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Technical report

Development of Fired Clay Bricks by Coal-Preparation Refuse

選炭廃石を用いた焼成粘土煉瓦の製造

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It is important to find a possibility for utilizing coal-preparation refuse (CPR) generated from coal mines in terms of environmental and economic points of view. In this study, a possibility for manufacturing fired clay bricks (FCBs) by CPR instead of clay was investigated. Firing shrinkage ratio, compressive strength, and water absorption ratio of test samples manufactured using CPR in the open air were measured. It was found that firing shrinkage ratio and compressive strength decrease with increasing the addition amount of CPR, while water absorption ratio is almost constant under the entire addition amount ranges of that. The test sample manufactured was nearly similar to the 1st grade clay bricks of Korea Standard L (ceramics) 4201.

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***Key-words :** Coal-preparation refuse (CPR), Fired clay bricks (FCBs), Coal mine, Coal refuse*

1. Introduction

Large amount of coal-preparation refuse (CPR) has been discharged from coal mines all over the world. Currently, hundreds of millions of tons of CPR have been dumped on the surface around coal mines in Korea. This disposal of CPR is somewhat toxic to living organism since though CPR contains small amounts of heavy metals, sulfur, and organic materials, they contaminate the surface and ground water through the leachate generated at the dump-sites. Furthermore, CPR contains lots of carbon and clay mineral. So, some methods to utilize clay mineral from CPR were suggested.[1)-6)] However, CPR has not been effectively utilized because of its low quality. It is thus highly desirable to find an effective utilization method of CPR.

In general, CPR contains the SiO_2/Al_2O_3 weight ratio of 2.0-4.0, which is nearly similar to that contained in fired cay bricks (FCBs).[7),8)] Thus, in this study, a possibility for utilizing CPR as a raw material of FCB replacing of clay was investigated. It is here expected that the utilization can save firing energy because of the amount of carbon contained in CPR. However, the utilization of CPR as a raw material of FCB has not yet been developed; this article describes research into developing such utilization.

Keeping this in mind, the present research is concerned with examining the properties of FCB manufactured using CPR that is generated from a coal mine in Korea. The properties examined include plasticity, color change, surface morphology and microstructure, phase transformation, ignition loss, porosity, firing shrinkage ratio, water absorption ratio, and compressive strength. The study is part of a major research project aimed at developing an effective utilization method of CPR.

2. Experiments

2.1 Raw materials

The raw material used in the experiments was CPR generated from Hwa-Sun coal mine in Korea. An X-ray diffraction pattern of the CPR is shown in **Fig. 1**. Shown in this figure represents that the CPR contains quartz, chlorite, muscovite, kaolinite, talc, anorthite, and so on.

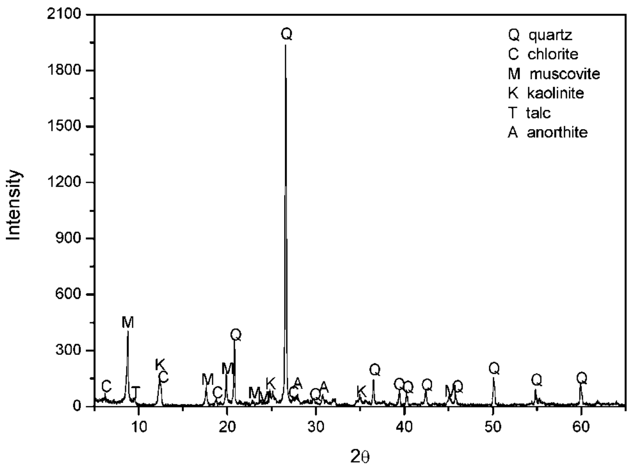

Fig. 1. XRD patterns of CPR.

Table 1. Chemical Compositions of the Sample

SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	K_2O	Na_2O	TiO_2	MnO_2	P_2O_5	Igloss
61.16	17.28	3.38	1.01	0.94	2.27	0.22	0.84	0.04	0.06	12.14

Table 1 shows the chemical composition of the CPR analyzed by XRF (XRF-1700, Shimadzu, Japan). The table shows that the main ingredients are SiO_2 and Al_2O_3, and the weight ratio of SiO_2/Al_2O_3 is 3.6 that are in the range (2.0-4.0) of that of clays used as raw materials of FCB in general. Especially, it is shown that the ignition loss is 12.14 mass%. The CPR was thus analyzed by the coal industrial analysis method in order to investigate carbon content and calorific value. The result is shown in **Table 2**. The results represent that the CPR contains 88.30 mass% ash and 6.17 mass% fixed carbon, and the calorific value is 494 cal/g.

