Utilization of coal fly ash as a raw material for refractory production

Rinyapat SUKKAE[1], Somkeat SUEBTHAWILKUL[2], and Benya CHERDHIRUNKORN[1,*]

[1]*Thammasat University, Klong Luang, Pathum Thani, 12120, Thailand*
[2]*The Siam Refractory Industry Co., Ltd, Ban Mo, Saraburi, 18270, Thailand*

***Corresponding author e-mail:** benya@tu.ac.th

Received date:
15 September 2017
Accepted date:
20 January 2018

Keywords:
Fireclay brick
Coal fly ash
Fire clay

Abstract

In this research, fireclay bricks were produced using fire clay and coal fly ash as raw materials. Coal fly ash was added to the mixture from 10-50 wt%. The disk shape samples were sintered at the temperatures of 1,100-1,400°C. The mechanical property, thermal property, phase structure and microstructure of the fireclay bricks were studied. It was found that coal fly ash can be used in the fabrication of fireclay bricks. The lower linear shrinkage and lower thermal conductivity were obtained as the higher amount of coal fly ash was added. However, bulk density and strength were lower in the samples contained coal fly ash. The effects of coal fly ash on the phase structure and microstructure of the fireclay bricks were investigated using XRD and SEM technique, respectively. Mullite phase decreased with the higher coal fly ash added, whereas anorthite phase increased.

1. Introduction

Refractory are widely used in many industries such as glass, iron, and ceramic industries. Because of high refractoriness, good corrosion resistance and good thermal shock resistance, fireclay bricks are one of the most popular refractories. Fireclay bricks are made of clay minerals consisting of 18-44% alumina and 50-60% silica. Fireclay bricks are inexpensive and easy to produce. Therefore, the production of fireclay bricks has been constantly increased due to the high demand. The reduction in production cost is very important. One of the ways to lower the production cost is to use the cheaper raw materials.

The large amount of industrial wastes have been generated over the last few decades, causing the environmental problem. Among all industrial wastes, coal fly ash is one of the solid waste produced during the coal combustion process. Kalyoncu [1] reported that in USA, during 2001, about 20% of by-product was generated from coal combustion process. Coal fly ash is the major combustion product component (more than 58%) [1]. which is collected using a electrostatic precipitator and a mechanical filter before being ejected to the atmosphere. Landfill is a general practice for disposal this coal fly ash. The major chemical compositions of coal fly ash are SiO_2, Al_2O_3, CaO, Fe_2O_3 and SO_3 [2]. The use of coal fly ash has been found in many products such as cement, concrete, brick, geopolymer and light-weight aggregate for construction industry [2-6]. However, less than 40% of the total amount of coal fly ash has been utilized [3]. Anja et al. [7] report that coal fly ash can be used as the mechanically activated microfiller in the production of refractory bauxite shotcrete. Hassan et al. [8] studied the effect coal fly ash on some refractory properties of alumino-silicate clay for furnace lining. The results showed that the addition of coal fly ash in alumino-silicate clay could decrease the physical property such as linear shrinkage and apparent porosity of the refractory. In addition, Gonzalez et al. [9] reported that the addition of fly ash in refractory insulating bricks possessed good mechanical strength and low thermal conductivity.

In this research work, the fabrication of fireclay brick using coal fly ash and fire clay as raw materials was carried out. The physical, thermal and mechanical properties, phase structure, and microstructure of fireclay brick were investigated in order to study the feasibility of using coal fly ash as the raw material in fireclay brick production.

2. Materials and methods

2.1 Raw materials and characterization

In this work, the commercial fire clay (from the Siam Refractory Industry Co., Ltd.) and coal fly ash (waste from SCG Packaging Public Co., Ltd.) were used as raw materials for fireclay brick fabrication. Phase structures of raw materials were characterized

DOI: 10.14456/jmmm.2018.16

using X-ray diffractometer, XRD (PaNalytical, X' Pert PRO). The samples were measured at 2θ of 10° to 70° with step size of 0.02° and step time of 2 s. The chemical compositions of these raw materials were analysed using X-ray fluorescence spectroscopy, XRF (RIGAKU, MEXCG).

