Investigation of the refractory bricks used for the flue wall of the horizontal anode baking ring furnace

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Abstract

The anode manufacturing, particularly the baking process, is an important part of the primary aluminum production process. The anode baking is carried out in a closed or open top ring furnace. In these furnaces, the anodes are placed in pits and surrounded by packing coke for preventing their oxidation with infiltrated air and mechanical support. The anodes are baked with indirect contact with the hot gas flowing in flue on both sides of the pit. The flue walls are made of commercial refractory bricks. These are subjected to chemical (high temperature corrosion), mechanical (creep, walls, anode loading and unloading) and thermal (high temperature, thermal shock) conditions during the baking process. The resulting stresses cause chemical and physical alterations across the wall width. Theses stresses generally manifested themselves by collapsing, cracking and bending of the flue walls. The degraded flue walls refractory bricks from industrial plant were investigated via their chemical composition and physical properties. It was shown that regular redressing and maintenance of the flue wall can prevent or reduce additional energy consumption, due to pit deformation, consequently, reduce the cost of the anode production.

Keywords: Refractory corrosion, thermal properties, flues walls, anode baking, energy consumption.

1. Introduction

Refractory materials are used in many industries and they are selected in accordance with the desired application, to ensure their resistance against chemical corrosion and externals mechanical loads [1]. These materials are usually called aluminosilicates differed by their alumina content [2]. Furthermore, in anode baking furnaces, refractory bricks of the flue walls are subjected to various destructive forces, both chemical and thermo-mechanical in nature, during their operation. These include high temperature environment, stresses induced by inadequate expansion allowance, thermal cycling, air infiltration, reaction with alkali and aluminium fluorides present in recycled anode butts, mechanical damage during anodes loading and unloading, damage from removal of graphitized carbon build-up on pits walls [3-5]. These effects cause cracking, bending and eventually the collapse of the flue walls. The economic challenge is to find the best way to improve the resistance and durability of flues wall refractory bricks without increasing the total cost (initial, operation, maintenance). These depend probably on the thermomechanical and chemical constraints that flues walls are subjected to and the nature of the refractory bricks. There are studies reported in literature on the utilization of refractory waste from anode baking furnaces as a potential raw material for the refractory industry [6-9]. It was found that chemical contamination, which occurred after a number of thermal cycles, was the major obstacle for the utilization of waste for this purpose [6]. The chemical attack is usually determined by various processes such as migration of gases, reduction of oxides in brick components at the anode side of the bricks and recrystallization processes [10]. During its lifetime, the refractory bricks of the flue wall are exposed to high temperature and intensive chemical corrosion, leading to the alteration of its thermomechanical properties [11]. Many works are present in the literature, which highlight the importance of refractory brick deterioration in avoiding or minimising the failure of the flue walls [12 13]. This deterioration mechanism is very complex and it takes place generally at high temperature. Some authors studied the thermomechanical properties of industrial refractories at high temperature using microstructural characterisation to evaluate the corrosion resistance of various refractories, and to determine the chemical attack mechanisms [14 19]. It is difficult to interpret the changes in microstructures of corroded refractories due to its heterogeneous nature and presence of multiple components. Various methods were developed in order to analyse and interpret the refractories microstructures after use [20 21], to determine and to simulate the corrosion mechanisms on the macro/microscale [22] for the better understanding of the alteration of refractories. Prigent et al. (2008) found that the corrosion of refractory material by gaseous sodium can be reduced by adding fine andalusite particles into the refractory bricks [23]. Allaire showed that the thermal expansion behaviour and creep is also strongly influenced by the sodium deposition [24]. Furthermore, it is very difficult to predict the thermal and mechanical behaviour of the refractory materials at a macroscopic scale. The mechanical behaviour of the refractories were analysed and the thermal damage was quantified by different researchers in order to determine the mechanism responsible for this damage [25-29]. The tensile, compressive, flexure and bending tests are used to measure elastic modulus and mechanical resistance [26, 27] while the nano-indentation and ultra-son methods are used to determine Young modulus and stress-strain laws [25]. It was highlighted that the refractory exhibit non-linear mechanical behaviour and the analysation of the measured Young modulus allow to show that microcraks present in material are not fully closed at the maximum temperature of the thermal cycle [25]. The thermal stability of the commercial refractories depends strongly on their chemical composition specially the presence of oxide impurities and increasing thermal damage significantly decreases the tensile strength and increases the fracture energy [25 29]. It can be concluded that the corrosion of refractory material, its origin and effects on the refractory wall have been widely investigated. In the present study, the focus was on the effect of flue wall refractory aging on the anode baking process. Different types of industrial refractory bricks (new and unused) were investigated. Their chemical compositions and physical properties were determined and analysed. A dynamic process model, recently published by Oumarou et al. [30 31], was used to investigate the impact of refractory corrosion (particularly the flue wall curvature) on the anode baking furnace process.

2. Materials and Methods

Two types of commercial refractory bricks namely Brick A and Brick B, both new and used, were chosen (Figures 1 and 2) for this study. The new ones were received from the supplier. The used refractory bricks were obtained from the flue wall of the industrial horizontal anode baking furnace after 115 and 111 thermal cycles for the Brick A and Brick B, respectively. To understand the effect of repeated thermal cycles on the brick refractory walls, their chemical and physical properties were determined along the sample thickness at each 10 mm from anode (pit) side to gas (flue) side. The X-ray fluorescence (XRF) technique was used to determine the chemical composition of the above mentioned refractory bricks. Detail of this technique can be found elsewhere [32]. The thermal properties were measure using the Laser Flash method and more information on this technique can be found in the literature [33]. The refractory samples, shown in Figure 3, were subdivided into various regular small samples of around 4 mm for the Laser Flash measurement. Three measurements of thermal properties were made for each small sample and the average value is used. Finally, a recently developed numerical tool was used to study various configurations of flue wall refractory and pit dimensions in order to highlight the direct impact of refractory corrosion on the anode baking process [30, 31].