On the other hand, a standard clay mixture (SCM) obtained from a ceramic company in Korea was used as a refer-

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Table 2. Various Properties of CPR

Ash	Moisture	Volatile Matter	Fixed Carbon	cal/g
88.30	0.21	5.32	6.17	494

Table 3. Chemical Compositions of a Standard Clay Mixture (SCM) Obtained from a Ceramic Company in Korea

SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	K_2O	Na_2O	TiO_2	MnO_2	P_2O_5	Igloss
58.33	24.22	2.86	5.70	0.57	1.44	1.52	0.49	0.04	0.03	5.07

ence composition of FCB, of which composition is shown in **Table 3**.

Based on the raw material analyses, it was found that the weight ratio of SiO_2/Al_2O_3 contained in the CPR might be suitable for manufacturing FCB. In addition, it was expected to be able to save the consuming energy in firing process by using CPR as a raw material of FCB replacing of clay.

2.2 Fired brick making and testing methods

In the experiments, SCM and CPR were first pulverized by a jaw and cone crusher. The particle size of the powder ranged under 1 mm. The SCM and CPR were then mixed as designed proportions. The amount of CPR mixed was varied by from 10 mass% to 100 mass%. And then, 30 g of the samples was mixed with distilled water of 7 mass%, molded to bricks in a diameter of 50 mm by a compress pressure of 1,000 kg/cm^2 and dried in the room temperature for 1 d, and then the bricks were kept at 60°C for 3 h. And then, the bricks were again kept at 105°C for 24 h. After that, the bricks were burnt in the electrical furnace at desired temperatures for 2 h, and then cooled in the furnace. The heating rate was 1.5°C/min, and the firing temperature range between 1100°C and 1200°C in air condition.

The plasticity of the raw materials of FCB was evaluated by means of Atterberg's consistency limits: plastic index was calculated by plastic limit and liquid limit which were measured by KS F 2303.

Color change of the specimens was measured by using spectrophotometer (CM–2002, Minolta, Japan) with D65/10° geometry and indicated by CIE $L^*a^*b^*$ system.[9)]

The surface morphology and microstructure of the specimens were observed by SEM (JSM6380LA, Jeol, Japan), and the phase transformation by XRD (Pillips X'pert MPD, Philips, USA).

The porosity of the specimens is measured by Mercury Porosimeter (Poresizer 9320, Micromerities, USA) which is able to detect the pore size distribution between 0.007 μm and 300 μm.

Firing shrinkage ratio, water absorption ratio and compressive strength of the test specimens were also calculated by KS L 4201.

3. Results and discussions

Figure 2 shows the plastic index of bricks with the additional ratio of CPR. The results shows that the plastic index of bricks manufactured by SCM only is about 7.8, while the plastic index of bricks done by adding CPR decreases according to $y=12.23–0.063x$ as the additional amount of CPR increases. The plastic index was also about 11.03 at the addition of CPR 20 mass%, as shown in the figure. The results indicate that bricks can be manufactured by addition of CPR up to 60 mass% without a plasticity problem.

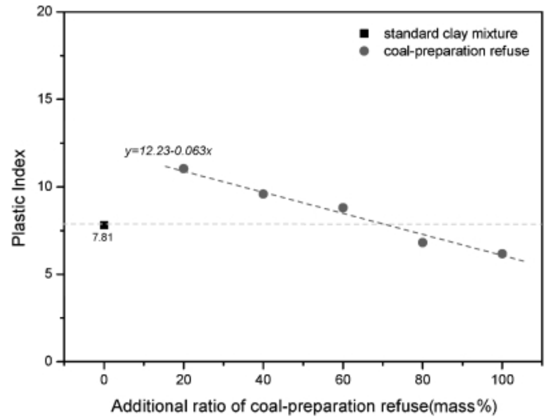

Fig. 2. Plastic index of raw material for ceramic firing body as a function of additional ratio of CPR.