2.2 Sample preparation

The fire clay and coal fly ash were weighed and mixed according to the compositions as shown in Table 1. The dry mixtures were planetary milled at the rotational speed of 200 rpm for 60 min. The milled powder was sieved through the sieve number 100 (150 μm), and was then uniaxially pressed into a die with a diameter of 31.50 mm at the pressure of 150 MPa in order to form a disk shape samples. After that the samples were sintered in an electric furnace at the temperatures of 1,100 to 1,400°C for 60 min with the heating rate of 5°C/min. The flow chart for the sample preparation is shown in Figure 1

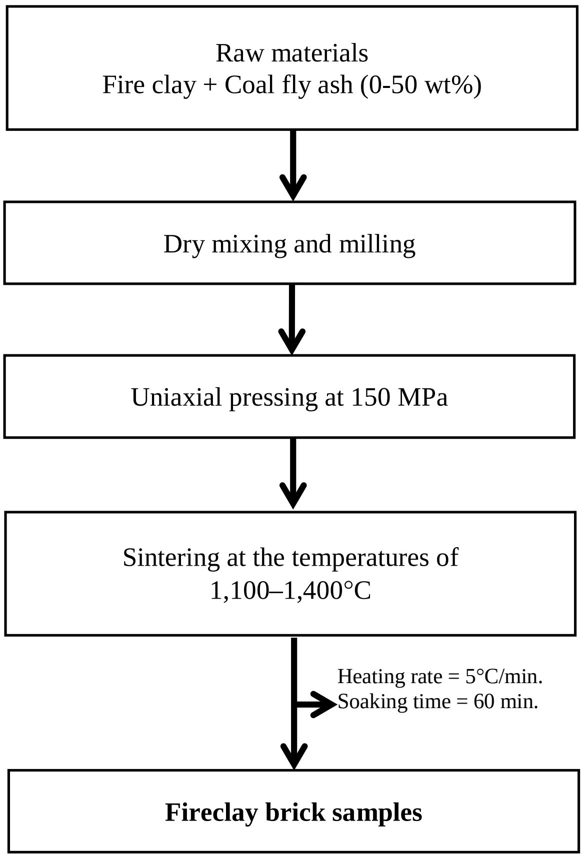

Figure 1. The fabrication of fireclay brick samples.

Table 1. The compositions for the mixtures of coal fly ash and fire clay.

Samples	Raw materials	
	Coal fly ash (wt%)	Fire clay (wt%)
F0	0	100
F10	10	90
F20	20	80
F30	30	70
F40	40	60
F50	50	50

2.3 Characterization of sintered samples

The bulk density and water absorption were determined according to Archimedes principle (ASTM standard C20). The linear shrinkage of the fireclay brick samples was measured according to ASTM standard C326-09 [10]. The universal testing machine (Tinius Olsen, H50KS) was used for mechanical test. In this study, Brazilian test (Indirect tensile test) was employed in order to study the mechanical property of the sintered samples. The disk shape sample was placed under the load cell as shown in Figure 2. The strength of the samples was determined according to equation (1) where P is applied load, D is diameter of the sample and L is thickness of the sample.

$$\sigma = \frac{2P}{\pi DL} \tag{1}$$

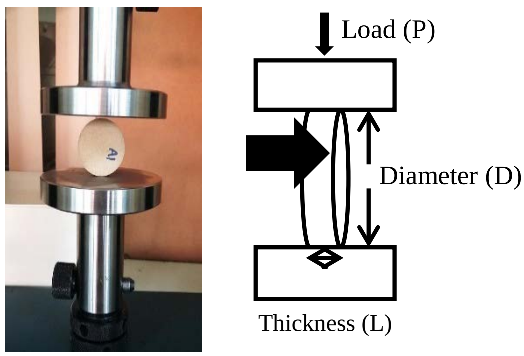

Figure 2. Illustration of disk sample placed under the load cell for Brazilian test.

