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Extracting Alumina from Coal Fly Ash with Ammonium Sulfate Sintering Process

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Abstract

The whole process of extracting alumina from coal fly ash was trailed by experimentation. Alumina was prepared with sintering, leaching, separating, precipitation with ammonia water and calcination units. The effects of the sintering temperature and the sintering time, molar ratio of ammonium sulfate and alumina in fly ash and the size of fly ash on the extracting efficiency of alumina in fly ash were studied. The extracting efficiency of alumina can reach 95% under the optimum conditions.

Introduction

Coal-burning power plants consume pulverized solid fuels and produce large amount of coal fly ash (CFA) as a residue [1]. Through chemical analysis, the main compositions of CFA are alumina and silica. Metallurgical alumina is the raw material of aluminum; its value is much higher than concrete. The use of CFA as a feedstock to make alumina is one potential application. Alumina has been produced using the limestone sintering process [2–5], soda-lime sintering process [6,7], leaching by acid with ammonium fluoride process[8,9]. A number of problems in these processes existed, such as the volume of residue caused from the sintering processes is very large—normally 8 to 10 times to initial CFA, it would cause the secondary pollution; HF caused from adding ammonium fluoride process is harmful to workers and local resident, etc.

CFA sourced from Inner Mongolia and processed using ammonium sulfate in this paper. The advantages compared with the limestone sintering process and soda-lime sintering process, this process causes fewer residues, and the use of the residue is more. And the advantages compared with the acid leaching process, ammonium sulfate and ammonium aluminum sulfate are much less corrosion than acid. The residue of limestone or soda-lime sintering processes is calcium hydrate garnet, which can only be used in cement industry. The main composition of the residue of the process of this paper is silica, which can be used to make white carbon black and other chemical products. The output of metallurgical alumina was 28.96Mt in 2010 in China, it was 34% of the global alumina output. The experiments of fly ash and ammonium sulfate mixture compounding sintering unit and sinter leaching unit of the process were finished and the optimum conditions were carried out in this paper.

1 Experimental

1.1 Materials and Reagents

The main composition of CFA is given in Table 1.

Table 1 Composition of the fly ash of somewhere in Inner Mongolia province

Item	Al_2O_3	SiO_2	Fe_2O_3	CaO	TiO_2	MgO	MnO	others
Content (%, ω)	48.20	42.49	1.89	3.31	1.30	0.20	0.01	2.60

The water in the experiments is redistilled water. The reagents are analytical.

2.2 Experimental and Analytical Instruments

A box resistance furnace was used in the sintering experiments, and the 4-mouth flasks and a heating jacket(2NHW-1000 ml, precision is ±1℃) were used in sinter leaching experiments. The phase composition and microstructure were observed by XRD (D/max-2500PC, Rigaku, Japan) and SEM(SSX-550, Shimadzu, Japan). Thermogravimetry and weight loss were observed by SDT 2960 Simultaneous DSC–TGA (TA Instruments, USA).

2.3 Methodology

The mixture, which had been prepared with CFA and ammonium sulfate by the planet mill, was sintered by the box resistance furnace. Ammonium aluminum sulfate and ammonia were generated. The ammonia was absorbed by water. The sinter and hot water were placed into the 4-mouth flask and heated with the heating jacket and agitated for leaching ammonium aluminum sulfate. The ammonium aluminum sulfate solution was separated from the residue with filtration. Aluminum hydroxide was precipitated from the solution by adding ammonia water which was from sintering unit.

The optimum conditions of sintering unit were defined through single factor and orthogonal experiments.

The CFA, mid-production (ammonium aluminum sulfate) and alumina were identified by XRD.

The main compositions, including alumina, silicon oxide, ferric oxide and etc, of the CFA and the residue of acid leaching were detected with XRF machine.

The Al and Fe content of solutions were detected with chemical analysis methods[10].

2 Results and Discussion

The alumina extracted rate can be stabilized about 95% under the optimum conditions. Alumina could be gotten by leaching the sinter, precipitating the leaching solution and calcinating the aluminum hydroxide. The Figure 1 shows the XRD spectra of the CFA. Figure 2 shows the XRD spectra of the products. The results showed that the product of sintering the mixture of CFA and ammonium sulfate was ammonium aluminum sulfate, and the product was α-Al_2O_3, which was calcinating the precipitation of adding ammonia water to the ammonium aluminum sulfate solution at 1200 degree C.