3. Results and discussion

The chemical composition and thermo-physical properties of used bricks were measured, and compared with those of the new bricks. An industrial campaign was carried out to investigate the deformation caused by refractory brick corrosion. Simulations were carried out, using the process model developed previously, to study the effect of pit deformation on anode temperature during baking. The results are presented in the following sections.

3.1 Chemical composition

The properties of aluminosilicates and their potential applications depend on their alumina content [2]. The chemical compositions of new Brick A and Brick B are presented in Table 1 whereas the compositions of used Brick A and Brick B are given in Tables 2 and 3, respectively. The chemical compositions of two refractory bricks are found to be slightly different (Table 1). The compositions of both used bricks varied from the anode (pit) side to the gas (flue) side (Tables 2 and 3). The used Brick A was subjected to 115 thermal cycles whereas Brick B was exposed to 111 thermal cycles. It was observed that the compositions of both used bricks were modified mostly on anode side (Figure 3) where the brick was in contact with the packing coke. Figure 4 shows the change in Silica (S_iO_2) and alumina (Al_2O_3) contents of the used refractory Bricks A and B in the region close to the anode side. The results show that the Silica (S_iO_2) decreases and that of alumina (Al_2O_3) increases compared to those of the new bricks. The Silica (S_iO_2) depletion on the anode side leads to a grain separation, decrease in strength and finally destruction of the microstructure, consequently, a materials spalling during loading and unloading of the anodes or a failure of the flue walls [5, 10]. The alkali content (Na_2O+K_2O) was also found to increase significantly in the same region (Tables 1, 2 and 3). Usually, a high sodium contamination can be related to utilization of high quantity of recycled anode butts containing sodium. This causes significant decrease in mechanical stability of refractory bricks [6]. The increase in alkali content is due to the migration of gaseous sodium from anodes at high temperature, which reacts with the refractory brick and leads to the formation of the vitreous phase during the cooling step [12, 22]. Consequently, the formation of glassy phase will reduce the resistance of the brick against the stresses imposed on the flue wall (increase in creep) and can lead to cracking as the wall is repeatedly heated and cooled down [3]. The SO_3 component was also significantly increased in the used refractory particularly in the area close to the anode side (Figure 5). This is probably due to the combustion of the fuel and the recovering coke which are the main source of sulphur content [34].

3.2 Thermo-physicals properties

The thermal properties of the new and used refractory bricks were measured with the Laser Flash method and presented in the Figures 6 and 7, Table 4 and 5. These properties varied along the used refractory thickness and was different than that of the new samples, for both the Brick A and Brick B. The highest change of thermal conductivity was observed in the area close to the anode side (Figure 6). This could be explained probably by the change in chemical composition on this side. The density was also affected by the brick aging (Figure 7). The density was highest on the anode side. Similarly, heat capacity was also highest on this side (Table 5). The calculated thermal diffusivity increased along the brick width from anode side towards flue side for Brick A and Brick B. As observed, the thermal conductivity is high close to the anode side (Figure 6) while the thermal diffusivity is lower in this area (Table 5). This can be obviously explained by the observed increase of heat capacity and density. Despite the fact that the thermal diffusivity of used refractory varied along brick width, its resulting mean values ($1.097\ 10^{-6}$ J/kg K and $1.258\ 10^{-6}$ J/kg K for brick A and B respectively) are closed to that of the new one (Table 4).

3.3 Impact of refractory corrosion on flue wall deformation

The most serious consequence of the flue wall corrosion is its failure, curvature formation and collapse in the extreme cases after a number of thermal cycles [5, 35]. An industrial measurement campaign was carried out. During this campaign, the pit widths were measured in four sections of the furnace before the planned destruction of the flue wall. An open anode baking furnace is composed of many sections. Each section contains a number of pits each pit is sandwiched between two flues. When the flues walls are new, all pits have the same standard dimension. After they are subjected to repeated thermal cycles, it was found that the pit width varied from one section to another as well as along the length of the same pit for a given section (Figures 8 and 9) which resulted in flue wall curvature. The most variation was observed in the pit number 3 followed by the pit number 4, for all the studied sections. These pits were in the middle of the sections and were put through more anode loading and unloading compared to the side pits

3.4 Impact of refractories corrosion on the anode baking

To highlight the effect of refractory corrosion, consequently, the flue wall deformation of on the anode temperature evolution during baking, simulations were carried out with the previously developed process model [30 31] for three cases (case 1, 2 and 3). The case 1 is the reference case with standard pit dimensions and flue wall made of new Brick B (Figure 2a). The pit dimensions of the most deformed pit (pit 3 of section A, Figure 9) and flue wall made of used Brick B (Figure 2b) were considered for the case 2. Standard pit dimensions and used Brick B (Figure 2b) were utilized for case 3. For the all three cases, the same gas temperature profile was taken to avoid the effects other than those of the brick properties and the pit dimensions. Figure 10 shows the change of gas as well as the anode average temperatures with baking time for the three cases studied. The pit deformation leads yields to lower anode temperatures during the baking cycle (case 2) compared to that of the reference case (case 1). The curvature of the flue wall resulted in wider pit width, consequently, increase in packing coke thickness since the anodes dimensions were kept constant. The packing coke acts as a thermal barrier due to its lower thermal diffusivity with respect to that of the anode. Increase in packing coke thickness requires, decreases the anode temperature (Figure 10). Therefore, longer cooling time is required to cool the anodes before they can be taken out of the pit. Nevertheless, if the pit is not deformed but the refractory bricks are used (case 3). The anode temperature is the same as the temperature of the reference pit wit new bricks. This means that the aging of refractory brick does not alter the process. Regular redressing and maintenance of the flue wall can the need for additional time and energy for the anode baking operation.

