STUDY ON UTILIZING FLY ASH FOR CASTABLE REFRACTORY

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ABSTRACT

Fly ash could be utilized as alumina and silica sources material. It could be treated by the following processes of demagnetisation, sinterization, grinding, mixing with crushed bricks as well as fired cement to create acid based on castable refractory. Sinterization changes alumina and silica significantly into mullite. The standard measurement of pyrometric cone equivalent (PCE) is used to understand the temperature resistance of the studied castable refractory based fly ash. The temperature resistance seems to increase after contacting with high temperature at longer time or repeatedly until reaching its Si-Al stabilization phase.

Keywords: Fly ash, castable refractory, temperature resistance, PCE number

1. INTRODUCTION

Fly ash generated during combustion of coal for energy production is one of the industrial residues and it is recognized as an environmental pollutant mainly due to its fineness. Because of environmental problem of fly ash, the utilization of fly ash as minerals resources has been studied and applied for many areas such as in ceramic products of bricks, cement, aggregate, land filled, etc (Anonimous, 1997 and Hwang, 1991). More over, there are other potential markets in the future for utilization of quality controlled fly ash products such as ceramic refractories, plastic fillers, metal matrix composites, carbon adsor-bents, etc (Hwang, 1991). An enormous amount of fly ash has been produced every year, however, the utilization of the fly ash in Indonesia is relatively low.

A major factor preventing utilization of fly ash might be due to the government regulation that fly ash is categorized as hazardous materials. Another factor might be due to the difficulty of producing high quality fly ash materials. The aim of, this study is to utilize the fly ash for ceramic product of castable refractory.

Refractory is one kind of inorganic ceramic materials which is resistance to high temperature, and it is required in furnace as furnace linings and for metal-melting pots. Castable refractory is loosed-fine-refractory materials like mortar that it needs casting like concrete in its functions and so called as monolithic refractory.

Prior to the present works, it was preceded by studying the characteristic of typical Suralaya fly ash, followed by studying the high temperature resistance of mixed materials composition of the fly ash with other silica-alumina source materials (Aziz, et.al, 2006; Aziz and Ardha, 2006). Based on the standard measurement of pyrometric cone equivalent (PCE) number, high temperature resistance of the experimental mixed materials was measured up to 1460°C. On the other hand, the high temperature resistance of the referenced commercial castable refractory of CAJ-16 is found at 1750°C (Aziz and Ardha, 2006). Thus, the results were incomparable yet. However, it is worth to say that the typical Suralaya fly ash has a positive tendency to be utilized as raw material of Si-Al based on castable refractory by further study on its characteristics.

In this work, the fly ash was processed to increase the temperature resistance of experimental castable refractory based on fly ash that is at least approaching the temperature resistance of the commercial castable refractory of CAJ-16 as a reference material. There are many aspects influ-

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encing the specification of castable refractory, however, the temperature resistance and the micrograph texture of the castable refractory materials would be an focused in this study.

2. METHODOLOGY

2.1 Principles

The fly ash that was studied in this works dominantly has chemical composition of silica and alumina. Hence, the type of material discussed in this paper would be an acid based on refractory.

During formation of acid based on refractory, thermo-chemical reaction occurs between SiO_2 and Al_2O_3 at temperature above 1400°C. The silica transforms into tridymite and cristobalite, alumina transforms into corundum, while both silica and alumina transform as solid solution of mullite ($3Al_2O_3.2SiO_2$). All of these artificial minerals have high temperature resistance. As shown in Figure 1a, there is one compound in the system that mullite melts at about 1800°C, and the eutectic lies close to the silica. For example, a refractory brick with 70% alumina might contain mullite with glassy forms on cooling down to the eutectic temperature of 1550°, while a refractory brick of above 70% alumina would have hard composites of mullite and corundum up to temperature 1800°C. From this phenomenon, it is clear that a refractory brick contains a small amount of mullite, would deform at about 1500°C; while if it contains higher alumina, it would stand up well to temperature of 1800°C. As a general rule, it is endeavor to choose refractory materials of such composition that it has high alumina content with high melting point. A composite containing mullite and corundum in certain amount is desirable as good acid based on refractory materials.

As shown in Figure 1b, the refractoriness or temperature resistance of the brick in SiO_2-Al_2O_3 system was tested based on standard PCE number in which the maximum cone number is 42 with maximum temperature resistance is 2015°C. Theoretically, the temperature resistance of a refractory brick changes by changing its composition of SiO_2/Al_2O_3. The composition of about 5% Al_2O_3 is known as eutectic point, where it would be the lowest temperature resistance. It means that a refractory containing alumina between 2% to 12% would be undesirable. The higher the Al_2O_3 content, the higher the PCE number. Figure 1b reveals that if the refractory brick contains 20% Al_2O_3, its PCE number is 29.5 or it has temperature resistance of about 1660°C, while, if the refractory brick contains Al_2O_3 more than 50%, its PCE number is 35.6 or it has temperature resistance of 1800°C. Accordingly, the present fly ash contains Al_2O_3 of about 20% that should fulfil the acid based on refractory materials with temperature resistance of 1660°, but theoretically the fly ash must have no impurities at all (Kumar et al, 2003).

