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Preparation and Properties of Sintered Brick Based on Coal Gangue and Fly Ash†

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Coal gangue and fly ash were selected as raw materials to prepare building sintered brick. The influence of ratio of coal gangue and fly ash and sintering temperature on mineralogical composition, compressive strength, linear shrinkage and pore size distribution of building sintered brick were investigated. The results indicated that mineralogical compositions of sintered brick were mullite, quartz and β-cristobalite. With the ratio of coal gangue and fly ash decreased and sintering temperature increased, the compressive strength and linear shrinkage of sintered brick increased. For ratio of coal gangue and fly ash = 1:1, the sintered brick with compressive strength of 63.88 MPa, linear shrinkage of 8.84 %, porosity of 15.28 % and average pore diameter of 203.9 nm was prepared at 1250 ºC and the porosity and average pore diameter reduced by 42.7 and 28.2 % with the temperature increased from 1050 to 1250 ºC, respectively.

Keywords: Sintered brick, Coal gangue, Mineralogical composition, Pore size distribution.

INTRODUCTION

Coal gangue is one kind of complex solid waste, which is discharged in the production process of coal mining and washing[1,2]. Its major chemical composition is SiO_2 and Al_2O_3, followed by Fe_2O_3, CaO, MgO, K_2O and Na_2O. About 1600 sites of coal gangue hills have been formed, which are even called "coal mining cancer"[3,4]. Fly ash is a major by-product obtained from the flue gas of coal fired power plants[5]. The main chemical composition is SiO_2 and Al_2O_3 that total amounts is about 70-90 %. According to a statistical report, in 2010, the amount of coal gangue and fly ash was 1.07 billion tons in China. In addition, there will be up to 1.3 billion tons in 2015. More and more farmlands have been occupied by coal gangue and fly ash, resulting in environmental issues and even engendering geological disasters, *etc*[6]. In recent years, applied researchers have become increasingly interested in comprehensive utilization of coal gangue and fly ash. Although there a number of studies have been used coal gangue and fly ash, the utilization ratio of them is far low. In 2012, the Government of China has limited the utilization of clay sintered products and even prohibited in some areas. Based on coal gangue and fly ash have similar chemical composition to clay. They can be used as a substitute for clay in sintered brick. In the paper, coal gangue and fly ash are used as main raw materials to prepare sintered brick. The mineralogical composition, compressive strength, linear shrinkage and pore structure of sintered brick are also investigated.

EXPERIMENTAL

The main raw materials used in the paper included coal gangue, fly ash and plasticizer. Coal gangue and fly ash were obtained from a coal mine and power plant in Huainan, Anhui province of China, respectively. The chemical compositions of the raw materials were presented in Table-1. Their mineralogical compositions are kaolinite and quartz, mullite and quartz, respectively.

Five mixtures were selected for experiments according to Table-2. The mixtures were homogenized in a blender and water at a weight percentage of 25 % was added to the mixture.

TABLE-1
CHEMICAL COMPOSITION OF THE RAW MATERIALS (wt %)

Raw materials	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	K_2O	Na_2O	SO_3	LOI
Coal gangue	57.71	19.14	6.12	1.97	0.90	1.02	0.41	0.41	12.02
Fly ash	58.15	29.39	6.14	1.62	0.64	0.90	0.80	0.46	1.39

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TABLE-2
MIX PROPORTIONS OF RAW MATERIAL (wt %)

No.	Coal gangue	Fly ash	Plasticizer
M-1	85	0	15
M-2	68	17	15
M-3	63.75	21.25	15
M-4	56.60	28.40	15
M-5	42.5	42.5	15

Under a certain pressure, cylindrical samples with Φ 25 mm × L30 mm were prepared by PVC pipes. The samples were dried at room temperature for several days to drive off the moisture. Subsequently, sintered at a heating rate of 5-6 ºC/min at 1050, 1150 and 1250 ºC for 0.5 h in KSL-1400X furnace, respectively. Finally, these were naturally cooled to room temperature in the furnace. The samples were coded as M-1, M-2, M-3, M-4 and M-5.

Compressive strength of sintered brick was conducted on nine samples of each mixture. It was calculated following the eqn. 1:

$$P = \frac{4F}{\pi d^2} \tag{1}$$

where P is the compressive strength (MPa), F is the load (N) on the sample and 'd' is the diameter (mm) of cylindrical samples after sintering.

Linear shrinkage was determined using the eqn. 2:

$$\alpha = \frac{L_1 - L_2}{L_1} \times 100\ \% \tag{2}$$

where 'α' is the linear shrinkage, 'L_1' and 'L_2' are the lengths of cylindrical samples before and after sintering, respectively.

