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Original Research Paper

Synthesis of alumina with coarse particle by precipitating aluminum ammonium sulfate solution with ammonia

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ABSTRACT

The uniformity particle size distribution (PSD) and coarse particle size of alumina were synthesized by precipitating aluminum ammonium sulfate solution with ammonia. The effect of temperature and ammonia concentration on precipitation products was studied by using X-ray diffraction (XRD), field emission scanning electron microscopy (SEM), laser particle size analyze and fourier transform infrared (FTIR) spectroscopy. The results show that the particle size distribution of the precipitations is greatly affected by temperature and ammonia concentration. The precipitation product belongs to boehmite and its particle size has little variation after calcining. The optimal precipitation condition is 7.0 mol L^{-1} ammonia at 90 °C. Under the optimal precipitation condition, the D10, D50, and D90 of the precipitation particles are 12.54 μm, 41.28 μm, and 78.19 μm, respectively.

1. Introduction

Alumina has been widely used as high performance raw materials in the fields of electron, chemical engineering, ceramic, petroleum, and so on. Meanwhile, it is also the main raw material in electrolytic aluminum industry [1]. Smelter Grade Alumina (SGA) has been mainly produced by using bauxite ores as raw in Bayer process or the sintering process. Bayer process is basically used for the extraction of alumina from bauxite ores with mass ratio of alumina to silica (A/S) above 9, and sintering process is widely used to process the poor-grade diasporic bauxite ores with A/S below 7 [2].

Coal fly ash is the major coal solid waste, beside coal waste and coal slime. Due to low utilization rate, coal fly ash is generally dumped in lands and poses a serious threat to the environment. Efficient and safe disposal of coal fly ash is of environmental concern in the whole world [3–7]. Coal fly ash contains about 10–55 wt.% of Al_2O_3 and is a potential substitute of bauxite for alumina production [8–12]. Therefore, extracting of aluminum from coal fly ash is environmentally and scientifically significant for disposing and utilizing waste materials and exploring new aluminum source, and has attracted extensive attention recently [13–15].

Recently, a new process for extracting alumina from coal fly ash has been established and it contains basically four steps [16,17]. The first step corresponds to sintering fly ash with ammonium sulfate. Secondly, the sintered fly ash is leached in the hot water. In the third step, the insoluble residues are separated. Afterwards, the dissolved aluminum in the solution is mainly precipitated in the form of boehmite (AlOOH) by putting ammonia water into leaching liquor containing 60∼85 g L^{-1} ammonium aluminum sulfate. The final step is the calcination of the precipitate to produce alumina. Compared with acidic, alkali and acid-alkali methods, this process results in less residue and ammonium sulfate as a raw material with much less corrosive. In addition, the residue left in this process is mainly silica-rich by-product that can be used as a resource for white carbon black and other chemical production. The lowest energy consumption is also the highlighting advantages of the new process. The average energy consumption calculated based on the experimental results for acidic method, alkali method, acid-alkali method and the new technology one presented here is 28.0 GJ/t- Al_2O_3, 46.6 GJ/t- Al_2O_3, 50 GJ/t- Al_2O_3, and 26.5 GJ/t-Al_2O_3, respectively. Precipitation of leaching liquor with ammonia is the one of key steps in this process, which significantly affects the economic efficiency and circulation efficiency.

Ammonium aluminum sulfate (Alum) is a potential starting material for preparation of high purity reactive alumina. Alum is commonly synthesized by cooling crystallization. The decomposition of alum and transformation of resultant product to fine or nano-particles alumina have been reported in the literatures [18–22]. Very few references had mentioned "synthesis of alumina particles (including nanoparticles) from aluminum sulfate aqueous

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solutions". H.C. Park et al. had prepared high purity alum by reaction of Ammonium aluminum sulfate derived from coal fly ash and finer α-Al_2O_3 with Particle size at 0.1–0.5 μm obtained by calcining alum [18]. A.F. Zhang et al. had prepared nano alumina powder with spherical shape and particle size at 5–6 nm by precipitation ammonium aluminum sulfate with ammonium carbonate [22]. However, the literatures about synthesizing alumina by precipitating aluminum ammonium sulfate solution with ammonia have not been found. As raw in electrolytic aluminium industry, the alumina should have uniformity particle size distribution (PSD) and coarse particle size.

Motivated by the analysis above, the purpose underlying the present work is to explore crystallizing rule of aluminum hydroxide from aluminum ammonium sulfate solution and to produce alumina with coarse particle size. Hence the influences of reaction temperature and ammonia concentration have been investigated, and the precipitation product has also been characterized.

2. Experimental

Ammonia and all the other chemicals used in this study were of analytical grade and purchased from National Pharmaceutical Group, China. The 150 ml solution of containing 60 g L^{-1} aluminum ammonium sulfate was prepared by dissolve ammonium alum in hot distilled water. In a typical experiment condition, ammonia was pumped into 90 °C aluminum ammonium sulfate solution at a rate of 10 ml min^{-1} with stirring speed of 250 rpm until the pH value of the solution reach 6.8. The products were collected by vacuum filtration, followed by washing with hot distilled water for four times, and then dried in static air at 110 °C for 24 h. Calcination was carried out by increasing temperature from room temperature to 1100 °C (i.e., 15 °C min^{-1} ramping rate), and heating at 1100 °C for 2 h in air.