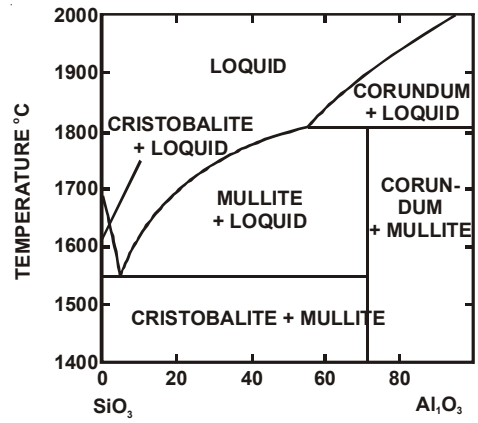

Figure 1a. The silica-alumina equilibrium diagram

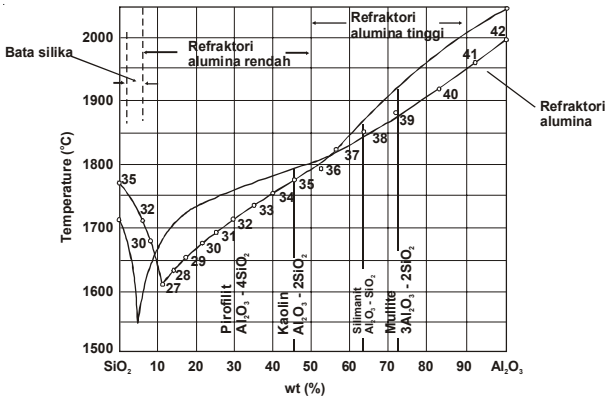

Figure 1b. Refractoriness in SiO_2-Al_2O_3 system (Surdia dan Saito, 1985)

Fly ash contains activated component of silica and alumina, when mixed with fired cement and a few grog materials in certain proportion and dry condition, it might develop as loose materials of ready used castable acid based refractory that is called monolithic refractory material (Hwang and Huang, 1995). However, fly ash generally contains some

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amount of oxide impurities such as Fe_2O_3, K_2O, Na_2O, including unburned carbon represented by lost of iguition (LOI), which would decrease the temperature resistance. Thus, processing of fly ash raw material to remove the impurities is considerably important.

To eliminate such impurities of iron oxide from the fly ash, a mineral processing technology of demagnetisation is required. In this study other impurities such as potassium, sodium and carbon were not removed yet from the fly ash.

2.2 Experimental Procedures

The introductory test results on utilizing fly ash for castable refractory has been published elsewhere (Aziz and Ardha, 2006); it was found that the best mixing material composition was 3, 2, 3, 2 for fly ash, fired cement, crushed brick, and oxide alumina by volume, respectively. However, the addition of oxide alumina did not significantly effected the high temperature resistance. Therefore, in this experiment, there was no oxide alumina addition, and the best mixing material composition would be 3, 2,3, for fly ash, fired cement, and crushed brick, by volume, respectively.

The experimental procedure was initialised by demagnetisation of the fly ash. The demagnetisedfly ash was mixed with typical binder of fired cement as well as typical grog of crushed brick with proportion of 3,2,3 by volume respectively. The mixed material was dried at temperature of 110°C for 24 hours and then kept it into a hermetical bag. A small amount of mixed material as a sample was taken and a small amount of water was added. The wet mixture was pressed by hydraulic pressure to turn into green composite-A. The green composite-A was then chemically and physically analysed including PCE number.

Burning tests of demagnetised fly ash were carried out to understand the changing of its micrograph texture and then it was compared with the micrograph texture of reference castable refractories of CAJ-16. This fly ash was called as sinterized demagnetisedfly ash. Further, the demagnetised sinterized fly ash was crushed and mixed with fired cement and crushed brick with proportion of 3,2,3 by volume, respectively to create a mixture of ready use castable refractory. The mixture was added a small amount of water and it was pressed to convert into green composite-B. The green composite-B was then analysed for PCE number.

Burning tests with time variation of the green composite-B were carried out to create composite-B, then the composite-B was analysed for PCE number. Finally, all of the test results were concluded.

2.3 Method and Equipment

Bulky fly ash samples were obtained from poured fly ash discard of the storage bin of PLTU-Suralaya. Then, the bulky samples for experimental works were divided by using splitter and coning-quartering method. Mixing materials were prepared by using a “Y” type mixer. Experimental data were collected from chemical analysis, while the data collected for micrograph textures were tested by SEM. Burning tests with high temperature difference was conducted in a muffle furnace, PCE numbers were tested based on the Indonesian standard tests of SNI 15-4936-1998.

3. RESULTS AND DISCUSSION

3.1 Summary of Previous Results

Prior to discussion of this experimental results, let the introductory experimental results that has been published elsewhere (Aziz and Ardha, 2006) is summarized. This summary might be closely in correlation with the present study. The commercial castable refractory called as CAJ-16 has been used as a reference material of castable refractory, which contains Al_2O_3/SiO_2 of 1.62; the major minerals content of corundum, mullite and cristobalite; the PCE number was 34 with melting point of 1750°C. The typical Suralaya fly ash was added directly into CAJ-16 with proportion of 1 : 1 by volume. The PCE was turned into little reduction with melting point of 1690°C. When the volume proportion of fly ash to CAJ-16 was 2 to 1 and 3 to 1, the PCE number or the melting point was turned into 1460°C and 1280°C respectively as presented in Figure 2. Generally, the higher the volume proportion of fly ash added into CAJ-16, the lower the PCE number, and the lower was the temperature resistance.

Further, the composites was prepared by mixing fly ash, fired cement, crushed brick, and oxide alumina with proportion of 3, 2, 3, 2 by volume, respectively; the PCE or the melting point was

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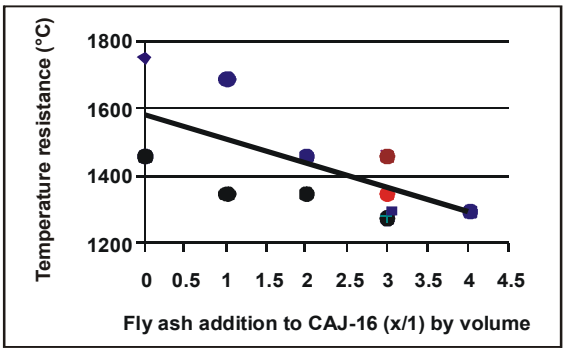

Figure 2. Direct use of fly ash added into CAJ-16 (Aziz and Ardha, 2006)

found at 1460°C. This micrograph texture was different from the micrograph texture of reference castable refractory of CAJ-16, where the composite consisted of sphere particles and found some holes between the spheres. The addition of oxide alumina did significantly effected the PCE number of the composite. It might be due to the oxide alumina consisted of inert high-grade corundum, difficult to react with water and fly ash to develop a composite (Aziz and Ardha, 2006). Therefore, in this study, it decided no oxide alumina to be added into the mixing materials.

