Kinetic and Thermodynamic Study of Aluminum Recovery from the Aluminum Smelter Baghouse Dust

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Abstract The recovery and utilization of metals from wastes are important for saving metal resources, as well as for protecting the environment. There are various types of solid wastes that have different origins and forms. Elemental and mineralogical composition of the wastes depends on the industry where they come from; therefore, the recovery processes for these materials are widely different. In general, the recovery of metals from ores and/or concentrates is carried out by extractive metallurgy methods comprising pyrometallurgy, hydrometallurgy, and electrometallurgy. In this research, sodium carbonate and sodium hydroxide leachings with different experimental conditions are tested for the aluminum dissolution. Also, a kinetic and thermodynamic study of sodium hydroxide leaching of the aluminum smelter baghouse dust is conducted. It is determined that when the solids content is 10 g/L, the overall reaction rate is controlled by pore diffusion and the activation energy is calculated to be about 4.8 kcal/mol. At temperatures below 100 °C, the stable form of the recovered compound after leaching is sodium aluminate.

Keywords Leaching · Hydrometallurgy · Waste recycling · Aluminum

Introduction

In metal production plants employing high temperatures, dust collection systems are often installed to control the emission of air pollutants from their off-gases. The air pollutants include particulate matter and harmful gases. In general, particulate matter which is collected in the dust collection systems may contain some amount of metals. The elemental and mineralogical composition of the dusts depends on the material being processed. Disposal of the particulate matter causes environmental problems and requires disposal fees. For these reasons, recycling metals from the dust can be valuable by reducing the environmental issues and saving the metal resources.

In this research, hydrometallurgical processes are studied for the recovery of aluminum from aluminum smelter baghouse dust since hydrometallurgical processes have several advantages over pyrometallurgical processes. In terms of ore types, a hydrometallurgical process is suitable for handling complex ores, low-grade ores, and secondary resources due to its selective leaching characteristics. On the other hand, a pyrometallurgical process is suitable to treat high-grade ores and rapid reaction can be accomplished due to the high temperatures involved [1]. Unit processes in hydrometallurgy generally consist of sample preparation, leaching, solid–liquid separation, concentration, and recovery. This research mainly focuses on the kinetic and thermodynamic study of aluminum leaching from aluminum smelter baghouse dust.

Experimental

In hydrometallurgical processes, particle size generally affects the overall processing rates. When the size of the particle decreases, the surface area exposed by the solvent

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greatly increases and it provides greater opportunity for the samples to be attacked by the solvent [2]. For this reason, size classification of the as-received samples is conducted before the experiments. Three different sizes of the metal sieves (45, 150, and 250 μm) are used, and samples with particle size smaller than 45 μm are considered for the hydrometallurgical materials processing in this research.

After size classification, an initial analysis of aluminum smelter baghouse dust is conducted. Samples with particle size smaller than 45 μm are fused with a lithium borate mixture (40 wt.% lithium tetraborate, 60 wt.% lithium metaborate) at 1000 °C. After the borate fusion process, the melt is dissolved in the nitric acid for ICP-AES analysis. This fusion process has several advantages over the other methods. First, minor refractory minerals are readily dissolved which would not be adequately attacked by hydrofluoric acid mixtures. Second, no pressure vessels are needed to perform this fusion process. Third, a clear aqueous solution is obtained that is suitable for a variety of analytical procedures [3]. In addition to the ICP-AES analysis, loss on ignition (LOI) of the samples with particle size smaller than 45 μm is also measured based on the ASTM D7348 [4].

For the effect of reagent and bath temperature tests, 1 M of NaOH and 1 M of Na_2CO_3 are used for aluminum smelter baghouse dust leaching. During leaching, solids content is fixed at 10 g/L and the bath temperature is changed from 45 to 60 °C during 4 h of leaching. For the kinetic study of 1 M NaOH leaching, the effect of the solids content on aluminum smelter baghouse dust leaching is tested with 10 and 50 g/L solids content. A magnetic stir bar is also used to mix the solution during leaching and the rotational speed is fixed at 250 rpm. After leaching, the solid residue is separated from the leachate by vacuum filtering and the leachate is analyzed by the ICP-AES