The phase structure of the sintered fireclay brick samples was determined using XRD technique (PaNalytical, X' Pert PRO). The sintered samples were measured at 2θ of 10° to 70° with step size of 0.02° and step time of 2 s. Microstructures of the sintered samples were investigated using Scanning electron microscope (SEM; Hitachi S-3400N). Thermal conductivity of the samples was determined using a thermal constant analyzer (Hot Disk TCA).

3. Results and discussion

3.1 Phase structure, chemical composition and particle size of raw materials

The XRD pattern shown in Figure 3 indicates that the major phases of the fire clay are quartz (SiO_2) and kaolinite ($Al_2Si_2O_5(OH)_4$). The major phases of coal fly ash are quartz, anhydrite ($CaSO_4$), calcite ($CaCO_3$), hematite (Fe_2O_3) and alumina (Al_2O_3) as shown in Figure 4. A hump peak was also observed at the scattering angle $2\theta \approx 20\text{-}35°$ in this XRD pattern indicating the amorphous phase.

The chemical compositions of these raw materials determined using XRF technique are shown in Table 2. The fire clay composed mainly of silica (55.4 wt%) and alumina (36.4 wt%). The coal fly ash consists of 34 wt% silica, 25.1 wt% calcium oxide and 25 wt% alumina.

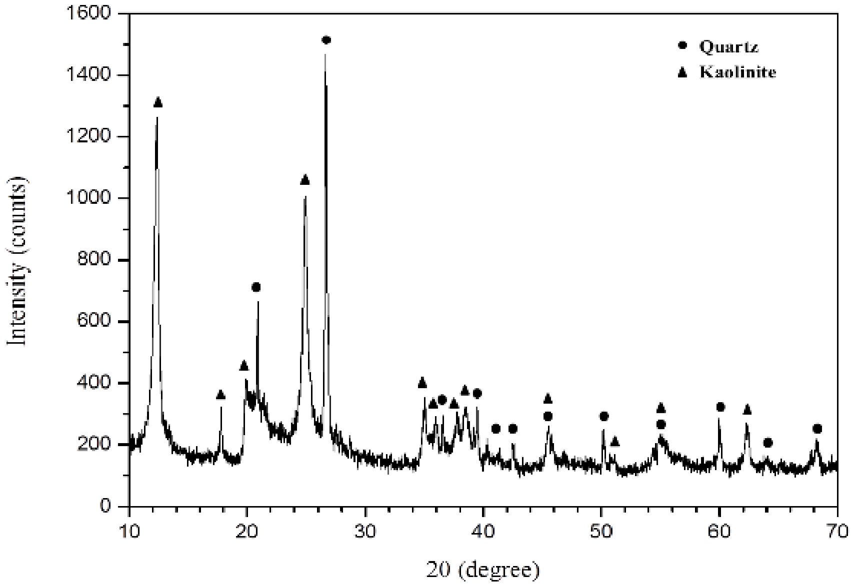

Figure 3. XRD pattern of the as received fire clay.