The mineralogical compositions of sintered brick were analyzed by X-ray diffraction (XRD) with 40 mA and 40 kV, CuK_α radiation. Porosity and pore size distribution were determined by mercury intrusion porosimetry (MIP).

RESULTS AND DISCUSSION

Mineralogical composition: Fig. 1 illustrated the mineralogical compositions of different sintered products at 1250 ºC for 0.5 h. The major mineralogical compositions were mullite, quartz and β-cristobalite. With the change of sintered brick from M-1 to M-5, the major peaks of quartz increased and β-cristobalite decreased. However, the peaks of mullite had no significant fluctuation. The reason caused this situation might be related with chemical composition of raw materials. The amount of SiO_2 and Al_2O_3 increased with the increase of ratio of coal gangue and fly ash. It caused too much SiO_2 formed glassy phase [Fig. 4(a)].

The mineralogical compositions of sample M-5 sintered at different temperatures were shown in Fig. 2. It could be observed that the mineralogical compositions were mullite and quartz at 1050 ºC. However, there were mullite, quartz and β-cristobalite at 1150 and 1250 ºC. The peaks of mullite increased with the increasing sintering temperature, accompanied with a reduction in the peaks of quartz. This because of mullite was obtained from the decompostion of kaolinite and that contained in fly ash. At higher temperature, the newly mullite was formed from solid-state reaction between Al_2O_3 and amorphous SiO_2 of raw materials. It was supported by preview research[7,8]. The higher the temperature, the reaction more fully, the more the generation amount of mullite. The mineral phase reaction during the sintering could be described as the equations:

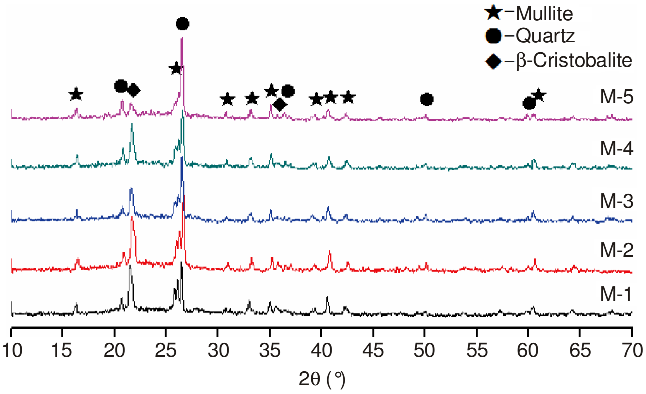


Fig. 1. XRD patterns of different samples sintered at 1250 ºC for 0.5 h

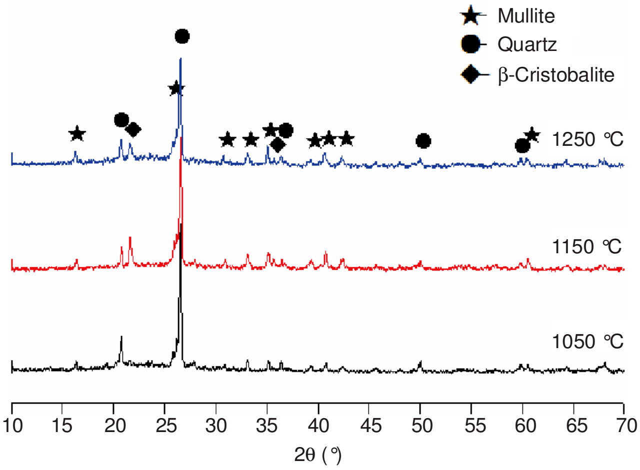


Fig. 2. XRD patterns of sample M-5 sintered at different temperatures

$$3[Al_2O_3{\cdot}2SiO_2{\cdot}H_2O] \rightarrow 3Al_2O_3{\cdot}2SiO_2 + 4SiO_2 + 6H_2O$$

$$2SiO_2 + 3Al_2O_3 \rightarrow 3Al_2O_3{\cdot}2SiO_2$$

Compressive strength: The compressive strength of sintered products was observed in Fig. 3. At the same sintering temperature, the compressive strength of sintered bricks increased with the change from M-1 to M-5. At 1250 ºC, the compressive strength increased from M-1 of 41.70 MPa to M-5 of 63.88 MPa. This may be due to that glassy phase filled in the pore of sintered bricks with ratio the of coal gangue and fly ash decreased. It could be seen that compressive strength markedly improved as the sintering temperature was increased from 1050 to 1250 ºC in Fig. 3. Its values, 27.16, 51.09 and 63.88 MPa, were obtained from M-5 sintered at 1050, 1150 and 1250 ºC, respectively. Moreover, M-5 sintered at 1250 ºC was 36.72 MPa higher in compressive strength than that sintered at 1050 ºC. This might be because that the amount of mullite increased with the increasing sintering temperature and the enamel layer was formed on the surface of sintered brick at 1250 ºC (Fig. 4b). It contributes to the increase in compressive strength.