Structural phase analysis of the sample was performed by an XRD-7000 equipped with Cu $K\alpha$ radiation. A continuous mode was used to collect 2θ data from 10° to 70° with a sampling pitch of 0.02° s^{-1}. FTIR spectrum was collected by a Bruker IF S66V spectrometer in a frequency range of 4000–450 cm^{-1} at the frequency step size of 4 cm^{-1}. The morphology of the sample was studied with a Field Emission Scanning Electron Microscope (SEM, SU8010) operated at an accelerating voltage of 20.0 kV. Particle size distribution (PSD) was measured by Malvern Mastersizer 2000. D_{10}, D_{50} and D_{90} correspond to the particle size of the sample whose cumulative particle distribution reaches 10%, 50%, and 90%, respectively.

3. Results and discussion

The PSD curve and some characteristic parameters of the precipitations under different temperatures are shown in Fig. 1 and summarized in Table 1. In Fig. 1, the curve of the PSD moves to right along with the increase of temperature from 60 °C to 90 °C, which indicates the particle size becomes coarse with increasing temperature. The variations of D_{10}, D_{50} and D_{90} in Table 1 are also the same trend, and the D_{10}, D_{50} and D_{90} of the product increase 7.02 μm, 18.54 μm and 27.68 μm from 60 °C to 90 °C, respectively. Precipitation is mainly comprises of nucleation, crystal growth and agglomeration process. The size enlargement appeared in crystal growth and agglomeration process. At higher reaction temperatures, smaller number of nuclei formed and crystals grow up more rapidly. Increasing the precipitation temperature will enhance the agglomeration and effectively restrain the second nucleation process. As a result, the product with coarse particle size distribution was formed. At the lower temperature, fine product was formed due to the predominance of nucleation. Therefore, it is important to produce coarser alumina by increasing temperature.

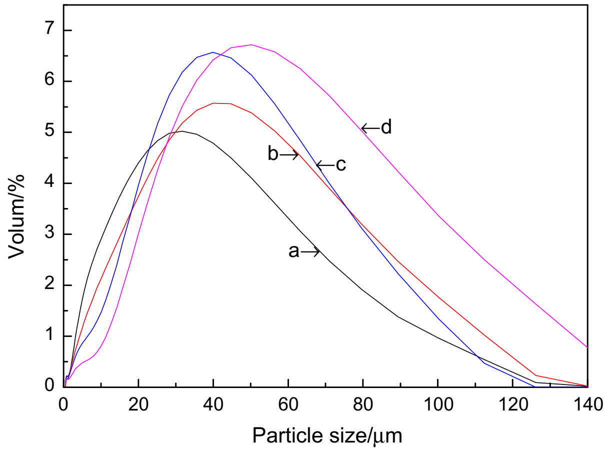

Fig. 1. Effect of temperatures on the precipitation PSD with 7.0 mol L^{-1} ammonia: (a) 60 °C, (b) 70 °C, (c) 80 °C, (d) 90 °C.

Table 1
Characteristic parameters of the precipitation PSD with different temperature.

Temperature (°C)	D_{10} (μm)	D_{50} (μm)	D_{90} (μm)
60	4.67	20.97	57.24
70	5.44	27.78	65.08
80	6.43	30.11	68.26
90	11.69	39.51	84.92

Fig. 2 and Table 2 show the PSD and some characteristic parameters of precipitations obtained with different ammonia concentration at 90 °C. Compared with Fig. 1 and Table 1, and it is found that influence of ammonia concentration on PSD is different from that of temperature. The curve of the precipitation PSD firstly moves to right and then moves to left with ammonia concentrations increasing from 3.5 mol L^{-1} to 14 mol L^{-1}. Crystal growth or agglomeration needs a suitable amount of aluminium hydroxide decomposed from aluminum ammonium sulfate solution to get coarse particles. The lower concentration of ammonia putted into the solution produces a smaller supersaturating degree in around. The decomposed aluminum hydroxide was less and resulted in the slower crystal growth rate and the lower agglomeration efficiency. At higher concentration, the supersaturating degree in around

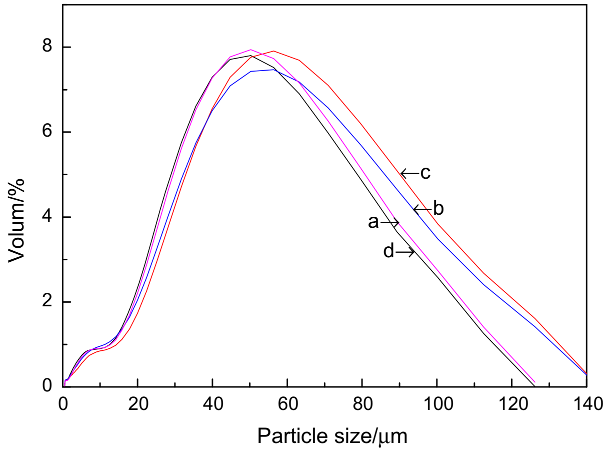

Fig. 2. Effect of ammonia concentration on PSD of the precipitations at 90 °C: (a) 3.5 mol L^{-1}, (b) 4.7 mol L^{-1}, (c) 7.0 mol L^{-1}, (d) 14.0 mol L^{-1}.