Study on the extracted process of mullite from coal fly ash by-product sodium silicate

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The mullite was directly extracted from coal fly ash through removing iron and silica without sintering process by-product sodium silicate. The effect of processing parameters on the leaching rate of iron and silica was investigated. The physical properties, phase composition and microstructure of the mullite extracted from the original fly ash were characterized by using X-ray fluorescence spectrometer, X-ray diffractometer, Fourier transform infrared spectroscopy, scanning electronic microscope and energy dispersive spectroscopy. The technical indicators of sodium silicate were also tested. The results show that the maximum iron extraction reaches 85.9 mass % when firstly with a magnetic separation process then leaching with 20 mass % of hydrochloric acid solution at 85°C for 3 h. The 40.4 mass % leaching rate of silica was presented after leaching with 25 mass % of sodium hydroxide at 95°C for 4.5 h. As a result, the beneficiated extracted sample exhibited a mullite content of 82 mass %, the corundum content of 18 mass %. This method provides a potential way on the utilization of coal fly ash to prepare mullite.

Key-words : Coal fly ash, Mullite, Scanning electron microscopy, X-ray diffraction

1. Introduction

Coal fly ash (CFA) is the unburned residue after the high temperature combustion of coal in thermal power plant. With the development of power industry, CFA becomes one of the largest industrial waste residues. The emissions mass of CFA is about 500 million tons per year in recent years in China, whereas only about 60% of CFA is utilized.1] It has been reported that more than 5 billion tons CFA has been disposed in solid controlled landfills or waste containment facilities in China.2] So much fly ash that has not been used not only causes serious environmental pollution but also brings storing problem.3,4] In view of demanding of the environmental protection and energy application, it is imperative to increase the utilization rate of massive fly ash. So far, CFA as a resource mainly has been used in building, agriculture, chemical industry such as extracting aluminum oxide or preparing ceramic.5–8]

The main components of CFA are aluminum oxide, silicon dioxide, some of these compounds accounting for about 5–50% in fly ash presence as crystal phase mainly including mullite and quartz, other parts appear as amorphous silica glassy phase. Among them, mullite has been recognized as an important material for high temperature structural applications9–12] due to its excellent properties such as good mechanical strength, excellent thermal shock, high creep resistance, a low thermal coefficient of expansion, and good dielectric properties.13] At present, mullite is mainly obtained by reaction of alumina and silica in certain conditions with different methods, and there are many reports about preparation of mullite using CFA as raw materials. Park et al. obtained orthorhombic mullite whiskers with a diameter of 0.6–1.8 μm by firing compacts of CFA and NH4Al(SO4)2·12H2O powders at 1300°C for 10 h.14] Ying et al. obtained low-cost mullite ceramics from natural bauxite and industrial waste fly ash after sintering below 1300°C.15] Tan prepared mullite whiskers from CFA using sodium sulfate flux by firing at 1000°C for 3 h.16] Jiang et al. investigated temperature feature on synthesizing mullite whiskers from CFA and andalusite-sericite phyllite with NaH2PO4·2H2O as molten salt media at 1600°C for 6 h.17] Chen et al. obtained mullite from high-alumina fly ash after sintering at 1500°C for about 4 h.18,19] All these studies, however, require to add aluminum resources into fly ash for preparation of mullite with sintering process. It dose not only consume large aluminum resources but also needs to consume a lot of electrical energy in the sintering process.

The objective of the present work is to facilitate the recycling of CFA and saving the energy in the production of mullite, the authors reported a simple process using CFA to directly extract mullite and by-product sodium
silicate without adding aluminum resources and sintering process. In this method, the iron ions in raw fly ash were firstly removed using a magnetic separation process then leaching with hydrochloric acid solution. Then amorphous silica in as-prepared solid phase was converted to sodium silicate after being treated by sodium hydroxide solution, which effectively extract and utilize amorphous silicon resources from CFA. Finally, the prepared mullite has been investigated. From the view of cost effectiveness and environment, the use of industrial waste fly ash for mullite preparation is very promising.