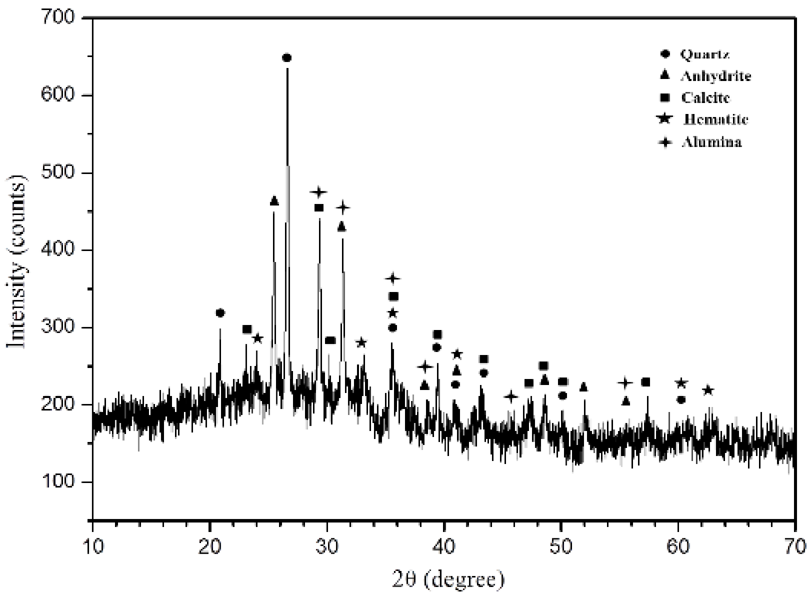

Figure 4. XRD pattern of coal fly ash.

Table 2. The chemical compositions of raw materials.

Chemical composition	Fire clay (wt%)	Coal fly ash (wt%)
SiO_2	55.4	34
Al_2O_3	36.4	25
CaO	0.152	25.1
Fe_2O_3	4.81	7.56
SO_3	0.107	4.56
K_2O	1.63	1.05
TiO_2	1.14	0.968
P_2O_5	0.105	0.885
Cl	-	0.246
MnO	0.0326	0.144
BaO	0.0085	0.0912
SrO	0.0068	0.0767
LOI	0.2081	0.3191

3.2 Physical properties of fireclay brick samples

3.2.1 Bulk density and water absorption

The plots of bulk density and percentage water absorption as a function of sintering temperatures are shown in Figures 5 and 6, respectively. The results indicate that the addition of coal fly ash could cause the reduction in bulk density of the samples. Since the density of coal fly ash (about 2.2 g/cm^3) is lower than the density of clay (about 2.6 g/cm^3), the addition of coal fly ash could cause the low density of the sintered samples. Figure 6 shows that the water absorption of the samples with 10 to 20 wt% of coal fly ash decreased as the sintering temperature increased up to 1,300°C. In addition, the water absorption of the samples with more than 30% coal fly ash decreased as the sintering temperature increased up to 1,250°C. The further increase in sintering temperature in these samples could cause the high water absorption and low bulk density as the forming of porosity and deforming of the samples.

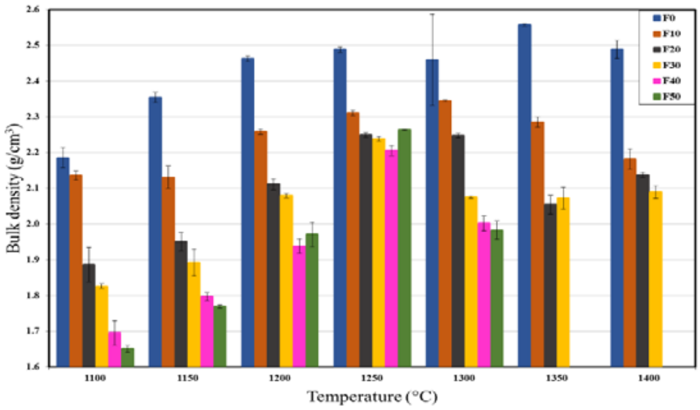

Figure 5. Relationship between the bulk density of the sintered samples and the sintering temperature for the fireclay brick samples with various coal fly ash contents.

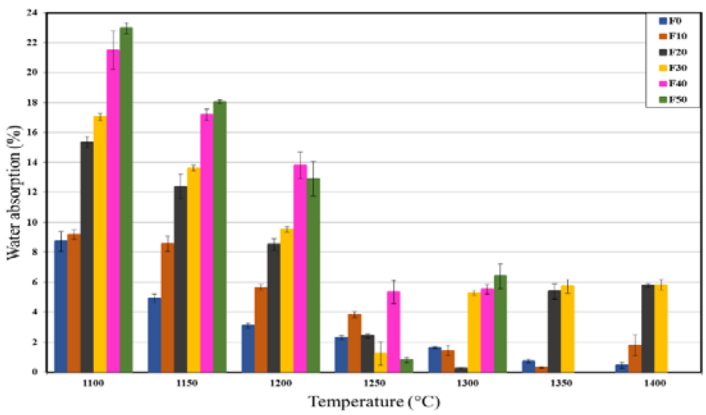

Figure 6. Relationship between the water absorption of the sintered samples and the sintering temperature for the fireclay brick samples with various coal fly ash contents.