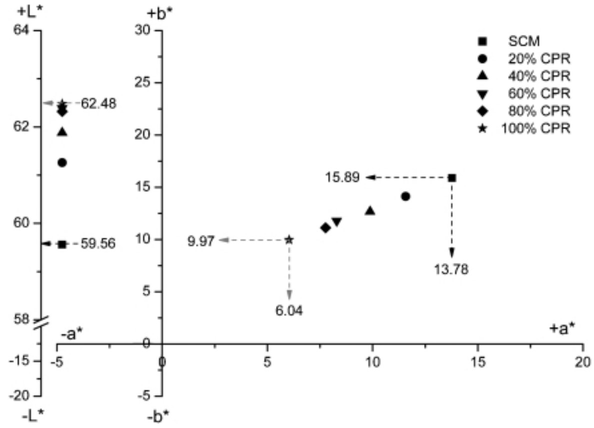

Fig. 3. Color change of the fired specimen as a function of additional ratio of CPR at 1150°C for 2 h.

The color change of bricks that are fired by varying the additional amounts of CPR at 1150°C for 2 h in air condition is shown in **Fig. 3**. The figure shows that the redness ($+a^*$) and yellowness ($+b^*$) of bricks manufactured by SCM only are lower than them of bricks manufactured by adding CPR, while the whiteness ($+L^*$) is high in the bricks manufactured by SCM only.

Thus, the bricks manufactured by adding CPR are brighter than them manufactured by SCM only. That might be explained by the fact that carbon in CPR reduces Fe^{3+} to Fe^{2+} at the firing process.

Figure 4 shows SEM microphotography of bricks that are fired by varying the additional amounts of CPR at 1150°C for 2 h in air condition. As shown in this figure, the surface morphology of bricks containing 20 mass% CPR seems to be a surface welding state. And, it was investigated by the naked eye that the distribution of surface porosity slightly increases with increasing the additional amount of CPR. Thus, the increase of porosity in FCB manufactured by adding CPR is forecasted.

Figure 5 shows the mineralogical compositions of bricks produced by SCM only and 40 mass% CPR. The bricks were fired at each temperature for 2 h in air condition. From the figure, it was known that both bricks were very similar in their mineralogical compositions, and mullite ($Al_6Si_2O_{13}$) and andesine ($Na_{0.499}Ca_{0.491}(Al_{1.488}Si_{2.506}O_8)$) were produced in

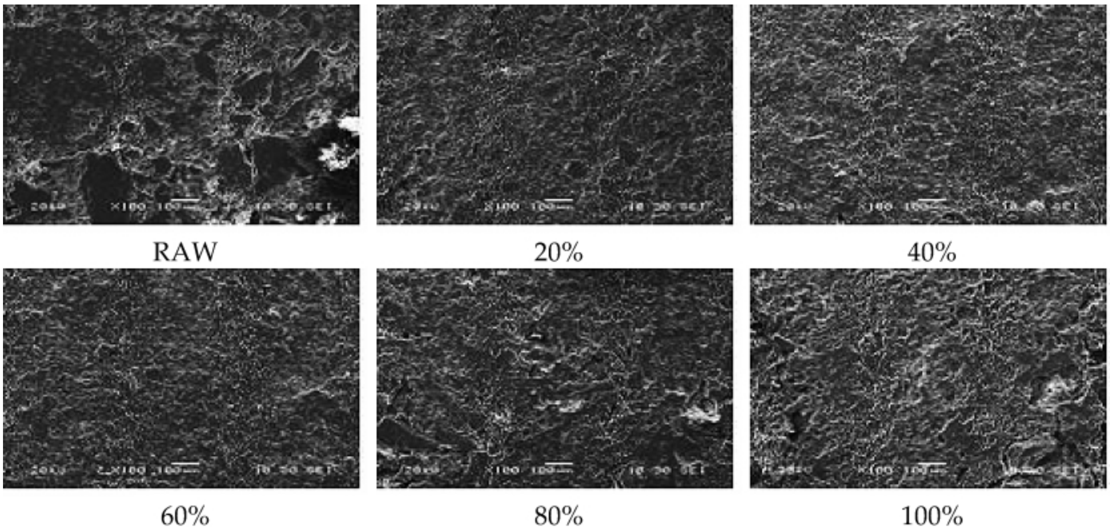

Fig. 4. SEM photography of the fired specimen as a function of additional ratio of CPR at 1150°C for 2 h.