4. Conclusions

Two types of commercial refractory bricks, both new and used, were studied. The bricks were mostly corroded on the anode side. Increase in alkali concentration in bricks are probably due to the recycled anode butts while increase in SO_3 concentration come likely to the fuel and recovering coke source. The flue wall deformation, which is the result of refractory brick corrosion, decreases the anode temperature, consequently, increases the energy consumption and the baking time. The regular flue wall redressing and maintenance can prevent or to reduce these unwanted effects.

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Table 1 Chemical composition of new refractory bricks A and B (OCC means other component content)

New Refractory	Al_2O_3 (%)	SiO_2 (%)	Na_2O (%)	K_2O (%)	CaO (%)	MgO (%)	Fe_2O_3 (%)	TiO_2 (%)	SO_3 (%)	OCC (%)
Brick A	45.90	48.87	0.50	0.32	0.10	1.32	1.00	1.25	0.22	0.52
Brick B	45.64	49.16	0.65	0.25	0.06	1.32	0.87	1.42	0.15	0.48

Table 2 Chemical composition of used flue wall refractory brick A (OCC means other component content)

Brick A Samples	Width (mm)	Al_2O_3 (%)	SiO_2 (%)	Na_2O (%)	K_2O (%)	CaO (%)	MgO (%)	Fe_2O_3 (%)	TiO_2 (%)	SO_3 (%)	OCC (%)
1	0 - 10	48.49	29.67	1.21	0.07	1.05	0.86	1.58	2.01	14.54	0.52
2	10 - 20	42.96	47.19	1.08	0.17	0.38	0.95	0.848	2.69	3.15	058
3	20 - 30	41.97	48.89	0.98	0.12	0.36	0.90	0.89	2.66	2.87	0.36
4	30 - 40	42.55	45.45	1.58	0.17	0.44	0.86	1.08	3.81	3.66	0.40
5	40 - 50	41.37	45.50	1.63	0.18	0.47	0.79	1.12	4.63	3.88	0.43
6	50 - 60	40.29	45.41	1.53	0.13	0.50	0.77	1.11	5.71	4.13	0.42
7	60 - 70	40.37	44.49	1.27	0.11	0.56	0.76	1.17	6.31	4.52	0.44
8	70 - 80	40.23	44.64	1.21	0.11	0.47	0.77	1.18	6.78	4.15	0.46
9	80 - 90	40.00	45.36	0.91	0.08	0.58	0.75	1.15	6.01	4.72	0.44
10	90 - 100	42.48	48.93	0.94	0.13	0.43	0.92	0.92	1.61	3.02	0.62

Table 3 Chemical composition of used flue wall refractory brick B (OCC means other component content)

Brick B Samples	Width (mm)	Al_2O_3 (%)	SiO_2 (%)	Na_2O (%)	K_2O (%)	CaO (%)	MgO (%)	Fe_2O_3 (%)	TiO_2 (%)	SO_3 (%)	OCC (%)
1	0 - 11	47.88	38.87	1.23	0.18	0.73	1.79	0.66	1.21	7.21	0.24
2	11 - 22	43.02	51.96	0.81	0.32	0.13	1.18	0.62	0.99	0.69	0.28
3	22 - 33	41.23	47.75	1.15	0.30	0.21	0.97	0.77	4.96	2.36	0.30
4	33 - 44	41.98	47.03	1.15	0.32	0.20	0.90	0.82	4.95	2.29	0.36
5	44 - 55	41.36	47.75	1.18	0.33	0.19	0.99	0.82	5.00	2.11	0.27
6	55 - 66	41.74	48.30	1.15	0.28	0.16	1.02	0.78	4.59	1.75	0.23
7	66 - 78	42.89	49.49	0.95	0.25	0.10	1.13	0.71	3.17	1.01	0.30
8	78 - 89	41.76	50.11	0.91	0.26	0.11	1.06	0.74	3.60	1.13	0.32
9	89 - 100	40.26	46.94	0.97	0.27	0.40	0.90	0.87	5.27	3.74	0.38

Table 4 Thermal properties of new refractory brick A and B

Brick A (New)		Brick B (New)	
Heat Capacity (J/kg K)	Diffusivity (m2/s)	Heat Capacity (J/kg K)	Diffusivity (m2/s)
850	$1.046\ 10^{-6}$	900	$1.229\ 10^{-6}$

Table 5 Thermal properties of used flue wall refractory brick A and B

Refractory	Brick A (Used)		Brick B (Used)	
Width (mm)	Heat Capacity (J/kg K)	Diffusivity (m2/s)	Heat Capacity (J/kg K)	Diffusivity (m2/s)
0-4	939.21	$1.001\ 10^{-6}$	1130.23	$1.189\ 10^{-6}$
5.5-9.5	915.77	$1.029\ 10^{-6}$	1111.23	$1.177\ 10^{-6}$
11-15	866.26	$1.068\ 10^{-6}$	1091.30	$1.013\ 10^{-6}$
16.5-50.5	879.70	$1.038\ 10^{-6}$	895.90	$1.219\ 10^{-6}$
22-26	876.19	$1.056\ 10^{-6}$	877.07	$1.262\ 10^{-6}$
27.5-31.5	864.47	$1.099\ 10^{-6}$	895.20	$1.232\ 10^{-6}$
33-37	772.38	$1.231\ 10^{-6}$	868.30	$1.052\ 10^{-6}$
38.5-42.5	814.58	1.11110^{-6}	759.01	$1.286\ 10^{-6}$
44-48	844.46	$1.072\ 10^{-6}$	795.77	$1.351\ 10^{-6}$
49.5-53.5	821.57	$1.040\ 10^{-6}$	806.33	$1.177\ 10^{-6}$
55-59	786.53	$1.140\ 10^{-6}$	778.93	$1.315\ 10^{-6}$
60.5-64.5	784.19	$1.118\ 10^{-6}$	818.47	$1.154\ 10^{-6}$
66-70	799.69	$1.172\ 10^{-6}$	707.40	$1.481\ 10^{-6}$
71.5-75.5	823.95	$1.104\ 10^{-6}$	790.53	$1.381\ 10^{-6}$
77-81	832.62	$1.099\ 10^{-6}$	837.03	$1.308\ 10^{-6}$
82.5-86.5	845.52	$1.155\ 10^{-6}$	840.35	$1.343\ 10^{-6}$
88-92	826.76	$1.106\ 10^{-6}$	820.51	$1.330\ 10^{-6}$
94.5-98.5	821.63	$1.099\ 10^{-6}$	830.46	$1.367\ 10^{-6}$