3.2.2 Linear shrinkage

The plots of linear shrinkage for the samples contained various coal fly ash contents as a function of sintering temperature are shown in Figure 7. The linear shrinkage of the samples with 10-20 wt% of coal fly ash (F10 and F20) increased gradually as the sintering temperature increased up to 1,300°C, whereas the linear shrinkage of the samples with more than 30 wt% of coal fly ash (F30-F50) increased up to the sintering temperature of 1,250°C. This indicated that the densification occurred at the sintering temperature of 1,300°C for F10 and F20 and 1,250°C for F30 to F50. The further increase in sintering temperature caused the reduction of linear shrinkage and the shape deforming of the sample (F40 and F50). As reported in the previous section, the maximum bulk density and lower water absorption were obtained in these samples at the same temperatures (1,250°C for F30-F50 and 1,300°C for F10-F20). Consequently, the optimum sintering temperature would be 1,300°C for low coal fly ash samples (F10-F20) and 1,250°C high coal fly ash samples (F30-F50). The present of alkaline (Ca) in coal fly ash could act as flux causing the densification at lower temperature.

3.3 Mechanical property

Figure 8 shows the effect of sintering temperature on Brazilian tensile strength of fireclay brick samples. The highest strength was obtained in the sample without coal fly ash. For the low sintering temperatures (lower than 1,250°C), the strength of all samples was relatively low due to the high porosity of the samples. For the sample contained high amount of coal fly ash (F30- F50), the lower strength was obtained in all sintering temperatures. The F40 and F50 samples sintered at the temperature more than 1,300°C were deformed and could not be measured.

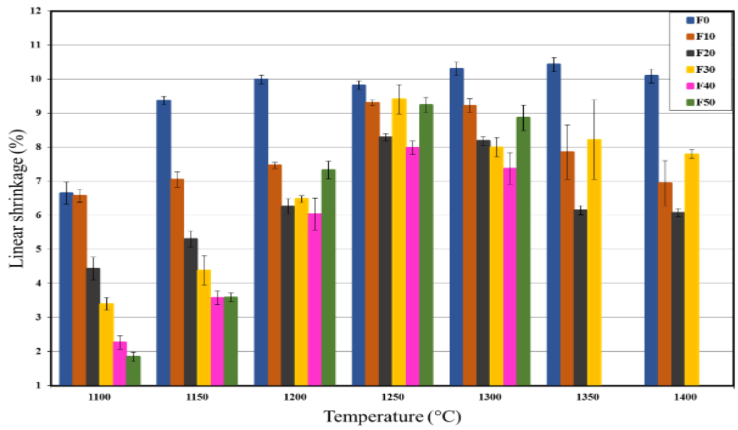

Figure 7. Relationship between the linear shrinkage of the sintered samples and the sintering temperature for the fireclay brick samples with various coal fly ash contents.

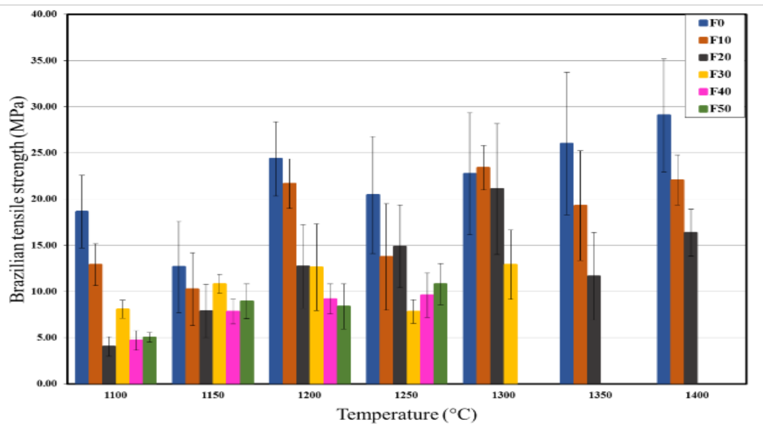

Figure 8. Relationship between the Brazilian tensile strength of the sintered samples and the sintering temperature for the fireclay brick samples with various coal fly ash contents.

