The effect of sintering time on recycled magnesia brick from kiln of the cement plant

B B Aji¹, D Rosalina², Azhar² and M Amin¹

¹Research Division for Mineral Technology, Indonesian Institute of Sciences, Jalan. Ir. Sutami Km 15, South Lampung 35361, Indonesia
²Department of Chemical Engineering, University of Lampung, Jalan. Prof. Dr. Soemantari Brodjonegoro No.1, Bandar Lampung, Lampung, 35145, Indonesia

Email: bramantyo.bayu.aji@lipi.go.id

Abstract: This research aim was to investigate the effect of sintering time on reuse waste of magnesia brick from the rotary kiln of the cement plant. Reuse of the magnesia brick was carried out by mixed the kaolin as the binder. Spent refractory was used as aggregate with the composition of 85% spent refractory and 15% kaolin clay, respectively. The reuse brick then was molded with the size of 5x5x5 cm using hydraulic press under a load of 10 tons in order to forms green body. Green body then dried and sintered at 1200 °C with time variation of 2 hours, 4 hours, 6 hours, 8 hours and 10 hours, respectively. Thus, for comparison reuse brick was tested to its apparent porosity, the bulk density, and Cold Crushing Strength (CCS). The effect of kaolin addition as binder was also discussed.

1. Introduction
Refractory can be classified as ceramic materials which have ability to hold its physical and chemical properties at high temperature and extreme condition. Beside steel industry, cement industry also consume refractory in great amount such as for isolator on rotary kiln to protect kiln from high temperature, chemical reactions, and abrasion during cement production. As reported by Wiharja, refractory was used almost in all part of kiln from preheating zone, calcination, sintering, to cooling zone, respectively [1]. After service life reached, usually brick will dispose to landfill introduce to environmental problem. As reported, around 60 million tons monolithic and brick were consumed in typical cement plant [2]. Not only health problem, refractory waste also contributed to high cost to industry since refractory was disposable material.

Issues to minimize cost and environment problem related to refractory waste initiate research in reuse spent refractory. Several researches have been reported, Kwong et al reported successful recycled Mg-O refractories from EAF as raw material for slag conditioner [3], Arianpour et al [4] investigated reuse spent Magnesia brick from Steel plant as aggregate have adequate properties at composition up to 30 % and slightly better corrosion resistance than prepared sample with natural sintered magnesia, and recently, study by Avelar et al reported that recycle MgO-C refractories with dolomite addition could be satisfying industrial inquiry in order to better production [5].

Although there are already research conduct reuse spent refractories, however it still limited information provide how sintering temperature can affect to performance of recycled refractories as new brick. In ceramic manufacturing, physical properties were formed during sintering. Several factor that affect are, temperature and sintering time, particle size, composition and green body density,
respectively [6]. Propitious composition, particle size, and typical temperature of sintering were the main factors to get high green body density. Green body was made by compacted the raw material with certain load pressure. Disadvantage reuse spent refractories fact that valuable mineral was declined due to oxidation and overheated. Kaolin was known for their high contain of Alumina which refers to refractory materials. When heated, kaolin will formed spinel which is seed of Mullite phase [7,8]. Mullite is refractory materials that contribute to high strength which is associated with its excellent thermal shock resistant [9]. Uses of kaolin as additive showed, it can enhance the mechanical properties of the ceramic due to changes of its crystalline phase during sintering time [7,10,11]. Therefore in this research, the aim was to investigate the effect of sintering temperature on reuse waste of magnesia brick and the effect of kaolin addition as binder.

2. Experimental

2.1. Material

Material which used was magnesia used refractory of rotary kiln from cement plant in West Java, Indonesia. Kaolin clay was added as adhesive was taken from Kemiling, Bandar Lampung City.

| Component | wt% in Used refractory | wt% in Kaolin clay |
|-----------|------------------------|--------------------|
| MgO       | 86.83                  | 0.35               |
| Al₂O₃     | 1.59                   | 28.90              |
| SiO₂      | 1.16                   | 42.32              |
| Fe₂O₃     | 0.34                   | 1.76               |
| CaO       | 2.76                   | 1.89               |

2.2. Recycle refractory brick preparation

Used refractory was grinded and shifted into two sizes aggregate. The large aggregate size was 40# to 80 # (at range 150-480 µm), smaller refractory aggregate size was -80 # (less than 180 µm) and kaolin powder was size of 100 # (150 µm), respectively. All materials were completely mixed, then molded in size 5x5x5 cm using hydraulic press under a load of 10 ton and forms green body. Total composition a refractory recycle sample (Rr) was 300 grams with 85% of used refractory and 15% kaolin clay and water. The time of sintering varies for 2, 4, 6, 8, and 10 hours, respectively. As for comparison, the refractory without kaolin clay was made as standard with composition was 100% of used refractory sintered at 1200 °C for 6 hours.

2.3. Physical properties measurement

Apparent porosity and bulk density is tested by using standard method ASTM C20-00 [12]. crushing strength or pressure strength is the maximum load when cracking occur or material destruction per unit surface area when pressured at room temperature [13]. Cold Crushing Strength test was based on ASTM C133 procedure [14]. Surface morphology was characterized using scanning electron microscopy (SEM, SU-35000, Hitachi, Japan).