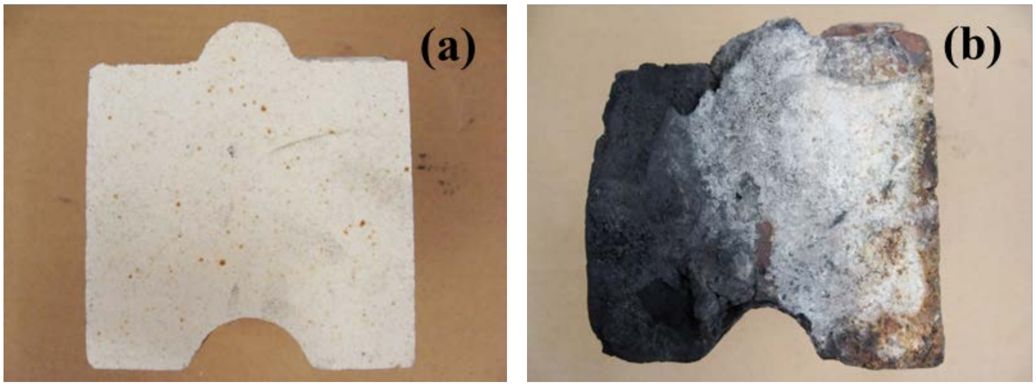

Figure 1: Brick refractory (Brick A): a) unused and b) after 115 thermal cycles.

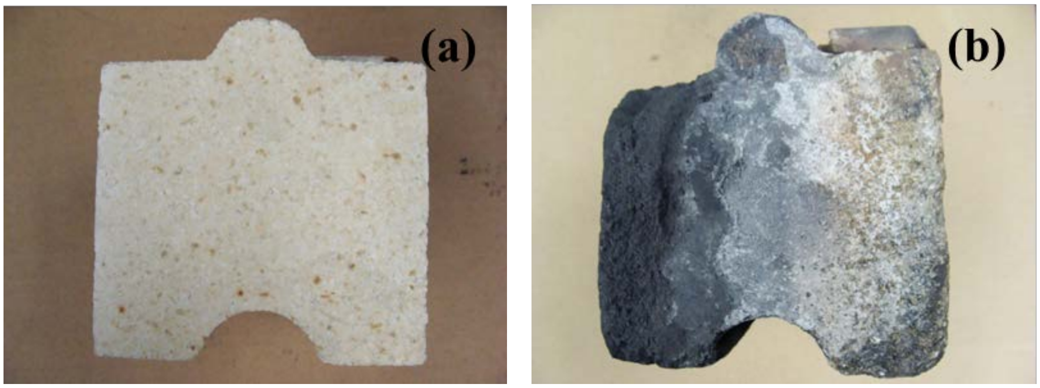

Figure 2: Brick refractory (Brick B): a) unused and b) used after 111 thermal cycles.

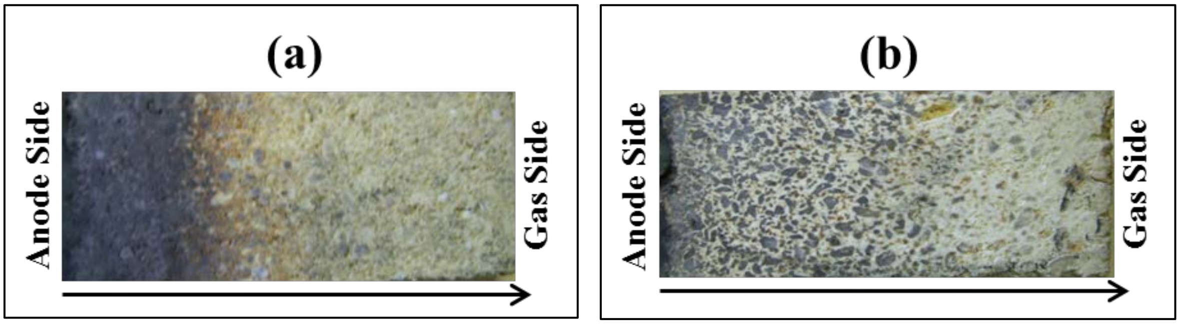

Figure 3: Refractories samples including the direction from anode side to gas side for a) Brick A and b) Brick B.

