate [36]. (3) MPs might also result from the collision between the evaporated coating species during transportation, leading to their agglomeration before arriving on the substrate [37]. Meanwhile, the craters are considered the outcome of geometric shadowing by the MPs and become visible after the flaking of loosely bound MPs [35]. They could also result from an incomplete reaction between the reactive gas and coating atoms [38].

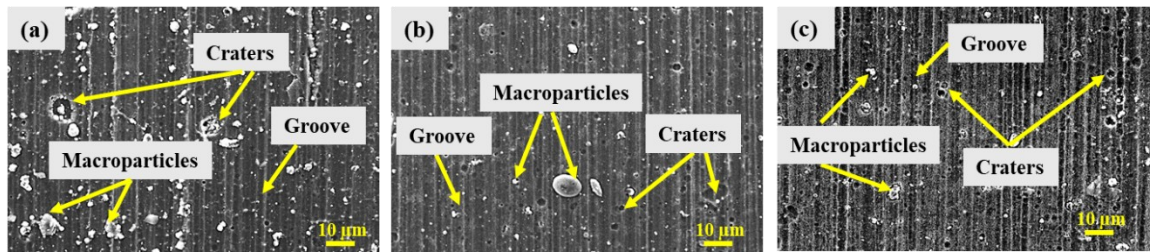


Figure 5-2. SEM micrographs showing the surface of the different AlTiN/AlTiCrN-based coating architectures: (a) AlTiCrN I; (b) AlTiCrN II; (c) AlTiCrN III.

Figure 5-3a shows the density and average size of the MPs. From Figure 5-3a, AlTiCrN I architecture showed the highest MP density of about $0.14 \text{ particle}/\mu\text{m}^2$. This value was observed to be reduced by 50 % and 42 % in AlTiCrN II and AlTiCrN III architectures, respectively. The average size of the MPs is also the largest for the AlTiCrN I ($0.74 \mu\text{m}^2$). It reduced to 0.59 and $0.39 \mu\text{m}^2$ in AlTiCrN II and III, respectively. Further analysis of the MP size distribution is presented in Figure 5-3b. The analysis revealed that

some of the MPs reached dimensions of $1 \mu\text{m}^2$. However, the majority have dimensions typically $< 0.3 \mu\text{m}^2$. Similar MP statistics have been previously reported for CAE-PVD coatings [35], [39]. Figure 5-3b also revealed that the AlTiCrN I architecture presented the highest MP counts for all the size ranges. This justifies the greater MP density of the AlTiCrN I as compared to the other architectures. This is followed by the AlTiCrN II, except for the $0 - 0.3 \mu\text{m}^2$ size range, where the AlTiCrN III showed a higher MP count. Consequently, the greater MP count in this size range ($0 - 0.3 \mu\text{m}^2$) observed in the AlTiCrN III could have contributed to its greater MP density as compared to the AlTiCrN II architecture.

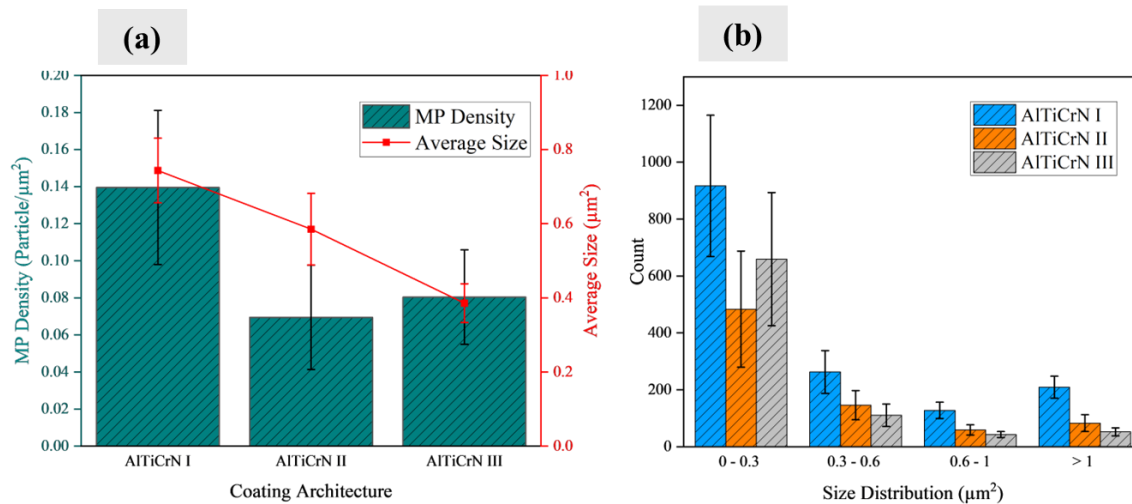


Figure 5-3: MP statistics of the different AlTiN/AlTiCrN-based coating architectures:

(a) MP density and average size and (b) MP size distribution.

The observed variation in the MP densities of the coating architectures could be linked to the deposition parameters. Explicitly, the 50 % and 42 % reductions in MP densities observed in AlTiCrN II and III architectures respectively, could be connected to the dominating effect of the 40 % increase in substrate biasing in these coatings [3]. Researchers have extensively reported the reduction in coating defects (i.e. macroparticles) with

increasing substrate bias in CAE-PVD coatings [37], [40], [41]. Two mechanisms put forth by researchers to explain MP reduction with increased biasing are the ion bombardment-resputtering phenomenon and the electrical repulsive mechanism. With respect to the former, increased substrate biasing energizes the MPs, resulting in a more momentous substrate bombardment. Eventually, this leads to the resputtering of loosely bound MPs [41]. Meanwhile, according to the electrical repulsive mechanism, the increase in collision frequency between MPs and electrons in the plasma stream at higher biases results in more negatively charged MPs. These negatively charged MPs are subsequently repelled upon arriving at the negatively biased substrate [42].

On the other hand, the 8 % lower MP density observed in the AlTiCrN II compared to AlTiCrN III, having similar substrate bias, could be connected to the 3 % higher N₂ gas supply in the former, particularly during the deposition of the top AlTiCrN layer (see Table 5-2). This is in agreement with the findings of Warcholinski et al. [25] who observed a reduction in macroparticles at higher reactive N₂ gas flow rates. In this regard, the MP reduction could be explained by the “target poisoning” phenomenon [3]. Accordingly, at a higher supply of N₂ reactive gas, compound layers comprising the nitrides of the target material are formed on the surface of the target. These compound layers “poison” the target, thereby reducing the evaporation rate of the coating species, including the MPs [43], [44]. Hence, the lower MP density observed in the AlTiCrN II architecture. However, the average size of the MPs Figure 5-3a) shows considerable correlation with the N₂ supply of the top AlTiCrN layers of the coatings. Explicitly, the increase in average MP size from 0.39 to 0.74 μm^2 (from AlTiCrN III to I) could be correlated with the corresponding 6 % increase in N₂ supply. The observation could be attributed to the increase in collision frequency of the

coating species facilitating their agglomeration, thus resulting in larger MPs at higher N₂ supply [37]. In summary, substrate biasing and N₂ gas supply significantly influenced the MP statistics, with the greatest value obtained in the AlTiCrN I architecture.

Figure 5-4 shows the surface roughness parameters (R_a and R_z) of the coating architectures. Moving from AlTiCrN I to III, the R_a and R_z values reduced from 1.89 to 1.62 μm and 12.4 to 10.8 μm , respectively. Compared to previously reported roughness parameters for AlTiCrN coatings deposited on tool steel [45], the relatively higher values obtained in this study are probably due to the rougher condition of the substrate ($R_a \sim 1.6 \mu\text{m}$) before the deposition process. Nonetheless, the observed variation in the roughness parameters of the coatings could be explained by the MP statistics.

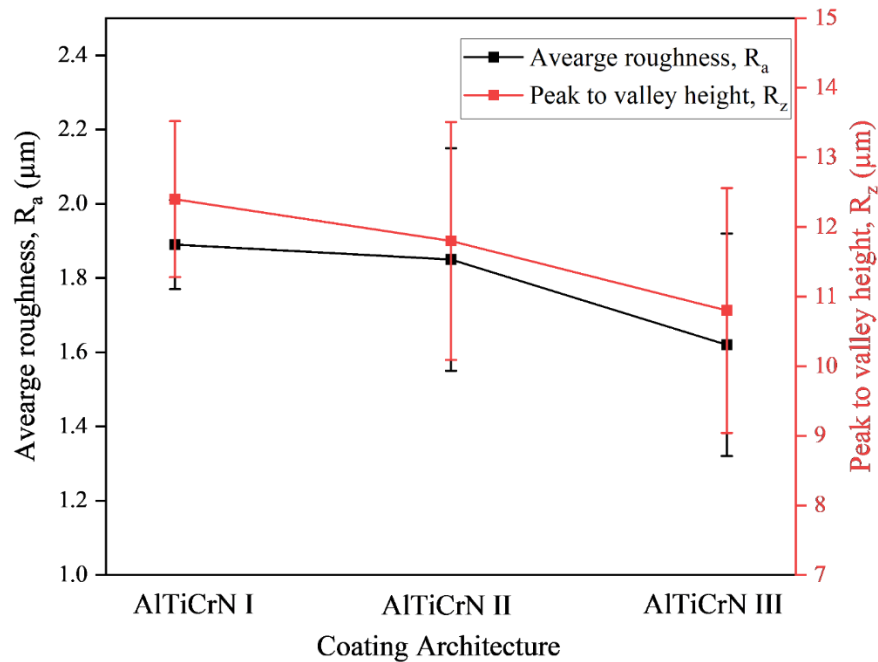


Figure 5-4: Surface roughness parameters of the different AlTiN/AlTiCrN-based coating architectures.

Figure 5-5 depicts the relationship between the coating roughness parameters, MP density, and average MP size. From the figure, a distinct positive correlation is observed

between the roughness parameters (R_a and R_z) and the average MP size. This is evident in the increase in both roughness parameters with increasing average MP size from AlTiCrN III to I. Meanwhile, the MP density fluctuated with the roughness parameters. Firstly, an increase in R_a and R_z accompanied by a reduction in MP density is observed from AlTiCrN III to II, followed by an expected increment in both parameters with MP density from AlTiCrN II to I. Nonetheless, the general observed trend is the increment in the roughness parameters at higher MP statistics and vice versa.

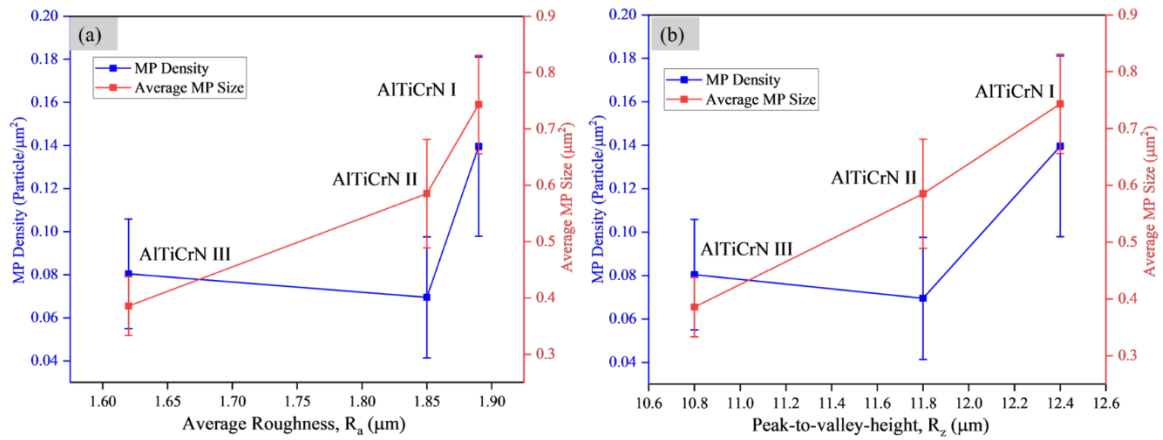


Figure 5-5: Relationship between MP density, average MP size of the AlTiN/AlTiCrN-based coating architectures, and: (a) Average Roughness (R_a); and (b) Peak-to-valley height (R_z).

Generally, higher MP statistics are an indication of an increase in surface asperities, leading to rougher coatings. In this regard, the higher MP density with larger sizes associated with the AlTiCrN I architecture accounts for its highest R_a and R_z values. Additionally, researchers have reported a connection between lower substrate biasing and columnar coating structure, resulting in rougher coatings [46]. Consequently, this could be another

contributing factor accounting for the higher roughness parameters observed in AlTiCrN I architecture deposited under lower substrate bias. Meanwhile, in the case of AlTiCrN II and III architectures deposited under similar bias, the greater average MP size (about 27 %) of the AlTiCrN II exceeds the higher MP density of the AlTiCrN III (about 8 %). This implies that there are larger MPs in AlTiCrN II than there are more MPs in AlTiCrN III. As a result, higher roughness parameters were observed in AlTiCrN II architecture. Furthermore, considering all three architectures, further analysis revealed that about 1 % change in roughness parameters would result from about 6.35 % and 5.9 % corresponding changes in the MP density and the average size, respectively. This suggests that the coating roughness is more sensitive to changes in average MP sizes than the density. In summary, the roughness parameters generally reduced from AlTiCrN I to III, showing a good correlation with the average MP sizes.

5.3.1.2 Coating cross-section and compositional analysis

Figure 5-6 presents the cross-sectional view of the AlTiCrN I to III coating architectures. The coatings generally presented a dense structure characterized by a fine columnar morphology with minimal visible defects. For the AlTiCrN I architecture (Figure 5-6a), two distinct layers are visible: a dark, 2 μm AlTiN (I) base layer and a bright, 1.7 μm AlTiCrN top layer, as identified by the EDX analysis. The AlTiCrN II architecture (Figure 5-6b) presented a similar morphology, but with a 0.5 μm thicker AlTiCrN top layer and an additional relatively darker 0.3 μm AlTiN (II) base layer. Meanwhile, the AlTiCrN III architecture (Figure 5-6c) was a replica of the AlTiCrN II morphology, except for 0.2 μm and 0.1 μm thicker AlTiN (I) and AlTiCrN layers, respectively in the former. Consequently,

the total coating thickness was highest for the AlTiCrN III architecture, and relatively lower for the AlTiCrN II and I, by 5 % and 25 % respectively. The higher coating thickness of the AlTiCrN III coating architecture is due to the longer deposition time of the coating layers. The total thicknesses of AlTiCrN-based layered architectural designs developed in this study are comparable to that of the AlTiN/AlTiCrN coatings reported by Santana et al. [21], who achieved a thickness of 4.5 μm .

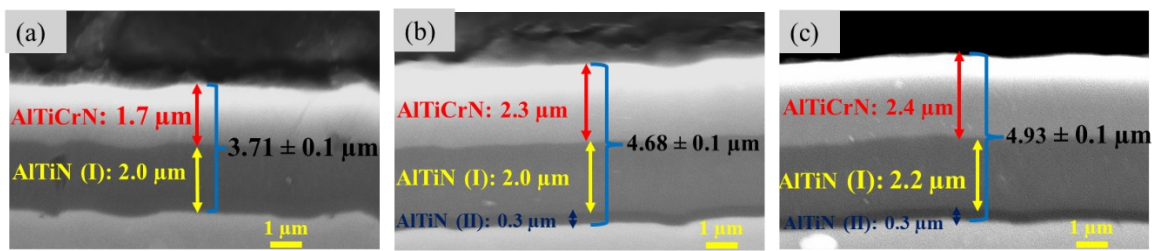


Figure 5-6: SEM micrographs of the cross-sectional view of the AlTiN/AlTiCrN-based coating architectures: (a) AlTiCrN I; (b) AlTiCrN II; (c) AlTiCrN III.

Figure 5-7 presents the EDX elemental mapping of the cross-sectional morphology of the coating architectures. In all three architectures, a uniform elemental distribution of Al (Figure 5-7a₁ – c₁), Ti (Figure 5-7a₂ – c₂), and N (Figure 5-7a₄ – c₄) is evident throughout the coating thickness. Similarly, Fe (Figure 5-7a₅ – c₅), the primary alloying element in the AISI A8 tool steel substrate, exhibits a uniform distribution across all coating architectures. However, the elemental mapping of Cr (Figure 5-7a₃ – c₃), revealed its presence in trace amount within the AlTiN (I) layers and a uniform distribution in the top AlTiCrN layers across all coating architectures. Cr was also uniformly distributed in all the substrates.

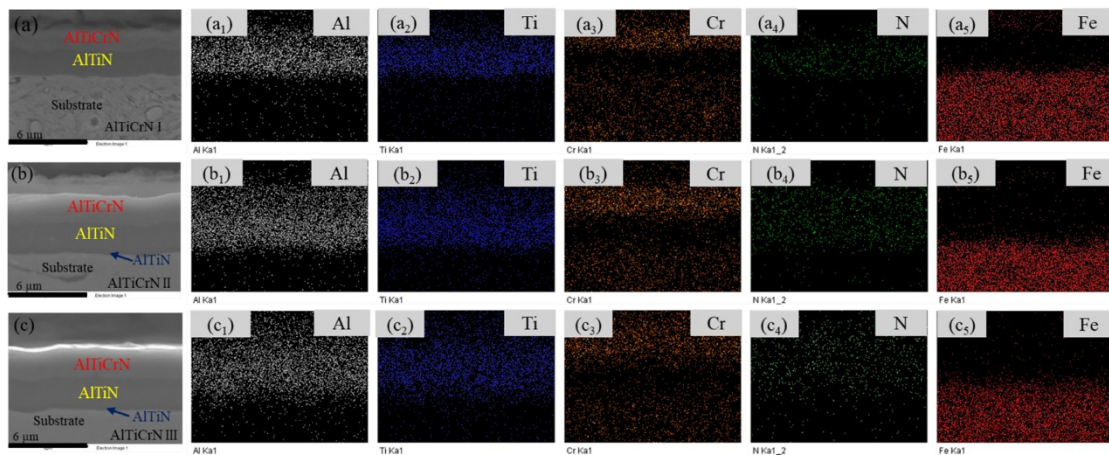


Figure 5-7: EDX mapping of elements of the cross-section of AlTiCrN I to III (a–c) coating architectures: (a₁), (b₁), (c₁): Al; (a₂), (b₂), (c₂): Ti; (a₃), (b₃), (c₃): Cr; (a₄), (b₄), (c₄): N; (a₅), (b₅), (c₅): Fe.

This reduction in the Cr content in the AlTiN (I) layer is also evident in the EDX line scan of the coating architectures (Figure 5-8a–c). Differentiating the elemental mapping of the coating elements, particularly between the AlTiN (I) and (II) layers unique to the AlTiCrN II and III architectures, proved challenging. It is also worth mentioning that trace diffusion of coating elements into the substrate as well as between coating layers, was generally observed in all three architectures. This is essential for promoting adequate coating adhesion [3]. Another significant observation from the EDX mapping is the generally higher distribution density of the metallic elements (such as Al, Ti, and Fe) compared to the non-metallic N. This occurrence might be attributed to the higher electrical conductivity of the former [12].

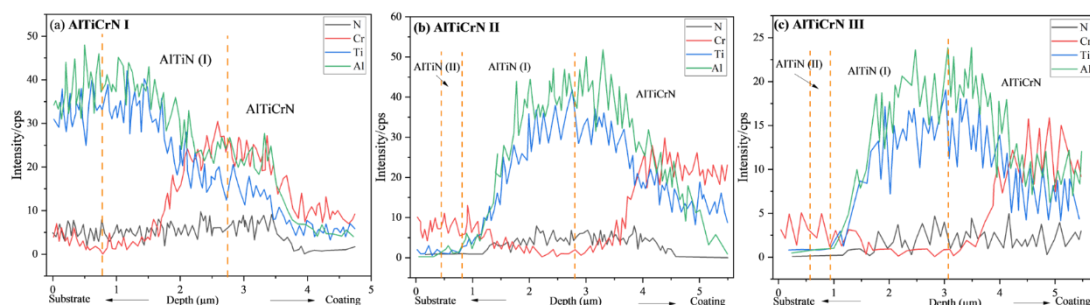


Figure 5-8: EDX line scan of the AlTiN/AlTiCrN coating architectures: (a) AlTiCrN I; (b) AlTiCrN II; (c) AlTiCrN III.

Table 5-4 shows the result of the EDX elemental analysis of the coating architectures. The result of the analysis of the AlTiN (I) layer confirms the presence of Al, Ti, and N for the three architectures. Additionally, trace amount of Cr appeared particularly for the AlTiCrN I architecture, may be likely due to diffusion from the substrate or the Cr-containing AlTiCrN top layer, as evident in the EDX maps (Figure 5-7a₃ – c₃) and line scan (Figure 5-8a–c). The analysis of the AlTiN (II) base layer peculiar to the AlTiCrN II and III architectures presented a similar finding. However, the metallic content (Al and Ti) of this layer is observed to be relatively higher than that of the AlTiN (I) layers in both AlTiCrN II and III coating architectures. This higher metallic content correlates with the relatively darker contrast of the layer compared to the AlTiN (I) layers (Figure 5-6b and c). Meanwhile, for the top AlTiCrN layers in all three architectures, expectedly, there were notable reductions in the Al and Ti compositions compared to the other layers, following the addition of Cr. The introduction of Cr to form the AlTiCrN top layers correlates with the relatively brighter contrast observed for this layer in the cross-sectional micrographs (Figure 5-6a – c). Furthermore, the ratios of N to other metal element compositions of the AlTiN (I) and

AlTiCrN layers were stoichiometric (i.e., > 1) for all architectures. This is an indication of the formation of different metallic nitrides [16]. However, non-stoichiometric compositions were observed for the AlTiN (II) base layers in the AlTiCrN II and III architectures. This observation, reflected in the lower N composition (< 50 at. %, Table 5-4), and evident in the line scan (Figure 5-8b and c), is connected to the lower supply of N_2 gas during the deposition of the base layer in the AlTiCrN II and III architectures [47].

Table 5-4: EDX Elemental composition of the layers of the different AlTiN/AlTiCrN-based coating architectures.

Architec ture	Layer	Position	Composition (at. %)				$\frac{[Al]}{[Al] + [Ti]}$	$\frac{[Al]}{[Al] + [Ti] + [Cr]}$	$\frac{[N]}{[Al] + [Ti] + [Cr]}$
			Al	Ti	Cr	N			
AlTiCrN I	AlTiN (I)	Base	27.40	20.70	1.20	50.70	0.57	0.56	1.03
	AlTiCrN	Top	16.70	8.60	18.00	56.70	0.66	0.39	1.31
AlTiCrN II	AlTiN (II)	Base	28.60	21.60	5.00	44.80	0.57	0.52	0.81
	AlTiN (I)	Intermediate	27.70	21.40	–	50.90	0.56	0.56	1.04
	AlTiCrN	Top	18.50	10.40	18.90	52.30	0.64	0.39	1.10
AlTiCrN III	AlTiN (II)	Base	29.80	22.70	5.30	42.20	0.57	0.52	0.73
	AlTiN (I)	Intermediate	27.00	19.90	–	53.00	0.58	0.58	1.13
	AlTiCrN	Top	18.80	10.10	21.00	50.10	0.65	0.38	1.00

Additionally, considering a given layer between the architectures, the atomic ratios of Al and Ti (i.e., $Al / (Al + Ti)$) appeared to be fairly constant but notably higher for the AlTiCrN top layers. This is connected to the Al/Ti ratio of the target employed in the deposition of the respective layers [38]. For instance, in the case of the AlTiN (I) and (II) layers, the ratio is about 0.56 – 0.58, intermediate between those of the targets (i.e., 0.5 and 0.67) employed during the deposition process. But for the AlTiCrN top layers, it is between 0.64 and 0.66, which is very close to that of the targets (i.e. 0.67). This is an indication of

the efficiency of the deposition process with minimal loss of coating species [12], [48]. Furthermore, the ratio of Al to other metal elements (i.e., Al/ (Al +Ti + Cr)) could significantly impact the coating's tribological properties. Improved tribological performance has been reported for ratios up to 0.6 – 0.7 [8], [10]. In this study, the ratio was observed to be between 0.5 – 0.6 for the AlTiN (I) and (II) layers in all the architectures. However, it was relatively lower for the top AlTiCrN layers (about 0.4) due to the introduction of Cr atoms substituting Al atoms [12]. It is also worth mentioning that the higher substrate biasing applied in the AlTiCrN II and III architectures did not significantly impact the elemental coating composition. This is in agreement with previous studies [48]. In summary, apart from the AlTiN (II) base layers in AlTiCrN II and III architectures, the three coating architectures generally exhibited stoichiometric compositions with similar Al/ (Al +Ti + Cr) ratios.

Figure 5-9 presents the XRD patterns of the different coating architectures. The broken lines in the figure correspond to the reference peak positions according to the Crystallography Open Database [49]. The XRD patterns shown in the figure are a combination of peaks emanating from the different coating layers (i.e., $2\theta = 37.2^\circ$ and 43.6°) and the tool steel substrate (i.e., $2\theta = 44.5^\circ$). The notable reduction in the substrate peak intensity from AlTiCrN I to III correlates with the increase in the coating thickness [38]. Furthermore, the diffraction peaks of the coatings appear to be positioned between the reference c-TiN (COD 01-087-0632) and c-AlN (COD 00-046-1200) positions. However, compared to the reference, the diffraction peaks of the coatings generally appear to be shifted to higher and lower angles, respectively. This indicates the formation of a single-phase solid solution whereby coating atoms (such as Al, Ti, and Cr) are assumed to occupy random positions within the fcc NaCl-type structure, promoting lattice strain leading to the

positioning of the peaks between referenced standards [12], [47]. For all three architectures, two coating diffraction peaks are evident: a (111) preferred orientation and a minor (200) orientation, apparent in the peak intensities. This is in agreement with previous reports [11], [47], [50]. Some authors have linked the preferred texturing in the (111) plane to the higher reticular density [24].