3. Results and discussion

3.1. Apparent porosity and bulk density

Sintering was the main process during ceramic manufacturing and controlled its final physical properties. Diffusion of atom during particle growth, pore composition, and neck formation are among possible typical that affect to bulk density and porosity of green body.
Figure 1. Bulk density and apparent porosity behaviour of recycle refractory (Rr) with increase of sintering time at temperature of 1200°C.

As shown in figure 1, illustrated bulk density and apparent porosity of recycle refractory with the sintering time. It can be seen that, bulk density slightly increased from 1.035 gr/cm³ to 1.058 gr/cm³ at 10 hr. The result of the apparent porosity of refractory was tend to slightly decreased from 19.8 % at 2 hr to 19.6 % at 6 hr, then sharply decreased to lower level 19.18 % at 10 hr of sintering time. On molding process, the given loaded pressure will shape the initial green body. The small particle filling to the empty spaces between the larger particle so the refractory will become more compact. Densification then started after heat involves. Longer time gives more time to particle growth therefore density of green body increase.

Porosity of recycle refractories decreases with increases of sintering time. During sintering, diffusion of atoms which move from particle surface to pores thus, fusion one particle to another particle occurs that called as neck. As diffusion become more intense densification occurred and denser refractory can be achieved. Discontinue of neck growth on specific temperature will promote of pore formation. The pore formed due to unbalanced diffusion and trapped gas in material [15]. The neck growth will influence the shrinkage of material pores. Therefore, as longer time of sintering densification of refractory is high and porosity composition is less.

3.2. Cold crushing strength (CCS)

Cold crushing strength (CCS) is standard of refractory which stated the strength of refractory to the given maximum load. CCS does not indicate the characteristic of refractory when it is used and more represent state the strength of refractory when it is transported. Refractory with the good strength is not easily crack or damage when it is transported. As well as with thermal conductivity, CCS also has relation with density and porosity of materials.

As illustrated in figure 2 CCS of recycle refractory (Rr) was increased from 5.25 MPa at 2 h to 6.8 Mpa at 10 hr of sintering. Similar with bulk density, longer time of sintering make green body denser and finally CCS as the state strength of refractory increase as longer time of sintering.
3.3. The effect of kaolin clay addition

Kaolin is one of clay that has good plasticity when it is mixed with water [16]. Uses of kaolin as additive expected can enhance the mechanical properties of the ceramic by exploits its crystalline phase changing during heating process. As a material with high contain of alumina, it can considerable of mullite raw material. Mullite is considered as a binding phase in most of refractory brick and it has a high resistance to melting and minimum thermal expansion as well as low thermal conductivity [17].

Table 2. The comparison of recycle refractory (Rr) and used refractory sintered at 1200°C for 6 hr.

| Sample | CCS, (Mpa) | Porosity, (%) | Bulk Density, gram/cm³ |
|--------|------------|---------------|------------------------|
| Used kiln | 48.38 | 5.49 | 2.82 |
| Rr | 5.75 | 19.65 | 1.048 |

As presented in table 1, addition of kaolin in recycle refractory (Rr) did not give better properties compare to used refractory. CCS for Rr sample was only 7.75 Mpa much lower than used refractory sample with CCS is 48.38 Mpa. It can be explain that, Rr sample was not dense enough as represented the bulk density is only half than used kiln, were the bulk density 1.048 gram/cm³ and 2.82 gram/cm³, respectively. CCS value was small since density of material is low then the loaded pressure that can be
accommodated is also low. This reason due to densification was not occurred during sintering. As longer time, kaolin expected can enhance densification process through mullite formation. It shown from the higher porosity of Rr than used refractory, 19.65% and 5.49%, respectively. At around temperature 450°C, combined water in kaolin is release and leaves a large pore within refractory body, resulting in the formation of metakaolin clay. Metakaolin clay is heated around 980°C, spinal phase is formed, an amorphous siliceous phase. As the temperature increased, the spinnel is transformed into mullite and, simultaneously SiO₂ is expelled to form of glassy phase. Then glassy silica promotes sintering, which densification initiated, and formation of third shrinkage phase at temperature above 1200°C [18,19]. However, recycle refractory (Rr) has higher porosity than used refractory which means rather than densification another phenomenon could occur such as coarsening.

As illustrated in figure 5(a), SEM image result show that Rr has larger particle than used refractory. It is proved that rather than particle has grow and then densified, coarsening occurred resulting in imperfect grains and its various size. The growth of very large grain will eliminate the bond between particles which located at the edge of grain borders (grain boundaries), thus will decrease the strength of refractory, and it influence the refractory to become weak. Besides, the imperfect grains growth will affect formation of pores because and contribute to high percentage of refractory porosity. Meanwhile, in figure 5 (b), it can be seen that particle has more uniform in shape and size, however recycle brick still has lower bulk density than new magnesia brick [20]. It can be explained due extreme condition, used refractory has decline its properties by oxidizing or mechanical contact during processes.

4. Conclusions
Investigation variations of sintering times have been done in order to recycle waste magnesia brick into new refractory. As increasing of the sintering time, bulk density of recycle refractory (Rr) was slightly increased. However, as addition of kaolin that used as binder did not successfully enhance the final properties such as increase of CCS number. High porosity percentage of Rr indicates that may be from preparation of green body, the load pressure is not only high enough but also sintering temperature should be higher to ensure mullite formation from kaolin and increase of recycle refractory properties. Therefore, future research utilization of waste brick with kaolin addition with higher load pressure and sintering temperature could be applied in order to achieve excellent properties.

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