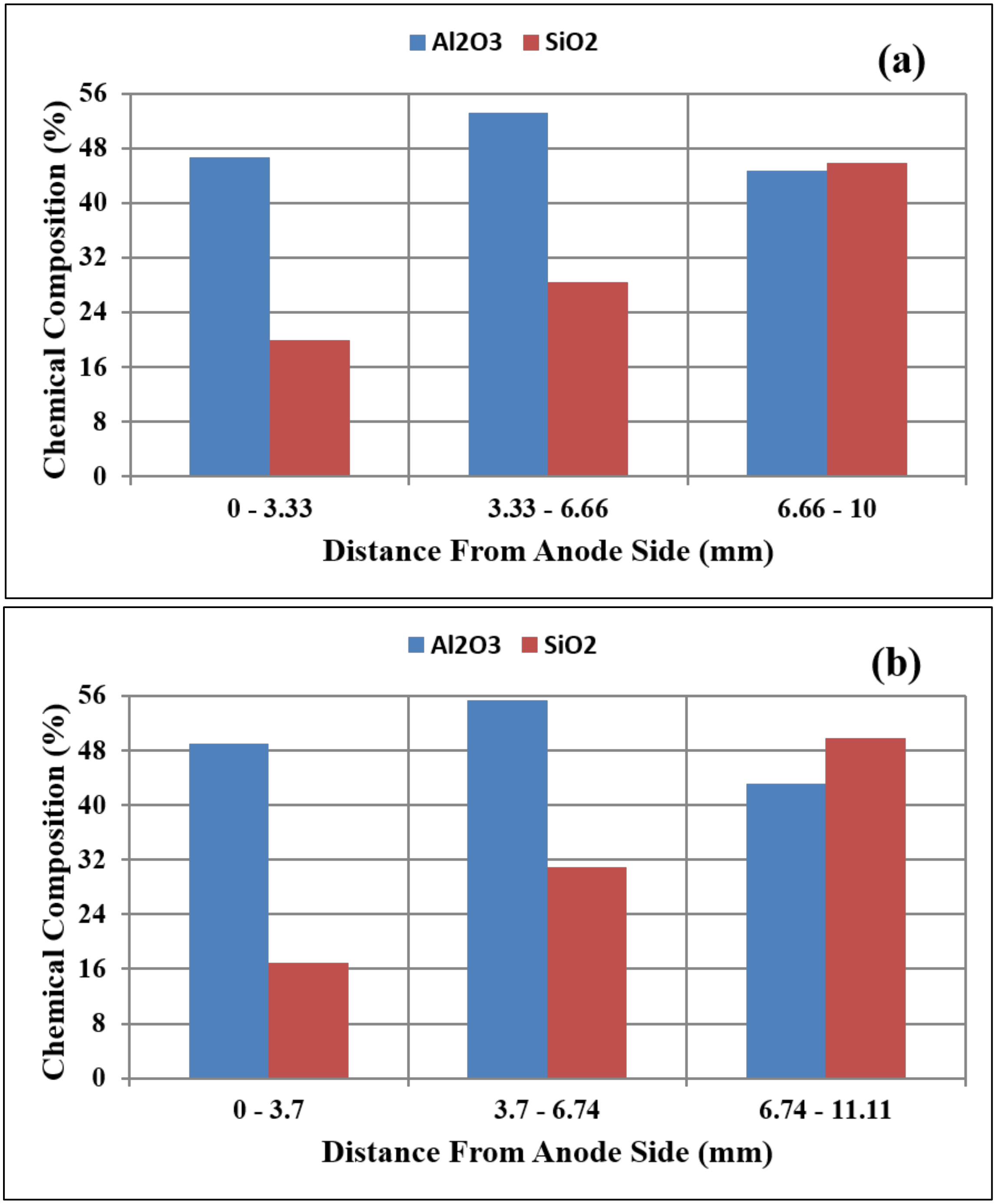

Figure 4: Aluminosilicate refractories content for the used a) Brick A and b) Brick B