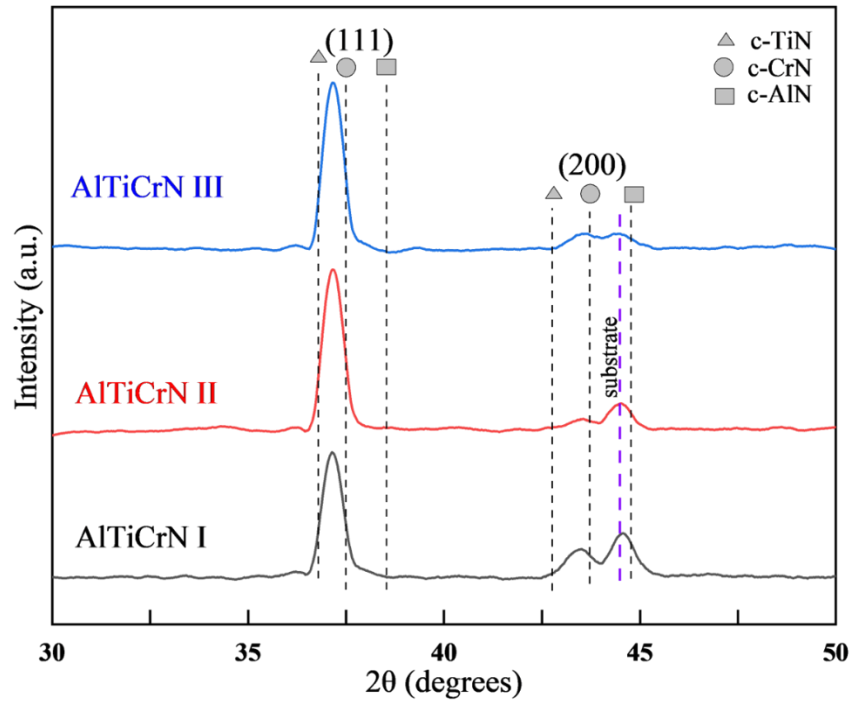


Figure 5-9: XRD patterns of the different AlTiN/AlTiCrN-based coating architectures.

Additionally, it is worth mentioning that diffraction peaks associated with the detrimental h-AlN phase were not observed. This was expected, as the Al/ (Al + Ti + Cr) ratios obtained from the EDX analysis (Table 5-4) were generally less than 0.7 for all three architectures. Furthermore, the (111) orientation is more enhanced in AlTiCrN II and III architectures, accompanied by a reduction in the intensity of (200) peaks compared to

AlTiCrN I. This could be attributed to the effect of the higher negative biasing in the AlTiCrN II and III coatings [13], [14]. Meanwhile, between AlTiCrN II and III with a similar substrate bias, the increased texturing of coating crystals in (111) and (200) planes in the AlTiCrN III could be due to the greater thickness of the coating layers, resulting in more coating atoms being textured in these orientations [38]. Furthermore, the (111) and (200) peak positions were observed to shift slightly to higher angles (37.159 to 37.167 °) and (43.495 – 43.585 °), respectively, from AlTiCrN I to III. The shift in the position of the peaks is an indication of changes in the residual stresses in the coatings [51]. This occurrence could be likely attributed to the generally lower N₂ supply (i.e., for AlTiCrN II and III), increasing metallic components dissolving in the coating's crystal, leading to lattice distortion [52]. Additionally, the greater energetic metallic bombardment at elevated bias could be another contributing factor [48]. This also suggests that the unit cells of the coating become smaller [53], justified by the reduction in the lattice constant (i.e., 0.4171 to 0.4167 nm) from AlTiN I to III, as shown in Table 5-5.

Table 5-5: Average lattice constants, peak positions, microstrain, and crystallite size of the different AlTiN/AlTiCrN-based architectures.

Architecture	Average lattice constant (nm)	(111) plane		(200) plane		Average Microstrain (%)	Average crystallite size (nm)
		2θ (°)	FWHM (°)	2θ (°)	FWHM (°)		
AlTiCrN I	0.4171	37.159	0.481	43.495	0.955	0.834	13.186
AlTiCrN II	0.4168	37.161	0.581	43.556	0.874	0.854	12.101
AlTiCrN III	0.4167	37.167	0.572	43.585	1.684	1.290	9.860

Further examination revealed a significant broadening of the peaks from AlTiCrN I to III, particularly for the (200) orientation. This is indicated by the increase in full-width at half-maximum (FWHM) values from 0.481 to 0.582, and 0.954 to 1.683, for the (111) and (200) orientations, respectively, as shown in Table 5-5. The broadening of the peaks might result from the effect of the compositional gradient and interfaces of the coating architectures, with each layer having different lattice parameters [47]. The additional third layer in the AlTiCrN II and III makes this effect more prominent in these architectures. Generally, peak broadening suggests coating crystallite size reduction and microstraining [54]. The crystallite size (D) was calculated using the Scherrer Equation (Equation 4), while the microstrain (ϵ) was computed using Equation 5 [55].

$$D = \frac{K \cdot \lambda}{\beta \cdot \cos \theta} \quad (4)$$

$$\epsilon = \frac{\beta \cdot \cos \theta}{4 \cdot \sin \theta} \quad (5)$$

where K is the Sherrer constant, λ is the wavelength of the X-ray, β is the FWHM, and θ is connected to the peak position.

The results presented in Table 5-5 confirm the reduction in the average crystallite size (from 13.2 to 9.9 nm) and the increase in microstrain (0.83 to 1.29 %) from AlTiCrN I to III. The reduction in crystallite size and increase in microstrain are likely due to the cumulative effect of: (1) the accompanying increase in ion bombardment of the growing coating following a 40 % increase in the bias, resulting in deformation (2) hindrance of crystallite boundary migration by the impinging energetic coating species (3) the aftermath

of coating lattice distortion resulting from the formation of solid solution [46], [50]. In summary, moving from AlTiCrN I to III, the (111) orientation is enhanced, the average crystallite size is reduced, and the average microstrain is increased.

5.3.2 Mechanical properties

5.3.2.1 Hardness, plasticity Index, and resistance to plastic deformation

Figure 5-10 presents the results obtained from the nanoindentation test. Figure 5-10a shows the load-displacement curves for the three coating architectures obtained by plotting the force and displacement data recorded during the test. The curves are divided into two parts representing the loading and unloading sections that characterize the coatings' elasto-plastic behavior [37]. From the figure, during the loading stage, there is a gradual increase in displacement (plastic behaviour) with increasing loading followed by gradual displacement reduction (elastic behaviour) during the unloading stage. The maximum coating deformation corresponding to the maximum displacement is reached at the maximum indenting load (i.e., 20 mN). It was observed that the displacement of the indenter into the coating was highest for the AlTiCrN I and least for the AlTiCrN III. This was evident in the shift of the force-displacement curves to the left, moving from AlTiCrN I to III. This is an indication that the latter exhibited the greatest resistance to plastic deformation by the indenter and increased hardness [38], [51].

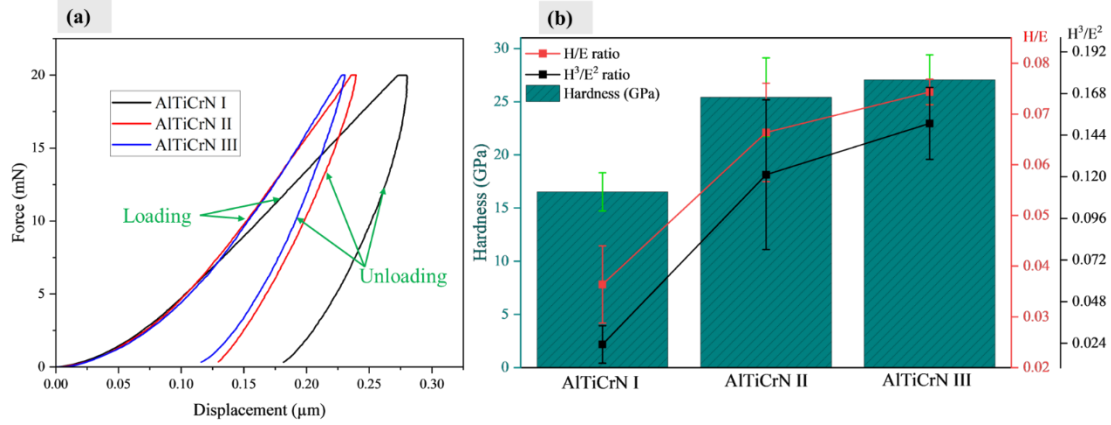


Figure 5-10: (a) Load-indentation depth curves for the different coating architectures and (b) Hardness, resistance to elastic strain (H/E), and plastic

Figure 5-10b presents the hardness, H/E, and H^3/E^2 of the three coating architectures. The results confirm an increase in hardness (16.5 to 27.1 GPa) from AlTiCrN I to III. These hardness values are slightly lower compared to those reported by Santana [21] for a similar AlTiN/AlTiCrN coating. This difference may stem from the difference in coating composition and deposition parameters. Generally speaking, the hardness of CAE-PVD coatings largely depends on the competitive interplay of several factors, including composition, morphology, crystallite size, and residual stresses, among others [16]. Dense and compact coatings characterized by lower MP density have been correlated with increased coating hardness [3]. This implies that the highest MP density exhibited by the AlTiCrN I architecture might have contributed to its relatively lower hardness compared to the other architectures. However, no correlation was observed in the case of AlTiCrN II and III, as the latter, contrarily, showed a 6 % higher hardness despite its 8 % higher MP density.

Furthermore, the enhancement of the (111) orientation in the AlTiCrN II and III architectures is an indication of an increased effect of solid solution strengthening of coating atoms in the closed pack plane [47]. This might have also contributed to the higher hardness

in these architectures. This is similar to previous reports by Zhou et al. who observed an increase in the hardness of AlTiCrN coatings with an increase in the peak intensity of the (111) plane accompanied by a reduction in (200) intensity owing to a reduction in Cr composition [12]. Additionally, the superlattice effect associated with layered coating designs could also account for the higher hardness of the AlTiCrN II and III coatings. In this regard, the coherency strain generated by the shear moduli differential and lattice mismatch between individual layers hinders dislocation movement. Thus, resulting in increased hardness [17], [51]. This mechanism presents a significant correlation with the coating hardness. This is evident in the substantial 58 % difference in hardness between the bilayer AlTiCrN I and the trilayer AlTiCrN II. Meanwhile, only a marginal 6 % difference is observed between AlTiCrN II and III, both having trilayer architectures. This marginal difference highlights the contribution of other factors to the hardness of the coatings.

Moreover, crystallite size strengthening based on the Hall-Petch phenomenon is also applicable. Figure 5-11 shows a plot of the hardness of the coating architectures against the average crystallite size. The figure reveals a distinct negative correlation between the coating hardness and the average crystallite size. This is evident in the increase in the coating hardness following a reduction in the average crystallite size from AlTiCrN I to III. Based on the Hall-Petch phenomenon, finer coating crystallite with more crystallite boundaries provides greater resistance to dislocation movement, thereby enhancing the coating's resistance to plastic deformation [56], [57]. The average crystallite size of the coatings reduced by about 25 % from AlTiCrN I to III. Therefore, the increase in hardness is expected with the reduction in the average crystallite size. The findings from previous studies revealed that a 1 % increase in coating hardness could result from about 0.9 – 1.2 % reduction

in the crystallite size [58], [59]. However, in this study, a 1 % increase in the coating hardness was obtained from about a 0.4 % reduction in crystallite size. The discrepancy could be attributed to the difference in coating composition and the deposition conditions.

Additionally, the strain hardening offered by the energetic coating bombardment by incoming adatoms with increased substrate biasing resulting in significant coating deformation could have also contributed to the higher hardness of the AlTiCrN II and III architectures [60]. As shown in Figure 5-11, a positive correlation exists between the hardness and the average microstrain of the coatings. This is evident in the increase in

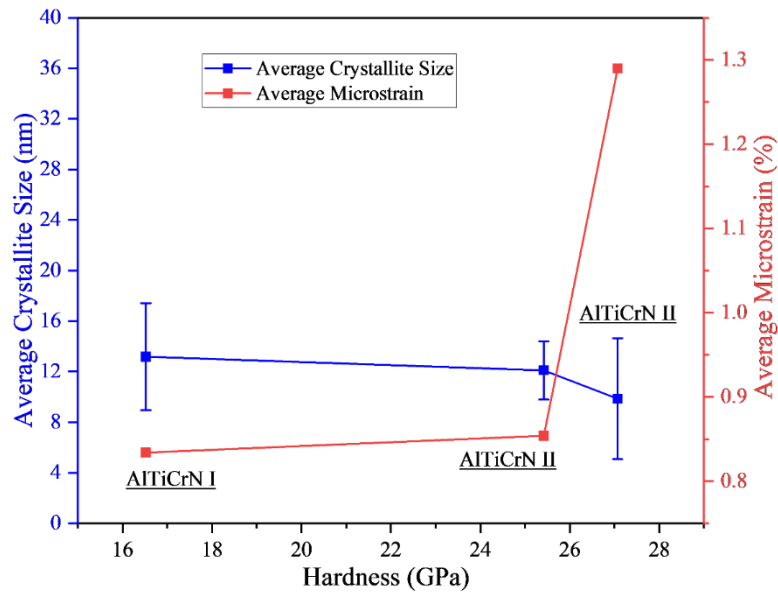


Figure 5-11: Relationship between hardness, average crystallite size, and average microstrain of the AlTiCrN I-III coating architectures.

microstrain from 0.8 % in AlTiCrN I to 1.3 % in AlTiCrN III. However, this mechanism is unlikely to have played a significant role in the hardness of the coatings. For instance, only a marginal 2 % difference in microstrain is observed between the AlTiCrN I and II architectures, despite the significant difference (54 %) in hardness. Similarly, despite the substantial (52 %) difference in microstrain between the AlTiCrN II and III architectures,

only a marginal 6 % difference in hardness is observed. Nonetheless, taking into account the effect of all factors, the coating hardness appears to be more correlated with the crystallite size, layered design, and crystal orientation than the other factors.

The H/E and H^3/E^2 ratios are widely used to evaluate the fracture resistance of PVD coatings. The former, also referred to as the plasticity index, provides valuable insight into the elastic behaviour of the coatings, while the latter indicates the coating's resistance to plastic deformation [61]. Generally, higher values of these ratios could correlate with improved tribological performance [3]. From Figure 5-10b, it is evident that both ratios followed a similar trend as the hardness. The H/E and H^3/E^2 ratios increased from 0.04 to 0.07 and 0.02 to 0.15 moving from AlTiCrN I to III, respectively. This suggests that the AlTiCrN III coating exhibits superior fracture resistance. This occurrence could be attributed to the combined effect of the increased crystallite boundaries (due to finer crystallites), interfaces (i.e., increased number of layers), and microstraining, which hinder crack propagation. Further analysis revealed that a 1 % change in H/E of the coatings would require about a 0.32 % change in crystallite size, 0.66 % in the number of coating layers, and 0.73 % in microstrain. A similar trend was observed for the H^3/E^2 ratio. This suggests that the H/E and H^3/E^2 are more impacted by changes in the crystallite size than the other factors. Thus, it could be inferred that the increase in crystallite boundaries resulting from the finer coating crystallite structure is the dominant mechanism responsible for the enhanced fracture resistance.

5.3.2.2 Adhesion strength

Figure 5-12 shows the results of the Daimler-Benz test for comparing the adhesion strength of the different coating architectures. The figure shows varying levels of coating

deformation around the submerged effect created by the Rockwell indenter. For AlTiCrN I (Figure 5-12a, a' and a'') and AlTiCrN II (Figure 5-12b, b' and b''), a combination of radial cracks extending from the indent and coating delamination are evident. Meanwhile, only radial cracks could be observed for the AlTiCrN III architecture (Figure 5-12c, c' and c''), suggesting superior adhesion strength [62]. The VDI 3198 standard for categorizing the coating adhesion quality for this test stipulates six grades (HF1 – HF6) of coating failures, depending on the extent of cracks and delamination [31]. Comparing the coating failures observed in this study with the standard, the adhesion strength of AlTiCrN I and II could be categorized as HF3, while AlTiCrN III could be assigned HF1. These failures are generally considered acceptable based on the standard and suitable for industrial applications. The high fracture resistance of the AlTiCrN III architecture might have played a significant role in its superior adhesion strength in this test [16].

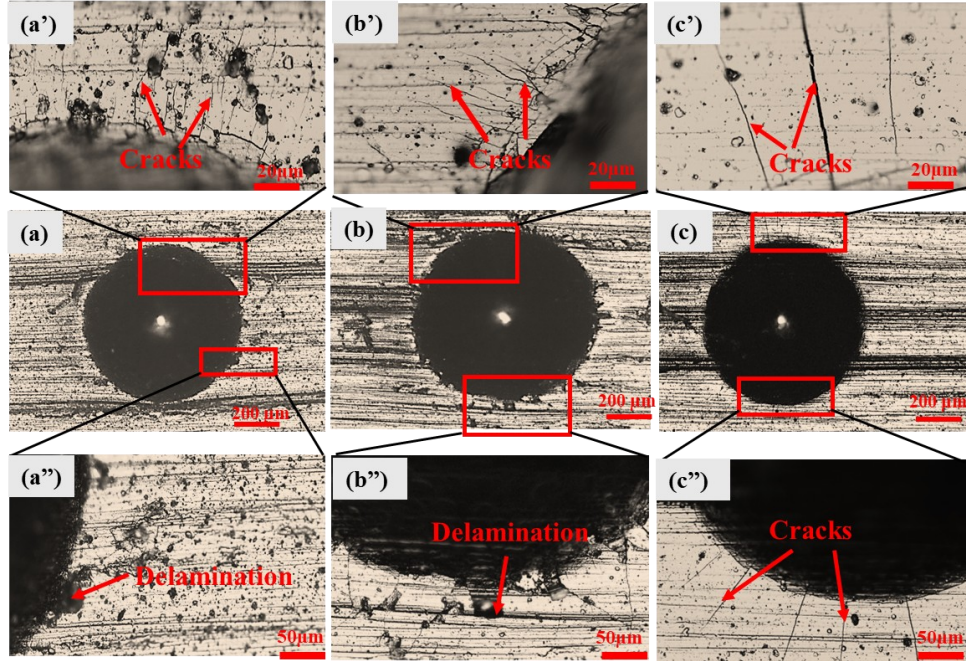


Figure 5-12: Optical micrographs showing the indentations from the Daimler-Benz adhesion test: (a), (a') and (a'') AlTiCrN I; (b), (b') and (b'') AlTiCrN II; (c), (c') and (c'') AlTiCrN III.

Further quantitative assessment of the coating adhesion strength was performed by scratch testing. Figure 5-13 shows the results obtained from the test for the different coating architectures. Figure 5-13a – c present plots of the normal force, frictional force, and acoustic emission alongside the scratch paths for the AlTiCrN I – III coating architectures. Two critical loads were defined: L_{c1} – critical load for cohesive failure or crack initiation and L_{c2} – critical load for adhesive failures or complete delamination from substrate [63]. These critical loads were ascertained by a complementary assessment of fluctuations in frictional forces, acoustic emission, and optical microscopy examination of the scratch path [55]. The locations of occurrence of these failures along the scratch length are indicated with the broken lines in the figures. From the figures, L_{c1} and L_{c2} increased from 5.2 to 14.6 N and 8.4 to 20 N, moving from AlTiCrN I to III, respectively. This indicates that AlTiCrN III

exhibited superior adhesion strength. It also agrees with the previous results obtained from the Daimler-Benz test (Figure 5-12). Additionally, regions of these critical load failures are characterized by significant fluctuations in acoustic emission [63], as evident in Figure 5-13a – c. The acoustic emission represents elastic waves generated due to the interaction between the coated sample and the stylus [64]. More so, the fluctuations in the frictional force provide useful insight into the coefficient of friction between the coatings and the scratch stylus. The frictional force fluctuations and acoustic emissions were generally minimal for AlTiCrN III coating architecture, suggesting a lower coefficient of friction recorded for the coating [46], [65], [66]. This correlates with the results obtained for surface roughness (Figure 5-4), as the lower roughness of the AlTiCrN III coating resulted in lower acoustic emissions and frictional forces. Furthermore, the crack propagation resistance (CPR) of the coatings was evaluated using the expression proposed by Zhang et al., i.e., $CPR = L_{c1}(L_{c2} - L_{c1})$ [67]. The result of the CPR of the coatings presented in Figure 5-13d shows

that the CPRs increase from AlTiCrN I to III. This is consistent with the previous results from the fracture resistance (H/E and H^3/E^2) obtained by nanoindentation (Figure 5-10b).

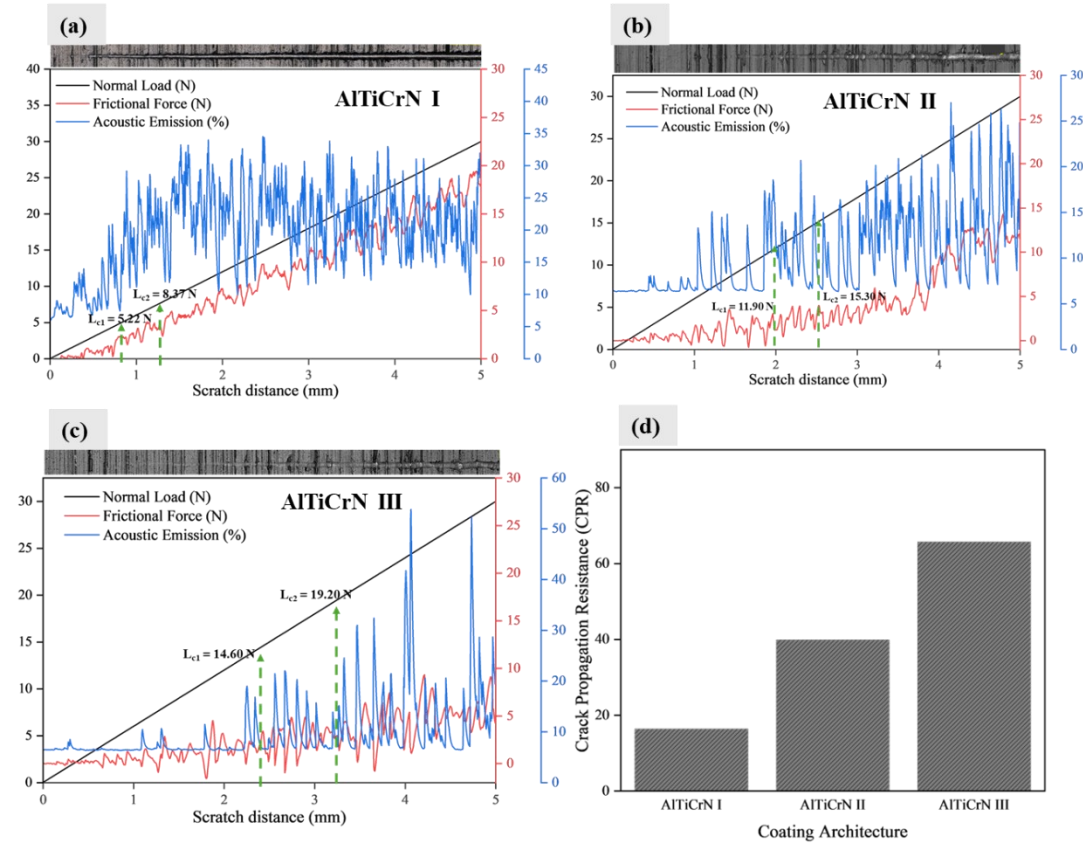


Figure 5-13: Scratch test results of different AlTiN/AlTiCrN-based coating architectures: (a) AlTiCrN I; (b) AlTiCrN II; (c) AlTiCrN III; (d) Crack propagation resistance.

The adhesion strength of the coatings could be influenced by the fracture resistance and the layered architectural design. In order to better understand the relationship between the fracture resistance and the adhesion strength of the coating architectures, Figure 5-14 was graphed. The figure shows a plot of the coating's adhesion strength against the indices of the coating's fracture resistance obtained from the nanoindentation (Figure 5-10) and scratch tests (Figure 5-13). The figure reveals a positive correlation between the coating's adhesion

strength and the fracture resistance indices. This is characterized by the increment in the critical load (L_{c2}) with increasing H/E , H^3/E^2 , and CPR from AlTiCrN I to III. Consequently, the higher fracture resistance of the AlTiCrN III coatings suggests enhancement in the coating's ability to absorb energy from plastic deformation during tribological interactions without fracturing. This also translates to enhancement in the coating's resistance to crack initiation and propagation, particularly towards the coating-substrate interface. Thus, reducing the likelihood of delamination during external loading.