2. Experimental

2.1 Raw materials

The raw CFA was from thermal power plant located in Inner Mongolia, China. Table 1 lists representative chemical composition data for CFA. As can be seen, the primary components of CFA are alumina (Al₂O₃), silica (SiO₂) and iron oxides (Fe₂O₃) with little amounts of calcium, magnesium, sodium and potassium. The phase composition and microstructure of CFA were characterized by using X-ray diffractometer and scanning electronic microscope respectively, which was described in section 3.1.

2.2 Prepared method

Firstly, CFA solution with a concentration of 30 mass % was passed through magnetic separator machine to remove magnetic iron in the condition of 13000 Gs of magnetic flux density with two magnetic separation processes. The residues were mixed with a measured 20 mass % of hydrochloric acid solution and mass ratio of liquid/solid was 5:1 at 85°C for 3 h for further removing iron. The as-prepared solid phase was further treated with 25 mass % of sodium hydroxide and mass ratio of fly ash/alkali was 1:0.5 at 95°C for 4.5 h to remove amorphous silica. After it was filtered and washed by distilled water, the mullite powders were obtained after the residues were dried at 105°C for 24 h and the sodium silicate was extracted from the filtrate.

2.3 Characterization

The chemical compositions were examined by X-ray fluorescence spectrometer (XRF, X-calibure, Xenemetrix, America). The phase compositions were examined by X-ray diffraction (XRD, D/MAX-2500/PC, Rigaku, Japan). The quantitative analysis of the phase composition in the prepared material was conducted by using Rietveld method with TOPAS 4.2 software.²⁰,²¹ The morphology of the samples were examined by scanning electron microscope (SEM/EDS, SU8010, Hitachi, Japan). The fourier transform infrared spectrosophotometer (FTIR) were collected using NEXUS 670 (Nicolet, America) in the 4000~400 cm⁻¹ region using potassium bromide as the diluents and binder. The characteristic of mullite powder was tested strictly comply with GB/T 7322-2007 named Refractory products - Determination of pyrometric cone equivalent (refractoriness) and YS-T469-2004 named Determination of whiteness of alumina and aluminum hydroxide which are widely used in China. The test of sodium silicate is complied with GB/T 4209-1996 named Sodium silicate for industrial use and GB 3049-86 named Chemical products for industrial use general method for determination of iron content o-phenanthroline spectrophotometric method.

3. Results and discussion

3.1 Characterization of raw materials

Figure 1 presents the XRD pattern of the original CFA. As can be seen, the major crystalline phases detected were mullite and corundum. The characteristic diffraction peak of amorphous aluminosilicate glass phase was also detected as a broad reflection at 18 and 25° in the XRD pattern. Mullite existing in the fly ash belongs to primary mullite, which was formed from aluminosilicate clay minerals during the combustion process of raw coal.²² The content of mullite was about 40.2 mass %. Amorphous silicate glassy phase derived from uncrystallized silica-rich substance during the process of rapid cooling. The heavy background in the XRD pattern is due to the existence of iron oxide in fly ash.

Figure 2 displays SEM photographs of the original CFA. As shown in Fig. 2, a large number of CFA spherical particles whose surface was covered with some amorphous components parcel were observed, these particles contained an exterior aluminosilicate glass hull with some small grain deposits followed by a near surface layer of mullite and corundum crystalline phase. The majority of particles were in the range of 2.4~14.3 μm.

3.2 Removing magnetic iron from raw fly ash

In view of poor liquidity of fly ash, the fly ash was mixed with water to pass through magnetic separator machine to remove magnetic iron. The influence of process parameters such as concentration of fly ash solution,
magnetic flux density of magnetic separation machine and magnetic separation times on removing effect of iron was investigated.