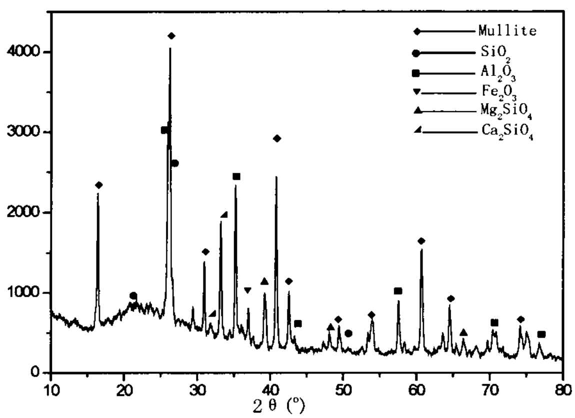

Fig.1 XRD spectrum of the fly ash

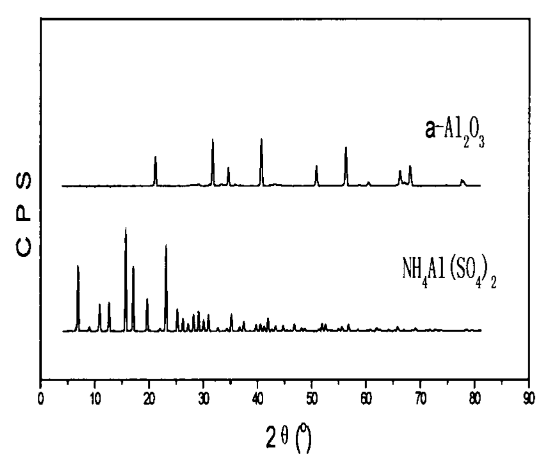

Fig.2 XRD spectrum of products

2.1 Influence of the size of fly ash on Alumina Extracted

The relation between the size of fly ash and the alumina extracted rate is shown in Fig.3, with the rate dependant on the contact area of fly ash and ammonium sulfate. It showed that the size smaller the extracted rate higher.

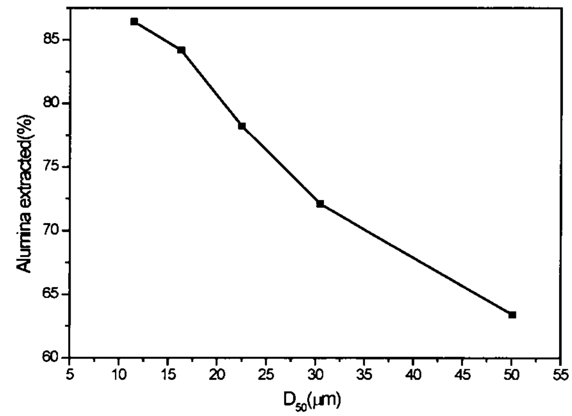

Fig.3 Influence of the size of fly ash on Alumina extracted

2.2 Influence of the sintering temperature on Alumina Extracted

The ammonia starts to be released from ammonium sulfate at 280℃. The ammonium aluminum sulfate starts to break down to aluminum sulfate and ammonia at 500℃. So the sintering temperature should be controlled between 280℃ to 500℃. The relation of temperature and extracted rate is shown in Fig. 4, with the temperature higher the extracted rate higher and the curve is flatting after 400℃. The result showed that it needed enough energy to break the link of Al-Si-O and the diffusion rate was affected a lot by the temperature.