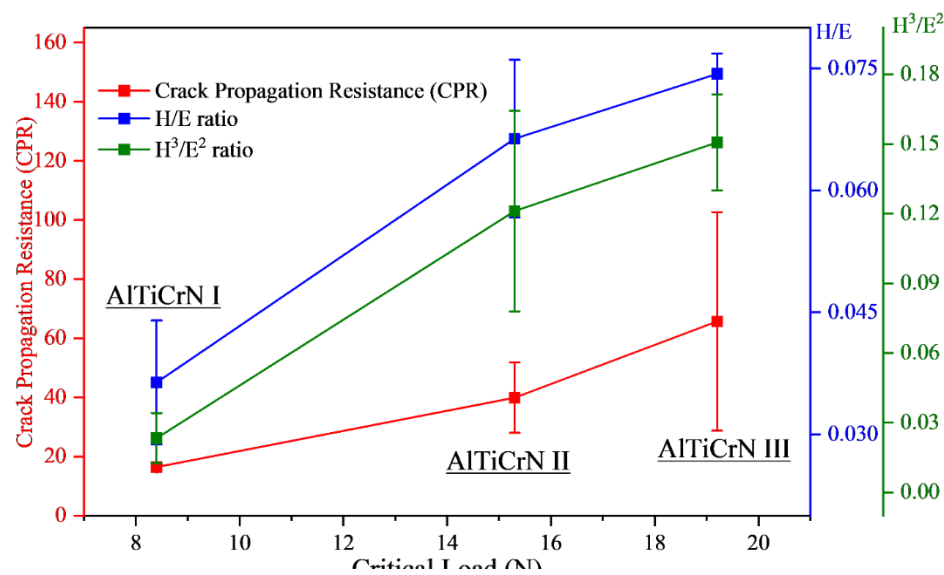


Figure 5-14: Relationship between critical load, crack propagation resistance, H/E , and H^3/E^2 of AlTiCrN I to III coating architectures

Furthermore, in layered coatings, the composition of the base layer plays a key role in the adhesion strength [3]. Most often, for improved coating adhesion, it is desirable to have a smaller hardness difference between the base layer and the substrate to minimize interfacial stress generation which promotes coating delamination during external loading [68]. Previous studies have shown that non-stoichiometric coating compositions are usually characterized by non-equilibrium phases which have a detrimental effect on the coating hardness [26]. Consequently, the non-stoichiometric AlTiN (II) base layers present in AlTiCrN II and III coatings might be characterized by a lower hardness owing to the associated non-equilibrium phases, compared to the stoichiometric AlTiN (I) base layer in the AlTiCrN I architecture. Thus, the likely lower hardness differential between the substrate and the non-stoichiometric AlTiN (II) base layers of AlTiCrN II and III coatings might have culminated in their superior adhesion strength. Furthermore, the coating hardness could even be further impacted by the extent of its non-stoichiometry [25], [37]. Consequently, the less stoichiometric AlTiN (II) base layer in AlTiCrN III relative to AlTiCrN II architecture, is expected to further minimize the substrate-base layer hardness differential. More so, the likely reduction coating/substrate hardness mismatch induced by the non-stoichiometric AlTiN (II) base layer, may lower interfacial stresses, thereby improving the interfacial shear strength and reducing the susceptibility of coating delamination from the substrate during tribological interactions [26], [69]. Additionally, the more metal-rich nature of the non-stoichiometric AlTiN (II) base layer (i.e. higher compositions of Al, Ti, and Cr; see Table 5-4), could promote the formation of stronger metallic bonds with the substrates, further enhancing coating adhesion [24]. This might also explain the superior adhesion of the exhibited by the AlTiCrN III architecture. In summary, the adhesion strength generally

increased from AlTiCrN I to III and was largely influenced by the fracture resistance and stoichiometric properties of the base layer.

5.3.3 Wear performance

Figure 5-15 presents the wear rates for the different coating architectures obtained from the dry sand rubber wheel test. From the figure, AlTiCrN I coating exhibited the highest wear rate ($2.2 \times 10^{-4} \text{ mm}^3/\text{Nm}$), corresponding to the least wear resistance. These wear rates are significantly higher than those reported for other AlTiCrN-based coatings. The higher wear rates observed in this study could be attributed to the relatively higher contact force (about 130 N, see Table 5-3) used in the ASTM G65 test in this study, compared to lower forces ($< 40 \text{ N}$) reported in other studies [12], [50]. The wear rates were reduced by 14 % and 29 % in AlTiCrN II and III architectures, respectively. The observed wear performance of the coatings could be explained in terms of the hardness, H/E , H^3/E^2 (Figure 5-10b), adhesion strength (Figure 5-13), thickness (Figure 5-6), and surface roughness (Figure 5-4).

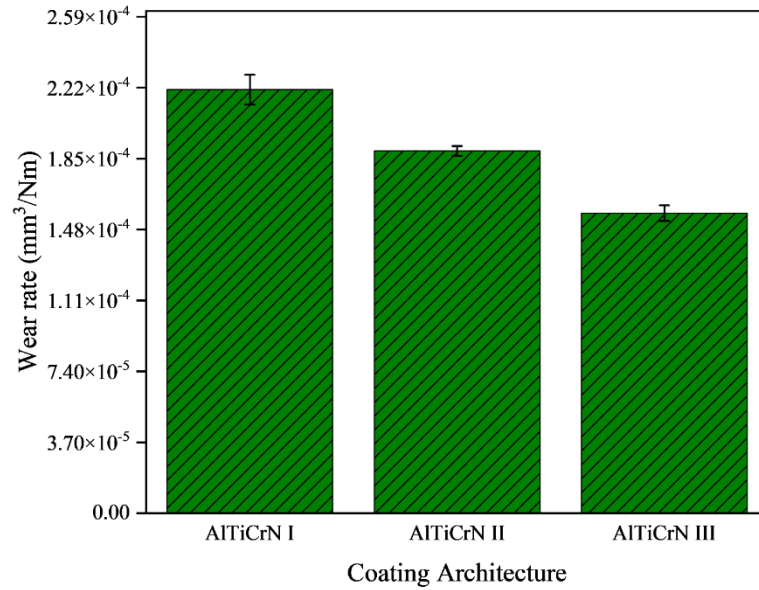


Figure 5-15: Wear rates of the different AlTiN/AlTiCrN-based coating architectures obtained from the DSRW test.

Figure 5-16 shows the relationship between the wear rates, hardness, H/E , and H^3/E^2 ratios of the coating architectures. From the figure, an expected negative correlation is observed between the coating wear rates, hardness, H/E , and H^3/E^2 ratios. This is characterized by reductions in the wear rates with increasing coating hardness, H/E , and H^3/E^2 ratios from AlTiCrN I to III. The lower wear rates with increasing hardness could be attributed to the improvement in the coating's resistance to plastic deformation. Meanwhile, the enhancement of the coating's fracture toughness indicated by higher H/E and H^3/E^2 ratios results in improved resistance to crack initiation and propagation during tribological interactions [16]. Hence, the lower rates observed in the AlTiCrN III architecture with higher H/E and H^3/E^2 ratios. Further analysis to ascertain the impact of the properties on the coating wear rates revealed that 1 % changes in the hardness, H/E , and H^3/E^2 ratios would result in corresponding 0.64 %, 0.39 %, and 0.075 % changes in the wear rates respectively. This

suggests that the wear rate of the coatings is more impacted by the hardness than the fracture toughness (i.e. H/E and H^3/E^2 ratios).

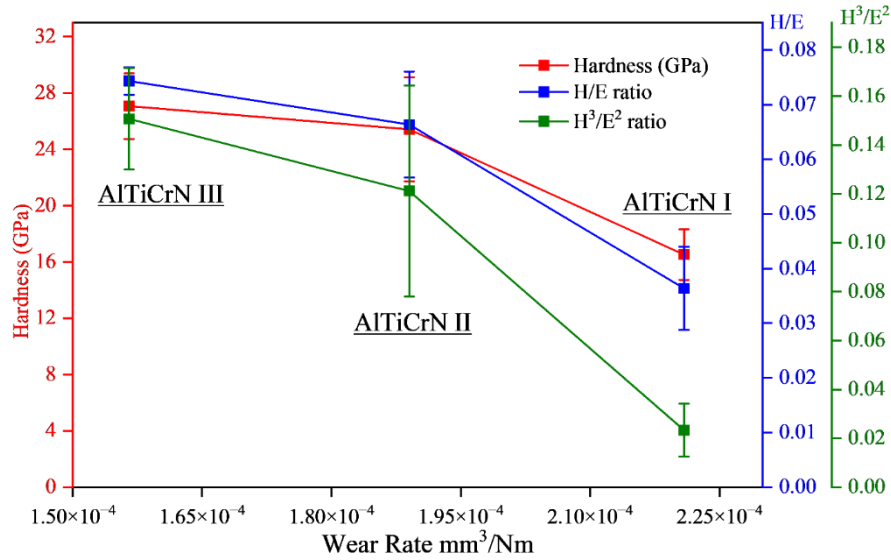


Figure 5-16: Relationship between wear rate, hardness, H/E , and H^3/E^2 ratios of the AlTiN/AlTiCrN-based coating architectures.

Figure 5-17 shows the relationship between the adhesion strength (critical load), surface roughness, thickness, and the wear rates of the coatings. The adhesion strength (i.e. critical load) and thickness exhibited a negative correlation with the wear rates, as previously observed for hardness, H/E , and H^3/E^2 ratios. The lower rates at higher critical loads (from AlTiCrN I to III) could be attributed to the improvement in coating bond strength, resulting in less likelihood of premature delamination during tribological interaction [63]. Additionally, the thicker AlTiCrN III coatings provide greater load support, resulting in lower shear force, owing to load distribution over a greater volume as compared to AlTiCrN I coating with lower coating thickness, thus resulting in lower wear rates in the former [70]. Meanwhile, a general positive correlation is observed between the wear rates and roughness.

This is evident in the general increase in the wear rates with increasing average roughness from AlTiCrN III to I. The expected lower wear rates observed for the AlTiCrN III coating exhibiting lower roughness could be attributed to the increase in the contact area, thus lowering the stress concentration during tribological contact and eventually culminating in lower material loss [3]. Further analysis to quantify the impact of the parameters on the wear rates revealed that 1 % change in the critical load, R_a and coating thickness resulted in corresponding 0.39 %, 2.47 % and 1.25 % changes in the wear rates, respectively. This suggests that the wear rate of the coatings in the DSRW test is more sensitive to changes in the roughness than the other parameters.

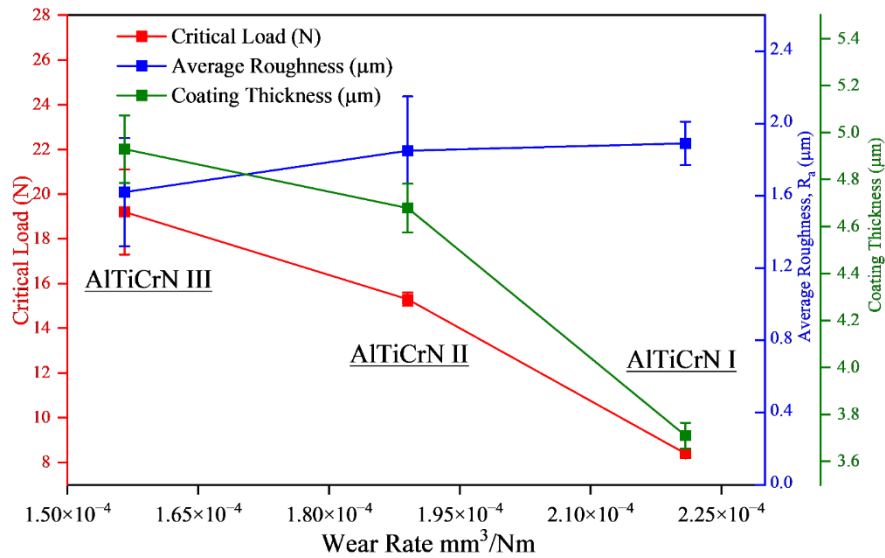


Figure 5-17: Relationship between wear rate, adhesion strength, surface roughness, and thickness of the AlTiN/AlTiCrN-based coating

Overall, the resulting wear performance of the AlTiCrN coating architectures is influenced by the cumulative impact of the coating properties, with the AlTiCrN III

architecture presenting superior tribological performance. It was also found that the coating wear rate was negatively correlated with the hardness, H/E , H^3/E^2 , adhesion strength, and coating thickness. This translated to better tribological performance at higher values of these properties, as observed in AlTiCrN III architecture. Meanwhile, the average roughness showed a positive correlation with the wear rate, translating to poor tribological performance at higher roughness values, as seen in the AlTiCrN I architecture. Furthermore, the quantitative analysis revealed that the impact of the coating properties on the wear performance ranks in the order: roughness > thickness > hardness > H/E > adhesion strength > H^3/E^2 .

Figure 5-18 shows the SEM micrographs of the wear track of the coatings after the wear test. The post-wear analysis is important to gain insight into the prevalent wear mechanisms. The figure shows three distinct zones (A, B, and C) in the wear track. The zones represent the region of entry of the abrasive sand into the wear track, the center of the wear track, and the exit of the abrasive sand from the wear track, respectively. The mechanisms leading to the formation of these distinct zones have been extensively discussed in our previous work [28]. From the figure, adhesive and abrasive wear appear to be the predominant wear mechanisms. In zone A (Figure 5-18a – c), regions of abraded coating without substrate exposure (darker region) and complete coating delamination (brighter region) are evident. The EDX analysis of the darker regions (P1 – 3) presented in Table 5-6 confirms the presence of coating elements (i.e., Al, Ti, Cr, and N), while analysis of the brighter regions (P4 – 6) revealed trace amounts of coating elements and high amounts of Fe confirming substrate exposure. The region also exhibits adhesive wear characterized by randomly located areas of coating delamination across all coating architectures (Figure 5-

18a – c). The relatively lower contact force in this region might contribute to this occurrence. Meanwhile, the center of the wear track (Figure 5-18a' – c') revealed the occurrence of abrasive wear in the form of microcutting grooves. The microcutting grooves result from the high-angle indentation of coated samples by the abrasive silica sand under pressure [71]. The wear track exit, zone C (Figure 5-18a'' – c''), exhibited a similar occurrence as zone A. The EDX analysis confirming the regions representing substrate exposure (P7 – 9) and abraded coating (P10 – 12) are presented in Table 5-6. Additionally, the presence of O on the wear track is likely from the ambient air during the wear test [12], [72]. Furthermore, Si contamination (i.e., abrasive sand) was also detected in the wear tracks of the samples. Si contamination has been reported to facilitate abrasive wear through tribochemical reactions [19]. From Table 5-6, the Si amounts were observed to be generally higher in the regions of exposed substrate (P4 – 9), highlighting the protective role of the coatings. Further analysis with Image J to quantify the area of the exposed substrate ($\bar{A}$, Figure 5-18' – c') revealed that the least substrate area ($\bar{A} = 216.75 \text{ mm}^2$) was exposed in the AlTiCrN III while the greatest ($\bar{A} = 246.94 \text{ mm}^2$) was observed in AlTiCrN I architecture. Additionally, the increase in the volume of the worn material from $32.8 \pm 0.6 \text{ mm}^3$ to $36.1 \pm 0.8 \text{ mm}^3$, as measured by profilometry from AlTiCrN III to I, correlates with the higher wear rates exhibited by the latter (Figure 5-15).

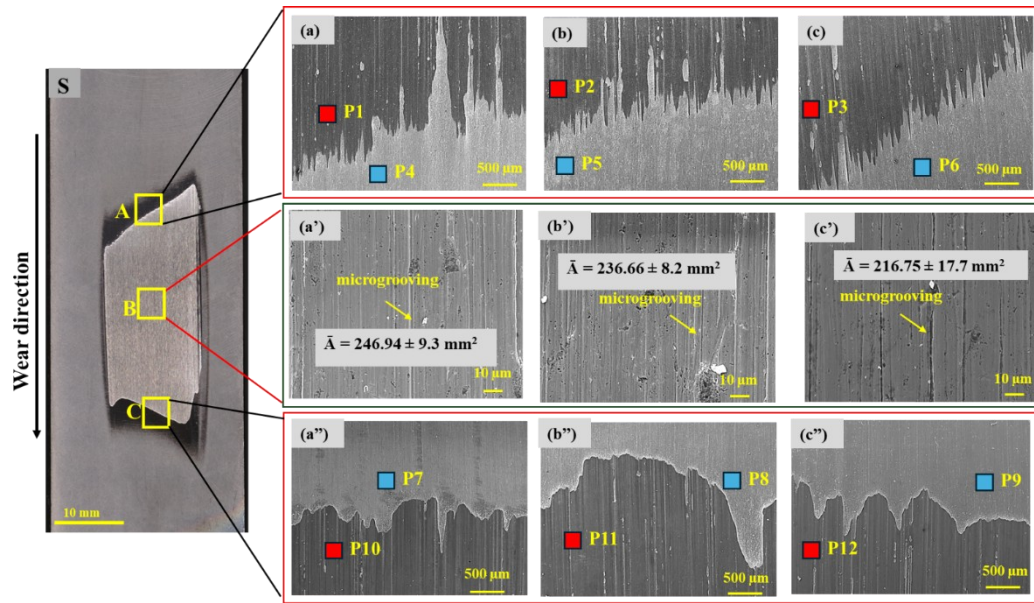


Figure 5-18: (S) Micrograph showing the different zones of the wear track of the coated tool steels for the different architectures: (a), (a') & (a'')

Table 5-6: Elemental composition of spectrum points in the wear track (i.e. Figure 5-18).

Points	Elemental composition (at. %)						
	Al	Ti	Cr	N	O	Si	Fe
P1	17.8	13.4	14.0	45.8	8.5	-	0.5
P2	23.2	15.1	4.1	46.3	8.4	-	2.8
P3	25.2	17.7	0.2	41.5	14.5	0.2	0.7
P4	0.6	0.2	9.7	-	16.3	4.1	69.1
P5	0.5	0.1	9.6	-	13.5	3.7	72.6
P6	0.2	0.3	8.1	-	20.4	6.6	64.6
P7	0.1	0.3	9.9	-	6.9	2.4	80.4
P8	0.1	0.2	9.4	-	13.5	2.6	74.2
P9	0.4	0.2	8.8	-	12.4	3.0	75.4
P10	18.6	14.1	11.0	49.3	6.7	0.1	0.3
P11	17.7	12.4	14.9	40.4	14.0	0.2	0.5
P12	19.2	13.7	12.0	42.1	12.6	0.1	0.4

5.4 Conclusion

The following key conclusions were derived from the investigation of the effect of layered architectural design on the tribological behaviour of different AlTiN/AlTiCrN-based CAE-PVD coatings:

1. The reduced surface roughness observed in AlTiN (I)/AlTiN (II)/AlTiCrN coating architectures (i.e. AlTiCrN II and III) contributed to their improved tribological performance compared to the AlTiN (I)/AlTiCrN architecture (i.e. AlTiCrN I). This was evident in the lower MP statistics in the former, resulting from the effect of substrate biasing and supply of reactive N₂ gas.
2. The compositional analysis indicated the enhancement of the (111) orientation from AlTiCrN I to III coating architectures. Furthermore, while the AlTiCrN I exhibited a stoichiometric AlTiN (I) base layer, AlTiCrN II and III were characterized by non-stoichiometric AlTiN (II) base layers, which significantly improved the adhesion strength
3. The adhesion strength of the coatings showed significant correlations with the crack propagation resistance, H/E, and H^3/E^2 ratios. This was evident in the general increase in the coatings' adhesion strength with the values of the parameters from AlTiCrN I to III.
4. The enhancement of the (111) preferred orientation, increase in microstrain, reduction in average crystallite size, and increase in the number of coating layers from AlTiCrN I to III coating architecture, led to an increase in hardness, H/E and H^3/E^2 .

5. The tribological performance was influenced by the hardness, fracture resistance, adhesion strength, thickness, and roughness of the coatings. Explicitly, lower rates were obtained at higher values of hardness, H/E , H^3/E^2 , adhesion strength, thickness, and lower roughness values. Further quantitative analysis revealed that 1 % changes in the hardness, H/E , H^3/E^2 ratios, critical load, R_a , and coating thickness resulted in 0.64, 0.39, 0.075, 0.39, 2.47, and 1.25 % changes in the wear rates, respectively. This indicated that the wear rate is most significantly impacted by the roughness and least affected by the H^3/E^2 ratio.

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Chapter 6

Influence of deposition temperature on the microstructure and wear behaviour of cathodic arc evaporated-ALTiN/ALTiCrN ceramic coatings on AISI A8 tool steel

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Abstract

Deposition temperature is a known pivotal process parameter influencing the tribological performance of physical vapour deposited ceramic coatings. Yet, the impact of the parameter remains highly system-specific, limiting generalization of observed trends across different coating-substrate systems. Therefore, this study investigates the effect of deposition temperature (350 – 450 °C; designated as T350 – T450) on the microstructural evolution, mechanical properties, and wear behaviour of ALTiN/ALTiCrN coatings deposited on AISI A8 tool steel for woodworking applications. Coatings were characterized using scanning electron microscopy, energy dispersive X-ray spectroscopy, X-ray diffraction, nanoindentation, scratch testing, and ASTM G65 abrasive wear testing. Results showed that T350 exhibited the lowest wear rate ($\sim 2 \times 10^{-4} \text{ mm}^3/\text{Nm}$); 36 and 56 % lower than T400 and T450, respectively. This superior wear resistance was attributed to its higher adhesion (critical load $\sim 11 \text{ N}$), thicker coating (2.8 μm), smoother surface ($R_a = 0.25 \mu\text{m}$), and higher H/E ratio (0.072), despite an intermediate hardness (31 GPa). Although maximum hardness (36 GPa) occurred at T400 due to finer crystallite ($\sim 10.4 \text{ nm}$) and elevated microstrain (0.77

%), its higher macroparticle density ($0.18 \text{ particle}/\mu\text{m}^2$) and roughness ($R_a = 0.31 \mu\text{m}$) compromised performance. Overall, these findings demonstrate that a deposition temperature of 350°C provides an optimal wear resistance in AlTiN/AlTiCrN-coated A8 tool steels, offering valuable insights for tailoring PVD process parameters for woodworking applications.

Keywords: Deposition temperature, AlTiN/AlTiCrN; Cathodic arc evaporation; Physical vapour deposition; Wear.

6.1 Introduction

AlTiN/AlTiCrN physical vapour deposition (PVD) ceramic coatings deposited on A8 tool steels have gained significant attention as promising protective materials for woodworking applications. The synergy between the cost-effectiveness of the A8 tool steel substrate and the superior hardness and thermal stability of the AlTiN/AlTiCrN layered coatings renders this system particularly suited for highly abrasive and high-temperature conditions typical of wood machining operations [1], [2], [3]. Nevertheless, the outstanding performance of the layered coatings is not solely determined by their multilayer architecture [4] or the substrate condition [5], [6]. However, process-related parameters, especially the deposition temperature, also play a critical role in defining the coating properties and overall functional performance [7].

Several studies have demonstrated that deposition temperature exerts a complex and often coating/system-dependent influence on the tribological performance of PVD ceramic coatings. For instance, Ling et al. [8] reported that increasing the deposition temperature of AlCrN coatings on tungsten carbide from 250 to 450°C resulted in lower wear rates. The

improvement in wear resistance was attributed to enhanced coating hardness, thickness, and adhesion strength at elevated temperatures. Similarly, Gautier and Machet [9] observed improved wear resistance of Cr coatings on stainless steel with increasing deposition temperature (250 – 500 °C), attributing the improvement to the annihilation of lattice defects and consequent hardness enhancement. Meanwhile, contrasting observations were reported by Warcholinski et al. [10] for a similar AlCrN coating investigated by Ling et al. [8], but on HS6-5-2 steel substrate. The authors reported a decline in hardness and adhesion strength of AlCrN coatings with increasing deposition temperature from 350 – 450 °C. These deteriorations in hardness and adhesion strength were linked to crystallite growth coupled with the associated residual stress relaxation at higher temperatures [7], [10]. Collectively, the findings from these studies suggest that the influence of deposition temperature on the tribological performance of ceramic coatings is largely dependent on the coating-substrate system.

The foregoing discussion highlights the dependence of the effect of deposition temperature on wear behaviour of PVD ceramic coatings on the coating-substrate system. Consequently, this makes the generalization of the findings reported across different coating-substrate systems challenging. Notably, a literature survey reveals the existence of no prior studies investigating the effect of deposition temperature on the wear behaviour of AlTiN/AlTiCrN-coated A8 tool steels, despite the widespread industrial usage of the material in the woodworking industry. To address this knowledge gap, this present study investigates how varying the deposition temperature influences the microstructural, morphological, and mechanical properties of AlTiN/AlTiCrN coatings on A8 tool steels, and how this cascade of property changes, in turn, affects the wear behaviour. The results from

the study aim to provide valuable insights for optimizing the deposition temperature of AlTiN/AlTiCrN-coated A8 tool steel for enhanced wear performance.

6.2 Experimental details

6.2.1 Substrate preparation and coating deposition

The chemical composition of the AISI A8 tool steel substrate, commonly employed in woodworking applications, is presented in Table 6-1.

Table 6-1. Chemical composition of the AISI A8 tool steel substrate (wt. %).