Eventhough the quality of refractory depends on many aspects such as PCE number, compressive strength, mechanical strength at high temperature, thermal expansion, shrinkage, spalling resistance, chemical and mineral composition, etc., this study was focused on minimizing impurities in fly ash, and then the processed fly ash was mixed with binder and grog to be a composites. The PCE number of composites was an important aspect to be tested in this study.

3.2 General Characteristics of the Fly Ash

Generally, the chemical composition of fly ash all over the world is relatively the same, the different is only in the percentage of chemical content. In the case of the Suralaya fly ash, the percentage of chemical content changes by changing the raw coal used during the combustion. Table 1 shows the chemical content of the fly ash produced at different time, in which the sample was taken one year after the previous sample. The sample contained low silica but higher alumina than that of the previous sample. The impurities of iron oxide of the present fly ash was high, while calcium that may be good as binding agent, was also high. In contrast, the previous sample contained low alumina and high silica as well as high unburned-coal (LOI). The different chemical composition of fly ash at different time definitely due to different source of coal that used in combustion process, i.e. the typical coal accepted by PLTU-Suralaya was delivered from PT. Bukit Asam Coal Mine at 13th March 2006 containing SiO_2 of 48.94%, Al_2O_3 of 21.42%, and Fe_2O_3 of 18.76%, while, the typical coal accepted by PLTU-Suralaya was delivered from PT. Artha Daya Coalindo at 14th March 2006 containing SiO_2 of 34.96%, Al_2O_3 of 24.49%, and Fe_2O_3 of 19.47% (The data obtained from certificate of analysis, which was showed by PT. Indo Power PLTU Suralaya at the end of March 2006). Therefore, the percentage of chemical content of the Suralaya fly ash would be fluctuated and uncontrollable.

Based on XRD analysis as illustrated in Figure 3, the fly ash dominantly contained minerals of quartz and mullite. When it was compared with the chemical content in Table 1, the fly ash may contains quartz, iron oxide, carbon, which was not being yet to get the refractory characteristics. The particles of fly ash had the shape of sphere as depicted in Figure 4 with particles size ranging between 0.31 and 300.74 μm. The size distribution was 80% within sized of 0.31 to 40.99 μm and mean size (d_{50}) of 6,22 μm.

The chemical proportion of Al_2O_3/SiO_2 in the fly ash was 0.34 dominantly containing quartz. This chemical proportion cannot be compared with the reference castable refractory of CAJ-16 in which

Table 1. Chemical composition of Suralaya fly ash at different time

	%SiO_2	%Al_2O_3	%Fe_2O_3	%TiO_2	%CaO	%MgO	%K_2O	%Na_2O	%LOI
Present sample	56.1	19.09	12.29	0.77	5.74	4.34	0.80	0.34	0.36
Previous sample	72.9	11.37	5.93	0.76	3.19	1.99	0.46	1.45	1.04

