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Comparison of leaching behaviors of aluminum in ash from combustion and catalytic gasification

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Abstract: The leaching behaviors of aluminum in ash from combustion and catalytic gasification, and effects of Na_2CO_3 addition (0–15%), temperature (600–1000°C) on Al leaching behaviors and mineral composition were investigated. The compositions of ash from combustion and gasification, and acid leaching residue were investigated by XRD. The results show that the combustion ash is predominantly composed of mullite, while that from catalytic gasification is sodium aluminum silicate ($(Na_2O)_{0.33}NaAlSiO_4$). The Al extraction yield of combustion ash only reaches 40% at leaching conditions of 6 mol/L H_2SO_4, 60°C and 30 min, while that of catalytic gasification ash with 10% Na_2CO_3 addition can reach 88%. The catalytic gasification with Na_2CO_3 addition can achieve higher Al extraction yield.

Key words: catalytic gasification; coal ash; alumina extraction

Coal ash, acting as one of the products of high temperature coal conversion, is produced by fragment, concreting, homogeneous and heterogeneous deposition of the minerals in raw coal. Al and Si containing minerals in raw coal can decompose to form inert mullite, sillimanite, corundum, quartz and amorphous Si-Al matters[1]. The content of Al_2O_3 and SiO_2 in coal ash can reach above 80%, and can be potential substitute of other Si and Al resource. Storage in pond is one of the ways to deal with coal ash, which costs lots of land and poses potential hazards to the environment. Meanwhile, the harmful elements, such as Cr, As and Pb, can be leaked by rainfalls and contaminate soil and groundwater[2], subsequently do harm to human's health. Therefore, re-utilization of coal ash is meaningful.

There are many ways for utilization of coal ash, such as being roadbed of highway, backfill waste of mine, preparation of cement, recovery of alumina[3], preparation of molecular sieve[4] and silica aerogel[5]. Alumina in high-alumina coal ash can reach over 40%, so extraction alumina is one of the effective ways for utilization of high-alumina coal ash. Extracting Al from coal ash collected from power plant has been comprehensively investigated by alkali hydrothermal method[6], direct acid leaching[7], roasting with soda lime[8], acid leaching after roast[9], two steps alkali leaching[10] and roasting with ammonium[11]. The results show that high recovery of Al is not achieved by direct acid leaching and alkali hydrothermal method. The inert mullite and sillimanite have high degree polymerization of Si-Al mesh structure which is not destroyed by acid and alkali at ambient temperature, resulting in low recovery of Al from coal ash. Roasting method is as follows: mixing coal ash with roasting activator and reacting at high temperature, and then leached by dilute acid, dilute alkali and water. After that, precipitation, filtration and carbonization are used to extract Al from coal ash. During roasting, mullite and sillimanite in ash react with Na^+, K^+, Ca^{2+} and NH_4^+, achieving high Al recovery. When Na_2CO_3 and NaOH are used as roasting activators, the activating products are acid-dissolved aluminosilicate[12] and nepheline[13]. However, the most excellent performance of catalysts are Na and K in catalytic gasification[14,15]. Ding et al[16] and Wang et al[17] found that the added Na and K can react with minerals in coal to form deactivated aluminosilicate and nepheline which are the activating products of roasting method to extract Al from coal ash. Therefore, it is meaningful to use catalytic gasification ash to extract Al, which is a novel way and can lower energy consumption of roasting process. Extracting Al from coal ash usually focuses on the ash collected from coal power plant, but the research about extraction Al from gasification ash and comparison between them are rare.