Linear shrinkage: The linear shrinkage of sintered products is represented in Fig. 5. It could be clearly observed from Fig. 5 that the linear shrinkage increased with the change

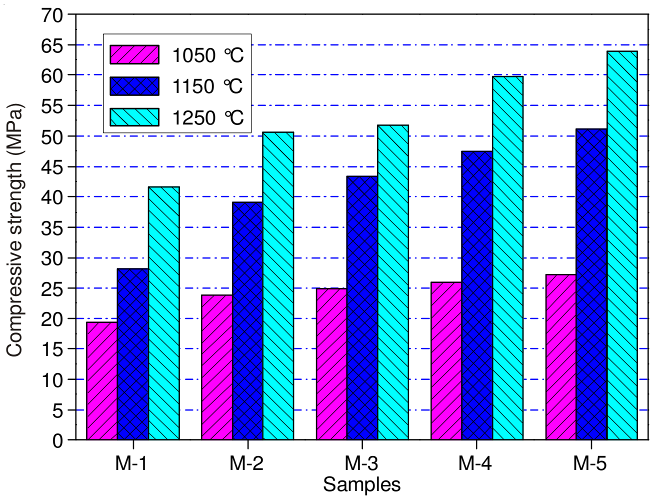


Fig. 3. Compressive strength of samples

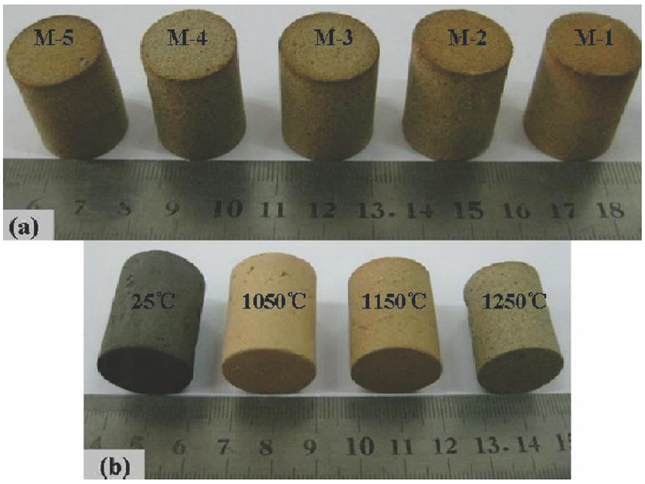


Fig. 4. (a) Appearance of various samples sintered at 1250 ℃. (b) Appearance of M-5 sintered at various temperatures

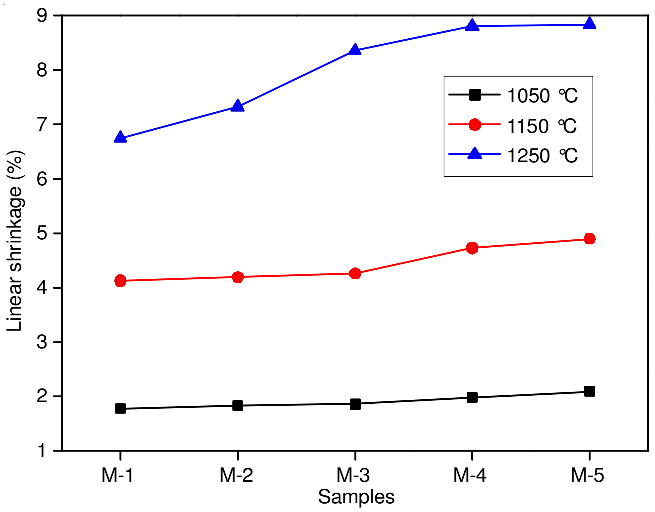
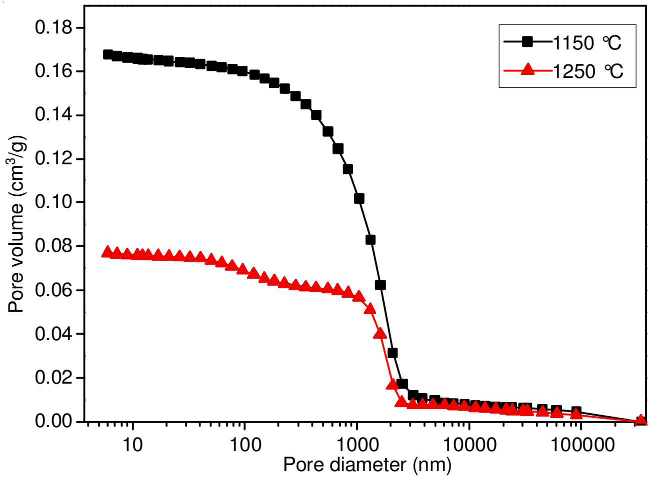