C	Cr	Mo	W	Si	Mn	Fe
0.5	8.1	1.2	1.1	0.8	0.4	Balance

The substrates with dimensions $75 \times 25 \times 10$ mm were ground to achieve an average roughness (R_a) of $0.1 - 0.2 \mu\text{m}$, then cleaned sequentially with mild detergent, rinsed with water, and dried under vacuum. The coatings were deposited using a cathodic arc evaporation system, equipped with planar targets including pure Cr and Al/Ti hybrid compositions. The deposition process involved loading the prepared substrates into the deposition chamber, followed by evacuation to a vacuum to minimize coating contamination and enhance deposition efficiency [11]. This was followed by Al/Ti ion and argon gas etching to remove residual surface contaminants [5]. The AlTiN/AlTiCrN coatings were then deposited following established procedures in previous studies [2], [4], with the deposition temperature varied between 350, 400, and 450 °C, which were designated as T350, T400, and T450, respectively. Continuous substrate rotation was maintained throughout the process

to ensure uniformity during deposition [7]. After deposition, the system was cooled before unloading the samples for subsequent characterization.

6.2.2 Coating characterization

The surface morphology and coating cross-section were analyzed using a scanning electron microscope (SEM, JSM-6480LV, Japan). Thereafter, quantitative post-imaging analysis for Macroparticle (MP) density, size, and coating thickness was carried out using Image J software [12]. For the MP analysis, 10 SEM micrographs of consistent magnification and contrast were evaluated, while 10 measurements were recorded for coating thickness determination. The elemental composition of the coatings was examined by energy dispersive X-ray spectroscopy (EDS, INCA energy 250). The phase composition was characterized by X-ray diffraction (XRD, D8 Discover, Bruker AXS, Germany) utilizing Co α radiation. Data were collected at a step size of 0.05° with a counting time of 0.5 s/step.

Nanoindentation tests (Anton Paar MCT3) were performed to measure the hardness and elastic modulus of the coatings under a maximum load of 20 mN. A minimum of five indents were obtained for each condition. Adhesion strength was determined using a progressive load scratch test (Anton Paar MCT3) with a Rockwell-type diamond stylus of 50 μm radius. The load increased linearly from 0 to 30 N during the test. The loading rate, scratch speed, and track length were 20.98 N/min, 3.5 mm/min, and 5 mm, respectively. Each test was repeated three times per coating condition. The wear resistance was evaluated following ASTM G65, procedure B [13], based on the mass loss method [2]. Each test condition was repeated three times, and the resulting wear volume was corrected to account for rubber wheel wear using Equation 1 [7]:

$$\begin{aligned}
& \text{Adjusted wear volume (mm}^3\text{)} \\
& = \text{wear volume (mm}^3\text{)} \\
& \times \frac{228.6 \text{ mm}}{\text{Wheel diameter after test (mm)}}
\end{aligned} \tag{1}$$

Thereafter, the adjusted wear volumes were used in the computation of the wear rates [14]. Post-wear examination of the wear tracks was also performed using SEM-EDX to identify the dominant wear mechanisms.

6.3 Results

6.3.1 Microstructural evaluation

6.3.1.1 Effect of deposition temperature on surface morphology, MP statistics, and surface roughness

Figure 6-1a-c presents the micrograph showing the surface morphology of the AlTiN/AlTiCrN coatings. The surface of the coated tool steels is characterized by deposition defects, namely, macroparticles (MPs) and craters. These defects are typical for PVD coatings, and a lot of research is ongoing on the subject, due to their detrimental impact on the quality and performance of the coatings [15]. The MPs may result from a combined effect of hydrodynamic effects and thermal shock on the target, leading to the ejection of molten particles, which eventually solidify on the substrate as MPs. The craters, on the other hand, may arise from incomplete reactions between the coating species and the reactive gas (N₂) and often become evident following the flaking of loosely adhered MPs [4]. Further examination revealed the presence of profiling grooves from substrate preparation, highlighting the significant role of the substrate topography on the morphology of the deposited coatings [5]. More so, the grooves appear to be more visible with increasing

deposition temperature, with the coatings deposited at 400 and 450 °C (Figure 6-1b and c) exhibiting the most pronounced grooves. This could suggest a reduction in coating thickness with increasing deposition temperature, as substrate irregularities are expected to be more prominent in thinner coatings [11].

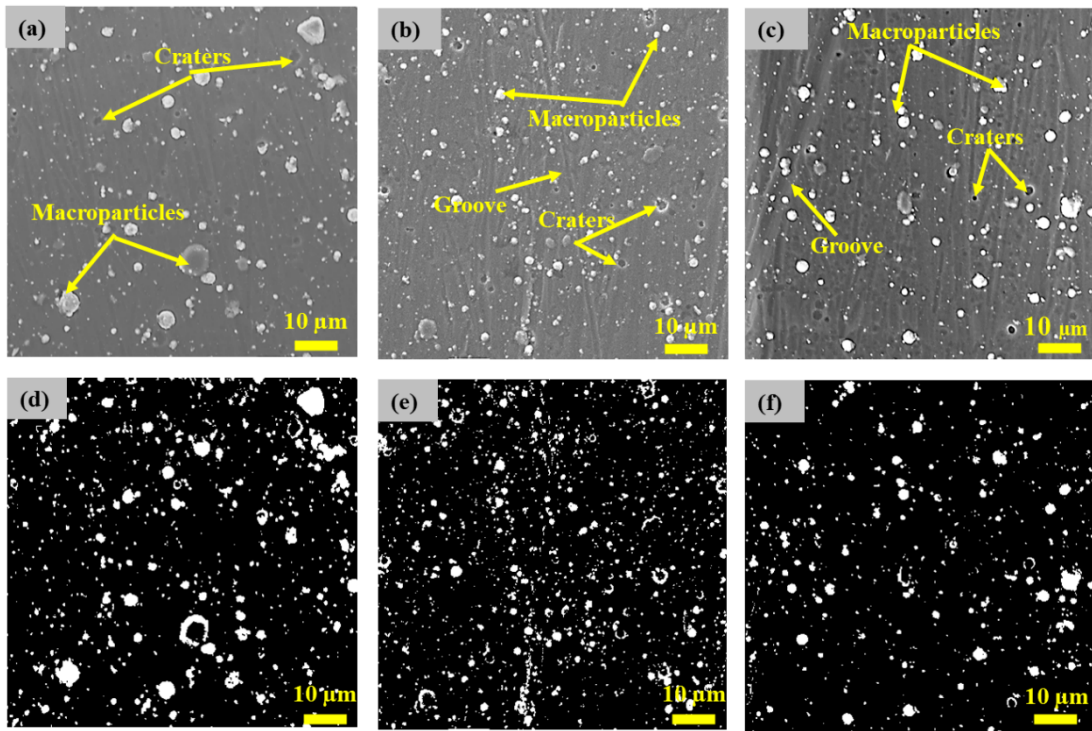


Figure 6-1. SEM micrographs and corresponding ImageJ-processed images of AlTiN/AlTiCrN-coated AISI A8 tool steel surfaces deposited at different temperatures: (a, d) 350 °C; (b, e) 400 °C; and (c, f) 450 °C.

Figure 6-1d-f presents the ImageJ-processed micrographs, which were used for the MP quantification. The relationship between the MP statistics and deposition temperature presents a novel finding, as it has not been explored in previous studies [8], [16]. The result of the analysis is presented in Figure 6-2. The figure reveals that the MPs reached a maximum density of about 0.18 particle/ μm^2 at T400, while T350 and T450 conditions

exhibited comparable MP density values of about 0.09 particle/ μm^2 , which is about 50 % lower than that at T400. Meanwhile, the average MP size was generally higher at T350 (about 0.75 μm^2), followed by about 43 and 17 % reductions with increasing temperature at T400 and T450, respectively.

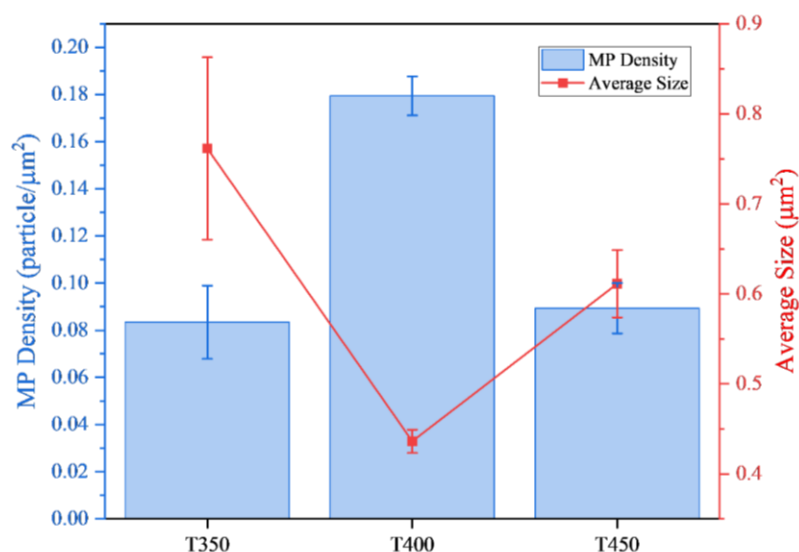


Figure 6-2. Macroparticle density and average size of AlTiN/AlTiCrN coatings deposited at different temperatures.

The evolution of the MPs with deposition temperature could be explained by considering MP agglomeration-coalescence and bombardment-resputtering mechanisms, as illustrated in Figure 6-3. Explicitly, at the relatively low deposition temperature of 350 °C, MPs are less energetic, which reduces their ability to bombard and disrupt previously condensed particles. This lack of interruption favors MP agglomeration and subsequent coalescence, producing fewer MPs with larger sizes, as observed in the T350 condition.

When the temperature increases to 400 °C, the MPs acquire greater kinetic energy, leading to a higher arrival rate of MPs on the substrate surface. However, the enhanced bombardment of incoming particles disrupts agglomeration and coalescence, thereby producing a higher MP density but with smaller particle sizes. Upon further increasing the temperature to 450 °C, the arrival rate of the MPs is further increased, but the accompanying increased energetic bombardment leading to resputtering decreases the MP density compared to T400. Nonetheless, the higher particle mobility at this temperature (i.e. T450) could facilitate the coalescence of the remaining MPs, resulting in an increase in average MP size relative to T400 [7], [11]. In summary, the MPs are generally larger at 350 °C due to the dominance of MP agglomeration-coalescence, while the highest MP density with the least size occurs at 400 °C due to the influence of MP bombardment interrupting coalescence. Meanwhile, at 450 °C, MP density decreases but the size increases relative to 400 °C, attributed to the combined effects of resputtering and enhanced MP mobility promoting coalescence.

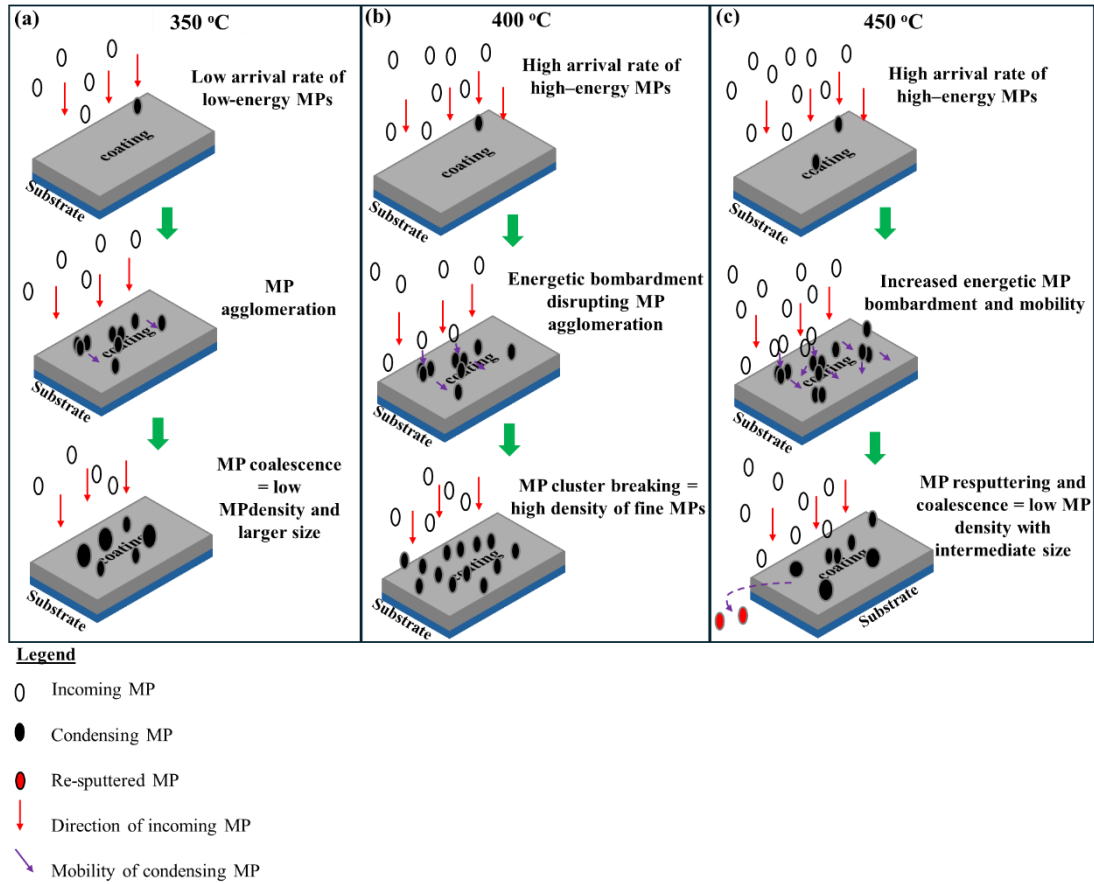


Figure 6-3. Schematic illustration of macroparticle evolution mechanisms at different deposition temperatures for AlTiN/AlTiCrN coating.

Figure 6-4a presents the surface roughness parameters (R_a and R_z) of the coatings deposited at different temperatures. From the figure, both parameters exhibit a similar trend. For instance, the R_a increases from 0.25 μm (T350) to 0.31 μm (T400), followed by a reduction to 0.27 μm (T450). Notably, the roughness values obtained in this study are relatively higher than those reported for other AlTiCrN-based coatings [17], primarily due to the comparatively rougher substrate condition ($R_a \sim 0.1 \mu\text{m}$) prior to deposition. Moreover, previous studies have shown that substrate topography significantly influences the roughness of the deposited coatings [5]. Nonetheless, the coating roughness evolution

with deposition temperature could be correlated with the MP statistics. As shown in Figure 6-4b, R_a and R_z exhibit a positive correlation with MP density. For instance, the increase in MP density from T350 to T450 coincides with higher R_a and R_z values, while the reduction in MP density from T400 to T450 corresponds to lower R_a and R_z values. Consequently, the relatively higher roughness of the T400 condition could, therefore, be explained by its maximum MP density. In this regard, the accumulation of MP defects increases surface asperities, thereby producing a rougher coating surface [4]. In summary, although the roughness did not follow a distinct trend with temperature, it shows a clear correlation with the MP density, with the highest roughness values at T400 attributed to its highest MP density.

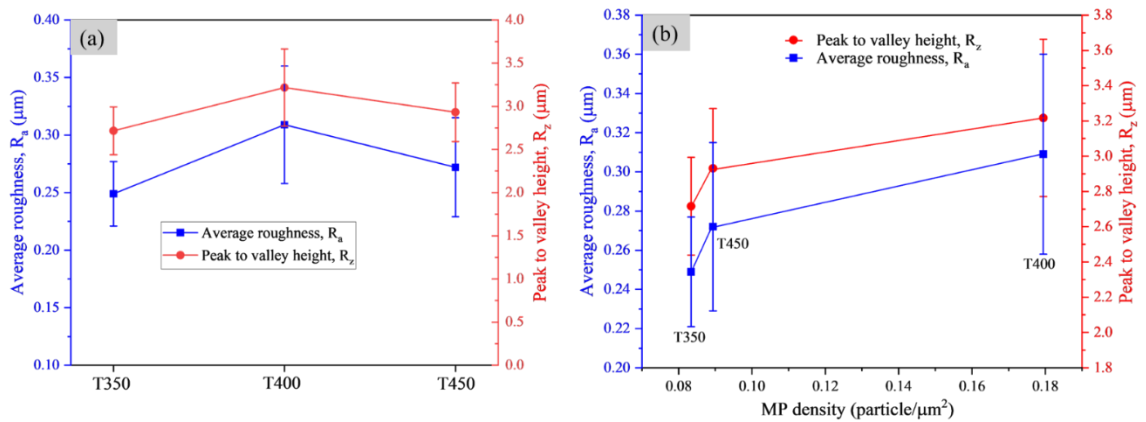


Figure 6-4. (a) Surface roughness parameters (R_a and R_z) and (b) correlation between R_a and R_z and macroparticle density of AlTiN/AlTiCrN coatings deposited at different temperatures.

6.3.1.1 Effect of deposition temperature on cross-sectional morphology, thickness, and elemental composition.

Figure 6-5 shows the cross-sectional morphology of the AlTiN/AlTiCrN coatings deposited at different temperatures. From the figure, overall, the coatings exhibit a dense structure and minimal visible defects. The coating thickness is also observed to reduce progressively with increasing deposition temperature. Explicitly, the thickness was found to be $2.8 \mu\text{m}$ at 350°C , followed by 4 % and 14 % reductions upon increasing the temperature to 400 and 450°C , respectively. These values fall within the range ($2 - 4 \mu\text{m}$) typically reported for AlTiN/AlTiCrN coatings under similar deposition conditions [2], [4]. Moreover, comparable trends have also been reported by Warcholinski et al. [10], who observed about a 20 % reduction in AlCrN coating thickness with increasing temperature from $350 - 450^\circ\text{C}$.

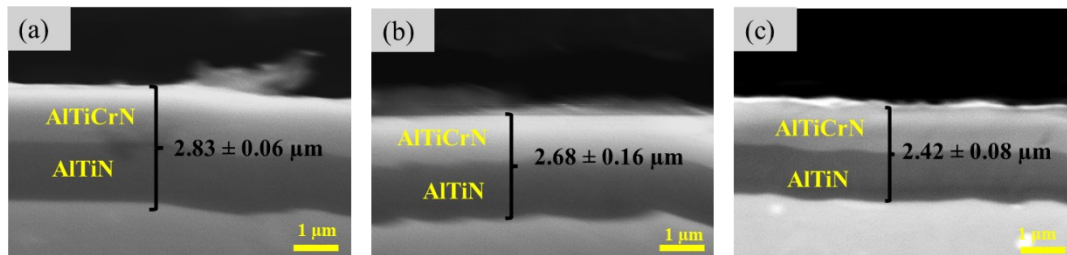


Figure 6-5. Cross-sectional SEM micrographs of AlTiN/AlTiCrN coatings deposited on tool steel substrate at temperatures of: (a) 350°C ; (b) 400°C ; (c) 450°C .

In this regard, the reduction in the deposition rate leading to lower coating thickness with increasing temperature could be explained by two phenomena: ion bombardment-re-sputtering mechanism and the so-called “target poisoning”. With respect to the ion-bombardment mechanism, at higher deposition temperatures, the incoming coating adatoms

exhibit higher mobility, leading to more energetic bombardment of the substrate. Consequently, this leads to an increased tendency of adatom resputtering, thereby reducing the deposition rate and resulting in thinner coatings, as observed in the T450 condition [4]. Meanwhile, according to the target poisoning phenomenon, at elevated temperatures, there is an increased tendency for the formation of compound metallic nitrides on the surface of the targets, leading to target poisoning. These metallic nitrides formed on the targets reduce the effective evaporation of coating adatoms from arc spots, thereby reducing deposition rate, leading to thinner coatings [10].

Further examination of the coating cross-sectional morphology reveals a bilayer structure, comprising a darker AlTiN base layer and a brighter AlTiCrN top layer. This morphology is consistent with previous findings by Danek et al. [18], who reported brighter contrast in Cr-rich AlTiCrN coatings. The EDX elemental mapping of the coating shown in Figure 6-6a confirms the presence of Al, Ti, Cr, and N in the top AlTiCrN layer, while the AlTiN base layer contains Al, Ti and N, along traces of Cr. The EDX line scan (Figure 6-6b) suggests that the trace Cr detected in the AlTiN base layer likely originates from interdiffusion of Cr from the AlTiCrN top layer or from the Cr-containing substrate.

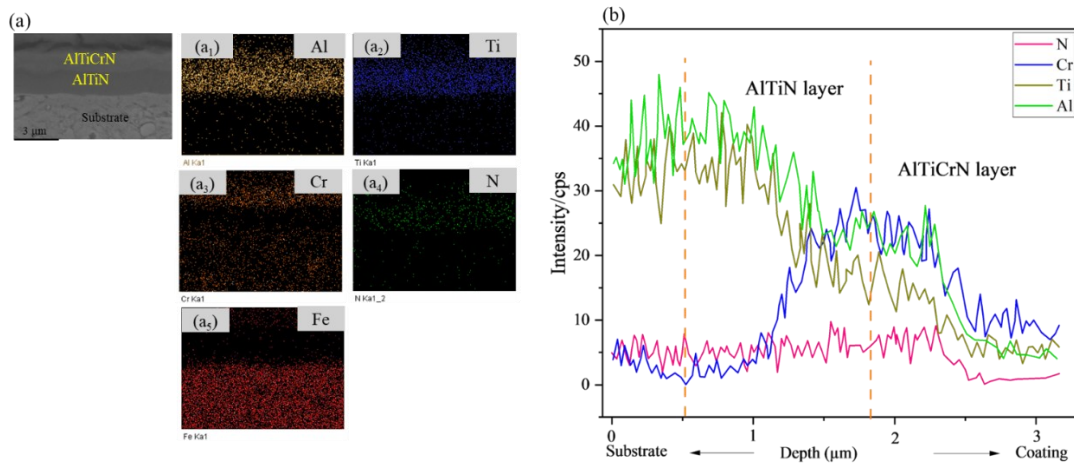


Figure 6-6. (a) EDX elemental mapping and (b) corresponding line scan profile of the AlTiN/AlTiCrN coating deposited at 350 °C.

Additionally, the EDX compositional results shown in Table 6-2 further reveal that the coatings exhibit stoichiometric compositions at all deposition temperatures, indicated by an $N/(Al + Ti + Cr)$ ratio greater than 1. This suggests that there is a supply of reactive N_2 gas to promote the formation of stable metallic nitrides [8], [19]. Furthermore, researchers have observed a correlation between higher ratios of $Al/(Al+Ti+Cr)$ and improved tribological performance of PVD coatings [20]. In this study, for the AlTiN layer, the $Al/(Al+Ti+Cr)$ ratio decreases with temperature from 0.56 at T350 to 0.51 at T450 condition. However, much lower values of the ratios were observed for the AlTiCrN layer (0.39 to 0.41), which can be explained by the substitution of Al atoms with Cr atoms in the AlTiCrN layer [4]. Nonetheless, the bilayer architecture could benefit from the contributions of both layers to enhance the functional performance of the coating. Overall, considering both layers cumulatively, the deposition temperature appears not to have a significant effect on the $Al/(Al+Ti+Cr)$ ratio, with average values generally in the range of 0.46 – 0.48. In summary, increasing the deposition temperature resulted in: (1) a reduction in coating thickness, (2)

coatings with generally stoichiometric compositions, and (3) no significant variation in the Al/(Al+Ti+Cr) ratio.

Table 6-2. EDX elemental composition of the AlTiN/AlTiCrN coatings deposited at different temperatures.

Layer	Condition	Composition (at. %)				$\frac{[Al]}{[Al]+[Ti]+[Cr]}$	$\frac{[N]}{[Al]+[Ti]+[Cr]}$
		Al	Ti	Cr	N		
AlTiN	T350	27.40	20.70	1.20	50.70	0.56	1.03
	T400	25.70	20.80	2.10	51.50	0.53	1.06
	T450	24.30	20.70	2.50	52.50	0.51	1.11
AlTiCrN	T350	16.70	8.60	18.0	56.70	0.39	1.31
	T400	21.0	12.30	15.60	51.20	0.43	1.05
	T450	20.60	12.20	17.10	50.20	0.41	1.01

6.3.1.3 Effect of deposition temperature on phase composition, crystallite size, and microstrain

Figure 6-7 shows the XRD profiles of the AlTiN/AlTiCrN coatings deposited at different temperatures. In the figure, non-continuous lines are used to indicate reference peak positions according to the Crystallography Open Database [21]. The diffraction peaks presented by the coated samples consist of contributions from both the coating and the substrate. Specifically, three substrate peaks at $2\theta = 52.2^\circ$, 77° , and 99.4° were identified based on the reference code 96-901-3474. With an increase in deposition temperature (from T350 to T450), a reduction in the intensity of the main substrate peak ($2\theta = 52.2^\circ$) was observed. This observation is an indication of reduction in the coating thickness at higher deposition temperatures, as the X-rays more readily penetrate through the thinner coatings to the substrate [22]. This is corroborated by the cross-sectional micrographs (Figure 6-5), where the coating thickness consistently reduced from T350 to T450. Meanwhile, the

coating-related peaks were situated between the reference positions of c-TiN (96-101-1103), c-CrN (96-101-0975), and c-AlN (00-046-1200). However, the observed peak shifts towards either higher or lower 2θ angles relative to the reference peaks signify the formation of a solid solution of TiN, CrN, and AlN [4]. The typical resulting structure comprises coating atoms randomly occupying sites within the fcc NaCl-type lattice, causing lattice distortion, leading to the positioning of the peaks between referenced standards [14].