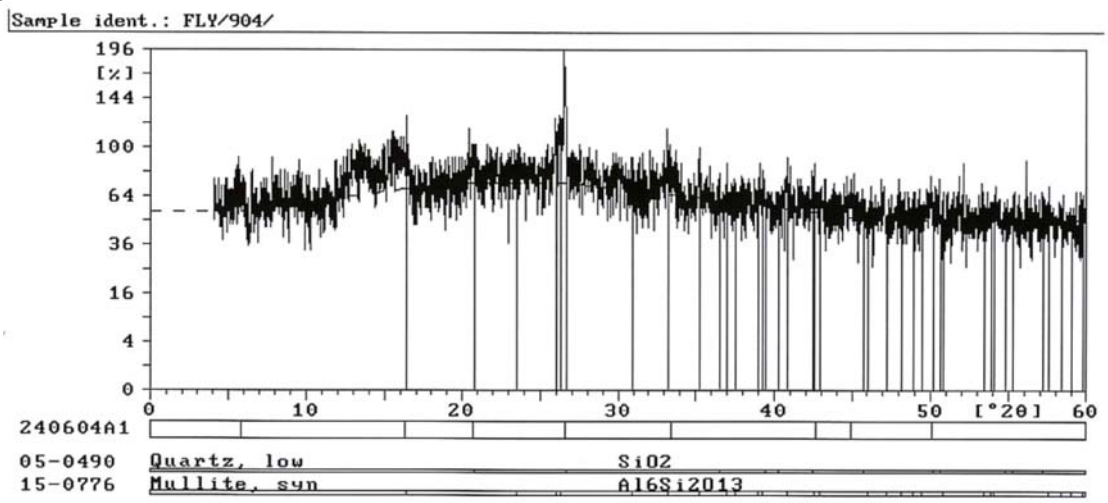

Figure 3. XRD analysis of Suralaya fly ash

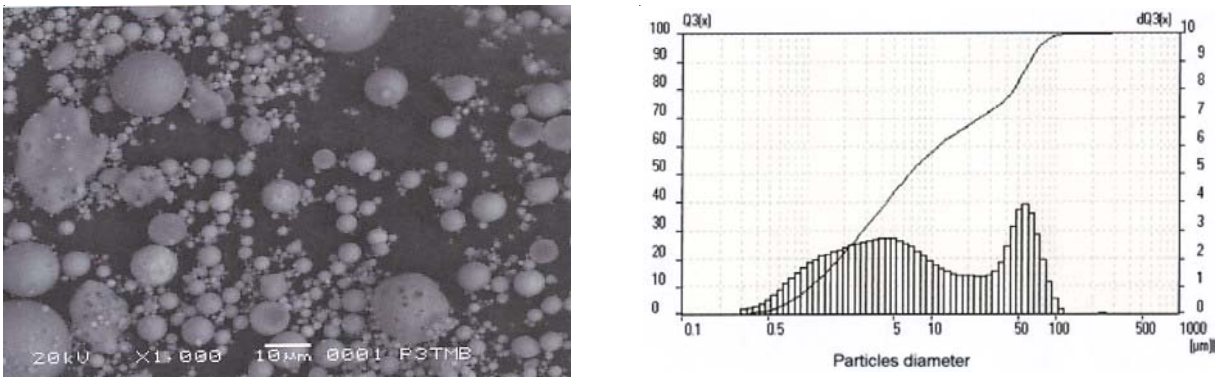

Figure 4. Particles shape and size distribution of Suralaya fly ash, d_{50} = 6,22 mm, (Aziz and Ardha, 2006)

the proportion of Al_2O_3/SiO_2 is 1.62 containing cristobalite and mullite. Lime content in the fly ash was considerably high (5,7%), hence the quality of the fly ash was preference as "C" class, match as cementing castable refractory at cold phase (Kumar et al, 2003). Iron oxide content was also high that it considerably affecting the temperature resistance, if it was used as refractory. Therefore, minimizing iron oxide content to create demagnetised-fly ash might be necessary.

3.3 Demagnetisation of Fly Ash

Demagnetisation of the fly ash was carried out to increase the quality of the fly ash by decreasing the content of iron oxide (Fe_2O_3). Using magnetic separation method with magnetic intensity of 830, 1100 and 1500 gauss would capable to decrease the iron oxide content significantly. Table 2 shows the results of decreasing of Fe_2O_3 content of the fly ash by magnetic separation.

Compared with the initial fly ash, iron oxide was discarded from fly ash by magnetic separation, improved slightly the content of alumina as well as silica; while the content of iron oxide in demagnetised-fly ash significantly decreased from 12% to about 5%. Although the content of silica-alumina was low with chemical proportion of Al_2O_3/SiO_2 of about 0.41, however, these important components in mixed material could be increased by the addition of silica and alumina from grog and fired cement. According to the data in Table 2, the demagnetised fly ash contained amount of silica-alumina relatively the same for any applied magnetic intensity during the experiment. For further

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Table 2. Demagnetisation results of fly ash by magnetic separation

Sample code	%SiO_2	%Al_2O_3	%Fe_2O_3	%TiO_2	%CaO	%MgO	%K_2O	%Na_2O	%LOI
Raw sample/ initial FA	56.1	19.09	12.29	0.77	5.74	4.34	0.80	0.34	0.36
Demagnetised FA-830 gauss	59.3	24.2	5.17	1.02	5.68	2.02	0.57	0.82	1.05
Magnetic - 830 Gauss	51.1	22.7	16.19	0.86	4.78	2.61	0.45	0.67	0.44
Demagnetised FA-1100 gauss	59.3	24.3	5.4	1.05	5.2	2.13	0.56	0.82	1.1
Magnetic - 1100 Gauss	49.1	23.3	18.36	0.73	3.75	2.84	0.52	0.72	0.49
Demagnetised FA-1500 gauss	58.6	26.5	5.01	0.80	4.44	2.05	0.58	0.81	1.1
Magnetic - 1500 Gauss	52.0	25.3	14.07	0.87	3.17	2.31	0.5	0.76	0.87

Note FA= fly ash

experiments, however, it decided to use demagnetised-fly ash that was experimentally processed from magnetic separation with magnetic intensity of 1100 gauss, because of the magnetizing cost possibility.

3.4 Mixed Materials and Green Composite-A

The demagnetised-fly ash was perfectly mixed with fired cement and grog within proportion of 3, 2, 3 by volume, respectively. The mixed material was then dried up to 110°C for 24 hours and then was kept in a hermetical bag. Grog was prepared from crushed bricks of 30 mesh containing minerals of corundum, mullite and cristobalite, while fired cement was used as a binder. This mixed materials could be considered as ready for use castable refractory. However, this ready for use castable refractory should be improved its characteristics in order to relatively resemble to the characteristics of CAJ-16.

According to the data on Table 3, the chemical composition of mixed material was lower than that of CAJ-16, in which Al_2O_3/SiO_2 was 0.62 for mixed material, and Al_2O_3/SiO_2 is 1.62 for CAJ-16. While the iron content dropped from initially 12.29% in fly ash to 2.9% in mixed materials. The content of alumina of 33.5% could theoretically improve the melting point up to 1700°C as depicted in Figure 1b. The potency of mixed materials to increase the melting point, can not be reach yet, due to the mixed materials containing high iron oxide (2.93%) and LOI (1.24%).

The characteristic of mixed material could be tested for some physical, chemical and PCE number as castable refractory. For this purpose, sample of the mixed material was taken for preparing compact mass material by adding a small amount of water (10-15%) and then it was pressed by hydraulic pressure of about 200 kg/cm^2 to turn it into green composite-A. Theoretically, when the mixed materials is added by some water, chemical reaction occurs to develop hydrated calcium alumina silicate as hard mass materials of concrete that resistance to high temperature (Hwang and Huang, 1995). The shape of the green composites-A is a cylinder with 5 cm in diameter and

Table 3. Chemical composition of mixed materials as castable refractory

%SiO_2	%Al_2O_3	%Fe_2O_3	%TiO_2	%CaO	%MgO	%K_2O	%Na_2O	%LOI
54.0	33.5	2.93	0.38	6.7	0.45	0.29	0.31	1.24

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10 cm in height. The green composite-A was cured for 14 days.