3.4 Thermal property

Thermal conductivity of the samples with coal fly ash sintered at the temperature of 1,250°C to 1,400°C is shown in Figure 9. The results indicated that the addition of coal fly ash could cause the lower thermal conductivity in the samples since the higher porosity was observed in the samples contained coal fly ash.

3.5 Phase structures

The XRD patterns of the fireclay brick samples sintered at temperature of 1250°C are shown in Figure 10. The main phases for the sample F0 and F20 were mullite ($3Al_2O_32SiO_2$), quartz, and cristobalite (SiO_2), whereas anorthite ($CaAl_2Si_2O_8$) and mullite phases were found in samples with higher coal fly ash contents (F30, F40, and F50). However, the intensity of mullite phase decreased with the higher coal fly ash contents in the samples, whereas the anorthite phase increased. In addition, anorthite phase was reported [11-12] to be a porous phase which could cause the lower thermal conductivity observed in the previous section.

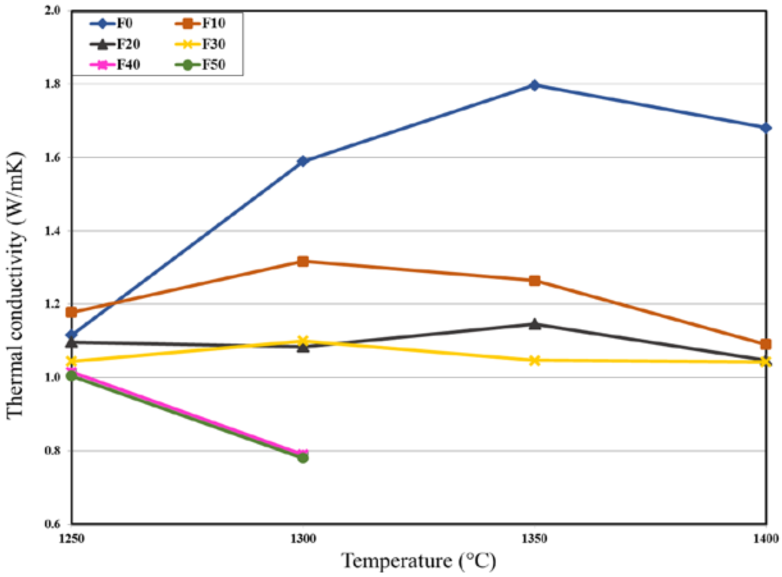

Figure 9. Thermal conductivity of fireclay brick at different coal fly ash: fire clay ratio.