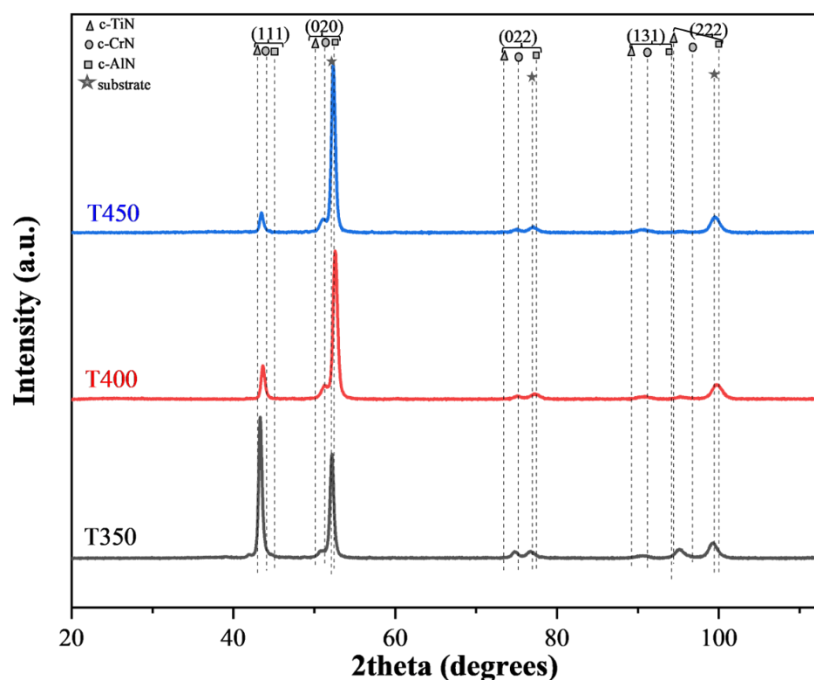


Figure 6-7. XRD patterns of AlTiN/AlTiCrN coatings at different deposition temperatures.

Furthermore, the coatings generally presented several crystallographic orientations, including (111), (020), (022), (131), and (222). Notably, at T350, the AlTiN/AlTiCrN coating exhibited an enhanced (111) preferential orientation. This is consistent with the findings from earlier studies, reporting a preferred (111) orientation for AlTiN/AlTiCrN coatings due to the high planar density of the closed-pack plane [4], [5]. However, as the deposition temperature increased to T450, (111) plane texturing diminished significantly, with its relative intensity decreasing from 84 to 47 % as shown in Figure 6-8.

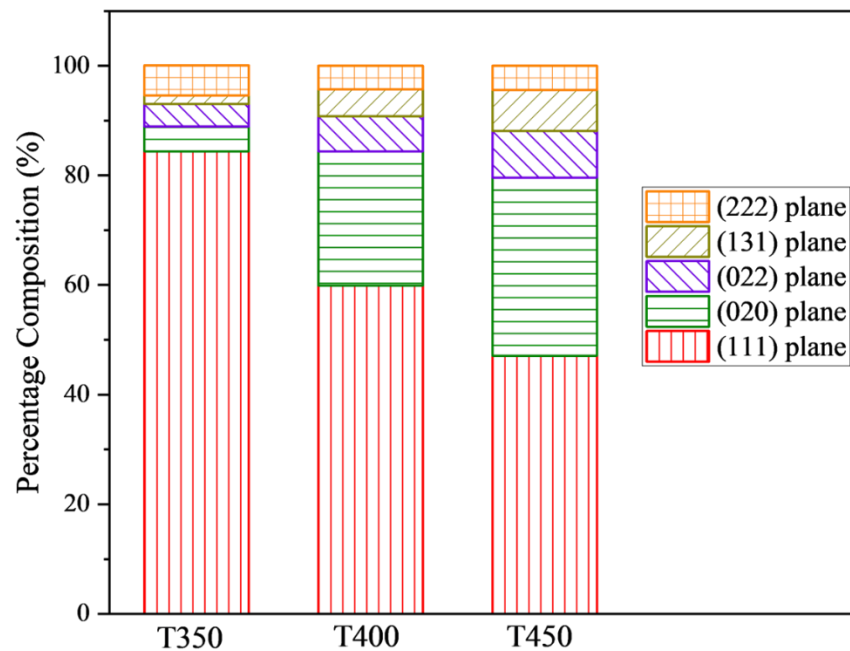


Figure 6-8. Plane distribution of AlTiCrN/AlTiN coatings deposited at different temperatures (Note: the percentage plane distribution was determined by normalizing the intensity of each diffraction peak to the total intensity of all coating peaks [5]).

The reduction in the (111) texture with increasing deposition temperature could be attributed to the effect of energetic collisions and bombardment of the growing film by incoming species at elevated temperature. Such an occurrence disrupts the coating crystallinity and promotes competitive growth of other crystallographic planes [5], [11]. This results in the re-distribution of the coating atoms in more random plane orientations, evident in the general increase in (020), (022), (222), and (131) plane compositions (Figure 6-8), at higher temperatures. This observation is consistent with findings reported by Gui et al. [23], where energetic bombardment in sputtered AlTiCrN coatings promoted a shift in the preferred orientation from (111) to (200). Additionally, higher deposition temperatures enhance adatom mobility and facilitate defect annihilation, which promotes stress relaxation. This in turn reduces the driving force for (111) texture, thereby leading to a more random distribution of the crystallographic orientations [24], [25]. The reduction in the (111) plane orientation with increasing deposition temperature suggests a reduction in the densification of the coating structure, which has been associated with reduced tribological performance [4], [5].

Apart from the impact of the (111) plane on coating densification, it could also affect the roughness. In this regard, the emergence of more random plane orientations at elevated temperatures (Figure 6-8) may contribute to increasing surface roughness. This is based on the underlying principle that different planes exhibit varying growth rates; hence, the resulting competitive growth can lead to a less uniform surface morphology [7]. This might also explain why AlTiN/AlTiCrN coatings deposited at a lower temperature (T350), which contained fewer random planes, exhibited lower roughness values (Figure 6-4). In summary,

an increase in deposition temperature resulted in a reduction in the (111) plane texturing, accompanied by a competitive growth of other planes.

Further examination of the FWHM (full-width at half-maximum) values presented in Table 6-3 reveals the following distinct trends with increasing deposition temperature (T350 – T450): (1) peak broadening was observed for the (111) and (022) planes, as evidenced by the general increase in their FWHM values. (2) Peak narrowing was observed for the (020), (131), and (222) planes, indicated by the general reduction in their FWHM values.

Table 6-3. Peak positions and corresponding FWHM values of AlTiCrN/AlTiN coatings deposited at different temperatures.

Condition	(111) plane		(020) plane		(022) plane		(131) plane)		(222) plane	
	2 θ (°)	FWHM (°)	2 θ (°)	FWHM (°)	2 θ (°)	FWHM (°)	2 θ (°)	FWHM (°)	2 θ (°)	FWHM (°)
T350	43.32	0.51	50.76	1.55	74.74	0.82	90.63	2.00	95.11	1.22
T400	43.67	0.58	51.27	1.32	75.16	1.13	90.88	1.99	95.21	1.35
T450	43.49	0.57	51.10	1.26	75.10	0.85	90.62	1.78	95.42	1.06

These variations in the FWHM values suggest corresponding changes in the crystallite size and microstrain of the coatings. In general, higher FWHM values are associated with finer crystallite sizes and higher microstrain, whereas lower values typically signify coarser crystallites and reduced microstrain [4]. Using established equations from the literature [26], the average crystallite size and microstrain were calculated, with the results presented in Figure 6-9.

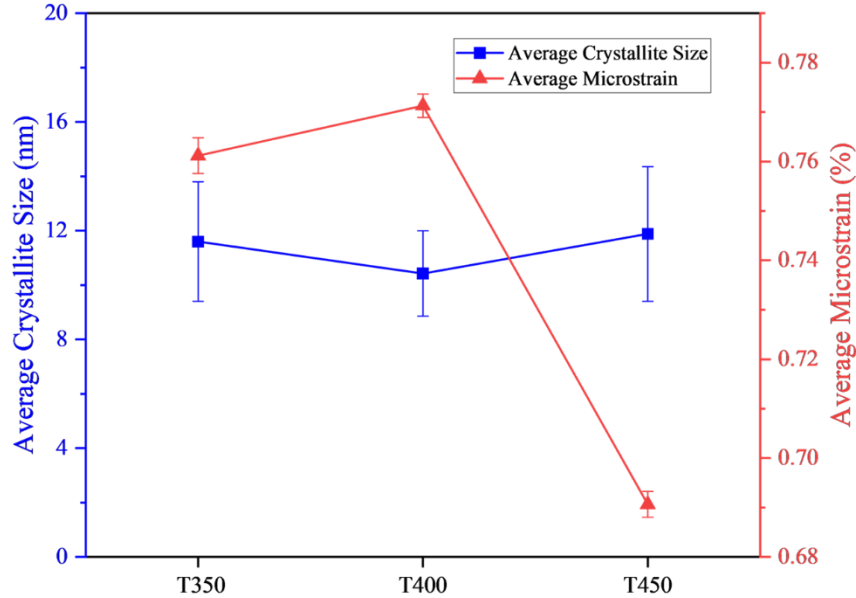


Figure 6-9: Average crystallite size and microstrain for AlTiN/AlTiCrN coatings with different substrate roughness.

The figure shows that increasing the deposition temperature (from T350 to T400) led to a general reduction in the average crystallite size from 11.6 to 10.4 nm, corresponding to an approximate 10.3 % reduction. However, a further increase in temperature in T450 resulted in an increase in the average crystallite size to 11.9 nm, representing a 14.4 % rise relative to T400. Comparable crystallite sizes, ranging between 9.9 – 13.2 nm, have been reported for AlTiN/AlTiCrN coatings different layered architectures [4]. The reduction in the average crystallite size (from T350 to T400) could be attributed to the enhanced coating bombardment by energetic species. This bombardment increases the impingement rate of the coating film, which suppresses the migration of crystallite boundaries, thereby leading to finer crystallites [8]. In contrast, the subsequent increment in the average crystallite size from T400 – T450 could be attributed to the combined effect of higher deposition temperature,

coupled with the additional heating resulting from increased energetic coating bombardment. These conditions promote adatom mobility and migration of crystallite boundaries, leading to crystallite coarsening [9], [10].

Furthermore, the figure reveals a slight increment in the average microstrain from 0.76 at T350 to 0.77 % at T400, followed by a dramatic decrease to 0.69 % at T450. Previous studies have reported microstrain values between 0.75 – 1.0 % for AlTiN/AlTiCrN coatings deposited on different substrate profiles. Generally, the occurrence of microstrain is attributed to defects such as vacancies, dislocations, and lattice distortions resulting from solid solution formation [25]. Using established equations from literature [27], the computed dislocation density also aligns with this trend. Explicitly, the dislocation density increased from 7.4 to 9.2×10^{15} lines/m² (T350 to T400), consistent with the increase in microstrain, driven by more intense energetic bombardment, following the increase in temperature. Meanwhile, the drastic strain relaxation upon further increase in temperature to T450 could be attributed to the annihilation of defects, evident in the reduction in dislocation density (to 7.0×10^{15} lines/m²). Additionally, the reduced contribution of the (111) plane at higher temperatures (Figure 6-8) may also diminish lattice distortions resulting from substitutional and interstitial defects [11], thereby lowering microstrain at T450. This interpretation is consistent with previous studies reporting that higher microstrain in AlTiN/AlTiCrN coatings deposited on smoother tool steel substrates originates from enhanced crystallographic ordering in the dense (111) plane, which promotes lattice distortion [5]. In summary, increasing the deposition temperature (T350 – T400) initially resulted in finer crystallites and higher microstrain attributed to intensified energetic bombardment, but

further temperature rise (T400 – T450) promotes migration of crystallite boundaries and defects annihilation, producing lower microstrain and larger crystallites.

6.3.2 Mechanical properties

6.3.2.1 Effect of deposition temperature on hardness, H/E and H^3/E^2 ratios

Figure 6-10a presents the force-displacement curves obtained from the nanoindentation test, showing the loading and unloading stages. The positioning of the curves appears not to follow a distinct trend with deposition temperature. For instance, the T400 curve is positioned furthest to the left, while the T450 appears on the far right, with the T350 curve between them. The rightward shift of the curve results from deeper indenter penetration into the coating, suggesting greater coating deformation for T350 and T450, compared to T400. The deformation energy, calculated as the integral of the area under the force-displacement curve [25], supports this observation. Explicitly, the T400 exhibited the lowest deformation energy (3.28 nJ), with the highest recorded for T450 (3.97 nJ). The lower deformation energy at T400 implies reduced work by the indenter, owing to greater coating resistance. In line with established understanding, lower deformation energy and a leftward curve shift are indicators of higher coating hardness, evident in the T400 condition [4], [27].

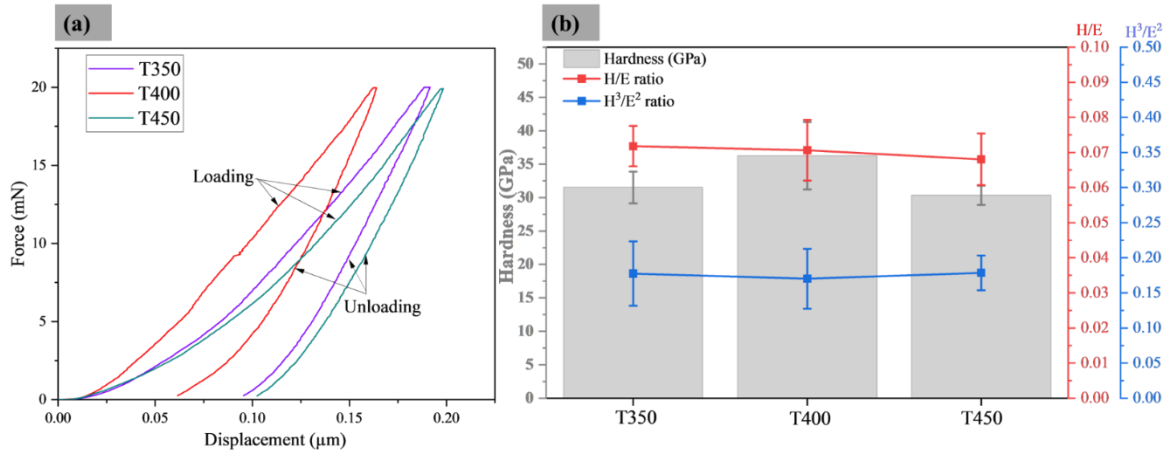


Figure 6-10. (a) Load-displacement curves and (b) Hardness and indices of fracture resistance of the AlTiN/AlTiCrN coatings deposited at different

The hardness results presented in Figure 6-10b further confirm that the T400 exhibited the highest hardness (36 GPa), reducing by about 13 % in T350 and 16 % in T450. These values are consistent with previous reports of AlTiN/AlTiCrN coatings, where hardness in the range of 27 – 34 GPa was observed under different substrate conditions and multilayer configurations [4], [5]. Additionally, the H/E and H^3/E^2 ratios are widely used indicators for evaluating the fracture resistance and tribological behaviour of PVD coatings, where higher values correlate with improved performance [7]. From Figure 6-10b, the H/E ratio decreases slightly with temperature, from 0.072 at T350 to 0.068 at T450, corresponding to about 6 % reduction. Meanwhile, the H^3/E^2 ratio was lowest at T400 (0.177), with comparable values at T350 and T450 (~ 0.178). Given the error margins, these differences can be regarded as negligible. Nonetheless, the ratios reported here align with those found in our previous study on AlTiN/AlTiCrN coatings deposited on different tool steels [2]. Overall, the higher H/E ratio at T350, indicate coatings deposited at T350 may offer superior fracture resistance [7].

Furthermore, concerns have been raised in prior studies that excessively high deposition temperatures can degrade the hardness of the tool steel substrate, thereby compromising cutting performance and tool life [28]. However, as shown in Table 6-4, the substrate hardness remained largely unaffected by the deposition temperature, with values consistently around 6 GPa. In summary, while the coatings achieved the maximum hardness at T450, a balance of hardness with fracture resistance-related indices was obtained at T350.

Table 6-4. Hardness of tool steel substrates of AlTiCrN/AlTiN coatings deposited at different temperatures.

Condition	Hardness (GPa)
T350	5.97 ± 0.12
T400	6.00 ± 0.09
T450	5.94 ± 0.16

The tool steel substrate hardness was obtained using $HV (GPa) = 0.00185 (P/d^2)$; P-load (N) and d-average diagonal length (mm) [29].

6.3.2.1.1 Effect of (111) plane, crystallite size, and microstrain on hardness and fracture resistance

The hardness trend exhibited by the coatings could be explained in terms of factors such as the crystallographic orientation, crystallite size, and microstrain [4]. Figure 6-11 shows the relationship between hardness and the (111) crystallographic plane. Previous studies have observed higher fractions of the densely packed (111) plane to enhance coating hardness through solid solution strengthening and impeding dislocation movement [14]. From the figure, no clear linear correlation between hardness and the (111) plane is observed in this study. For instance, the T350 condition, which contained the highest fraction of the (111) plane, presented a hardness value 13 % lower than that of T400, despite having 24 %

higher fraction of the (111) plane. This suggests that additional factors had a more dominant effect on the hardness of the coatings.

A clearer relationship is observed between crystallite size and hardness. Figure 6-11 shows that an increase of about 14 % in crystallite size (from T400 to T450) corresponded to approximately 16 % reduction in the hardness. This behaviour aligns with the Hall-Petch phenomenon, where finer crystallites, as in the T400 condition, provide more boundaries that act as barriers to dislocation movement, thereby enhancing resistance to plastic deformation [30]. In support of this, Muhammed et al. reported a ~ 64 % increase in hardness in AlTiN/AlTiCrN-based layered coatings, associated with about 25 % reduction in crystallite size. The more pronounced effect observed in their study was attributed not only to fine crystallite size, but also to strengthening from multiple interfaces of the layered structure, as well as coating densification and strain hardening due to increased substrate bias [4].

Furthermore, coating hardness could also be impacted by the microstrain. In this regard, increased microstrain introduces stress fields caused by lattice distortions during solid solution formation, which further hinders dislocation movement [31]. This effect is evident in the higher hardness of the T400, which exhibited about 11 % greater microstrain compared to T450. Further quantitative analysis revealed that a 1 % increase in microstrain and a 1 % decrease in crystallite size resulted in about 1.7 and 1.4 % increase in hardness, respectively. These results highlight that the microstrain exerted a stronger influence on hardness than the crystallite size. Additionally, the generally superior fracture resistance observed for T350, particularly in relation to T400, could be linked to its higher fraction of the densely packed (111) plane, which is effective at impeding crack initiation and

propagation [5]. In summary, the enhanced hardness at T400 could be attributed to the dominant effect of higher microstrain and finer crystallites, while the superior fracture resistance at T350 is associated with its higher (111) plane fraction.

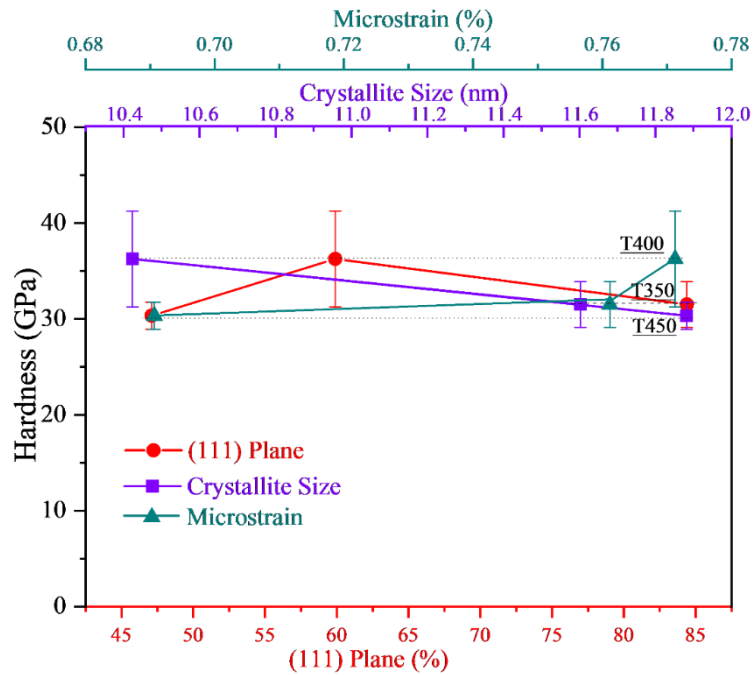


Figure 6-11. Relationship between hardness, (111) plane orientation, crystallite size, and microstrain of AlTiN/AlTiCrN coatings at different deposition temperatures.

6.3.2.2 Effect of deposition temperature on adhesion strength

The scratch test results presented in Figure 6-12a–c show the variation of frictional force, normal load, and acoustic emissions with the scratch length at different deposition temperatures. The acoustic emissions, which represent signals generated by the indenter-coating interaction, were minimal within the first 1.2 mm of the scratch length for the T350 compared to the other coatings. Explicitly, the peak acoustic emissions in the first 1.2 mm

reached approximately 6.5, 9.2, and 11.6 % for T350, T400, and T450, respectively. The increase in both the maximum peak intensity and their earlier occurrence with increasing deposition temperature indicates more severe coating failure and shorter time to failure [32]. Furthermore, fluctuations in the frictional forces were also observed at different deposition temperatures, particularly for T350, suggesting a higher coefficient of friction (COF) [33]. This observation is confirmed by the COF plot in Figure 6-12d, where T350 generally presented the highest COF, reaching a maximum of about 0.70, compared to 0.65 and 0.66 for T400 and T450, respectively. The higher COF exhibited at T350 could be linked to the relatively larger MP sizes (Figure 6-2), which promote stronger asperity interactions [7].

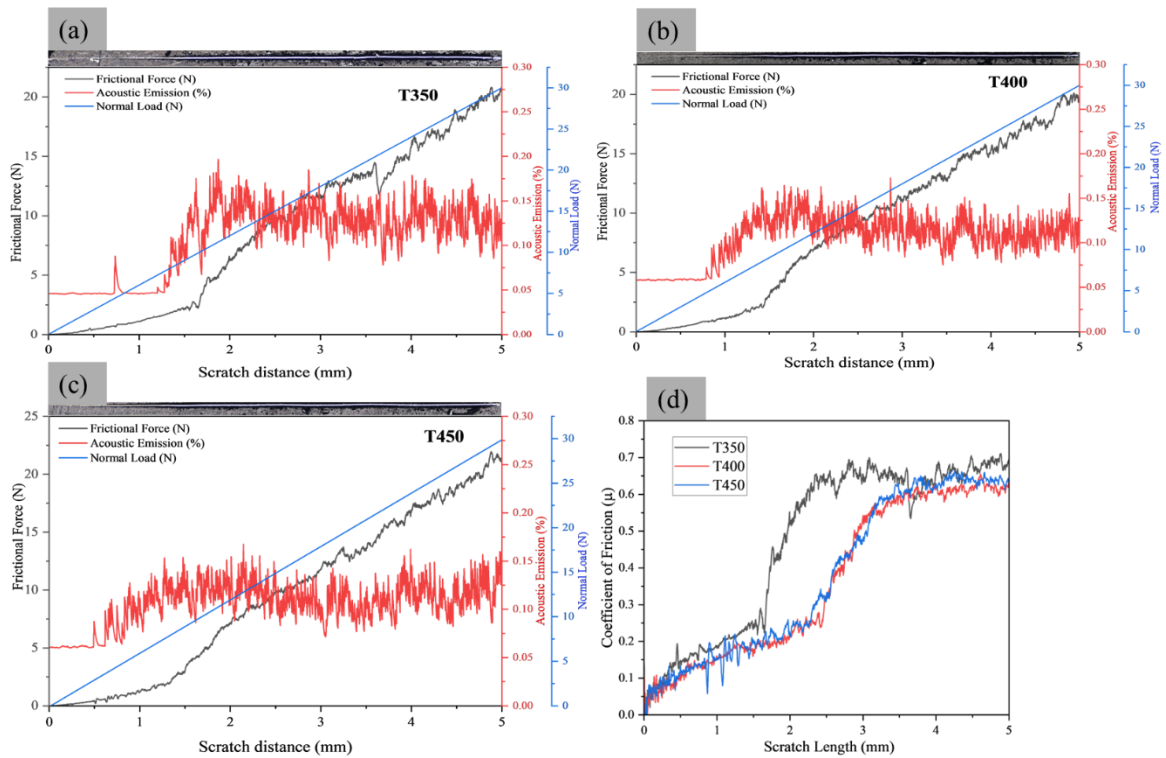


Figure 6-12. Frictional force, acoustic emission, and normal load as a function of scratch length for AlTiN/AlTiCrN coatings: (a) T350; (b) T400; (c) T450; and (d) coefficient of friction at different temperatures.

Further quantification of the coating adhesion strength was performed using the critical load corresponding to adhesive coating failure [4], with results presented in Figure 6-13. The maximum critical load (~ 11 N) was obtained at T350, while T400 and T450 exhibited ~ 17 and 29 % lower values, respectively. This trend suggests superior adhesion strength at lower deposition temperatures. This is in agreement with the findings of Warcholinski et al. [10], who reported about a 17 % decrease in the critical load of AlCrN coatings deposited on tool steels as deposition temperature increased from $350 - 450$ °C.