3.5 Characteristics of Green Composite-A

The characteristics of green composite-A was tested and observed for its change over micrograph texture and its PCE number to resemble the characteristics of CAJ-16. Chemically, the green composite-A was similar to the data on Table 3, while the physical characteristics and PCE numbers of some compositions of green composite-A can be seen in Table 4.

(Figure 5a) was much different from the micrograph texture of CAJ-16 (Figure 5b). Hence, the green composite-A was prepared by mixing the material of DFA: FC: GR of 3:2:3 was an unsuitable castable refractory material. Therefore, to improve the characteristics of green composite-A, a study of fly ash using thermal treatment was further conducted.

3.6 Thermal Treatment for Demagnetised-Fly Ash

The micrograph texture of green composite-A was

Table 4. Physical characteristics and PCE of fly ash and green composite-A

Materials Composition	Density (g/ml)	Compressive Strength (kg/cm^2)	PCE (SK.No)	Melting point (°C)
Fly ash (as is, Fe_2O_3-high)	1.45	52.1	SK.09	1.285
Demagnetized-fly ash (Fe_2O_3-little low)	1.18	51.0	SK.10	1.305
DFA:FC:GR = 3:1:3	2.65	85.0	SK.12	1.335
DFA:FC:GR = 3:2:3	2.75	90.5	SK.16	1.465
DFA:FC:GR = 3:3:3	2.83	106.5	SK.14	1.400

Note, DFA= demagnetized fly ash, FC=fired cement, GR=grog

Table 4 shows that the raw fly ash with high iron content had PCE number of 09 with melting point of 1285°C, while the demagnetised fly ash with low iron content, its PCE increased slightly to number 10 with melting point of 1305°C. When the demagnetised fly ash was mixed with fired cement and grog in composition of DFA: FC: GR were 3: 2: 3 by volume as a green composite-A, it is shown that the highest PCE number was 16 with melting point of 1465°C. The green composite-A contained 33.5% of Al_2O_3 as denoted in Table 3 should has temperature resistance of 1700°C as depicted theoretically in Figure 1b. However, the temperature resistance was low (1465°C). It might be due to the impurities and small amount of mullite. The effect of fired-cement on strength and PCE was known that the higher of fired cement would increase the strength and PCE. When the fired-cement was added more, the strength is also increased except for PCE. The bulk density and strength did not significantly affected the PCE.

The micrograph texture of green composite-A (see Figure 5a) at composition of DFA: FC: GR of 3:2:3,

Figure 5a. A micrograph texture of green composite-A at composition of DFA: FC: GR = 3:2:3

is shown like sphere and sugary particles of fly ash that sticking to other particles of grog and fired cement, which was easy to be detached. This composition had PCE number of 16 with melting point of 1465°C. This condition might be due to the containing impurities and small amount of mullite. The micrograph texture of green composite-A

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Figure 5b. A micrograph texture of the composite of reference castable refractory (CAJ-16)

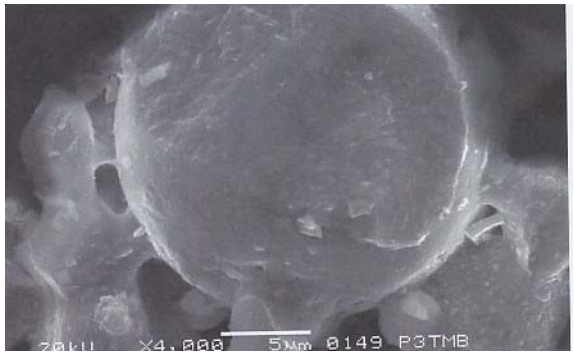

Figure 6a. Micrograph texture of fly ash burning at 1100°C, 0.5 hours, magnify 4000x, no crushing/ grinding

much different compared to the micrograph texture of CAJ-16 as depicted in Figure 5. Since the mixed materials was added with water, chemical reaction should be occurred to develop hydrated alumino silicate as hard mass material of concrete (Norton, 1974), but in this experiment in it did not occurred yet. This phenomenon might be due to the reactivity of silica and alumina in the demagnetised fly ash was not completely yet. Hence, in order to improve the reactivity of silicate and alumina in the fly ash, thermal treatment of the demagnetised-fly ash may be important to conduct at high temperature.

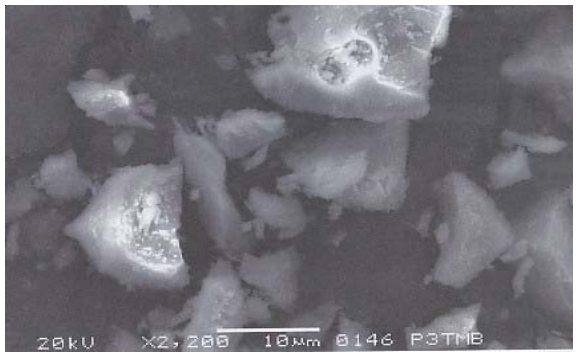

Figure 6b. Micrograph texture of fly ash burning at 1250°C, 0.5 hours, magnify 3000x, after crushing/ grinding

Burning tests of demagnetised fly ash were conducted at temperatures of 1100°C, 1250°C and 1350°C. When the fly ash was burned at temperature of 1100°C for 0.5 hours, the SEM showed the fly ash as original shape like sphere, but a little change occurred at the surface that appeared like glassy shape of fused silica as presented in Figure 6a. When the demagnetised fly ash was burned at temperature of 1250°C and 1350°C for 0.5 hours, the fly ash melted uncompletely and after cooling down, turned into hard mass sinter. The sinterized demagnetised fly ash was then crushed and was ground to create loose material within size less than –150 mesh. Under SEM, the particles of crushed sinterized demagnetised fly ash, which was burned at temperature of 1250°C and 1350°C were shown changing from sphere to irregular shapes with preferences lengthwise to sharp-edge as presented in Figure 6b and 6c. In this condition, quartz and alumina started to change into trydimite or cristobalite and mullite,

Figure 6c. Micrograph texture of fly ash burning at 1350°C, 0.5 hours, magnify 3000x, after crushing/ grinding

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which may be agreed with the silica-alumina equilibrium diagram that depicted in Figure 1a. While, if the fly ash was burned at 1350°C for 1 hour and left it cooling down to be hard mass sinter, under the SEM, the micrograph texture appeared like needles joining together as presented in Figure 6d. The texture of needles of silica-alumina was regarded as reactive mullite (Supomo dkk, 1997). Compared to the micrograph texture of mullite that made from burning topaz as revealed in Figure 7, it was clear that the shape of mullite is like needles (Supomo dkk, 1997). Therefore, the demagnetised fly ash must be burned at temperature at least 1350°C to get a good tendency as castable refractory containing mullite.