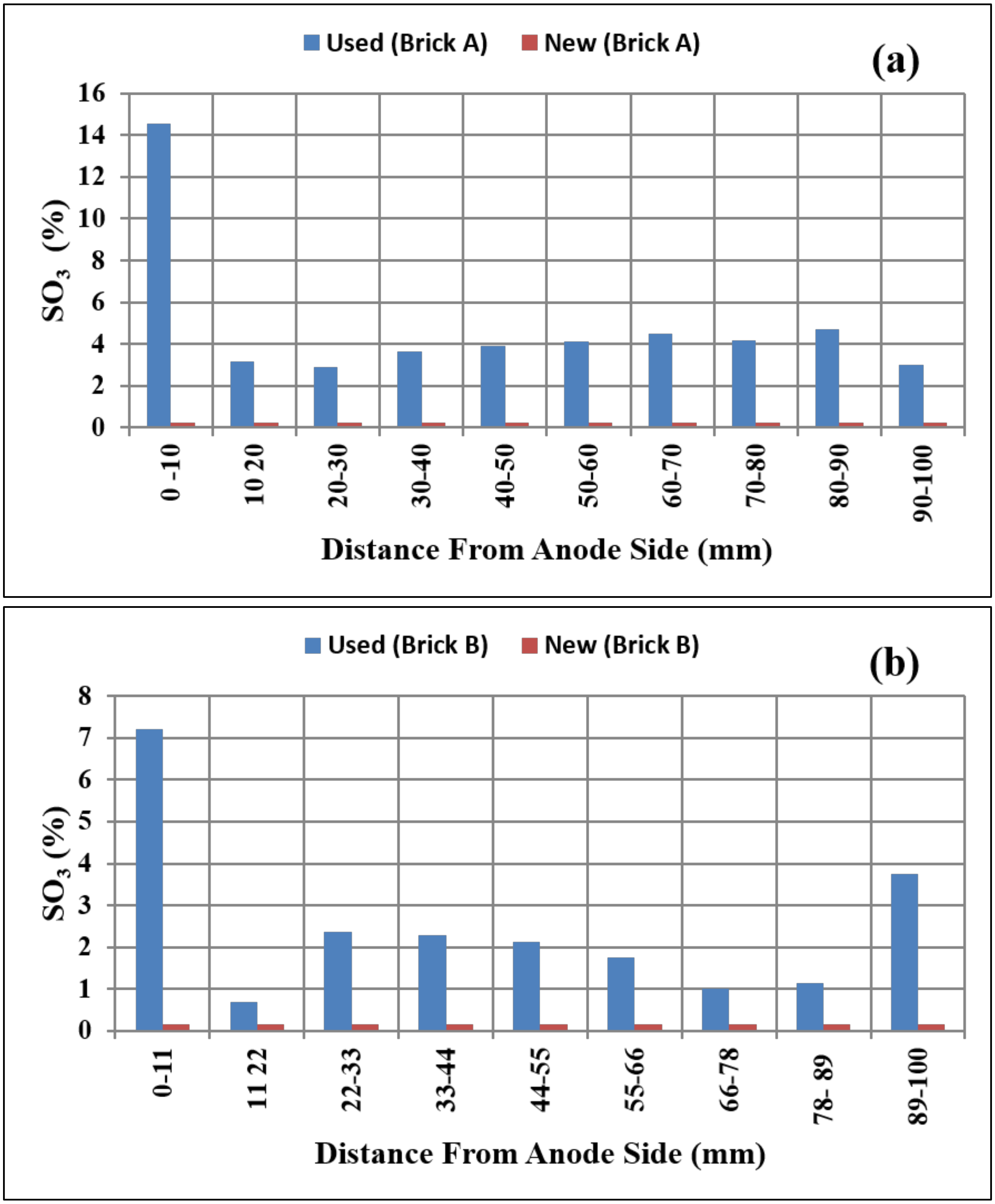

Figure 5: Sulfur oxide amount in the unused and used refractories a) Brick A and b) Brick B

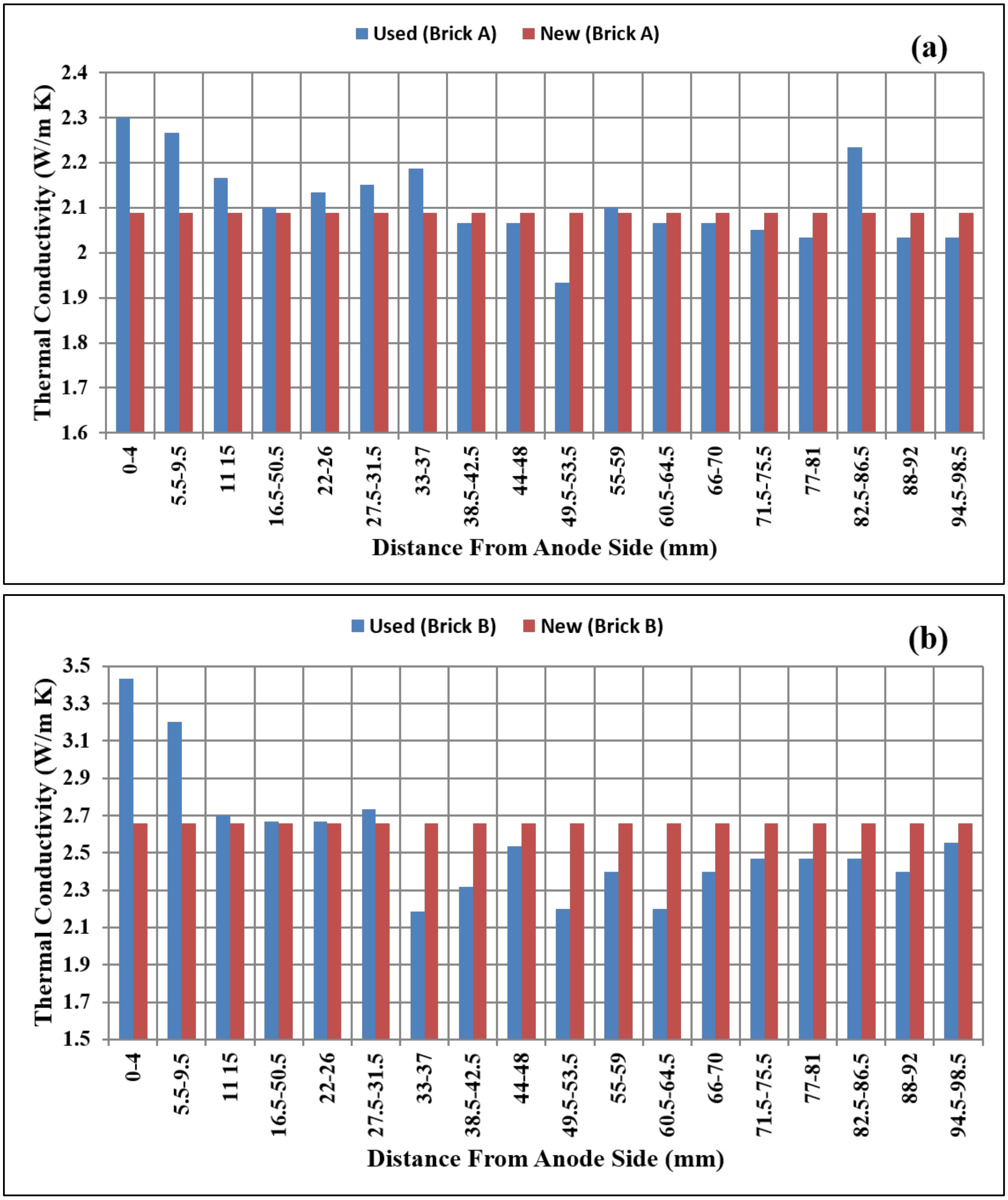

Figure 6: Thermal conductivity of the unused and used refractories a) Brick A and b) Brick B