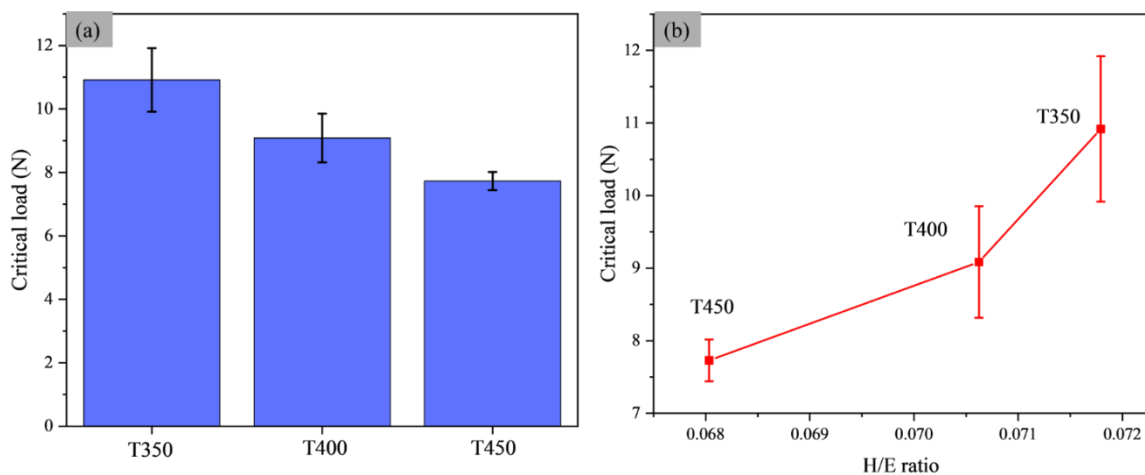


Figure 6-13. (a) Critical load and (b) Relationship between critical load and H/E ratio for AlTiN/AlTiCrN coatings deposited at different temperatures.

The superior adhesion strength observed at T350 could be explained by the combined effects of fracture resistance and coating roughness. As shown in Figure 6-13b, the critical load increases with the fracture resistance index (H/E ratio). Consequently, the enhanced

fracture resistance of T350 coatings enables them to absorb more energy during tribological interactions without fracturing. This increases the coating resistance to crack propagation, thereby lowering the likelihood of delamination under external loading [4]. Additionally, the lower roughness at T350 (Figure 6-4a) further contributes to the enhanced adhesion strength. This is consistent with previous reports on AlTiN/AlTiCrN coatings, where smoother surface morphology was associated with enhanced adhesion strength [5]. This occurrence is attributed to the reduction of roughness-induced asperities that generate localized stresses leading to premature failure [16]. In summary, superior adhesion strength was obtained at lower deposition temperature owing to lower roughness and enhanced fracture resistance.

6.3.3 Wear resistance

Figure 6-14 shows the wear rates of the coatings, with the T350 condition exhibiting the least wear rate ($\sim 2 \times 10^{-4} \text{ mm}^3/\text{Nm}$), suggesting superior wear resistance at lower deposition temperature. With increasing deposition temperature, the wear rate increased by about 36 and 52 % at T400 and T450, respectively. These wear rate values are comparable to those previously reported for cathodic arc evaporated AlTiN/AlTiCrN coatings, which were in the range of $1.8 - 2.2 \text{ mm}^3/\text{Nm}$ [5].

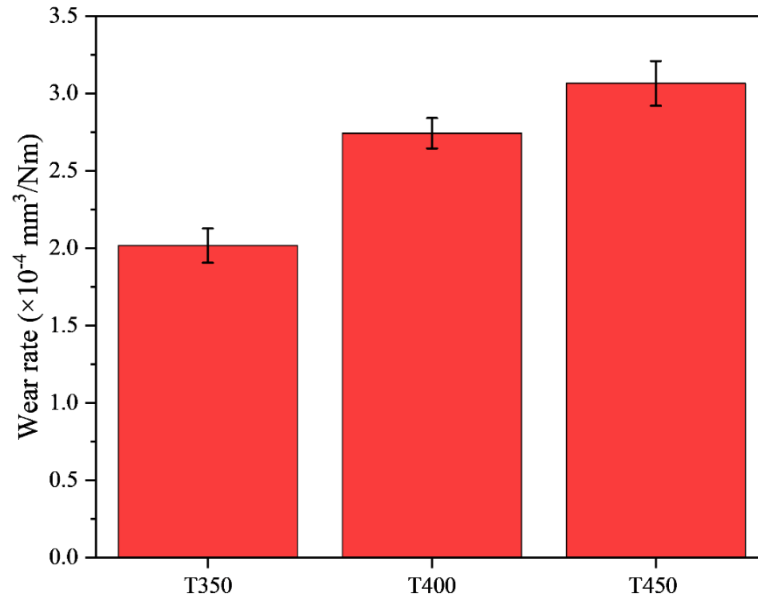


Figure 6-14. Wear rates of the AlTiN/AlTiCrN coatings at different deposition temperatures.

Further analysis of the wear track of the samples was conducted to gain insights into the wear mechanisms. The results of the analysis presented in Figure 6-15 reveal the presence of two distinct zones. The dark zone (Zone A; Figures 15a-c) contained predominantly coating elements (such as Al, Ti, and N) with relatively low Fe content, as confirmed by EDX analysis (P1-3, Table 6-5). This indicates the retention of coating material in these regions. Meanwhile, the bright zone (Zone B; Figures 15d-f) presented higher Fe content alongside reduced coating elements (P4-6, Table 6-5), signifying complete coating removal leading to substrate exposure [4]. Further examination of the exposed substrate region (zone B) revealed the occurrence of abrasive microcutting grooves at all deposition temperatures. This is likely due to the higher hardness of the abrasive silica particle (about 8.5 GPa [2]), compared to the tool steel substrate (about 6 GPa, Table), which provides the driving force for abrasive wear, in the form of microcutting grooves [34].

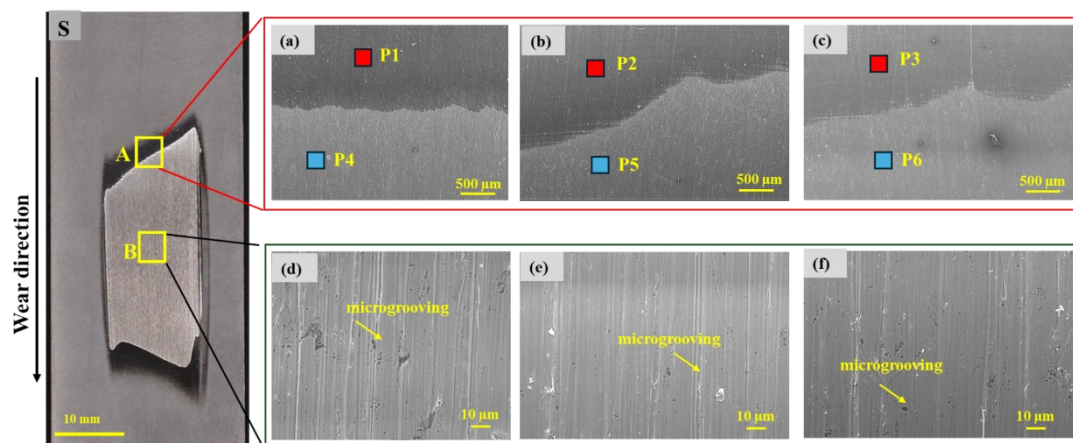


Figure 6-15. Micrographs of the wear tracks: (S) Overview; (a, d) T350; (b, e) T400; (c, f) T450.

In addition, silica contamination, known to facilitate tribochemical coating degradation [2], was also detected in both coating-containing and substrate-exposed regions. Notably, the relatively higher levels of Si contamination recorded in the region of the exposed substrate (0.3 – 1 wt. %, P4-6, Table 6-5) compared to the coating-containing region (P1-3, Table 6-5) highlight the protective role provided by the AlTiN/AlTiCrN coating in limiting silica-induced damage. More so, the presence of O detected in the wear track is most likely due to atmospheric exposure during wear testing, consistent with previous reports [35]. In summary, increasing the deposition temperature led to reduced wear resistance, as evidenced by the higher wear rates. The wear tracks were characterized by regions of partially abraded coating alongside exposed substrate with microcutting grooves, reflecting a transition from protective coating-dominated wear to substrate-driven abrasive wear.

Table 6-5. EDX analysis of the worn samples (corresponding to Figure 6-15).

Spectra	Composition (at. %)						
	Al	Ti	Cr	N	O	Si	Fe
P1	23.0	14.9	0.6	46.9	9.0	0.3	5.3
P2	20.8	15.4	1.7	34.6	19.0	1.0	7.6
P3	19.7	15.2	6.6	45.2	11.7	0.1	1.6
P4	4.6	1.9	7.6	1.0	11.5	2.8	70.5
P5	0.2	0.3	6.6	–	27.5	7.4	58.1
P6	0.3	0.1	9.4	–	3.4	2.5	84.3

6.4 Discussion

The variations in the wear behaviour of the AlTiN/AlTiCrN coatings with deposition temperatures could be explained by the differences in their morphological and microstructural characteristics such as MP statistics (Figure 6-2), coating thickness (Figure 6-5), coating composition (Figure 6-7, Table 6-2), microstrain and crystallite size (Figure 6-9), as well as their mechanical properties including hardness and H/E ratio (Figure 6-10), and adhesion strength (Figure 6-13). These discussions are elaborated under the following subsection.

6.4.1 Influence of coating properties on wear performance

The relationship between the wear rate, hardness, and H/E ratio of the AlTiN/AlTiCrN coatings deposited at different temperatures is presented in Figure 6-16. Previous studies have reported that coatings with reduced hardness typically exhibit higher wear rates [4], [14]. In the present study, although no clear linear correlation was observed between the wear rates and the hardness, the relatively lower hardness of the T450 coating

might have contributed to its higher wear rates. In this regard, the lower hardness diminishes the load-bearing capacity of the coating and resistance to plastic deformation, culminating in more material removal, leading to higher wear rates [25]. Meanwhile, the T350 coating demonstrated superior wear resistance despite having a lower hardness compared to the T400, suggesting the influence of additional factors.

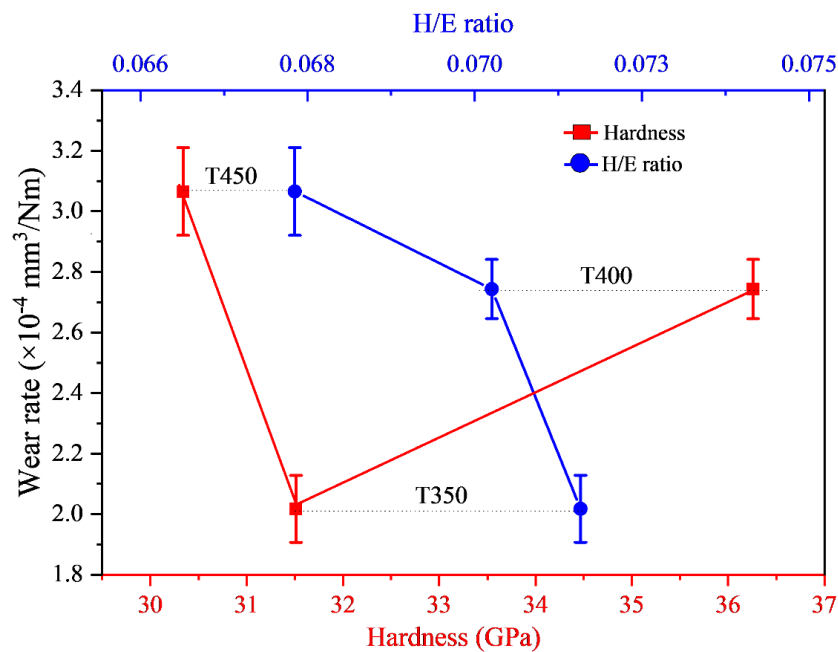


Figure 6-16. Relationship between wear rate, hardness, and H/E ratio of AlTiN/AlTiCrN coatings deposited at different temperatures.

Furthermore, as shown in Figure 6-16, the H/E ratio exhibited an inverse relationship with wear rates, with higher values corresponding to lower wear rates, as observed in the T350 coating. The current finding corroborates previous research, which established that PVD coatings with greater H/E ratios tend to exhibit better wear performance [14], [19]. Consequently, the superior wear resistance of the T350 coating could be attributed to its higher H/E ratio, which enhances the coating's elastic strain tolerance and fracture toughness. This improvement helps to resist crack initiation and propagation during abrasive loading, thereby reducing material loss [2], [36]. Further quantitative analysis to ascertain the sensitivity of the wear rates to changes in hardness and H/E ratio revealed that 1 % decrease in hardness and H/E ratio from T350 to T450 resulted in about 14.0 and 9.9 % increments in the wear rates, respectively, suggesting that the hardness exerts a greater influence on the wear behaviour compared to the H/E ratio.

The relationship between the adhesion strength (i.e., critical load), coating thickness, and surface roughness is presented in Figure 6-17. From the figure, a negative correlation is observed between wear rates and both critical load and coating thickness, where higher critical loads and thicker coatings are associated with lower wear rates (i.e., T350). This is consistent with findings from our previous study, where we observed AlTiN/AlTiCrN-based coating architectures with higher coating thickness and critical load consistently exhibited superior wear performance [4]. The lower wear rates observed at higher critical load in T350 coating could be attributed to stronger interfacial bonding, which minimizes the risk of coating delamination during abrasive contact [36]. In addition, the thicker T350 coatings offer improved load-bearing capacity, which reduces shear stress by distributing the applied

load over a larger material volume, thereby mitigating material removal compared to the thinner T400 and T450 coatings [2].

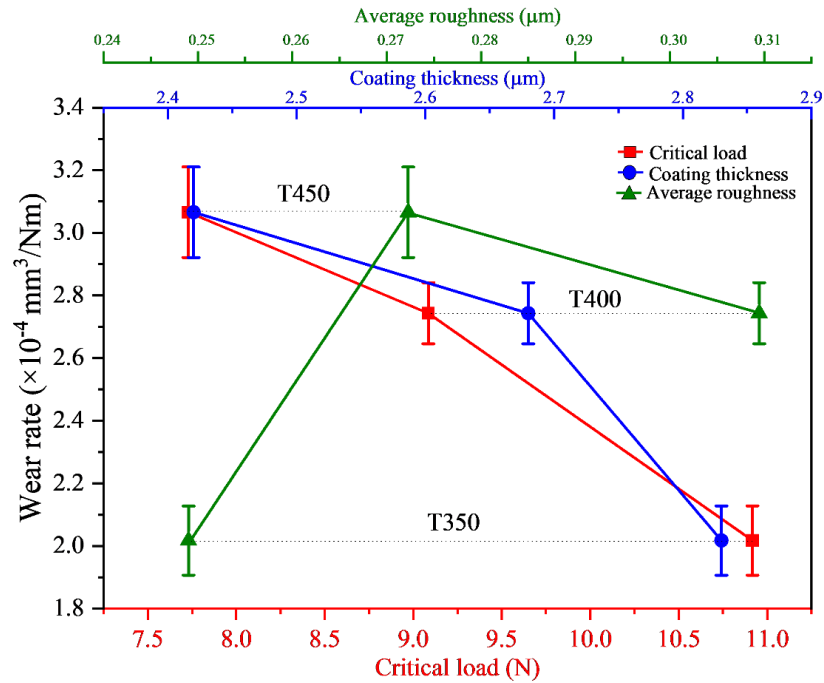


Figure 6-17. Relationship between wear rate, critical load, coating thickness, and roughness of AlTiN/AlTiCrN coatings deposited at

Furthermore, despite no distinct linear relationship being observed between the surface roughness and wear rate, coatings with higher surface roughness (such as in the T400 and T450) exhibited higher wear rates. This is consistent with findings from previous studies [2], [4], which associated increased surface roughness with more pronounced surface asperities that act as initiation sites for localized stress accumulation. Consequently, the T350 coating's smoother surface, on the other hand, minimizes stress concentration, leading to reduced material loss; hence, the lower wear rates [7]. Further quantitative analysis revealed that a 1 % decrease in critical load and coating thickness, and a 1 % increase in average

roughness from T350 to T450, resulted in 1.8, 3.6, and 5.6 % increments in the wear rate, indicating that the surface roughness has the most significant impact on the wear rates among the parameters. In summary, the wear rates decreased with the H/E ratio, coating thickness, and adhesion strength, translating to improved tribological behaviour at higher values of these parameters, as in T350 condition. Conversely, coatings with higher roughness (T400 and T450) displayed elevated wear rates, indicating poorer performance. The quantitative analysis revealed that the impact of the properties on the wear performance of the coatings ranks in the decreasing order: hardness > H/E ratio > roughness > coating thickness > adhesion strength. The superior wear resistance of the T350 coating can therefore be attributed to its intermediate hardness, higher H/E ratio, smoother surface, greater coating thickness, and stronger interfacial bond; all of which enhanced its load bearing capacity and resistance to crack propagation under sliding conditions.

6.5 Conclusion

The following are the key findings from the study on the effect of deposition temperature (350 – 450 °C; T350 – T450) on the wear behaviour of AlTiN/AlTiCrN ceramic coatings deposited on AISI A8 tool steel:

1. The MP characteristics varied with temperature; larger MPs formed at T350 due to MP agglomeration-coalescence, while T400 produced the highest density of fine MPs due to MP bombardment-induced fragmentation. At T450, MP density decreased, but coalescence led to larger MPs. Additionally, surface roughness correlated positively with MP density, yielding rougher surfaces and inferior wear performance at T400 and T450.

2. Increasing the deposition temperature reduced coating thickness and (111) plane texturing, while all coatings maintained stoichiometric compositions with nearly constant Al/(Al+Ti+Cr) ratio.
3. From T350 to T400, increased energetic bombardment produced finer crystallites and higher microstrain, but further temperature rise to T450 promoted crystallite growth and defect annihilation, resulting in larger crystallites and lower microstrain.
4. Maximum hardness occurred at T400, driven by finer crystallites and higher microstrain. However, T350 provided the optimal balance of hardness with fracture resistance indices (H/E and H^3/E^2) owing to its higher (111) plane fraction. Additionally, the superior adhesion strength of T350 was attributed to its smoother surface and enhanced fracture resistance.
5. Wear performance was influenced by mechanical and microstructural factors, with T350 exhibiting superior wear performance. The wear rates decreased with increasing H/E ratio, coating thickness, and adhesion strength, but increased with roughness. Quantitative analysis ranked the influence of coating properties on wear as: hardness > H/E ratio > roughness > coating thickness > adhesion strength.

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Chapter 7

Conclusions and recommendations

7.1 General conclusions

This research work systematically investigated the tribological performance enhancement of AlTiN/AlTiCrN-based coatings deposited on AISI A8 tool steel through three key process variables: substrate roughness, layered architectural design, and deposition temperature. The project combined extensive microstructural, mechanical, and tribological analyses to establish microstructure–property–performance correlations. The key conclusions drawn from this work are as follows:

1. The study demonstrated that substrate surface topography plays a decisive role in determining coating microstructural evolution, mechanical properties and tribological response. Increasing substrate roughness led to higher coating roughness, thicker coatings, and a decline in preferred (111) orientation and microstrain, resulting in reduced hardness and wear resistance. Conversely, smoother substrates enhanced coating adhesion, hardness-to-modulus ratios (H/E and H^3/E^2), and overall wear performance. The results established the (111) plane orientation, adhesion strength, hardness, and macroparticle size as the most influential parameters governing tribological performance.
2. The development of novel AlTiN/AlTiCrN-based multilayer coatings revealed that optimized architectural configurations achieved by adjusting substrate bias and reactive nitrogen gas flow significantly improved hardness, adhesion, and wear resistance. Enhanced

(111) orientation, finer crystallite structure, and reduced roughness in advanced architectures (AlTiCrN II and III) contributed to superior mechanical and tribological properties. Quantitative analyses confirmed that wear rate is most sensitive to surface roughness and least influenced by the H^3/E^2 ratio, underscoring the importance of smooth, well-adhered multilayer structures for high-performance cutting applications.

3. Variations in deposition temperature was found to critically influence macroparticle formation, surface morphology, and coating densification of AlTiN/AlTiCrN coatings. Coatings deposited at 350 °C exhibited the best combination of microstructural composition, fracture resistance, adhesion strength, and wear performance. Although T400 yielded maximum hardness due to finer crystallites and higher microstrain, T350 provided the most balanced mechanical and tribological performance, demonstrating that an optimal thermal energy input is essential to achieving structural stability and improved wear resistance.

4. Throughout the study, the tribological performance of AlTiN/AlTiCrN coatings was found to be a complex interplay of microstructural features (such as crystallite size, texture, and macroparticle density) and mechanical parameters (e.g., hardness, H/E , and adhesion strength). The research established clear quantitative correlations that enable predictive assessment of coating performance, thereby contributing to the broader understanding of structure–property relationships in PVD coatings.

7.2 Recommendations

Based on the findings from this study, the following recommendations are proposed to guide future research in the development and optimization of advanced PVD coatings for tribological applications:

1. Future studies should investigate the influence of carbon incorporation on AlTiN/AlTiCrN layered coatings. Controlled carbon doping has been reported to improve hardness, reduce friction, and enhance wear resistance. Optimizing the carbon content could reveal new pathways for improving the microstructural stability and wear resistance of these coatings under wood-machining conditions.
2. The present study maintained a constant arc current during deposition of the AlTiN/AlTiCrN coating. However, variations in arc current and target power are known to strongly influence ion energy and film density. Therefore, systematic exploration of these parameters would provide valuable insights into how plasma energetics affect the coating's microstructure, adhesion, and tribological performance.
3. Substrate etching potential during deposition plays crucial roles in determining coating adhesion and interfacial quality. Future work should therefore examine how variations in etching potential and durations affect interfacial bonding, load-bearing capacity of the AlTiN/AlTiCrN layered coatings, and overall tribological performance.
4. Beyond the AlTiN/AlTiCrN bilayer configuration studied here, further research should consider more complex multilayer architectures, such as those incorporating additional elements such as Si, Mo, V or C (e.g., AlTiCrSiN or AlTiCrVN). Comparative studies on these quaternary and quinary systems could help identify the optimal architecture for achieving superior tribological performance.
5. While this study focused on room-temperature tribological performance, real-world wood-machining and tooling operations often involve elevated temperatures and dynamic

contact conditions. Field trials and high-temperature wear tests would therefore be essential to validate laboratory findings and assess the coatings' long-term stability and durability.

List of publications and conference presentation

Journal publications

1. **Muhammed M**, Javidani M, Heidari M, Jahazi M. "Enhancing the tribological performance of tool steels for wood-processing applications: A comprehensive review." *Metals* 13.8 (2023): 1460. DOI: <https://doi.org/10.3390/met13081460>
2. **Muhammed M**, Javidani M, Ebrahimi Sadrabadi T, Heidari M, Levasseur T, Jahazi M. "A comprehensive review of cathodic arc evaporation physical vapour deposition (CAE-PVD) coatings for enhanced tribological performance." *Coatings* 14.3 (2024): 246. DOI: <https://doi.org/10.3390/coatings14030246>
3. **Muhammed M**, Javidani M, Ebrahimi Sadrabadi T, Heidari M, Levasseur T, Jahazi M. "Effect of Layered Architectural Design on the Tribological Behaviour of AlTiN/AlTiCrN-Based Cathodic Arc Evaporated Physical Vapour Deposition Coatings." *Metallurgical and Materials Transactions A* (2025): 1-24. DOI: <https://doi.org/10.1007/s11661-025-07802-8>
4. **Muhammed M**, Javidani M, Ebrahimi Sadrabadi T, Heidari M, Levasseur T, Jahazi M. " A Comparative Wear Analysis of Tool Steels: An Evaluation Based on ASTM G65 and G195 Testing Methods." *Journal of Materials Engineering and Performance* (2025): 1-21. DOI: <https://doi.org/10.1007/s11665-025-12019-2>
5. **Muhammed M**, Javidani M, Heidari M, Levasseur T, Jahazi M. " Effect of Substrate Roughness on the Tribological Performance of AlTiN/AlTiCrN-coated A8 Tool Steel.

- " *Metallurgical and Materials Transactions A* (2025): 1-19. DOI: <https://doi.org/10.1007/s11661-025-08013-x>
6. **Muhammed M**, Javidani M, Heidari M, Levasseur T, Jahazi M. " Influence of Substrate Surface Finish on the Wear and Corrosion Performance of AlTiN/AlTiCrN Coatings." " *Tribology Letters* (2025): 1-16. DOI: <https://doi.org/10.1007/s11249-025-02100-1>
7. **Muhammed M**, Javidani M, Heidari M, Levasseur T, Jahazi M. " Influence of Deposition Temperature on the Microstructure, Wear and Corrosion Behaviour of AlTiN/AlTiCrN Ceramic Coatings on A8 Tool Steel." *Ceramics International* (2025): 1-16. DOI: <https://doi.org/10.1016/j.ceramint.2026.04.059>

Conference presentation

1. **M. Muhammed**, M. Javidani, T. E. Sadrabadi, M. Heidari, T. Levasseur, and M. Jahazi, "Enhancing the tribological performance of tool steel through layered AlTiCrN PVD coating: Application to woodcutting tools," in *Proc. COM & LightMat, Montreal, QC, Canada*, Jul. 7–10, 2025.
2. **M. Muhammed**, M. Javidani, M. Heidari, T. Levasseur, and M. Jahazi, "Influence of Substrate Roughness and Coating Architecture on the Tribological Performance of AlTiN/AlTiCrN Coatings: Application to Wood Cutting Tools," in *Proc. Canadian Materials Science Conference (CMSC), Quebec City, QC, Canada*, May 4–7, 2026.