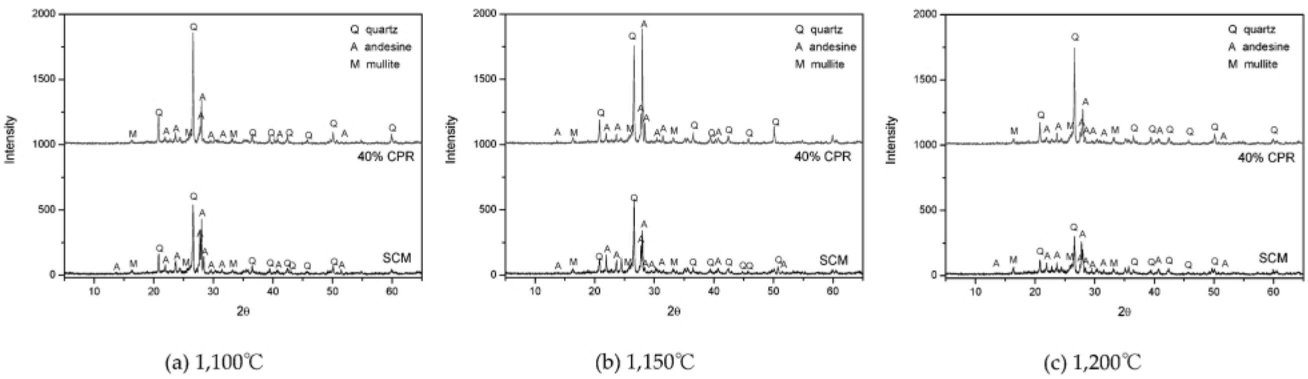

Fig. 5. XRD patterns of the fired specimens as a function of firing temperature for 2 h at various temperatures.

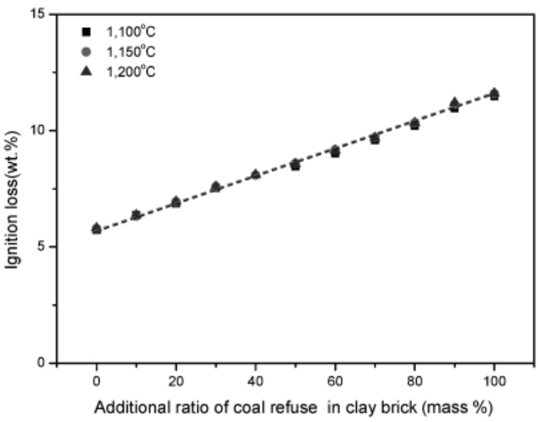

Fig. 6. Ignition loss of the fired specimen as a function of additional amounts of CPR at various temperatures.

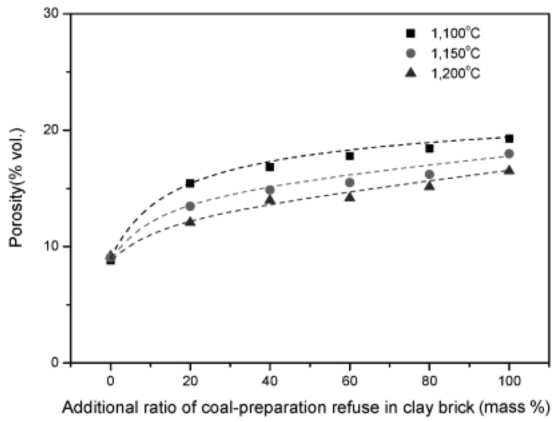

Fig. 7. Porosities of fired specimens as a function of additional ratio of CPR at various temperatures.

the bricks.

Figures 6 and **7** show the ignition loss and porosity of bricks that are fired by varying the additional amount of CPR between 10 mass% to 100 mass% and the firing temperatures of 1100–1200°C for 2 h.

From Fig. 6, it was investigated that the ignition loss of bricks manufactured by SCM only and adding CPR was independent with the firing temperature. It was also done that the ignition loss of bricks manufactured by SCM only was about 5.7 mass%, while that of bricks manufactured by adding CPR increased with increasing the additional ratio. And for bricks manufactured by CPR only, the ignition ratio of

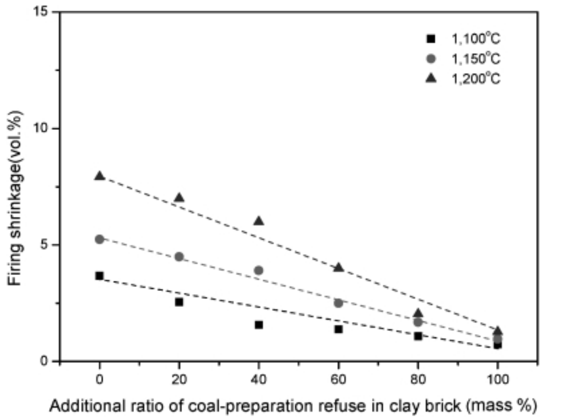

Fig. 8. Firing shrinkage of fired specimen as a function of additional ratio of CPR at various temperatures.

