



# Enhanced recycling and utilization of mullite from coal fly ash with a flotation and metallurgy process

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## A B S T R A C T

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Humic acid  
Flotation

The recycling and utilization of mullite in coal fly ash (CFA) gives rise to environmental and economic profits. The optimal experimental conditions for enhanced recycling of mullite with a flotation and metallurgy process are studied by factorial designs, and further analyzed by thermodynamic and dynamics calculations, and XRD/SEM characterizations. The unburned carbon is firstly removed from the CFA assisted with effective humic acid surfactant in the froth flotation step. The maximum removal rate of carbon is  $93.20 \pm 0.05\%$ . Then the digesting of glass phase in carbon-free product with mixed acid solutions (HF: H<sub>2</sub>SO<sub>4</sub> = 3:1) is employed in the metallurgy step. The influencing intensity of digesting factor on glass phase decreases in order of acid concentration, agitation intensity, reaction time, reaction temperature. The digesting kinetics of glass phase in mixed acid solution is belonging to diffusion-controlled reaction model. The maximum digesting rate of glass phase can be  $97.40 \pm 0.16\%$ . The mineralogical phase of the digested residue is mainly mullite. The performance of the refractory obtained from the water-cleaned mullite-enriched product meets the production requirements of refractory materials. This potential process would provide an effective approach for the separation and recycling of valuable components from CFA in further industry applications.

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## 1. Introduction

Solid wastes from industries that include mining, smelting operations or coal generating electricity contain various hazardous components, which are environmentally harmful when exposed without treatments (Li et al., 2017; Shibayama et al., 2010). Coal is one of the most important electricity sources in China, consuming about 3.96 billion tons of coal annually. Coal fly ash (CFA) has been widely generated during the firing process of coal in coal combustion power plants (Zyrkowski et al., 2016; Ahmaruzzaman, 2010). The CFA has become the largest displacement of solid waste. The annual amount of the CFA produced is approximately 600 million tons in 2016 (Benassi et al., 2016; Yao et al., 2015). The CFA emission load will further increase gradually with increasing demand for electric power. The hazardous CFA has occupied a vast amount of land and threatened the living environment of human

being because of the low utilization rate (Pandey and Singh, 2010; Lu et al., 2014; Tsang et al., 2014). Therefore, lots of efforts have been tried to find an effective and economical way for CFA recycling in China.

The CFA can be considered as a secondary resource for the recovery of valuable components (Blissett and Rowson, 2012). For the comprehensive utilization of CFA, many techniques such as flotation, stabilization/solidification approaches, hydrometallurgical/pyrometallurgical extraction and physical/chemical separation were developed to support other fields, such as iron and aluminum oxide separation, construction and building materials, catalysis and adsorbents for wastewater treating (Qi et al., 2015; Tennakoon et al., 2015; Franus et al., 2015; Visa and Chelaru, 2014; Li et al., 2014b; Olajire, 2013). The CFA generally consists of some unburned carbons and inorganic alumina and silicon minerals. The flotation recovering of unburned carbon from CFA had been studied by many researchers (Demir et al., 2008; Emre Altun et al., 2009; Zhou et al., 2017). Also, the recovering of alumina and silicon components from CFA had been widely investigated (Demir et al., 2008; Emre Altun et al., 2009; Zhou et al., 2017). The major methods of recovering these valuable components can be classified

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into acid leaching, alkali hydrothermal process, acid–alkali combination process, lime sintering, and desiccated lime–soda sintering processes (Li et al., 2014a; Yang et al., 2014; Yao et al., 2014; Komonweeraket et al., 2015; Guo et al., 2013). There are major reasons for the limitation of large-scale industrial applications for the above techniques, including high energy consumption, long process and secondary pollution (Cao et al., 2008; Hirajima et al., 2010). Thus, an alternative method for recycling and comprehensive utilization of CFA is desired and explored.

The CFA can also be considered as an important source of mullite. Mullite is a kind of structural ceramics material with a nominal composition of  $3\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2$  (Aksay et al., 1991). It has good thermal strength and thermal stress resistance owing to the low thermal expansion coefficient, low thermal conductivity and good dielectric behavior (Fischedick et al., 2017; Zhang et al., 2017). Natural mullite mineral is not very much. Mullite can be synthesized using many methods including spark plasma sintering, spray pyrolysis, chemical vapour deposition, sol–gel technique and hydrothermal synthesis (Xiao et al., 2017; Kool et al., 2015; Suriyanarayanan, N. et al., 2009). However, the high consumption of starting raw materials limits their further applications on commercial scale. In order to decrease the production cost, researchers have been devoted to preparing artificial mullite from low-cost kaolin, kyanite and fly ash (Pascual et al., 2000; Xu et al., 2017; Deutou et al., 2016). The utilization of CFA as starting resource material will not only obviously decrease the environment pollution, but also bring huge economic benefits. Thus, the study on the recovering of mullite from CFA is very necessary and critical.