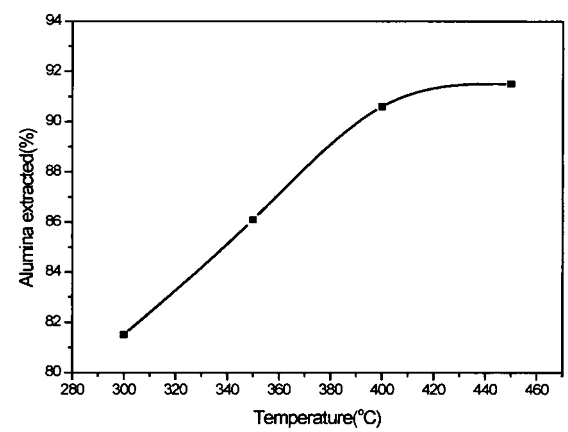

Fig.4 Influence of the sintering temperature on Alumina extracted

2.3 Influence of the sintering time on Alumina Extracted

The relation of sintering time and extraction rate is shown in Fig. 5, with the time longer the extracted rate higher and the curve is flatting after 2.5h.

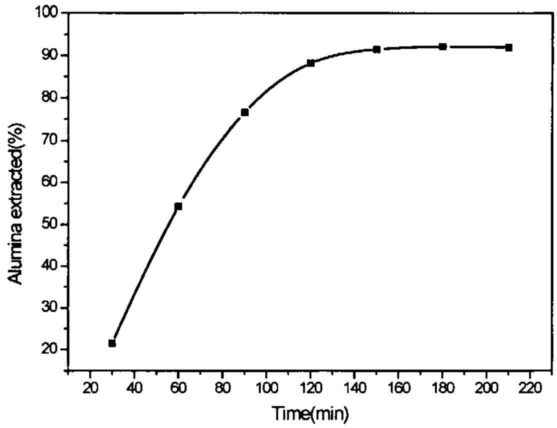

Fig.5 Influence of the sintering time on Alumina extracted

2.4 Influence of the molar ratio of ammonium sulfate and alumina in CFA on Alumina Extracted

According to thermodynamic calculation, the main reaction of sintering the mixture of CFA and ammonium sulfate at 400℃ is

$4(NH_4)_2SO_4(s)+Al_2O_3(s)=2NH_4Al(SO_4)_2(s)+6NH_3(g)+3H_2O(g)$.

The molar ratio is 4 in theory. The reaction is solid phase reaction, so it needs more ammonium sulfate to increase the extracted rate. The Figure 6 is the relation of the molar ratio and the alumina extracted rate. It showed that the molar ratio higher the extracted rate higher and the curve is flatting after the molar ratio is 7. The overdosed ammonium sulfate decomposes into ammonia and ammonium bisulfate.

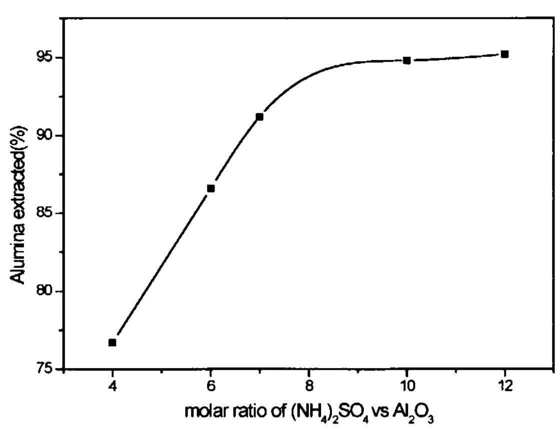

Fig.6 Influence of the molar ratio of ammonium sulfate and alumina on Alumina extracted

2.5 Confirmatory Experiments

The confirmatory experiments were at these conditions, the molar ratio 7, the sintering temperature 400℃, the sintering time 2.5 hours and the CFA size D50 ≤ 10 μ m. The alumina extracted rate was about 95%. Ammonia water was added into the solution that was separated from the slurry that had leached the sinter, and adjusted the pH=3.5~5.2 to precipitate aluminum hydroxide. The ammonium sulfate solution was recycled after evaporation.

3 Conclusions

(1) A new process is presented to extract alumina from coal fly ash. The raw materials are CFA and ammonium sulfate.

(2)Alumina had been prepared with sintering the mixture of ammonium sulfate and CFA, ammonium sulfate can be recycled.

(3) The alumina extracted rate can be stabilized about 95% at the conditions of the molar ratio 7, the sintering temperature 400℃, the sintering time 2.5 hours and the CFA size D50≤10 μ m.

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