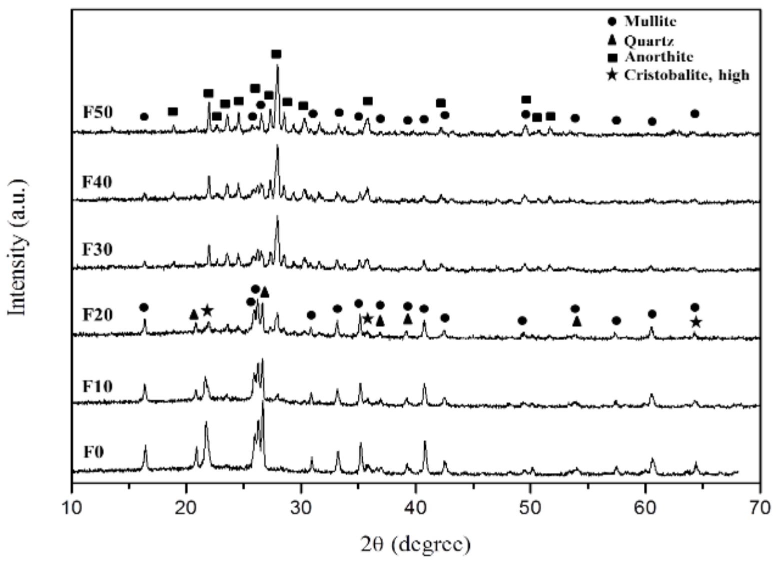

Figure 10. XRD patterns of fireclay brick samples sintered at the temperature of 1250°C.

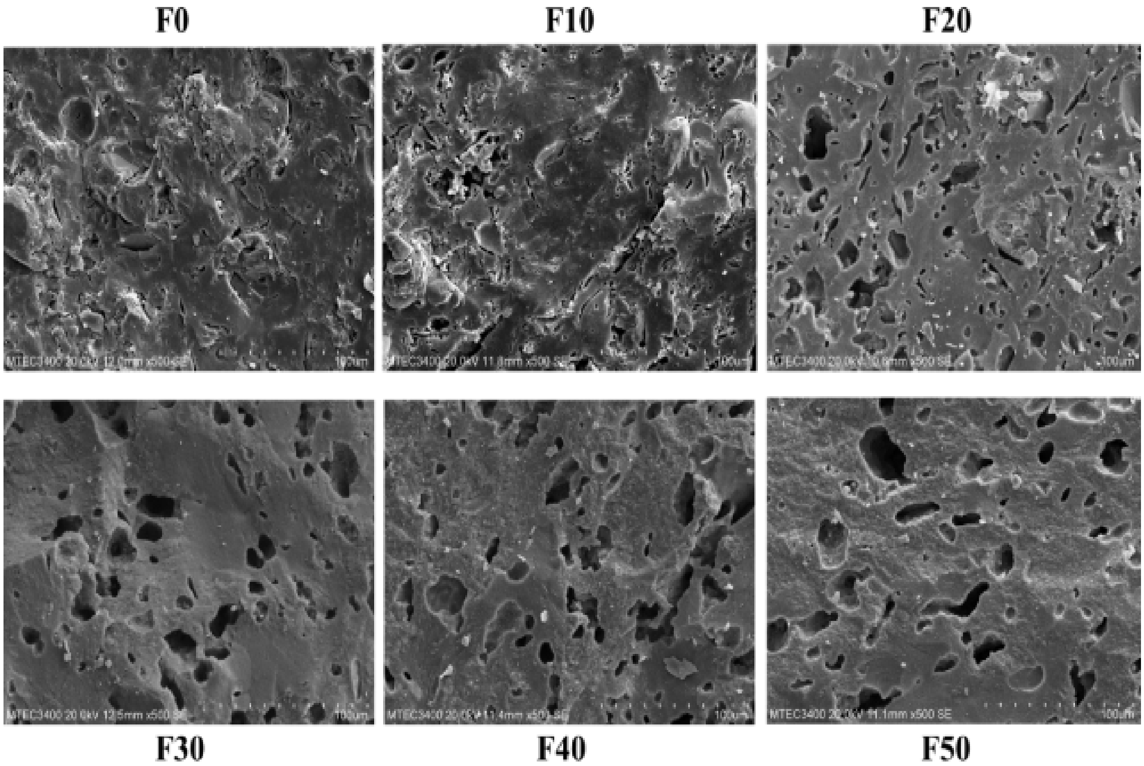

Figure 11. SEM images of fireclay brick samples sintered at 1,250°C.

3.6 Microstructure of fireclay brick

The microstructures of the fireclay brick samples (F0 to F50) sintered at 1,250°C are shown in Figure 11. The SEM images showed the heterogeneous structure in all samples. The pore wall was unconsolidated. The sample F0 and F10 has various pore and crack sizes. However, the larger pore size was found in the samples with higher amount of coal fly ash (F20 to F50). As the coal fly ash increased, the pore wall became denser, grains grew larger, and grain shape became obvious which might cause the lower strength in these samples. Generally, the exist of porosity could affect the properties of materials such as low density and low thermal conductivity because air trapped in the pore is a better thermal insulator [9].