A potential joint process for the recovering and utilization of mullite from coal fly ash is proposed by the authors, as a feasible approach to achieve nearly zero waste and recycling of the resources hidden in the fly ash. This study focuses on the enhanced recycling and utilization of mullite from coal fly ash with a potential flotation-metallurgy process. This work can be used as a simple, economic and effective method to produce refractory powder. That is, the purified mullite-enriched product can be used to fabricate high-quality structural refractory matters. The purpose of this work is to illustrate how the potential recovering technique can be used to design the process and determine the optimal operating conditions for the recycling of coal fly ash.

## 2. Experimental

### 2.1. Coal fly ash and chemical reagents

The CFA used in this study was collected from the Jungar Power Plant, Inner Mongolia, China. The CFA was obtained from pulverized coal combustion process, which had been burnt at  $1500^\circ\text{C}$ . The specific surface area of CFA was measured by the Brunauer–Emmett–Teller (BET) method using mode 3H-2000PS2 instrument, which is  $0.3237 \pm 0.0135 \text{ m}^2 \text{ g}^{-1}$ . The chemical composition of CFA was analyzed through X Ray Fluorescence (XRF) method. As shown in Table 1, the fly ash is significantly enriched in  $\text{Al}_2\text{O}_3$  of  $52.65 \pm 0.31\%$  and  $\text{SiO}_2$  of  $38.17 \pm 0.25\%$ , but poor in  $\text{Fe}_2\text{O}_3$ ,  $\text{TiO}_2$  and  $\text{CaO}$ .

The minerals in fly ash were quantified by powder X-ray diffraction (Bruker D8 Discover, Germany), and the XRD pattern of

CFA is depicted in Fig. 1. The results demonstrate that the main crystallized mineral phases in fly ash mainly consist of mullite, and bits of  $\text{Al}_2\text{O}_3$  and  $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$ . The results also show that the sample have a broad reflection peak at approximately  $23^\circ$ , which exhibits that the glass phase and the unburned carbon are amorphous. The mineral composition, glass and loss on ignition of the CFA can be found from Table 2. Combined with analysis of coal proximate analyzer, the content of unburned carbon is  $18.54 \pm 0.08\%$ .

The SEM images of CFA are given in Fig. 2. The main constituent for porous particles is carbon. The EDS analysis of bright spheres indicates the basic elements are Si and Al.

Emulsified kerosene and *n*-butyl alcohol (obtained from Sino-pharm chemical reagent Co., Ltd., China) were used as collecting reagents and frothing reagent, respectively. Humic acid was used as surfactant, which was isolated from a typical kind of Chinese lignite in the lab (Huang et al., 2016b). Emulsified kerosene and humic acid were dispersed by ultrasound prior to its usage.

Hydrofluoric acid (23.0 mol/L) and concentrated sulfuric acid (18.4 mol/L) were employed as digesting reagents, which were obtained from Tianjin Aokatet Chemical Co., Ltd., China. Other reagents were of analytical grade, and deionized water was used as well.

### 2.2. Enhanced recycling process of coal fly ash

The schematic diagram of enhanced recycling process of CFA is given in Fig. 3.

#### 2.2.1. Flotation process

The carbon flotation removing were performed in a flotation apparatus under conditions similar to the procedure outlined by Yanfang Huang., et al. (Huang et al., 2016b). The schematic diagram of carbon flotation removing process is given in Fig. 4. Emulsified kerosene, *n*-butyl alcohol and modified humic acid were used as collector, frother and surfactant, respectively. In a typical process, 50 g CFA sample was introduced into a 1 L plastic container with

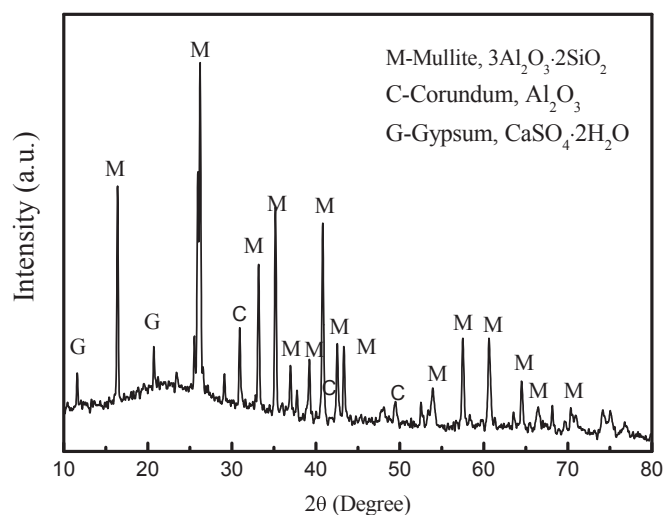


Fig. 1. The XRD pattern of raw CFA.

Table 1

Chemical composition of the raw CFA (wt.%).

Composition	$\text{Al}_2\text{O}_3$	$\text{SiO}_2$	$\text{Fe}_2\text{O}_3$	$\text{CaO}$	$\text{MgO}$	$\text{TiO}_2$
Content	$52.65 \pm 0.31$	$38.17 \pm 0.25$	$1.25 \pm 0.11$	$2.49 \pm 0.13$	$0.34 \pm 0.04$	$1.86 \pm 0.07$

Note: Values are the means and standard deviations.

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