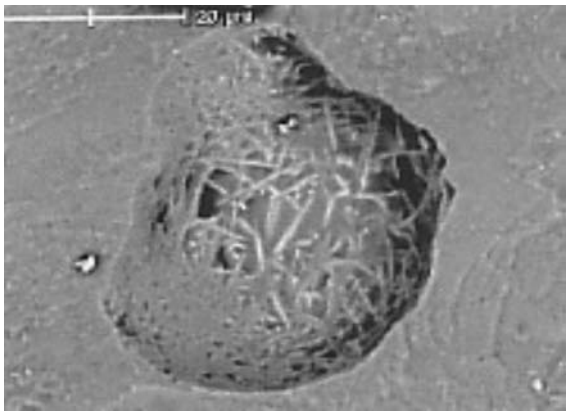

Figure 6d. Micrograph texture of fly ash burning at 1350°C, 1 hours, magnify 2000x, no crushing/ grinding

3.7 Mixed Materials and Green Composite-B

Green composite-B was prepared by perfectly mixing of crushed sinterized demagnetised fly ash (CSDFA) with fired cement and grog in the proportion of 3,2,3 by volume, respectively. Grog was prepared by crushed bricks of 30 mesh, while fired cement was used as a binder. The mixed material was kept in a hermetical bag. This mixed material was considered as ready for use castable refractory. However, the characteristics of this ready for use castable refractory must be improved to resemble the characteristics of CAJ-16. Prior to improving its characteristics, analysis of its physical, chemical and PCE number are important. For these purposes, sample of present mixed material was taken from the hermetical bag. The mixed material was added with a small amount of water (10-15%) and then pressed by hydraulic pressure of 200 kg/cm^2 to turn it into hard mass of green composite-B. The shape of the green composites-B was a cylinder with 5 cm in diameter and 10 cm in height. The green composite-B was cured for 3 days.

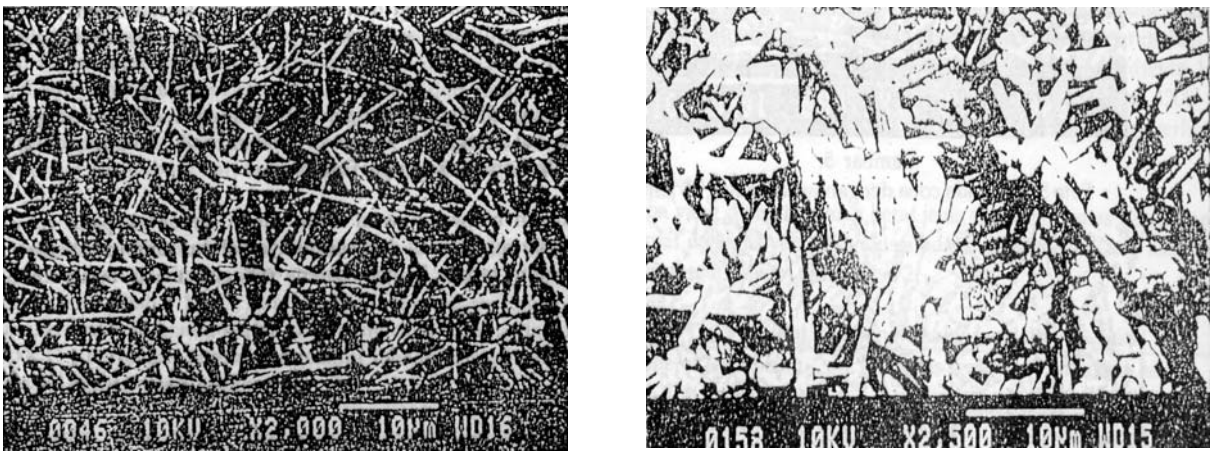

Figure 7a. Micrograph texture of mullite made from burning topaz 1200°C, high porosity (Supomo dkk, 1997)
b. Micrograph texture of mullite made from burning topaz 1700°C, less porosity (Supomo dkk, 1997)

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3.8 Characteristic of Green Composite-B

The chemical composition of the mixed material may relatively similar to that of the mixed materials as represented in Table 3. The size of CSDFA was about 150 mesh. The minerals content of CSDFA were not analysed. However, it should be better than minerals content of original fly ash as depicted in Figure 2. The cristobalite was changed from quartz, and reactive mullite was changed from original mullite. Grog contained minerals of corundum, mullite and cristobalite.

The green composite-B was prepared from CSDFA that was burned at temperature of 1100°C. Under SEM, the micrograph texture was presented in Figure 8a, where each material was in bonding into circled-flattened-composite. It means that the original sphere of the fly ash was uncompletely yet changing into lengthwise-needled-mullite. When the green composite-B was prepared from CSDFA that was burned at temperature of 1250°C, under SEM its micrograph texture is presented in Figure 8b, where each material was in bonding into irregular direction of lengthwise-needled-composite. This phenomenon might be due to some reactive mullite started changing into stable mullite that resistance to high temperature. Furthermore, when the green composite-B was prepared from CSDFA that was burned at temperature of 1350°C, under SEM its micrograph texture is presented in Figure 8c, where such bonding occured as a new composite that the shape of texture was relatively regular lengthwise in the same direction. In the composite, there were some holes that indicated the reaction of reactive mullite might be uncompleted yet. In general, compared to the micrograph texture of the reference material of CAJ-16 as depicted in Figure 8d, the micrograph texture of typical Al-Si composite-B 1350°C relatively resembled to the micrograph texture of reference castable refractory of CAJ-16.