Appendix: Response to Thesis Evaluation Committee Comments

Note: *Chapters 2–6 of this thesis are based on peer-reviewed published manuscripts. Direct revision of these chapters is not feasible; however, comprehensive scientific responses and clarifications are provided below for all comments raised.*

Response to Comments by Prof. Christian Moreau (Concordia University)

Thank you for the thorough and constructive evaluation of this thesis. The comments have helped clarify important aspects of the work. Please find my point-by-point responses below.

Chapter 1: Introduction

Comment 1.1: This chapter is very well structured and clear. It is unexpected to refer to a paper (Ref 11) published by the candidate in the section Background of study. Indeed, the candidate claims that:

“Recently, layered AlTiN/AlTiCrN-based coatings have emerged as promising candidates for woodworking applications. These coatings combine the benefits of a multilayered architecture with the superior properties of quaternary AlTiCrN components to achieve enhanced mechanical strength, oxidation resistance, and chemical stability, ultimately extending cutting tool life [11].

Response 1.1: Thank you for this insightful observation. I agree that citing the candidate’s own work within the Background section may not be appropriate in this context. Accordingly, the original reference has been replaced with a more suitable and independent source to better support the statement. The revised citation is:

“[11] A. E. Santana, A. Karimi, V. H. Derflinger, and A. Schütze, “Microstructure and mechanical behavior of TiAlCrN multilayer thin films,” *Surf. Coatings Technol.*, vol. 177–178, pp. 334–340, 2004, doi: 10.1016/j.surfcoat.2003.09.023.”

Comment 1.2: In this introductory chapter, one would have liked to understand the rationale behind selecting the CAE process and the nature of the deposited coatings and substrate

Response 1.2: Thank you for this observation. However, the objective of the introductory chapter was to introduce the general concept of PVD coatings and their preference over other tool life enhancement techniques, which was sufficiently addressed. The concept of cathodic arc evaporation was reserved for subsequent chapters. Consequently, the rationale for selecting the CAE-PVD process over other deposition techniques is extensively discussed in Chapter 3 (Sections 3.3 and 3.7), which constitutes a comprehensive review of CAE-PVD coatings. Specifically, CAE-PVD was selected for the following reasons: (i) it produces highly ionized vapor (ionization rate > 90%), yielding dense, adherent coatings with superior hardness; (ii) it enables deposition of multicomponent ceramic nitride coatings (AlTiN, AlTiCrN) with precise composition control; (iii) the process is well-established industrially for cutting tool coatings; and (iv) it operates at relatively low substrate temperatures (350 – 450 °C), compatible with tool steel substrates. The AISI A8 tool steel substrate was selected due to its widespread industrial use in wood-cutting tools (e.g., saw blades), as it offers a good balance of toughness, wear resistance, and cost-effectiveness (as detailed in Section 3.2 and Chapter 4).

Chapter 2: Review of Tribological Performance of Tool Steels

Comment 2.1: This chapter provides an in-depth review of the challenges associated with the wear performance of tool steels in the wood processing industry. It offers an exhaustive critical analysis, identifying gaps and suggesting future research directions. It appears that references to some tables and figures that have already been published are not included in the corresponding captions. For example, see Table 2-4 and Figure 2-8.

Response 2.1: Thank you for this careful observation. I would like to clarify that Table 2–4 and Figure 2–8 are original contributions developed by the authors based on the analysis presented within this thesis, as highlighted in the thesis. As such, no external citations are required.

Chapter 3: Review of CAE-PVD Coatings

Comment 3.1: In this chapter, the candidate provides an excellent review of CAE coatings used for wear resistance. Chapters 2 and 3 together are a very good literature review on the subjects addressed in this thesis. In Chapter 3, key elements relative to the process, coating microstructures and wear properties are very well summarised and presented. On page 78, it is mentioned that the evaporative PVD processes provides a high ionization rate of the produced vapor. This is likely true for CAE but not for the other evaporation processes such as electron beam (EB-PVD).

Response 3.1: Thank you for this comment. The statement on page 78 was indeed referring specifically to the cathodic arc evaporation (CAE) process, which is characterized by a very high ionization rate (typically > 95 % for metallic species), distinguishing it from other PVD evaporation methods such as electron beam evaporation (EB-PVD), which typically achieves ionization rates of only 1–5% without additional ionization assistance. The broader statement was intended to justify the advantages of the CAE process, not to generalize across

all evaporation-based PVD techniques. Since Chapter 3 has been published, a direct revision of the text is not feasible. However, I acknowledge that a more precise qualification, specifying ‘cathodic arc evaporation’ rather than ‘evaporative PVD’, would have improved scientific accuracy. This distinction is an important consideration for future review publications.

Comment 3.2: In Section 3.7.2, I assume that deposition temperature is actually the substrate temperature during the deposition process and not the target temperature. This should have been clarified in the text.

Response 3.2: Thank you for this valuable clarification. The reviewer’s interpretation is indeed correct. In the context of cathodic arc evaporation PVD processes, the term deposition temperature refers to the substrate temperature during coating deposition, rather than the target (cathode) temperature. However, the use of deposition in the thesis follows established convention in the PVD and tribology literature (<https://doi.org/10.1016/j.surfcoat.2022.128304>), where deposition temperature is commonly understood to denote the substrate temperature, as it is the controlled and experimentally relevant parameter governing adatom mobility, microstructural evolution, and residual stress development.

Chapter 4: Effect of Substrate Roughness

Comment 4.1: In Figure 4-7(b), the indicated positions for the substrate, AlTiN and AlTiCrN don’t look consistent with the element compositions determined in the line scan. This should be clarified or corrected. Additionally, the voltage used for the EDX analysis should be specified as it influences the lateral and depth resolution of the analysis as well as its precision.

Response 4.1: Thank you for this observation. Figure 4-7(b) presents the cross-sectional EDX line scan of the AlTiN/AlTiCrN bilayer coating. The layer boundaries indicated in the figure were determined based on the elemental transitions observed in the compositional profiles. Specifically, the AlTiN layer is identified by the absence of the Cr signal (characteristic of AlTiCrN), and the interface with the substrate is identified by the transition from coating-specific elements (Al, Ti, N) to the substrate-dominated composition (Fe, Cr, Mo). I acknowledge that the label positions may appear visually offset due to the figure scaling. In the published version, this may reflect a rendering artefact. The authors confirm that the layer assignments are consistent with the EDX compositional data and are corroborated by the cross-sectional SEM micrograph (Figure 4-7a). Detailed numerical composition values at selected positions are also provided in Table 4-2 of the thesis. Additionally, all EDX analyses were conducted at an accelerating voltage of 20 kV using a scanning electron microscope (SEM, JSM-6480LV, Japan) equipped with an energy-dispersive X-ray (EDX, INCA energy 250), operating in the secondary electron imaging mode. At this voltage, the electron interaction volume in the coating is estimated to be on the order of 0.5–2 μm , depending on coating density. For the coating thicknesses studied (2 – 5 μm), the interaction volume may partially extend into the substrate at the lower end of the coating thickness range, which is acknowledged as a limitation of EDX analysis for thin film characterization. Since the papers are already published, adding this detail to the published chapters directly is not possible, but we confirm this information for the record and note that it is consistent with standard practice in the field for this type of analysis.

Comment 4.2: The method used to determine the texture of the different samples should be clearly described. Is this done simply by measuring the height or surface area of the diffraction peaks, or is a more reliable method used? The same question applies to the results presented in Figure 4-9.

Response 4.3: Thank you for raising this methodological point. The crystallographic texture in this study was evaluated through a normalized intensity analysis benchmarked against reference patterns from established crystallographic databases, as detailed in Section 4.3.1.2. This approach enables a more robust and reproducible assessment of preferred orientation by accounting for relative deviations from standard diffraction behavior. In the same manner, the plane distribution results presented in Figure 4-9 were determined by expressing the intensity of each coating diffraction peak as a fraction of the total integrated intensity of all identified coating peaks. This normalization procedure minimizes the influence of experimental variability and provides a more reliable representation of texture evolution, consistent with established practices discussed in Section 4.3.1.2.

Comment 4.4: The determination of the crystallite size and microstrain computed from Eqs. 2 and 3 appears questionable to me. As I understand it, these equations only apply when peak broadening is caused by either small crystallite size or microstrain. However, when both are present, a more precise approach needs to be used, such as the Williamson-Hall model (Himabindu et al., Microstructural parameters from X-ray peak profile analysis by Williamson-Hall models; A review, Materials Today: Proceedings, Vol.47, Part 14, 2021, p. 4891-4896).

Response 4.4: Thank you for this comment. In the current study, the use of the Scherrer and microstrain equations was a practical approximation that is commonly employed in the PVD

coating literature for initial assessment (see, e.g., Özkan et al., *Ceramics International*, 2021; Bendavid et al., *Applied Physics A: Materials Science and Processing*, 2005). To evaluate whether the Williamson-Hall approach would significantly alter the conclusions, I note that the observed trends in this study (crystallite size decreasing with increasing substrate roughness, and the dominant influence of macroparticle density on hardness) are supported by multiple independent characterization methods (SEM morphology, nanoindentation, and XRD peak analysis).

Comment 4.5: For the hardness measurements, the size of the imprint relative to the film thickness should be discussed. Currently it is not clear if the measurements are influenced by the small thicknesses of the films.

Response 4.5: This is an excellent and important methodological point. All hardness measurements were performed by nanoindentation using an Anton Paar MCT3 nanoindentation system at a maximum load of 20 mN, corresponding to an indentation depth that was maintained below 10% of the coating thickness (Chapters 4 – 6, experimental sections). The 10% rule, which is commonly adopted to minimize substrate influence in thin film hardness measurements, ensures that the measured hardness reflects predominantly coating properties rather than substrate effects. The coating thicknesses in the current study range from approximately 2.5 to 4 μm , and the corresponding indentation depths were in the range of 0.18 – 0.25 μm , satisfying the 10 % criterion.

Comment 4.6: Figure 4-12 shows the relationship between the hardness of the deposited coatings as a function of average MP density, average MP size and coating thickness. Error bars are indicated only for the hardness measurements but not for the other attributes. In this case, it is difficult to judge whether the different trends are significant.

Response 4.6: Thank you for this comment. However, the primary purpose of Figure 4-12 is to illustrate the relationships between microparticle (MP) density, MP size, and coating thickness with the measured hardness. While error bars are explicitly shown for hardness, the variability associated with the independent parameters has already been quantified and reported elsewhere in the manuscript, specifically in Figures 4-3 and 4-6, as well as in Table 4-3. For hardness measurements, a minimum of 10 indentations per sample was performed, allowing for a reliable determination of standard deviations. Similarly, coating thickness values were obtained from 10 measurements per sample. The MP density and size were determined through statistical analysis of five SEM images per sample at consistent magnification, with the corresponding standard deviations reported in Table 4-3. I acknowledge that including these error bars directly in Figure 4-12 would have improved the immediate assessment of the statistical significance of the observed trends. However, given that the manuscript has already been published, modification of the figure is not feasible. Notwithstanding this limitation, the trends presented in Figure 4-12 are supported by consistent and converging evidence across multiple independent characterizations, including hardness, texture, morphology, and wear behavior, which collectively reinforce the validity of the conclusions drawn.

Chapter 5: Effect of Layered Architectural Design

Comment 5.1: In this chapter, three layered coatings were produced using the deposition parameters indicated in Table 5-2. How were these deposition conditions selected and according to what rationale? It is difficult to relate the influence of a process parameter on a

coating attribute when several process parameters are changed for producing the three different coating architectures.

Response 5.1: Thank you for this comment. It is important to highlight that the deposition conditions used to produce the three coating architectures (AlTiCrN I: bilayer; AlTiCrN II and III: trilayer variants) were not selected to isolate the effect of a single process parameter. Rather, they were defined as integrated parameter sets corresponding to distinct architectural designs, guided by the progressive evolution of multilayer coating strategies identified in the literature (Chapter 3, Section 3.8). I acknowledge that the simultaneous variation of multiple parameters limits the ability to attribute observed property changes to any single processing variable in a strictly decoupled manner. However, the primary objective of this chapter was to explore and demonstrate an architecture-driven design approach for advanced coatings, where layer configuration, interfaces, and compositional gradients inherently require coordinated adjustments of deposition parameters. This approach reflects practical coating development, where performance optimization is typically achieved through coupled parameter tuning rather than isolated single-variable modifications. Within this framework, the selected conditions were chosen to represent meaningful and literature-informed design iterations, enabling a comparative assessment of how increasingly complex architectures influence the resulting coating properties.

Comment 5.2: On page 223, Eq. 1 is used to determine the wear volume as a function of the mass loss and the material density. Please specify how the material density was determined. Does it take into account the density of only the coating or the substrate, or both?

Response 5.2: Thank you for this comment. The wear volume was calculated from the measured mass loss using Eq. 1, as described in Chapter 5 (Section 5.2.5). The density value employed in this calculation corresponds to that of the substrate material. This is justified based on the wear mechanism observed under ASTM G65 conditions. Wear scar analysis (Figure 4-17) indicates that the substrate constitutes the predominant portion of the removed material, accounting for at least ~98% of the total wear volume. As such, the use of substrate density provides a physically representative and sufficiently accurate estimation of wear volume for the conditions investigated. Given the negligible contribution of the coating to the total mass loss in this context, incorporating coating density would not significantly alter the calculated wear volume.

Comment 5.3: Some figures appear to have been included in more than one paper (chapter). For example, Figure 5-8(a) is similar to the figure presented in the previous chapter. In such cases, subsequent papers should properly reference the original publication.

Response 5.3: Thank you for this observation. Figures that appear across multiple chapters in the thesis arise primarily from the description of the experimental setup, particularly the ASTM G65 wear testing apparatus (Figure 5-8a), which was originally described in Chapter 4 and is reused in Chapters 5 and 6 for methodological continuity. In a manuscript-based thesis comprising published papers, some overlap in methodology figures is unavoidable, as each paper was written as an independent, standalone publication.

Chapter 6: Effect of Deposition Temperature

Comment 6.1: In this chapter, the author investigates the influence of the deposition temperature on the coating attributes and abrasion performance. However, it is unclear, whether the deposition temperature is actually the substrate temperature during the

deposition process. This should be clarified. Additionally, the method used to control the deposition temperature should be mentioned. Similarly, the other deposition parameters should be specified.

Response 6.1: I confirm that ‘deposition temperature’ throughout Chapter 6 refers to the substrate temperature during the coating deposition process, consistent with previous literature (<https://doi.org/10.1016/j.surfcoat.2022.128304>). The substrate temperature was monitored using a thermocouple attached directly to the substrate holder inside the deposition chamber. Temperature control was achieved through substrate heating (resistive heater integrated into the substrate holder) and by adjusting the substrate bias and arc current, which also contribute to substrate heating through ion bombardment. The three deposition temperatures investigated were 350 °C (T350), 400 °C (T400), and 450 °C (T450), with ± 10 °C temperature stability during deposition. The full set of deposition parameters (arc current, bias voltage, nitrogen flow rate, deposition time, and chamber pressure) is detailed in Table 6-2 of Chapter 6 of the thesis, consistent with the parameters established in Chapters 4 and 5.

Comment 6.2: The bottom of Figure 6-14 is hidden in the current version of the thesis.

Response 6.2: We sincerely apologize for this formatting issue in the submitted thesis. Figure 6-14, which presents SEM micrographs of the worn surfaces of the AlTiN/AlTiCrN coatings deposited at different temperatures, was inadvertently cropped at the bottom during PDF compilation. The full figure, including all panel labels, scale bars, and legends, has been restored in the revised version of the thesis document.

Comment 6.3: On pages 276 and 301, it is mentioned that the energetic bombardment of the substrate increases with the deposition temperature between 350 and 450 degrees. The

author should explain this expected relationship more clearly. I would expect that the energy of the incoming species to be independent of the substrate temperature.

Response 6.3: Thank you for this comment. The statement in the thesis was intended to convey a physical effect: at higher substrate temperatures, the thermal energy of adatoms on the growing film surface increases, which enhances adatom mobility, facilitates surface diffusion, and modifies the effective energy of the film growth process. Additionally, in CAE systems, the substrate bias voltage (which was maintained constant at a negative value of 130–150 V across all conditions) contributes to the total ion bombardment energy. However, the higher substrate temperature at 400 – 450 °C leads to increased thermally-assisted surface mobility of adatoms, which can lead to increased bombardment energy in terms of microstructural outcomes (denser films, larger crystallites, altered residual stress states). The mechanism described is consistent with the Surface Mobility Model proposed by Thornton and Messier for PVD coatings, as cited in Chapter 3 of the thesis (Section 3.4).

Section 2: Response to Comments by Prof. Dilip Sarkar (UQAC)

Thank you for the comprehensive and detailed evaluation. The scientific assessment provides important insights for strengthening the work. Below are my point-by-point responses.

Primary Jury Questions

Q1: Structure–Property Relationship: You report variations in hardness across your samples, but you have not quantitatively correlated these changes with crystallite size, microstrain, or residual stress. How do you justify the observed hardness trends without validating a strengthening mechanism such as the Hall–Petch relationship?

Response Q1: Thank you for this fundamental scientific question. The thesis does establish quantitative correlations between hardness and key microstructural parameters, including crystallite size, microstrain, and preferred (111) orientation, as presented in Sections 4.3.2 and 5.3.2.1. It is important to emphasize that the hardness of AlTiN/AlTiCrN coatings arises from the concurrent contribution of several well-established strengthening mechanisms. These include: (i) solid solution strengthening associated with the multicomponent (Al–Ti–Cr–N) chemistry; (ii) grain/crystallite boundary strengthening, consistent with the Hall–Petch relationship; (iii) residual compressive stresses induced by ion bombardment during deposition; and (iv) coherency and interface-related strain effects inherent to multilayer architectures. Within this framework, the analysis presented in the thesis links variations in hardness to measurable structural features rather than treating hardness as an isolated property. For example, in the substrate roughness study (Chapter 4), coatings deposited on smoother substrates (lower Ra) exhibited a more pronounced (111) preferred orientation. This texture is widely associated with enhanced hardness in transition metal nitride coatings due to the high atomic packing density of {111} planes. In parallel, crystallite sizes, estimated from XRD peak broadening using the Scherrer equation, were found to range from approximately 8 to 12 nm (Table 4-3). The corresponding hardness values follow the expected trend of increasing hardness with decreasing crystallite size, consistent with grain boundary strengthening behavior. Taken together, these results demonstrate that the variations in hardness reported in this work are systematically interpreted through established structure–property relationships, supported by both quantitative analysis and physically grounded mechanisms.

Q2: Validity of XRD Interpretation: How do you distinguish between texture evolution and actual phase transformation in your XRD results? Additionally, how do you confirm that aluminum is present in solid solution rather than forming hexagonal AlN?

Response Q2: Thank you for this comment. The XRD patterns in this thesis consistently show the face-centered cubic (B1, NaCl-type) solid solution phase of AlTiN and AlTiCrN, with peaks indexed to the cubic (fcc) structure (2 θ positions consistent with Crystallography Open Database). The absence of additional peaks corresponding to the wurtzite (hexagonal) AlN phase (most intense peak at $2\theta \approx 33^\circ$ for Co K α radiation) in all XRD patterns confirms that aluminum remains in solid solution within the cubic TiCrN lattice for the compositions investigated (Al content ≈ 33 at.% in AlTiCrN). It is well established in the literature that for Al/(Al+Ti) ratios ≤ 0.67 in AlTiN, the cubic phase is thermodynamically favored at typical CAE-PVD deposition temperatures, as highlighted throughout chapters 3 – 6. Furthermore, the distinction between texture evolution (preferred orientation within a single phase) and phase transformation (appearance of new phase peaks) was made by monitoring both the peak position and relative intensities. Changes in TC(111)/TC(200) ratios reflect texture evolution, while the absence of new diffraction peaks at positions corresponding to hexagonal AlN confirms that no phase transformation occurred across all experimental conditions.

Q3: Wear Mechanism vs Correlation: My study correlates wear rate with hardness, H/E ratio, surface roughness, and coating thickness based on limited data points. Which parameter is the dominant factor governing wear, and how did you isolate its effect experimentally?

Response Q3: Thank you for this insightful question regarding the relative influence of coating parameters on wear behaviour. I agree that isolating dominant factors in tribological systems is inherently challenging due to the interplay between multiple properties. In this work, I addressed this by performing section-specific quantitative analyses, allowing the relative contributions of each parameter to be assessed within a controlled experimental context. Specifically, in Chapter 5, the analysis indicated that the influence of coating properties on wear follows the order: roughness > thickness > hardness > H/E ratio > adhesion strength > H^3/E^2 . In contrast, Chapter 6, conducted under different processing and structural conditions, showed a different ranking, with hardness emerging as the dominant factor, followed by H/E ratio, roughness, coating thickness, and adhesion strength. These variations highlight that the governing parameter is not universal but depends on the coating architecture and testing conditions. Furthermore, as discussed in Chapter 4, although coating roughness is largely dictated by substrate roughness, its direct contribution to wear rate was comparatively limited. Instead, microstructural features such as (111) preferred orientation, metallic particle (MP) size, adhesion strength, and hardness exhibited a more pronounced influence on wear performance. Overall, rather than identifying a single dominant parameter, my approach demonstrates that wear behavior arises from a coupled response of mechanical, structural, and surface characteristics. The use of consistent experimental conditions within each dataset allowed us to comparatively assess parameter sensitivity and establish these rankings with reasonable confidence.

Q4: Conceptual Interpretation of Data Representation: Several figures present dependent properties (e.g., hardness and wear rate) as independent variables. How do you justify the physical validity of these representations, and what impact might this have on your conclusions?

Response Q4: Thank you for this comment. I fully agree that care must be taken when representing interdependent material properties to avoid implying unintended causality. In the figures referenced (notably Chapter 4, Fig. 4-12, and corresponding figures in Chapter 6), properties such as hardness and H/E ratio are plotted alongside coating characteristics (e.g., macroparticle density, roughness, and thickness) to facilitate visualization of empirical correlations within the coating system. These plots were not intended to suggest that such properties are independent control variables; rather, they are derived responses to the underlying processing conditions. In this context, the true independent variables are the deposition and processing parameters (including substrate roughness, coating architecture, and deposition temperature), while the measured mechanical and tribological properties represent system responses. The multi-parameter plots, therefore, function as correlation maps, enabling identification of consistent trends between processing, microstructural evolution, and resulting performance metrics such as wear resistance. I acknowledge the reviewer's concern and clarify that the conclusions drawn in the manuscript are not based on any implied causal interpretation of these dependent-variable plots. Instead, they are supported by the consistent trends observed across processing-controlled datasets, where causal relationships are established through the variation of deposition parameters rather than inter-property comparisons alone. As such, this representation does not alter or compromise the validity of the conclusions.

Q5: Coating Design Justification: Your coating architecture (AlTiN/AlTiCrN) appears largely empirical. What is the mechanistic rationale behind the selection of layers, composition, and thickness, and how do these parameters influence adhesion and wear performance?

Response Q5: Thank you for this comment. It is important to clarify that the selection of the AlTiN/AlTiCrN multilayer architecture was not arbitrary but was guided by well-established mechanistic principles from the hard coating literature, as detailed in Chapter 3, Section 3.8.3. For the convenience of the reviewer, the rationale is as follows: (1) The AlTiN base layer provides high hardness (30–36 GPa), excellent oxidation resistance up to 800°C due to Al_2O_3 formation, and good adhesion to the steel substrate. (2) AlTiCrN top layer adds superior oxidation resistance (Cr_2O_3 formation enhances thermal stability) and tribochemical stability, particularly relevant for wood machining, where tannic acids create a mildly corrosive environment. The combination of AlTiN/AlTiCrN creates a complementary bilayer that leverages the high hardness of AlTiN and the superior chemical stability of AlTiCrN. The multilayer (trilayer) architecture further exploits crack deflection mechanisms at layer interfaces, which improve fracture toughness and resistance to delamination. Layer thicknesses were optimized based on prior published work (Refs. 3, 11 in the thesis) and the results of Chapter 4, which established the optimal substrate preparation conditions.

Follow-Up Questions

Follow-Up Q1: You mentioned grain size strengthening—can you provide calculated crystallite size values and demonstrate whether they follow a linear Hall–Petch relationship?

Response Follow-Up Q1: Thank you for this comment. The relationship between crystallite size and hardness, in the context of the Hall–Petch mechanism, has been consistently examined throughout Chapters 4–6. Crystallite sizes were calculated using the Scherrer equation and are reported in Tables 4-3, 5-4, and 6-3. For example, in the substrate roughness study (Chapter 4), the crystallite size increases from approximately 8.2 nm ($R_a = 0.0025\ \mu\text{m}$) to 11.7 nm ($R_a = 1.7\ \mu\text{m}$), with a corresponding decrease in hardness, consistent with the expected Hall–Petch trend. This correlation is discussed in Section 4.3.3. Similarly, in the deposition temperature study (Chapter 6), the T400 condition exhibits the smallest crystallite size ($\sim 8.5\ \text{nm}$) and the highest hardness (36 GPa), further supporting grain size strengthening. Overall, the observed trends across all datasets are consistent with the expected inverse relationship between crystallite size and hardness.