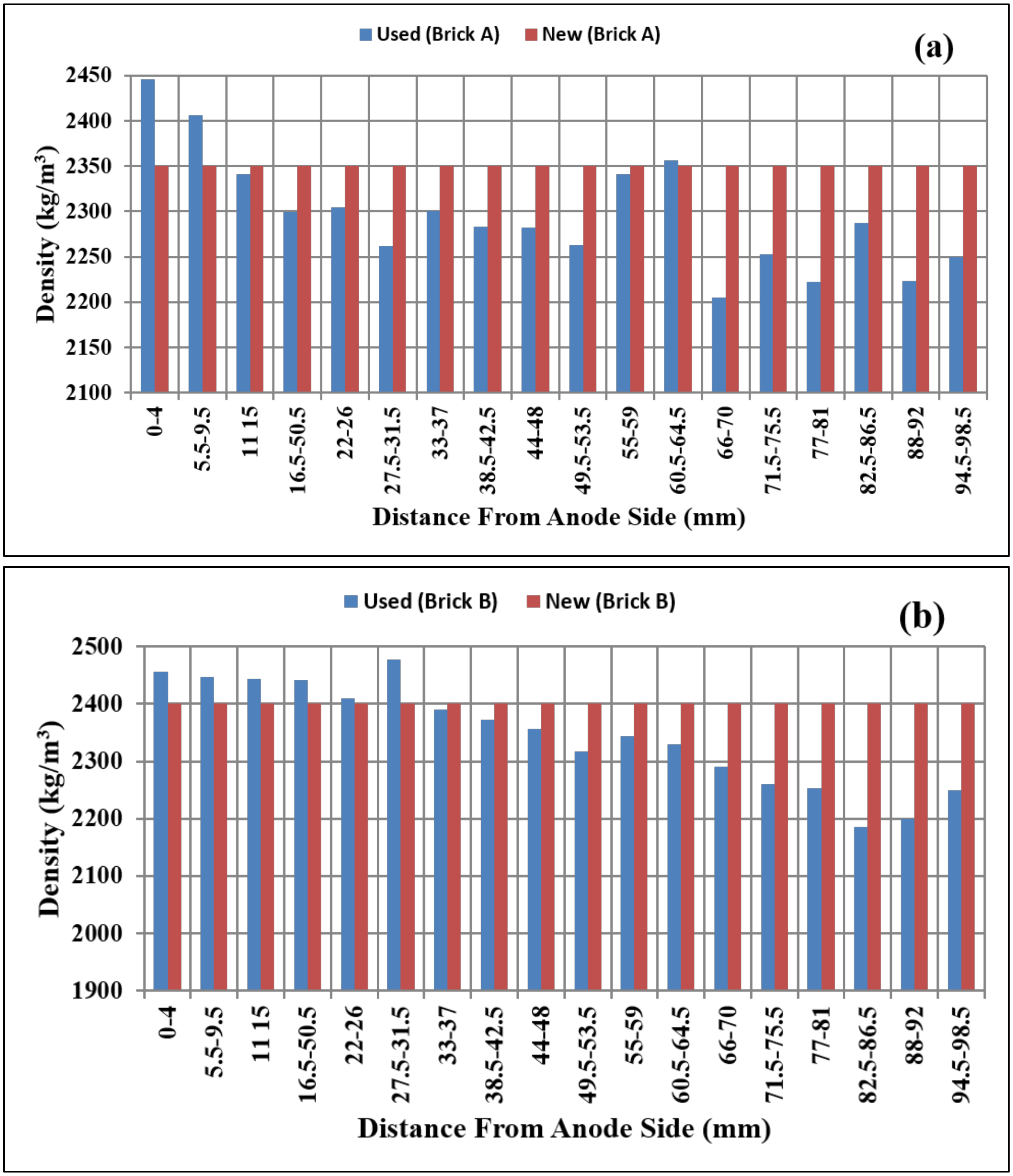

Figure 7: The density of the unused and used refractories a) Brick A and b) Brick B

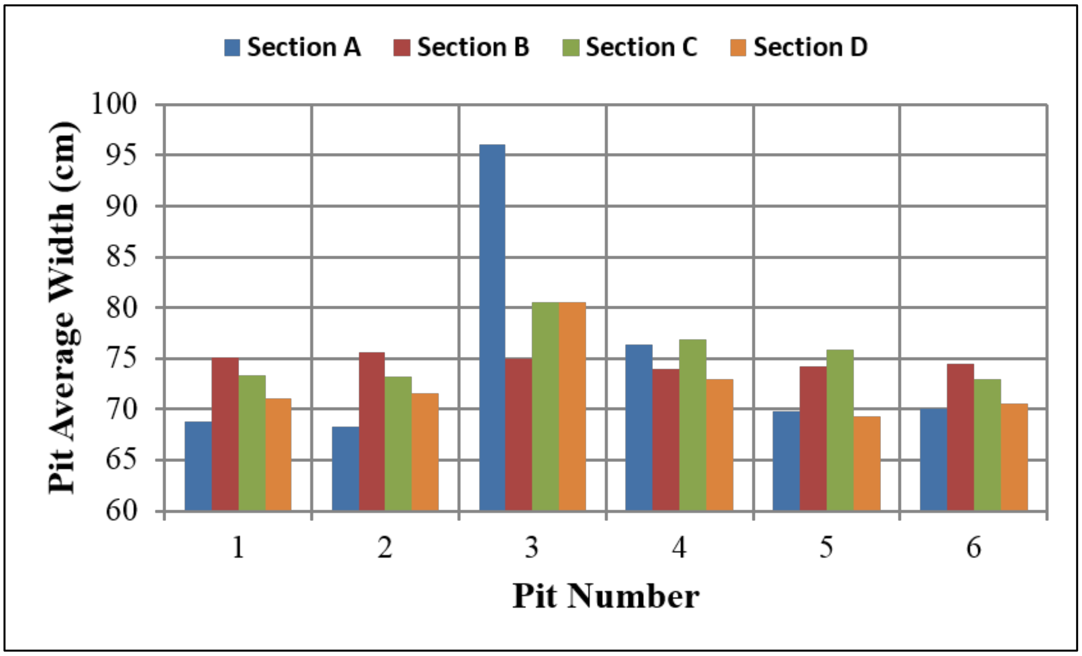

Figure 8: Pit average width of various pits for four different sections (A, B, C and D) of the furnace