### 3.2.1 The concentration of fly ash

Experiments were conducted in a magnetic flux density of 8000 Gs and a magnetic separation. Figure 3(a) reveals the influence of concentration of fly ash solution range from 10 to 40 mass% on the effect of removing iron. As can be seen, the iron removal ratio firstly increases then decreases. When concentration of fly ash solution was 30 mass%, the best iron removal effect was obtained and iron removal ratio reached up to 55 mass%.

### 3.2.2 Magnetic flux density of magnetic separation machine

Figure 3(b) shows the influence of magnetic flux density of magnetic separation machine on the effect of removing iron in the condition of 30 mass% (concentration of CFA solution) and a magnetic separation. The iron removal ratio increased when the magnetic flux density ranged from 8000 to 13000 Gs, while it has no change when the magnetic flux density was higher than 13000 Gs.

### 3.2.3 Magnetic separation numbers

The influence of magnetic separation times on the effect of removing iron in the condition of 13000 Gs of magnetic flux density and 30 mass% (concentration of CFA solution) was displayed in Fig. 3(c). The best iron removal effect was presented when two magnetic separations were adopted. The effect was not outstanding with the magnetic separation time increasing.

The 62.9 mass% leaching rate of iron was presented by using wet-removal magnetic separator method. The optimal process parameters as follows: the concentration of fly ash was 30 mass%, the magnetic flux density was 13000 Gs and magnetic separation times was twice.

### 3.3 Leaching of residual iron impurities from raw CFA

Fly ash, after magnetic separation, can only remove some of the magnetic iron minerals. The rest of the nonmagnetic or weak magnetic minerals need to be removed by using a chemical method. So, the residual iron impurities were further wiped out by acid. The reaction equations as follows (1) (2):

\[
\text{Fe}_2\text{O}_3 + 6\text{H}^+ = 2\text{Fe}^{3+} + 3\text{H}_2\text{O} \quad (1)
\]

\[
\text{FeCO}_3 + 2\text{H}^+ = \text{Fe}^{2+} + \text{CO}_2 + \text{H}_2\text{O} \quad (2)
\]

### 3.3.1 The type of acid

The experiment was conducted by using three type of acids: hydrochloric acid, sulfuric acid (H$_2$SO$_4$) and nitric acid (HNO$_3$) under three different concentrations to com-
pare the effect of removing iron, respectively. The experimental results were shown in Fig. 4. As can be seen, the removal effect of iron after hydrochloric acid treated is significantly better than that of H₂SO₄ and HNO₃ at various concentrations. The reason might be as follows: Firstly, this is mainly due to the formation of a variety of stable complexes between Fe³⁺ and Cl⁻, which accelerated the response process of iron and hydrochloric acid. Secondly, the decomposing of HNO₃ in the treated process leading to reduce the concentration of acid and produce harmful gas. Finally, the washing time of filter cake after treated with hydrochloric acid was about 2 min, while it was about 15 min after treated with H₂SO₄ under the same conditions. This might be related to the production of microsolvents such as CaSO₄. Therefore, hydrochloric acid is the best choice for removing residual iron impurities from raw CFA.

The influence of processing parameters such as leaching temperature, leaching time, concentration of hydrochloric acid and mass ratio of liquid/solid on leaching rate of iron was evaluated in order to determine the optimal process of the extracted procedure.

3.3.2 The concentration of hydrochloric acid

Experiments were conducted at 85°C, leaching time of 3 h and 5:1 (mass ratio of liquid/solid), respectively. The influence of hydrochloric acid concentration on leaching rate of iron was shown in Fig. 5(a). As can be seen, the concentration of hydrochloric acid has a significant influence on leaching rate of iron, with the increase of hydrochloric acid concentration (10–20 mass %) the leaching rate of iron was increased quickly and the maximum appeared when the concentration of hydrochloric acid was 20 mass %. After the concentration of hydrochloric acid was above 20 mass %, the leaching rate of iron declined...
due to the overflow of hydrochloric acid on account of co-evaporation of hydrochloric acid and water.