Table 1   Proximate and ultimate analyses of SJH coal

Sample	Proximate analysis w_{ad}/%				Ultimate analysis w_{daf}/%				
	V	FC	A	M	C	H	N	S	O*
SJH coal	29.6	51.2	16.9	2.3	78.8	4.9	1.5	0.8	14

*: by difference

Table 2   Ash compositions of SJH coal

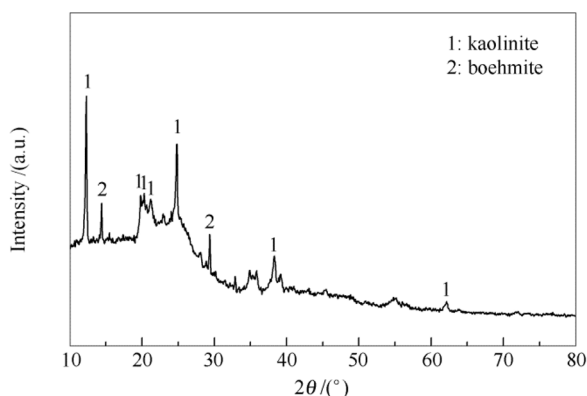| Composition w/%									
Al_2O_3	SiO_2	Fe_2O_3	TiO_2	CaO	MgO	Na_2O	SO_3	K_2O	P_2O_5
46.34	36.29	6.38	3.57	2.61	2.18	0.76	0.45	0.42	0.03

Fig. 1   XRD pattern of SJH coal

In this work, Sunjiahao high-alumina coal (SJH) was chosen as the sample, tubular furnace was used to prepare ash from combustion and catalytic gasification, and XRD was employed to investigate composition of the ashes. H_2SO_4 and NaOH were used as leaching medium, and ICP-OES was used to measure the leaching yield of Al.

1   Experimental

1.1   Preparation of ash from combustion and catalytic gasification

The SJH coal was ground and sieved to 120 standard mesh, and then dried in an oven at 110°C for 6 h. The proximate and ultimate analyses, and ash composition of SJH coal are listed in Tables 1 and 2. The alumina in coal ash is relatively high and reaches 46.34%. Figure 1 shows XRD spectrum of SJH coal, and the main minerals are kaolinite ($Al_2O_3 \cdot 2SiO_2 \cdot 2H_2O$) and boehmite (AlOOH) which contributes the high alumina content in coal ash.

High temperature tubular furnace was employed to prepare ash samples. The preparation of combustion ash was as follows: 2 g coal was evenly spread in a corundum boat which was pushed to flat-temperature zone of the furnace with continuous air flow of 200 mL/min. The furnace was heated at 10°C/min to 1000°C and kept 5 min to obtain the ash which was then ground and sieved to 120 standard meshes. The procedure of loading Na_2CO_3 on coal samples was as follows: a certain amount of Na_2CO_3 was completely dissolved in 50 mL deionized water to form Na_2CO_3 aqueous solution, and then 10 g coal was added and the mixture was kept at 60°C under stirring until it changed into viscous slurry which was then dried in an oven at 105°C for 5 h. The Na_2CO_3 loaded coal was ground and sieved to 120 standard meshes. The preparation of catalytic gasification ash was as follows: 2 g Na_2CO_3 loaded coal was evenly spread in a corundum boat, and then the furnace was heated to the desired temperature at 10°C/min and the boat was pushed to the flat-temperature zone of the furnace with continuous CO_2 flow of 200 mL/min. After 2 h, the boat was dragged to low temperature zone with N_2 purging. The obtained catalytic gasification ash was ground and sieved to 120 standard meshes.

1.2   Extraction Al from coal ash

H_2SO_4 and NaOH were used as leaching medium. The leaching procedure was as follows: suction pipet was used to quantify 10 mL H_2SO_4 to a cuvette, and a magnetic paddle was added to the cuvette fixed in a thermostat water bath. Then 0.5 g coal ash was added to the cuvette and leached for 30 min, then the hot solution was filtered, and washed by hot deionized water. The leaching solution and washing liquid were collected to a 100 mL volumetric flask and precisely set to 100 mL, and ICP-OES was employed to measure Al content in liquid. The filter cake was dried at 110°C and ground to obtain acid leaching residue. The alkali leaching procedure was the same as acid leaching except for changing the leaching medium to NaOH liquid.

1.3   Characterization means

An X-ray diffraction analyzer (D2, Bruker) using Cu $K\alpha$ radiation (λ = 0.15406 nm) was adopted to analyze the coal ash. The accelerate voltage of 40 kV, a tube current of 40 mA, and a step size of 4(°)/min between 2θ = 10° and 80° were used.

2   Results and discussion