4. Conclusions

4.1 Coal fly ash and fire clay could be used in the fabrication of fireclay bricks. It can improve the physical properties such as lower the linear shrinkage and thermal conductivity. However, it caused the reduction in mechanical property.

4.2 The SEM images showed the higher porosity and the larger pore size in the samples with coal fly ash increase, which could cause the low thermal conductivity, higher water absorption, lower bulk density and lower strength.

4.3 The suitable sintering temperature for the samples F0 to F20 would be 1,300°C whereas the samples F30 to F50 would be sintered at 1,250°C for the optimum properties.

5. Acknowledgements

The authors gratefully acknowledge the financial support provided by National Research Council of Thailand (NRCT) and the financial support provided by Thammasat University under the TU Research Scholar, Contract No. TN22/2018. In addition, we would also like to thank the Siam Refractory Industry Co., Ltd of Thailand, and National Metal and Materials Technology Center (MTEC) of Thailand for their supports.

References

[1] R. S. Kalyoncu, "Coal combustion products," World Coal Association, London, Report number 19, 2001.

[2] B. H. Bac, B. X. Nam, V. D. Hieu and N. T. Dung, "Characterization of a Vietnamese coal fly ash and its possible utilizations" present at The

2nd International Conference on Advances in Mining and Tunneling, Hanoi, Vietnam, 2012.

[3] I. Kourti and C. R. Cheeseman, "Properties and microstructure of lightweight aggregate produced from lignite coal fly ash and recycled glass," *Resources, Conservation and Recycling*, vol. 54, pp. 769–775, 2010.

[4] E. P. Kearsleya and P. J. Wainwright, "The effect of high fly ash content on the compressive strength of foamed concrete," *Cement and Concrete Research*, vol. 31, pp. 105 -112, 2001.

[5] X. Lingling, G. Wei, W. Tao, and Y. Nanru, "Study on fired bricks with replacing clay by fly ash in high volume ratio," *Construction and Building Materials*, vol. 19, pp. 243–247, 2005.

[6] S. Andini, R. Cioffi, F. Colangelo, T. Grieco, F. Montagnaro, and L. Santoro, "Coal fly ash as raw material for the manufacture of geopolymer-based products," *Waste Management*. vol. 28, pp. 416–423, 2008.

[7] A. Terzić, L. Andrić, and V. Mitić, "Mechanically activated coal ash refractory bauxite shotcrete microfiller: Thermal interactions mechanism investigation," *Ceramics International*, vol. 40, pp. 12055-12065, 2014.

[8] S. B. Hassan and V. S. Aigbodion, "Effect coal ash on some refractory properties of alumino-silicate (Kankara) clay for furnace lining," *Egyptian Journal of basic and applied sciences*, vol. 1, pp. 107-114, 2014.

[9] J. G. Otero, F. Blanco, M. P. Garcia, and J. Ayala, "Manufacture of refractory insulating bricks using fly ash and clay," *British Ceramic Transactions*, vol. 103, pp. 181-186, 2004.

[10] American Society for testing and Material, "ASTM Designation C20-00, C326: General Product, Chemical Specialties, and End Use Products," in *Annual book of ASTM Standards*, USA: ASTM International, 2005, pp. 6-9.

[11] Y. Han, C. Li, C. Bian, S. Li,and C.-A. Wang, "Porous anorthite ceramics with ultra-low thermal conductivity," *Journal of the European Ceramic Society*, vol. 33, pp. 2573-2578, 2013.

[12] A. A. Priogov, V. P. Rakina, M. M. Mirakyan, and N. V. Volkov, "Anorthite insulating refractory," *Refractory*, vol. 11, pp. 36–40, 1970.