3.3.3 The leaching temperature

Figure 5(b) shows the influence of reaction temperature on leaching rate of iron in the condition of 20 mass % (concentration of hydrochloric acid), leaching time of 3 h and 5:1 (mass ratio of liquid/solid). With increasing of temperature, the leaching rate of iron increased rapidly below 85°C and almost no change above 85°C. This is attributed to decrease of concentration of acid owing to volatilization of hydrochloric acid when the reaction temperature was above 85°C.

3.3.4 The leaching time

The influence of reaction time on leaching rate of iron in the condition of 85°C, 20 mass % (concentration of hydrochloric acid) and 5:1 (mass ratio of liquid/solid) was shown in Fig. 5(c). As can be seen, the leaching rate of iron was gradually increased between 0–2 h and increased slowly 2 h later, so the optimal time is 3 h.

3.3.5 The mass ratio of liquid/solid

Figure 5(d) shows the influence of mass ratio of liquid/solid on leaching rate of iron in the condition of 85°C, leaching time of 3 h and 20 mass % (concentration of hydrochloric acid). The leaching rate of iron increased gradually before mass ratio of liquid/solid was 5:1, while there are almost no changes after 5:1, so the best mass ratio of liquid/solid was 5:1.

The optimal process with 85.9 mass % leaching rate of iron was presented when the concentration of hydrochloric acid was 20 mass %, the leaching temperature was 85°C, the leaching time was 3 h and the mass ratio of liquid/solid was 5:1.

3.4 Leaching of amorphous silica from fly ash

The author used sodium hydroxide solution to extract the amorphous silica, the reaction equation as follow (3):

\[
2\text{NaOH} + \text{SiO}_2 (\text{amorphous}) \rightarrow \text{Na}_2\text{SiO}_3 + \text{H}_2\text{O}
\]

The influence of process parameters on leaching rate of the amorphous silica was studied as shown in Fig. 6.

3.4.1 The leaching temperature

The experiments temperature range is 50–140°C, leaching time is 4.5 h, concentration of sodium hydroxide solution is 25 mass %, and mass ratio of fly ash/alkali is 1:0.5, respectively. The experimental results show that the leaching temperature has an important influence on the leaching rate of silica during alkali leaching process. With increasing of leaching temperature, the leaching rate of silica increased rapidly below 95°C and slowly above 95°C as shown in Fig. 6(a).

This is attributed to side reaction when the leaching temperature is higher than 130°C as Eq. (4), moreover, mullite will react to water vapor in the following reaction (5) at higher temperature, causing the mullite is damaged. Considering mullite and corundum almost do not react with sodium hydroxide when temperature is below 100°C, the reaction temperature of 95°C was adopted.

\[
6\text{H}_2\text{SiO}_4^- + 6\text{Al(OH)}_4^- + 8\text{Na}^+ \rightarrow \text{Na}_8\text{Al}_6\text{Si}_6\text{O}_{24}(\text{OH})_2(\text{H}_2\text{O})_2 + 10\text{OH}^- + 10\text{H}_2\text{O}
\]

\[
3\text{Al}_2\text{O}_3\cdot2\text{SiO}_2 + 4\text{H}_2\text{O} \rightarrow 3\text{Al}_2\text{O}_3 + 2\text{Si(OH)}_4
\]

3.4.2 The leaching time

The influence of leaching time on leaching rate of silica in the condition of 95°C, 25 mass % (concentration of sodium hydroxide solution) and 1:0.5 (mass ratio of fly ash/alkali) was shown in Fig. 6(b). As can be seen, the leaching rate of silica increased gradually with the increase of leaching time. After 3.5 h, the desilication rate increased slowly and the peak was presented at 4.5 h. This is because the side reaction leads to the extra consumption of amorphous silica, so the leaching time of 4.5 h was employed.

3.4.3 The mass ratio of fly ash/alkali

Experiments were conducted at 95°C, leaching time of 4.5 h and 25 mass % (concentration of sodium hydroxide solution), respectively. The leaching rate of silica increased gradually with a decrease of mass ratio of ash/alkali as shown in Fig. 6(c), when it was less than 1:0.5, the desilication rate almost did not increase, the best ash/alkali ratio appeared.