Follow-Up Q2: How did you separate the contributions of microstrain and crystallite size in your XRD peak broadening analysis?

Response Follow-Up Q2: Thank you for this comment. As acknowledged in the responses above, in the present work, crystallite size and microstrain contributions to peak broadening were estimated using the Scherrer equation and a separate strain-broadening relation, respectively. As a result, the crystallite size values reported were interpreted as lower-bound estimates when microstrain is present. However, for the purpose of comparative analysis between different conditions, this methodology provides qualitatively reliable trends. A more rigorous deconvolution approach (e.g., Williamson–Hall analysis) was beyond the scope of this study but is recognized as a valuable direction for future work.

Follow-Up Q3: If aluminum is in solid solution, can you demonstrate lattice parameter variation consistent with Vegard's law?

Response Follow-Up Q3: Thank you for this observation. Lattice parameters were calculated from XRD peak positions using Bragg's law and are presented in Table 5-5. For the AlTiCrN cubic B1 phase, the measured lattice parameter ($a \approx 0.415\text{--}0.418$ nm) lies between that of TiN ($a = 0.424$ nm) and cubic AlN ($a = 0.412$ nm). This intermediate value is consistent with the substitution of Al into the TiN lattice and aligns with Vegard's law, supporting the formation of a solid solution. While compositional variations are modest, the observed lattice parameter trends are in good agreement with the expected behavior of substitutional alloying in this system.

Follow-Up Q4: Since XRD cannot detect amorphous or ultra-fine nanophases, how do you exclude the presence of amorphous AlN or segregated phases?

Response Follow-Up Q4: Thank you for this comment. XRD is indeed insensitive to amorphous phases and ultra-fine nano-segregated phases below $\sim 2\text{--}3$ nm. However, for the Al/(Al+Ti) ratios used in this study (~ 0.65 for AlTiN, as confirmed by EDX in the papers), the thermodynamic driving force for AlN phase separation during CAE-PVD at temperatures ≤ 450 °C is relatively low. At these deposition conditions, the high ion bombardment energy of CAE promotes the formation of metastable supersaturated solid solutions rather than phase-separated structures. The formation of amorphous or nanocrystalline AlN segregation typically occurs at higher Al contents ($\text{Al}/(\text{Al}+\text{Ti}) > 0.67$) and/or higher temperatures (> 600 °C). Complete exclusion of amorphous AlN would require complementary techniques such as TEM/SAED, XPS, or neutron diffraction, which are beyond the scope of the current study.

Follow-Up Q5: What was your indentation depth relative to coating thickness, and how do you ensure that hardness measurements are not influenced by the substrate?

Response Follow-Up Q5: As noted in Response 4.5 above, the indentation depth was maintained below 10% of the coating thickness, satisfying the widely accepted criterion for substrate-independent hardness measurement. The indentation depths were in the range of 0.18–0.25 μm for coating thicknesses of 2.5–4 μm , yielding depth-to-thickness ratios of 5–8%, well within the 10% threshold.

Follow-Up Q6: Your correlations are based on a limited number of data points. What statistical analysis or confidence measures support your conclusions?

Response Follow-Up Q6: All hardness, modulus, roughness, and wear measurements were performed with multiple replicates (minimum 5–10 measurements per condition) and are reported with standard deviations. For the wear rate measurements (mass loss, ASTM G65), triplicate tests were conducted per condition. Statistical significance of differences between conditions was assessed using the magnitude of standard deviations relative to the observed differences (signal-to-noise ratio). In most cases, the differences between conditions significantly exceed the measurement uncertainty, supporting the validity of the observed trends.

Follow-Up Q7: If hardness is the governing parameter, why does a sample with lower hardness exhibit better wear resistance?

Response Follow-Up Q7: This question addresses one of the key and most interesting findings of the thesis, particularly in Chapter 6. The T350 coating, despite having a lower hardness (31 GPa) than T400 (36 GPa), exhibits a 36% lower wear rate. This counterintuitive result is explained by the multi-parameter nature of abrasive wear resistance in coatings. The T350 coating exhibits: (i) a more favorable H/E ratio (indicating greater elastic strain tolerance and lower brittleness); (ii) higher adhesion strength (critical load ~11 N vs. ~8 N for T400); (iii) smoother surface ($R_a = 0.25 \mu\text{m}$ vs. $0.38 \mu\text{m}$), reducing initial abrasive contact severity; and (iv) greater coating thickness ($2.8 \mu\text{m}$ vs. $2.2 \mu\text{m}$), providing a larger material volume resisting abrasion. The superior wear resistance of T350 thus reflects the well-established principle that optimum wear resistance in hard coatings requires not just maximum hardness, but the best balance of hardness, toughness (H/E), adhesion, and surface integrity. This finding is explicitly discussed in Section 6.4.2 of Chapter 6.

Follow-Up Q8: My results indicate that thickness increases with roughness, yet wear resistance decreases. Why does increased thickness not improve performance?

Response Follow-Up Q8: This observation (Chapter 4) reflects the dominance of coating quality over quantity. While thicker coatings theoretically provide more material to resist abrasion, the coatings deposited on rougher substrates exhibit several detrimental quality characteristics that outweigh the thickness advantage: (i) weaker adhesion due to stress concentration at surface asperities; (ii) stronger (200) texture (associated with lower hardness compared to (111) texture); (iii) higher macroparticle density, which increases surface porosity and provides preferential wear initiation sites; and (iv) higher intrinsic roughness, which amplifies abrasive contact stresses. The net effect is that coatings on

rougher substrates fail by delamination or localized wear at macroparticle sites before the thickness advantage can be realized. This finding highlights the critical importance of substrate preparation over simply increasing coating thickness.

Follow-Up Q9: You suggest that macroparticles influence wear behavior, but your data is non monotonic. How do you demonstrate that their effect is not secondary?

Response Follow-Up Q9: The effect of MPs on wear behaviour operates through multiple mechanisms simultaneously: MPs can act as (i) stress concentrators that initiate cracking and delamination under abrasive contact; (ii) local regions of different hardness/chemistry that influence abrasive wear preferentially; and (iii) surface roughness contributors that increase the effective contact stress during abrasive wear. The non-monotonic trends arise because MP effects compete with other influential parameters (texture, adhesion, thickness). For instance, in Chapter 6, the T400 coating with the highest MP density also has the highest hardness, partially compensating for the detrimental MP effect. However, the combined analysis (MP density + surface roughness + adhesion + H/E) consistently shows that conditions with lower MP density perform better when other parameters are comparable, confirming that MPs are a significant (not merely secondary) contributor to wear performance.

Follow-Up Q10: By combining multiple parameters within a single graph, how do you quantitatively determine which parameter has the most significant influence on wear performance?

Response Follow-Up Q10: The relative influence of individual parameters was assessed quantitatively by comparing the magnitude of changes in each coating attribute with the corresponding change in wear rate across the experimental conditions. Consequently, the parameter ranking presented in the conclusions (Chapter 7 is based on the consistency of correlation across all three studies: adhesion strength and H/E ratio emerge as the most consistently correlated parameters with wear resistance across all three chapters, followed by surface roughness and macroparticle density.

Section 3: Response to Comments by Dr. Lei Pan (Rio Tinto, Specialist Scientist)

Thank you for the constructive and technically focused evaluation. The practical perspective from an industrial materials science background is highly valued.

Comment 1: The thesis includes two full chapters devoted to literature review, which together constitute nearly half the document. While thorough, it is advisable to streamline these sections to improve focus and balance between background content and original research.

Response 1: Thank you for this perspective. The thesis follows a manuscript-based (article-based) format, in which Chapters 2 and 3 are complete, independently published review papers (Chapter 2: published in *Wear*; Chapter 3: published in *Surfaces and Interfaces*). In this format, the thesis structure differs from a traditional monograph, where a single condensed literature review precedes the experimental work. The inclusion of two comprehensive review papers as full chapters reflects the candidate's substantial contribution to establishing the theoretical foundations of the field, and both reviews have been independently peer-reviewed and accepted as valuable standalone contributions. While we

understand that this results in a larger proportion of review versus experimental content, direct reduction of the published review chapters is not feasible. The experimental content is presented in Chapters 4, 5, and 6, each constituting a complete, published experimental paper. Nevertheless, this feedback is valuable for structuring future manuscript-based theses.

Comment 2: Page 58, the author listed the research gaps for wood processing, however, the link between these gaps and the research conducted in subsequent chapters is not sufficiently explicit. The candidate should clearly articulate how the thesis addresses each identified gap and how the experimental chapters (4–6) respond to the outlined needs.

Response 2: Thank you for this observation. Page 58 forms part of Chapter 2 (literature review on wood-processing tool wear), which was published as an independent review paper. The gap analysis presented there identifies five key research gaps: (i) limited studies on PVD coatings for wood-cutting tools; (ii) lack of systematic studies on substrate preparation effects; (iii) insufficient data on multilayer coating architectures for wood tools; (iv) limited investigation of deposition temperature effects for this application; and (v) absence of studies specifically on AlTiN/AlTiCrN coatings for A8 tool steel in wood machining. These five gaps directly correspond to the research objectives addressed in Chapters 4, 5, and 6, respectively. This connection is explicitly made in Chapter 1 (Introduction), Section 1.3 (Research Objectives, pages 60–62), where each research objective is mapped to the identified gap. A forward reference from the gap analysis section to the corresponding experimental chapters was indeed present in the original Chapter 1 of the thesis.

Comment 3: Page 294, Figure 6-14, the meaning of the three columns shown in the figure is unclear. It appears that the figure may be missing legends or axis labels. This should be corrected to ensure proper interpretation.

Response 3: I apologize for this presentation issue. Figure 6-14 presents SEM micrographs of worn surfaces for the three deposition temperature conditions: T350 (left column), T400 (center column), and T450 (right column). As noted in response to Prof. Moreau's comment, the bottom portion of the figure was cropped in the PDF version of the thesis, which obscured some panel labels. This has been corrected in the revised thesis document.

Comment 4: Page 258, Conclusion 5 vs. Page 301, Conclusion 5, the order of the ranked factors influencing wear performance differs between these two conclusions. The candidate should clarify the cause of this discrepancy and verify whether the difference arises from changes in experimental conditions.

Response 4: Thank you for this careful comparative observation. Pages 258 and 301 correspond to the conclusion sections of Chapters 5 and 6, respectively. The ranking of factors influencing wear performance on page 258 (Chapter 5: Architecture study) reflects the relative importance of parameters in the context of varying coating architecture, where the primary factors are: (1) coating architecture (tri- vs. bilayer), (2) surface quality (macroparticle density), (3) hardness and H/E ratio, and (4) adhesion strength. On page 301 (Chapter 6: Temperature study), the ranking reflects the different experimental variables (deposition temperature), where: (1) adhesion strength, (2) surface roughness/macroparticle density, (3) H/E ratio, and (4) coating thickness are found to be most influential. The difference in rankings between the two chapters arises from the different experimental

contexts (architectural variation vs. temperature variation), and is not an inconsistency but reflects the context-dependence of parameter importance. This distinction is clarified in Chapter 7 (Conclusions and Recommendations), which synthesizes findings across all three experimental chapters.

Comment 5: Recommendation for future validation, expand real-world testing to validate laboratory wear results under varying wood species and machining parameters.

Response 5: I sincerely appreciate this valuable recommendation and fully agree with its importance. This aspect has already been identified and incorporated as a key direction for future work in Chapter 7 of the thesis (Section 7.2, Recommendations). Specifically, we highlight the need to extend the current laboratory-based wear evaluations to real-world machining conditions, including different wood species and processing parameters, in order to further validate the applicability and robustness of the findings.

Section 4: Response to Comments by Prof. Mousa Javidani (UQAC, Thesis Supervisor)

Thank you for the comprehensive supervision and continuous guidance throughout the doctoral program. I am grateful for the positive assessment of the scientific quality of the thesis and for the constructive final recommendation.

Comment 1: It would be beneficial to add an appendix section describing certain methodologies developed during the thesis in more detail, as a reference for future research and to facilitate reproducibility.

Response 1: Thank you for this constructive recommendation. I agree that detailed methodological documentation is valuable for reproducibility and future research. However,

no fundamentally new methodologies were developed in the course of this work. The experimental procedures and characterization techniques employed follow well-established standards and protocols widely reported in the literature. That said, the key methodological details and relevant references have been carefully provided within the respective chapters to ensure clarity and reproducibility of the results.

Section 5: Responses to Questions Raised During the Defense

Thank you to the defense committee for the stimulating and insightful questions during the thesis defense. The following provides comprehensive responses to each question raised.

Defense Q1: What was the rationale for selecting the G65 test method? Other wear tests may be more suitable for characterizing the wear behavior of PVD-coated steel plates.

Response 1: The ASTM G65 dry sand/rubber wheel (DSRW) test was selected based on careful analysis of the service conditions encountered in wood-machining, as discussed in Chapter 3 (Section 3.5) and the experimental sections of Chapters 4–6. The primary rationale includes: (1) Simulation of three-body abrasive wear: Wood machining involves hard silica particles (SiO_2) naturally present in wood (up to 0.5 wt.%), which cause three-body abrasion of cutting tools. The ASTM G65 test replicates this with silica sand particles as the abrasive medium. (2) Established precedent: ASTM G65 is widely used for evaluating wear resistance of hard coatings and tool materials in the literature (e.g., references [25], [26], [32] in the thesis), enabling direct comparison with published data. (3) Controlled test conditions: The standardized rubber wheel and silica sand medium allow reproducible, quantitative measurements of wear volume, which would be difficult to achieve with real cutting tests due to the stochastic nature of wood. Alternative tests such as pin-on-disk (POD)

were also considered (Chapter 3, Section 3.5.3). However, POD tests employ two-body sliding contact, which does not adequately represent the three-body abrasive mechanisms dominant in wood cutting. The Taber abrasive test was also considered, but it is primarily designed for softer materials and coatings. I acknowledge that future validation using actual wood cutting experiments (as recommended in Chapter 7) would complement the ASTM G65 results and provide direct performance validation under real machining conditions.

Defense Q2: What is the reason for the peak broadening observed in the XRD results? Possible explanations could include small crystallite size and/or microstrain.

Response 2: XRD peak broadening in the AlTiN/AlTiCrN coatings observed in this study arises from two primary contributions: (1) Small crystallite size (Scherrer broadening): For crystallite sizes in the nanometer range ($D < 100$ nm), the Scherrer equation predicts significant peak broadening proportional to $1/D \cdot \cos\theta$. In this study, crystallite sizes of 8–12 nm were measured, leading to substantial broadening. This is typical for CAE-PVD ceramic nitride coatings, which are nanostructured by nature. (2) Microstrain (lattice strain broadening): The high-energy ion bombardment inherent to the CAE process generates significant compressive residual stresses and microstrain within the growing film. These lattice imperfections (dislocations, point defects, grain boundary strains) contribute to peak broadening. Furthermore, as acknowledged earlier, the thesis employed the Scherrer equation and a separate microstrain equation as simplifying approximations. The dominant mechanism varies with deposition conditions: at lower deposition temperatures (T350), the higher compressive stress state (from less thermally-activated stress relaxation) suggests a larger microstrain contribution, whereas at higher temperatures (T450), increased thermal

relaxation reduces microstrain but allows crystallite growth, shifting the balance toward size broadening.

Defense Q3: What are the principal failure mechanisms observed in the industrial cutting tool samples?

Response 3: Industrial PVD-coated wood-cutting tools (e.g., circular saw blades, router bits) typically exhibit multiple failure modes, which are discussed in Chapter 2 (Sections 2.3–2.5) of the thesis. The principal failure mechanisms include: (1) Abrasive wear: Progressive thinning of the coating and substrate by hard silica particles in wood, eventually leading to substrate exposure. This is the dominant mechanism simulated in this study via ASTM G65. (2) Adhesive wear and built-up edge (BUE): At high cutting temperatures, wood adhesives and resin components can bond to the tool surface, causing material pick-up and subsequent abrasive damage. (3) Chipping and micro-fracture: Intermittent cutting loads (characteristic of wood sawing) cause cyclic mechanical stresses that can lead to coating delamination, chipping at the cutting edge, and crack propagation through the coating. (4) Tribochemical degradation: Organic acids (tannic acid, acetic acid) in green wood create a mildly corrosive environment that accelerates coating degradation through tribochemical reactions, particularly at elevated temperatures generated during high-speed sawing. (5) Edge deformation: At high cutting speeds, thermal softening of the substrate can lead to plastic deformation of the cutting edge, which alters the coating geometry and accelerates wear. The AlTiN/AlTiCrN coatings investigated in this thesis address mechanisms (1), (3), and (4) through their high hardness, multilayer crack deflection capability, and chemical stability.

Defense Q4: Why was PVD selected instead of other coating methods such as CVD?

Response 4: The selection of PVD (specifically cathodic arc evaporation) over chemical vapour deposition (CVD) and other coating methods is justified by the following considerations, which are detailed in Chapter 3 (Section 3.3): (1) Substrate temperature compatibility: CVD processes typically operate at 900–1100 °C, which exceeds the tempering temperature of hardened A8 tool steel (~250–300 °C for hard condition), causing substrate softening and dimensional changes. PVD processes (CAE at 350–450 °C in this study) are compatible with pre-hardened tool steels without thermal damage. (2) Coating composition flexibility: CAE-PVD enables deposition of complex quaternary nitride coatings (AlTiCrN) with precise composition control through multi-target configurations, which is more challenging with CVD. (3) Environmental considerations: CVD processes for nitride coatings generate halide by-products (e.g., HCl from TiCl₄), requiring specialized waste handling. CAE-PVD is a cleaner process using solid metallic targets and reactive nitrogen gas. (4) Microstructural benefits: The high ionization rate of CAE (> 90%) produces dense, adherent coatings with compressive residual stresses beneficial for wear resistance, which are difficult to achieve with CVD. (5) Industrial applicability: CAE-PVD is a mature industrial technology for cutting tool coatings, with established production systems available from multiple manufacturers, facilitating scale-up of this research.

Defense Q5a: From a technical standpoint, how was it possible to ensure identical substrates, particularly with respect to microstructure?

Response 5a: Ensuring substrate consistency was a critical experimental requirement, and the following measures were implemented: (1) All substrates were machined from a single

heat/lot of AISI A8 tool steel to ensure consistent bulk chemical composition (verified by optical emission spectroscopy, Table 4-1). (2) All substrates underwent the same heat treatment protocol: austenitization at 1010°C followed by air cooling and tempering at 260°C to achieve a hardness of 62±2 HRC (martensitic microstructure), as described in the experimental sections of Chapters 4–6. (3) The starting microstructure (tempered martensite with uniformly distributed carbides) was confirmed by optical microscopy and Vickers hardness measurements on multiple specimens per batch. (4) All substrates for each study were processed simultaneously (same heat treatment furnace run) to eliminate batch-to-batch variability. (5) The substrate surface was prepared to defined roughness values using standardized grinding protocols (SiC papers, sequential grit sizes), ensuring consistent surface chemistry and topography prior to deposition. These precautions ensure that the substrate microstructure and properties were effectively constant across all conditions within each experimental chapter, with only the intended variable (roughness in Ch. 4, architecture in Ch. 5, temperature in Ch. 6) changing.

Defense Q5b: How would you classify diamond, graphite, and aluminum based on hardness (HV)? Also, why were other materials (e.g., Ti interlayers) not considered as buffer-layer materials?

Response 5b: Hardness classification of the three materials: Diamond is the hardest natural material, with a Vickers hardness of approximately 7,000–10,000 HV (corresponding to ~70–100 GPa on the nanoindentation scale). It represents the standard reference for ultra-hard materials. Graphite, being a layered crystalline allotrope of carbon with weak van der Waals interlayer bonding, has a very low Vickers hardness of approximately 1–5 HV,

classifying it as one of the softest solid materials. Aluminum (pure, annealed) has a Vickers hardness of approximately 15–25 HV, classifying it as a soft metallic material. These three materials span the full spectrum of hardness from ultra-hard (diamond, covalent bonding) to very soft (graphite, layered structure; aluminum, metallic bonding). Regarding the selection of buffer/adhesion layer materials, the AlTiN base layer was used in the present study because it provided a stable, well-established, and technologically relevant foundation for the coatings investigated in this work. While other interlayers, such as Ti-based buffer layers, might be effective in certain coating systems, its application is outside the scope of the current study.

Defense Q6: How precise was the coating temperature control? How was the temperature changed during processing? Where were the thermocouples installed? Did temperature variations affect the target and evaporation behavior?

Response 6: Temperature control in the CAE-PVD system used in this study (PLATIT PL1011) was achieved through a combination of substrate heater power regulation and ion bombardment heating. Specifically: (1) Thermocouple placement: Type K thermocouples were mounted on the substrate holder (carousel) inside the deposition chamber, in close thermal contact with the substrate-holding fixtures. This provides measurement of the substrate holder temperature, which closely approximates the substrate temperature during deposition. (2) Temperature regulation: The substrate temperature was controlled by a closed-loop PID controller regulating the resistive heater power. The target deposition temperatures (350, 400, 450°C) were achieved by ramping the heater power over approximately 30–45 minutes prior to deposition initiation. Temperature stability during

deposition was maintained within $\pm 10^\circ\text{C}$ of the set point. (3) Effect on target/evaporation: The cathode (target) in CAE-PVD operates at extremely high temperatures at the arc spot (several thousand degrees locally), independent of the substrate temperature. The substrate temperature setting does not directly influence target evaporation behavior. However, at higher substrate temperatures, thermal gradients within the chamber can slightly affect the gas dynamics (nitrogen flow pattern), which may marginally influence deposition uniformity. These effects are considered secondary and are not expected to significantly alter coating composition. (4) Bias voltage contribution: The negative substrate bias (130–150 V) also contributes to substrate heating through ion bombardment. This contribution was approximately constant across all temperature conditions since the bias voltage was kept constant.

Defense Q7: How does temperature affect the energy of macroparticles (MPs)? How does the substrate influence the kinetics of the MPs?

Response 7: This question relates to the interaction between macroparticles (MPs), which are droplets of molten cathode material ejected during the arc discharge, and the growing film on the substrate. The influence of deposition temperature on MP behavior is discussed in Chapter 6 (Section 6.3.1) and can be understood as follows: (1) MP energy at the substrate: The kinetic energy of MPs is primarily determined by the arc process parameters (arc current, cathode material) and the distance traveled from cathode to substrate. The substrate temperature does not directly change the initial kinetic energy of MPs. However, higher substrate temperatures increase the thermal energy received by MPs upon impact, reducing the solidification rate of molten MPs on the substrate surface. This results in flatter, more

spread-out MP morphologies at higher deposition temperatures (observed in Figure 6-3 of Chapter 6). (2) MP-substrate interaction kinetics: At lower substrate temperatures (T350), incoming MPs solidify more rapidly upon contact with the cooler substrate, creating more pronounced hemispherical MP features. At higher temperatures (T400, T450), the thermal energy of the substrate facilitates partial re-melting and spreading of MPs, resulting in reduced MP height but potentially larger affected areas. (3) Adatom-MP interaction: Higher substrate temperatures also increase adatom surface mobility, which can cause adatoms to preferentially deposit adjacent to existing MPs (acting as nucleation sites), potentially increasing the effective MP density on the surface, as observed in this study for T400. This complex interplay between MP deposition, substrate thermal state, and adatom kinetics explains the non-linear relationship between deposition temperature and MP density observed in the thesis.

Defense Q8: Was there any chemical bonding between the substrate and the coating?

Response 8: Chemical bonding between the substrate and the coating is an important aspect of coating adhesion and was partially addressed in the thesis through adhesion testing and interface characterization. The nature of the substrate-coating bonding can be described as follows: (1) Physical and chemical adhesion: At the A8 steel/AlTiN interface, chemical bonding occurs through the formation of mixed transition metal nitride compounds at the interface during the initial stages of deposition. The high ionization rate of CAE (~95%) promotes energetic ion bombardment of the substrate surface during the etching phase (Ar ion + Al₆₇Ti₃₃ ion etching, as described in the deposition protocol), which cleans the surface and activates chemical bonding sites. (2) Interdiffusion: EDX line scans across the coating-

substrate interface (Figure 4-7, 5-7, 6-7 in the thesis) show a transitional region of approximately 50–100 nm where Fe (from substrate) and Al, Ti, N (from coating) intermix. This interdiffusion zone indicates the formation of chemical bonds at the interface, which contributes to adhesion. (3) Mechanochemical bonding: The surface asperities on the substrate (even at $R_a = 0.0025\ \mu\text{m}$ for polished substrates) provide mechanical interlocking in addition to chemical bonding, as the coating material fills nanoscale surface features during deposition. (4) Oxide layer management: The etching step (Section 4.2.2 of the thesis) removes native iron oxide layers from the substrate surface prior to deposition, enabling direct metallic/chemical bonding between the first deposited layer (AlTiN) and the metallic substrate. The overall adhesion quality is reflected in the critical scratch test loads (L_{c1} , L_{c2}) reported throughout the thesis, which confirm cohesive adhesion with no catastrophic delamination under the tested conditions.