Green composite-B that was prepared by different amount of fired cement can be seen in Table 5, where the higher the amount of fired cement was added, the higher the compressive strengths. However, this phenomenon was not followed by changing the PCE number, where the highest PCE number was found at composition of CSDFA: FC: GR = 3: 2: 3 by volume and its cone number of 17 with melting point of 1512°C (see Table 5).

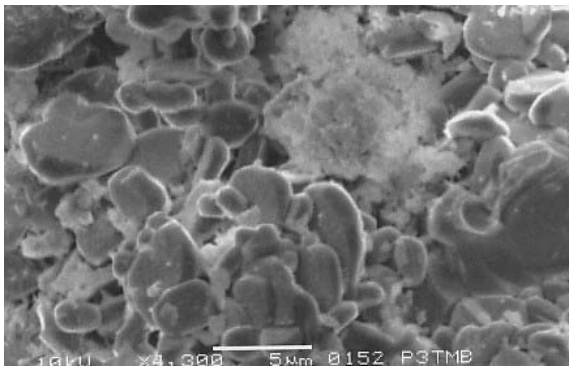

Figure 8a. Micrograph texture of green composite-B, 1100°C, 0.5 hours, circled-flattened

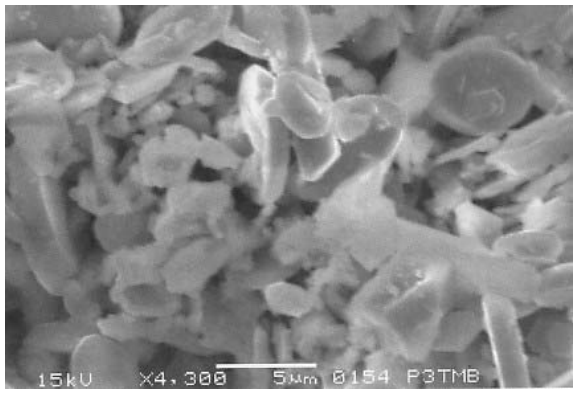

Figure 8b. Micrograph texture of green composite-B, 1250°C, 0.5 hours, irregular tends to lengthwise

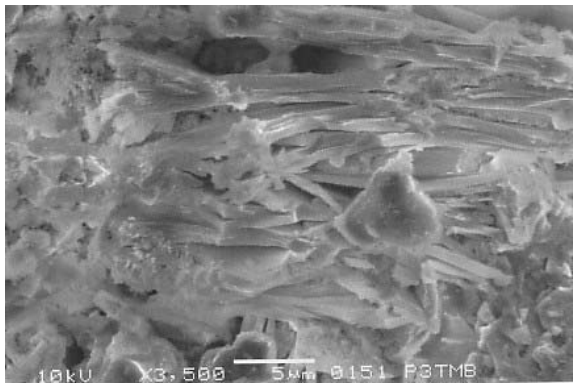

Figure 8c. Micrograph texture of green composite-B, 1350°C, 0.5 hours, relatively regular-lengthwise

Content

Figure 8d. Micrograph texture of the composite of reference castable refractory (CAJ-16), regular-lengthwise

Table 5. The compressive strength and PCE of green composite-B using CSDFA 1350°C, 0.5 hours

Materials Composition	Compressive Strength (kg/cm2)	PCE (SK.No)	Melting Point (°C)
CSDFA: FC: GR = 3: 1: 3	130	SK.14	1,400
CSDFA: FC: GR = 3: 2: 3	185	SK.17	1,512
CSDFA: FC: GR = 3: 3: 3	190	SK.15	1,430

Note: CSDFA = crushed-sinterized-demagnetization of fly ash, FA=fired cement, GR=grog

3.9 Characteristic of Composite-B by Thermal Treatment

Green composite-B was treated at high temperature to create composite-B. Each material composition as represented in Table 5 was burned at temperature of 1250°C for 0.5 hours. The products of composite-B with materials composition of CSDFA: FC: GR of 3: 1: 3; CSDFA: FC: GR of 3: 2: 3 and CSDFA: FC: GR of 3: 3: 3 by volume were analysed for the compressive strength as well as PCE. It revealed that both of the compressive strength and PCE slightly increased. The highest PCE was found at materials composition of CSDFA: FC: GR of 3: 2: 3 with melting point of about 1564°C as presented in Table 6. This phenomenon illustrates that the effect of temperature on the materials was proceeds until the definite high temperature.

Furthermore, due to thermal reaction of compo site materials was taken place during burning time, it was tried to vary the temperature to know its effect on the number of PCE of the materials composition of CSDFA: FC: GR of 3: 2: 3 by volume. Effect of burning temperature was presented in Table 7. After burning time of 1 hour at temperature of 1100°C, 1250°C, and 1350°C, it showed that the increase of the burning temperature, increases significantly the compressive strength as well as the PCE. The highest results was found

Table 6. The compressive strength and PCE of composite-B, 1250°C, 0.5 hours

Materials Composition	Compressive Strength (kg/cm2)	PCE (SK.No)	Melting Point (°C)
CSDFA: FC: GR = 3: 1: 3	144	SK.18	1,522
CSDFA: FC: GR = 3: 2: 3	178	SK.20	1,564
CSDFA: FC: GR = 3: 3: 3	193	SK.19	1,541

Note: CSDFA = crushed-sinterized 1350°C -demagnetization of fly ash, FA=fired cement, GR=grog

Table 7. The compressive strength and PCE of composite-B of CSDFA: FC: GR = 3: 2: 3 by volume with variation of burning temperature

Burning Temperature (°C)	Burning Time (minute)	Compressive Strength (kg/cm2)	PCE (SK.No)	Melting Point (°C)
1100	60	153	SK.21	1,574
1250	60	190,5	SK.23	1,605
1350	60	210	SK.26	1,621

after I hour burning at temperature of 1350°C, the compressive strength was 210 kg/cm^2 and PCE number was 26 with temperature resistance of 1621°C.