3.4.4 The concentration of sodium hydroxide solution

The influence of alkali concentration on leaching rate of silica in the condition of 95°C, reaction time of 4.5 h and 1:0.5 (mass ratio of fly ash/alkali) was shown in Fig. 6(d), the desilication rate increased with the increase of alkali concentration. After mass concentration of sodium hydroxide solution exceeded 20%, the change of desilication rate was not obvious, so the mass concentration of 25% was chosen.

The 40.4 mass % leaching rate of silica was presented with the optimal process of the alkali soluble desilication: the temperature was 95°C, the reaction time was 4.5 h, the mass concentration of sodium hydroxide solution was 25% and the mass ratio of ash/alkali was 1:0.5.

In summary, the optimum condition for feeding the extracted mullite were that, 30 mass % of CFA solution was passed through magnetic separator machine to remove magnetic iron in the condition of 13000 Gs of magnetic flux density with two times of magnetic separation processes. The residues with a mass ratio of 5:1 of liquid/solid were mixed with a measured 20 mass % of hydrochloric acid solution at 85°C for 3 h for further iron-removing. The as-prepared solid phase was further treated with 25 mass % of sodium hydroxide and the mass ratio of fly ash/alkali was 1:0.5 at 95°C for 4.5 h to remove amorphous silica. Herein, under the aforementioned optimum conditions, the leaching rate of iron and the leaching rate of silica were 85.9 and 40.4 mass % respectively.

3.5 Characterization results

3.5.1 The XRF result

Table 2 lists the chemical components of extracted mullite. As can be seen, after CFA was treated with magnetic separator, acid and alkali, the total content of iron is
below 0.3 mass %, the sum of Na$_2$O + K$_2$O reduced to less than 0.4 mass %, the content of SiO$_2$ from 37.21 mass % declined to 22.18 mass % while the content of Al$_2$O$_3$ from 51.88 mass % reached up to 69.13 mass % due to the dissolution of amorphous silicate glass. As a consequence, desilication rate reached up to 40.4 mass %, and the removal ratio of iron was 85.9 mass %. The weightlessness rate of extracted sample was about 48.2 mass %.

3.5.2 The XRD patterns of extracted mullite

Figure 7 demonstrates the XRD patterns of the extracted sample. Only diffraction peaks of mullite (PDF#73-1512) and corundum (PDF#10-0394) can be clearly observed. The broad characteristic diffraction peak between 18 and 25° in the XRD pattern disappeared completely. This is attributed to removing of amorphous silicate. The heavy background in the XRD pattern also disappeared due to the removal of iron oxide by using iron removal process. The diffraction intensity of mullite increased significantly. As a result, the content of mullite increased to 82 mass %, and the content of corundum was about 18 mass %.

Table 2. Chemical component of extracted samples (mass %)

| Sample   | Al$_2$O$_3$ | SiO$_2$ | TFe | CaO | TiO$_2$ | K$_2$O | Na$_2$O | MgO |
|----------|-------------|---------|-----|-----|---------|--------|---------|-----|
| Extracted mullite | 69.13 | 22.18 | 0.3 | 0.25 | 1.77 | 0.1 | 0.36 | 0.008 |

Fig. 6. Influence of (a) leaching temperature (b) reaction time (c) mass ratio of fly ash/alkali and (d) sodium hydroxide concentration on leaching rate of silica.

Fig. 7. XRD patterns and quantitative analysis result of extracted sample.
3.5.3 The SEM of extracted mullite

Figure 8 displays the SEM photographs of extracted sample. As shown in Fig. 8, there is a significant change in the microstructure of the extracted sample, the amorphous components particles on the surface of the spherical shape were decreased significantly. Meantime, the surface of fly ash particles became coarse and plenty of pristine mullite seeds appeared due to getting rid of amorphous glass. A number of anisotropic mullite microcrystals’ size ranged between 0.2 to 0.5 \( \mu \text{m} \) in diameter and the length of 3 \( \mu \text{m} \) with little glass phase were obtained.