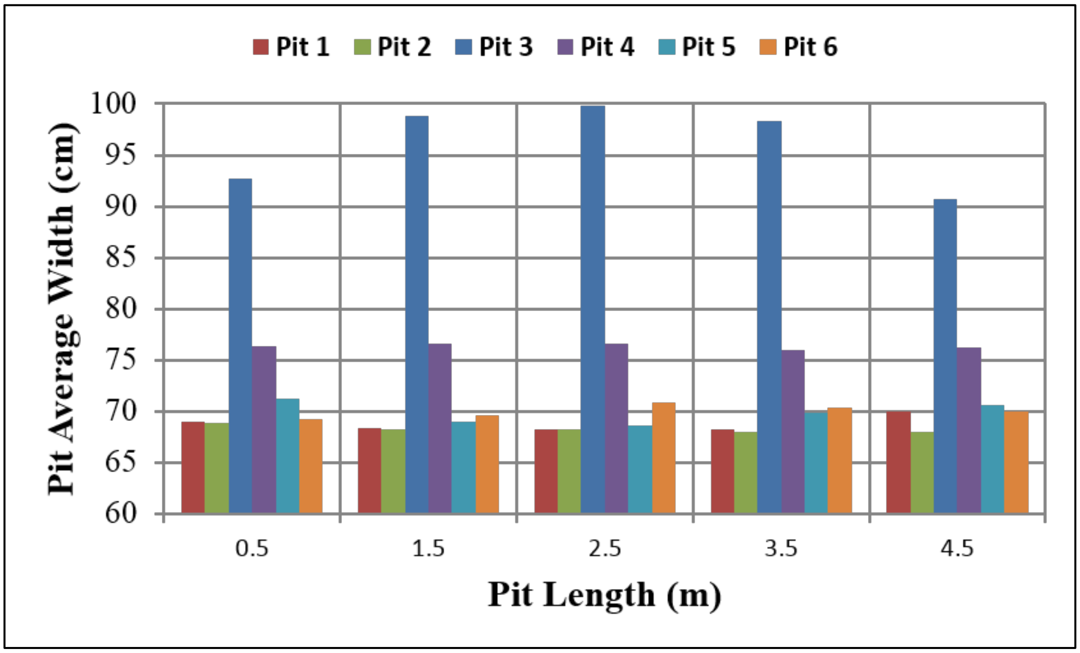

Figure 9: Pit average width along the pit length pits for section A.

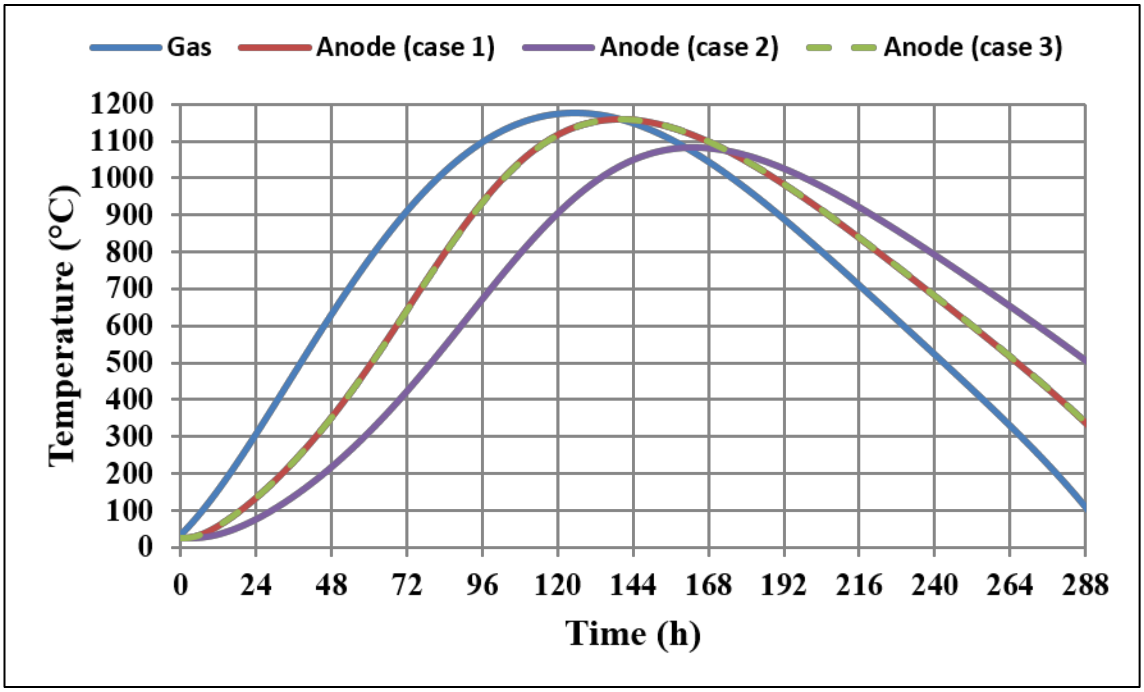

Figure 10: Temperature profiles of gas and anode for the three studied configurations of flue wall refractory – pit dimension (case 1, 2 and 3).