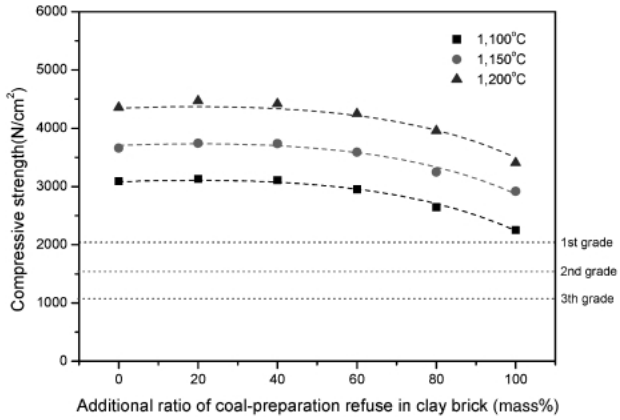

Fig. 10. Compressive strength of fired specimen as a function of additional ratio of coal-preparation refuse.

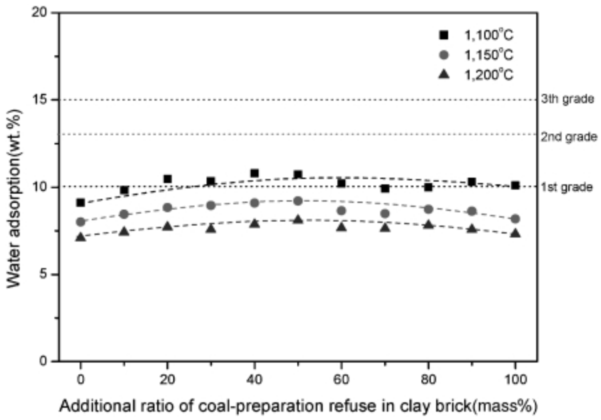

Fig. 9. Water absorption of fired specimen as a function of additional ratio of CPR at various temperatures.

11.5 mass% was shown in the figure. The increase of ignition loss is due to carbon contained in the CPR.

Shown in Fig. 7 also represents that the porosity of bricks manufactured by adding CPR increased with increasing the additional ratio, and decreasing the firing temperature. The decrease of porosity according to the firing temperature may be due to the surface welding.

Figure 8 shows the firing shrinkage ratio of each brick that is fired by varying the firing temperature for 2 h. The figure indicates that the firing shrinkage of each brick decreased with increasing the additional amounts of CPR and increasing the firing temperature. This is the reason why CPR contains much more silicate mineral than SCM, as shown in Tables 1 and 3.

Figure 9 also shows the water absorption ratio of each bricks that is fired by varying the additional amount of CPR at the firing temperatures of 1100–1200°C for 2 h. As shown in this figure, the water absorption ratio of bricks manufactured by adding CPR at over 1150°C satisfies that of 1st grade clay brick of KS L 4201. The water absorption ratio of below 10 mass% is the guideline of 1st grade clay brick.

Figure 10 shows the compressive strength of bricks that are fired by varying the additional amount of CPR between 10 mass% to 100 mass% and the firing temperatures between 1100 and 1200°C for 2 h. As shown in the figure, the compressive strength of bricks manufactured by adding CPR at the entire firing temperature ranges was 2250 N/cm^2–3400 N/cm^2, which is nearly similar to that of bricks manufactured by CPR only. Thus, it was expected that the compressive strength of all bricks manufactured by adding CPR is over 2059 N/cm^2 that is the standard criteria of 1st grade clay brick in KS L 4201.

From the above tests, it could be suggested that the use of coal-preparation refuse as a raw material to make fired cay bricks is possible.

4. Conclusions

In order to investigate the utilization possibility of CPR as a raw material of FCB, various properties such as plasticity, firing shrinkage, water adsorption, and compressive strength of bricks manufactured by CPR were examined in the study. The results indicated that firing shrinkage ratio and compressive strength decrease with increasing the addition amount of CPR, while water absorption ratio is almost constant under the entire addition amount ranges of that. It was also found that the properties of plasticity, firing shrinkage, water adsorption, and compressive strength of bricks manufactured by CPR satisfied them of the 1st grade clay bricks of Korea Standard L(ceramics) 4201. Therefore, it is expected that the use of coal-preparation refuse as a raw material to make fired cay bricks can save energy and decrease pollution.

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