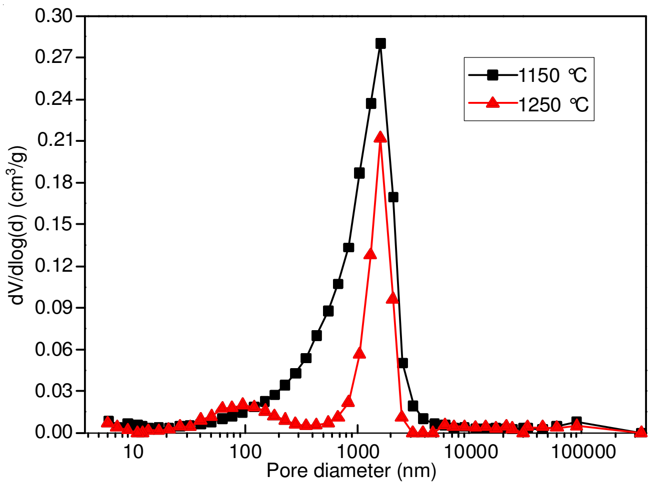
Fig. 5. Linear shrinkage of samples

of samples from M-1 to M-5 at the same sintering. The linear shrinkage of samples increased slightly at 1050 and 1150 ℃. However, it increased significantly at 1250 ℃, its linear shrinkage values changed from 6.75 to 8.84 %. In addition, we could know that the linear shrinkage values increased markedly with the increasing sintering temperature. The linear shrinkage values for sample M-5 sintered at 1050, 1150 and 1250 ℃ were 2.10, 4.90 and 8.84 %, respectively. This is due to the fact that the chemical composition and sintering temperature affect viscosity and sintering behaviour of sintered product. So when SiO_2 in the raw materials and sintering temperature increase, there will be more liquid phase. On one hand, it will accelerate the sintering; on the other hand, the particles were bonded together easily. Resulting in sintering bricks became much denser, accompanied with the volume shrinkage.

Pore structure: Fig. 6 showed that pore structures of sintered brick M-5 at 1050 and 1250 ℃. The porosity decreased sharply from 26.65 % at 1050 ℃ to 15.28 % at 1250 ℃, it reduced by 42.7 %. It concluded that pore volume decreased from 0.1675 to 0.0771 cm^3/g, reduced by 54.0 % with the temperature increased from 1050 to 1250 ℃ (Fig. 6a). The pore size distribution of M-5 sintered at 1050 and 1250 ℃ is shown in Fig. 6b. The pore diameter was mostly 1 μm at 1050 ℃. However, there pore diameter mostly located at 100 nm and 1 μm. The average pore diameter is 283.8 nm at 1050 °C to 203.9 nm at 1250 ℃, it reduced by 28.2 %. This means that the pore size of the sintered bricks becomes finer with the increase of sintering temperature. In other words, the sintered bricks become much denser with the increase of sintering temperature.



(a) Pore volume



(b) Pore size distribution

Fig. 6. Pore structures of M-5 sintered at 1050 °C and 1250 ℃

Conclusion

Coal gangue and fly ash were used as main raw material to prepare sintered brick. When ratio of coal gangue and fly ash = 1:1, the sintered brick with compressive strength of 63.88 MPa, linear shrinkage of 8.84 %, porosity of 15.28 % and average pore diameter of 203.9 nm was prepared at 1250 ºC. Its mineralogical compositions are mullite, quartz and β-cristobalite. With the ratio of coal gangue and fly ash decreased, the compressive strength and linear shrinkage of sintered brick increased. The mullite had no significantly change and the β-cristobalite decreased. With the increasing sintering temperature, the compressive strength and linear shrinkage of sintered brick increased markedly. For sintered brick with ratio of coal gangue and fly ash = 1:1, the porosity and average pore diameter reduced by 42.7 and 28.2 % with the temperature increased from 1050 to 1250 ºC, respectively.

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