Experimental results shown in Table 6 and 7 are clearly characterized that the higher temperature of burning process of the composite would increases the temperature resistance of the composite. This might be due to the burning time and burning temperatures were closely related to stabilization process of silica and alumina component reacting into christobalite and mullite phase as theoretically depicted in Figure 1a.

Alumina content in the composite-B of 33.5% was similar to the mixed material of green composite-A as revealed in Table 3. Hence, it is supposed to be resistant at temperature of 1700°C as depicted theoretically in Figure 1b. However, in this experiment the composite-B was only resisted to the temperature of 1621°C. This might be due to the composite-B contains significant impurities such as Fe_2O_3 of 2.9%, TiO_2 of 0,4%, K_2O+Na_2O of 0,6% and LOI of 1,24% as shown in Table 3. Moreover, compared with the chemical composition of the reference castable refractory (CAJ-16): SiO_2 of 29,1%, Al_2O_3 of 47,2%, and small amount of impurities (Aziz and Ardha, 2006), hence, the sample of the fly ash needs to increase Al_2O_3 content by decreasing SiO_2 content as well as decreasing impurities, or adding high grade Al_2O_3 sources into the fly ash.

Based on experimental works in this study it is confirmed that the fly ash generated during the combustion of coal can be utilized as castables refractory materials. The castable refractory based on fly ash had tendency to fulfil the criteria of two phases Si-Al diagram as depicted in Figure 1a. However, the fly ash required processes in series such as demagnetisation, sinterization up to temperature of 1350°C, crushing and grinding of sinter, and finally mixed with grog and binder as ready for use castables refractory. When the mixed materials of ready for use castables refractory was casted into mould as composite refractory, this composite material was resistance at temperature of 1564°C. The composite materials of refractory would increase its temperature resistance as it was treated at high temperature for longer time until reaching its Si-Al stabilization phase.

In these works, the composite materials of refractory were burned at temperature of 1350°C for 1 hour. Its temperature resistance increased up to 1621°C. It is raising questions as to whether should be solved, how long the composite must be burned and how much the high temperature degree must be applied until reaching its maximum Si-Al stabilization phase. From this study, the answers of those questions are not obtained yet. However, a simple correlation between burning temperatures and melting points might be regarded as shown in Figure 9. If the desired melting point is 1700C, hence the burning temperature of green composites should be approximately of 1760°C for 1 hour. This approximation did not tested yet. However, based on Figure 7b, it indicated that the best needles shape of the mullite was found when the

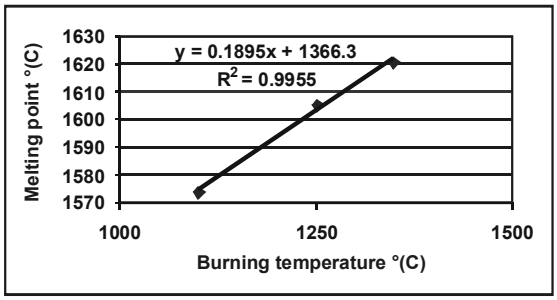

Figure 9. Simple correlation of burning temperatures and melting points

material was burned at temperature of 1700°C (Supomo et.al, 1997), where the micrograph texture had less holes with less porosity and the needles strongly tied up each others. Accordingly, the technology of making castable refractory based on fly ash could be succeed, but the economical is still questioning and needs further study.

There are many aspects in refractory such as physical, chemical and thermal affect its characteristics that are not discussed yet in this study. Some of them are porosity, bulk density, rheology of castables, thermal expansion etc. Therefore, this investigation should be continued.

4. CONCLUSIONS

The conclusions of this study are as follows :

- Fly ash generated during the combustion of coal does not have refractory characteristics yet, due to the minerals content is dominantly

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quartz and some impurities such as iron oxide and unburned coal. Fly ash should be processed prior to its utilization as castable refractory,

- Demagnetisation of fly ash using magnetic separator with intensity of 1100-gauss, reduced the Fe_2O_3 content from 12% to 5%. Then, this material was mixed with grog and fired cement, it had melting point of 1465°C,

- Demagnetized-fly ash was burned at 1350°C, can change the mineral content of quartz into mullite as indicated by changing the shape of materials texture from rounded to needles. This crushed sinterized demagnetized fly ash was mixed with grog and fired cement to develop into ready for use castable refractory. This mixed material had melting point of 1512°C,

- As the crushed sinterized demagnetized fly ash composite refractory was burned again up to 1350°C, this composit material refractory had melting point of 1621°C,

- Those phenomeno indicate that the composite materials of refractory would increase its temperature resistance as it was treated at high temperature for longer time until reaching its Si-Al stabilization phase.

Simple alternatives to utilize the fly ash are as follows :

- Direct use of the fly ash itself as castable refractory has melting point of 1285°C,

- Fly ash can be utilized as a mixed material into referenced commercial castable refractory (CAJ-16, melting point of 1750°C), by adding the fly ash into CAJ-16 with proportion of 1/1, 2/1, and 3/1 by volume. However, the melting point decreased to 1690°C, 1460°C, and 1280°C, respectively,

- Direct use of demagnetized-fly ash itself as castable refractory had melting point of 1305°C.

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