3.5.4 The SEM-EDS result

Figure 9 demonstrates the SEM-EDS analysis result of the surface of extracted mullite. This result revealed that the mullite whiskers fabricated in the present research showed a slightly higher molar ratio of Al to Si (\( \approx 3.30 \)) than the theoretical value of 3.0, which was close to the molecular components of mullite.

3.5.5 The FTIR of extracted mullite

Figure 10 displays the FTIR spectra of original fly ash and extracted mullite. As shown in Fig. 10, the band located at 1081 cm\(^{-1}\) in CFA was attributed to the vibration of quartz which disappeared in the extracted mullite. Hydrothermal treatment led to the disappearance of glass phase on the fly ash particle surface as well as the raise of silicate ions in alkali solution. From the curve of the extracted mullite, the wide bands appearing at 445 and 732 cm\(^{-1}\) were due to the vibration of [SiO\(_4\)]\(^{4-}\) and [AlO\(_4\)]\(^{5-}\). At the same time, the band located at 559 cm\(^{-1}\) was attributed to the vibration of [AlO\(_6\)]\(^{6-}\). The components appearing at about 890 cm\(^{-1}\) were attributed to the vitreous phase of original fly ash while the presence of mullite was responsible for the characteristic band at 1172 cm\(^{-1}\).28}

Fig. 8. SEM morphology of extracted sample.

Fig. 9. SEM-EDS micrographs of extracted sample. Accordingly, the whiskers have the following chemical composition (mass%): 41.27 O, 44.73 Al, 14.0 Si.

Fig. 10. FTIR spectra of fly ash and extracted sample (I) Si–O, (II) Al6–O, (III) Al4–O, (IV) Al4–O, (V) Si–O.
3.6 Physical properties of mullite powder

The physical properties of the extracted mullite were showed in Table 3. As can be seen, the excellent physical properties of mullite presented refractoriness, whiteness, bulk density and average particle size values of about 1850°C, 92, 0.60 g/cm³ and 0.045–0.075 mm, respectively.

3.7 Technical indicators of sodium silicate

Table 4 displayed the technical indicators of sodium silicate. As shown in Table 4, industrial sodium silicate with a modulus of 2.6 was prepared, which reached top grade of liquid sodium silicate-3 of National standard in China.

4. Conclusion

In summary, the 82 mass% of mullite was extracted from CFA after it was treated with magnetic separator, 20 mass% of hydrochloric acid solution and 25 mass% of sodium hydroxide solution. The refractoriness, whiteness, bulk density and the average particle size of extracted mullite were 1850°C, 92, 0.60 g/cm³ and 0.045–0.075 mm, respectively. Furthermore, industrial sodium silicate with a modulus of 2.6 was prepared, which reached top grade of liquid sodium silicate-3 of National standard in China. This method provides a potential way on the utilization of CFA to prepare mullite.

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Table 3. Physical properties of mullite powder

| Sample                  | Refractoriness (°C) | Whiteness | Average Particle Size (mm) | Bulk Density (g/cm³) |
|-------------------------|---------------------|-----------|---------------------------|----------------------|
| Extracted mullite       | >1850               | 92        | 0.045–0.075               | 0.60                 |

Table 4. Technical indicators of sodium silicate

| Grade                | Fe (mass%) | Insolubles (mass%) | Density (g/cm³) | Na₂O (mass%) | SiO₂ (mass%) | The modulus |
|----------------------|------------|--------------------|-----------------|--------------|--------------|-------------|
| Sample               | ≤0.018     | ≤0.17              | 1.443           | ≥10.3        | ≥26.7        | 2.6         |
| top grade            | ≤0.02      | ≤0.20              | 1.436           | ≥10.2        | ≥25.7        | 2.6–2.9     |
| of liquid-3          |            |                    |                 |              |              |             |

0.17 1.443 ≥10.3 ≥26.7 2.6