Results and Discussion

Initial Analysis of As-Received Aluminum Smelter Baghouse Dust

In general, the distribution of elements and the processing rate are greatly related to the mean particle size. As extraction of metals is related by their size, the size classification of aluminum smelter baghouse dust is conducted with three different meshes of sieve and the result is shown in Table 1. In the aluminum smelter baghouse dust, more than 84 wt.% of sample is larger than 150 μm and 6 wt.% of the sample is smaller than 45 μm . Leaching or dissolution of metals from the sample is the first step of a hydrometallurgical process. For the alkaline leaching experiments, samples with particle size smaller than 45 μm are used since the smaller size of particles guarantee a

Table 1 Size classification of as-received aluminum smelter baghouse dust

Aluminum smelter baghouse dust	
Size (μm)	Weight percent (%)
<45	6.3
45–150	9.5
150–250	32.1
>250	52.1

larger surface area to be attacked by the reagents. Also, the non-conductive fine particles can affect the efficiency of the separation process by the formation of aggregates, reducing the stability of the process, and accumulating on the surface of the corona or electrostatic electrode [5]. Therefore, ICP-AES analysis of the sample with particle size smaller than 45 μm is conducted and the result is shown in Table 2. Major elements of the aluminum smelter baghouse dust are Al, Si, Fe, Ca, and Mg and the amount of aluminum in the dust is about 13 wt.%. Based on the LOI measurements, the weight loss due to the loss of moisture, carbon, sulfur, and so forth were about 37 wt.%.

Effect of Reagent and Bath Temperature on Aluminum Smelter Baghouse Dust Leaching

In general, different reagents and bath temperatures affect the overall leaching characteristics [1]. Therefore, the effect of reagent and bath temperature on aluminum smelter baghouse dust leaching is investigated with 1 M of NaOH and 1 M of Na_2CO_3 based on the aforementioned experimental conditions. The percentage of aluminum dissolution after 4 h of leaching is shown in Fig. 1. Based on the results, it is concluded that bath temperature affects the percentage of aluminum dissolution and the value is increased from about 26 to 32 wt.% for the 1 M Na_2CO_3 ; on the other hand, for the 1 M NaOH, the percentage of aluminum dissolution is increased from about 82 to 100 wt.%. Therefore, leaching with higher bath temperature shows better aluminum dissolution. Additionally, the difference of the percentage of aluminum dissolution between 1 M NaOH and 1 M Na_2CO_3 at same bath temperature is about 56 wt.% at 45 °C and 68 wt.% at 60 °C. Thus, sodium hydroxide could be a better reagent for aluminum smelter baghouse dust leaching than sodium carbonate in terms of aluminum dissolution.

Effect of Solids Content on Aluminum Smelter Baghouse Dust Leaching With 1 M NaOH

From the previous results, aluminum leaching efficiency with sodium carbonate is only about 32 wt.% at 60 °C.

Table 2 Initial analysis of the aluminum smelter baghouse dust with particle size smaller than 45 μm

Elements	Al	Ca	Fe	K	Mg	Si	Zn	Ti	LOI
Weight percent (%)	13.0	2.3	2.5	1.2	1.0	5.2	0.4	0.2	36.6

Fig. 1 Effect of reagent and bath temperature on aluminum smelter baghouse dust leaching

Therefore, sodium carbonate is not a proper reagent for the aluminum smelter baghouse dust leaching due to poor aluminum dissolution. Thus, this study mainly focuses on the aluminum smelter baghouse dust leaching with sodium hydroxide. Sodium hydroxide is also used in the Bayer process for Al_2O_3 production from bauxite. In the Bayer process, after separating the insoluble red mud, which is a residue of the Bayer process, the pure solution is then seeded to precipitate crystalline aluminum hydroxide

which is subsequently filtered, washed, dried, and calcined to produce Al_2O_3 [6]. Since the percentage of aluminum dissolution with sodium hydroxide is about 80 wt.% at 45 °C after 4 h leaching, the effect of solids content on aluminum smelter baghouse dust leaching is tested with 1 M NaOH and the results are shown in Fig. 2. From the results, it can be concluded that a higher leaching temperature and longer leaching time guarantee better aluminum dissolution under both 10 and 50 g/L conditions. When the leaching time is longer than 5 min, the percentage of aluminum dissolution is changed from about 73 to 81 wt.% with 10 g/L solids content at 60 °C during 5 min of leaching. However, the difference of the percentage of aluminum dissolution during 5 min of leaching at the beginning of the reaction is about 73 wt.% with the same experimental condition. Thus, the reaction rate is relatively fast at the beginning of the reaction and it gradually decreases with time. Additionally, 1 M NaOH leaching with 10 g/L solids content shows relatively higher aluminum dissolution than that of 50 g/L.

Kinetic Study of the NaOH Leaching of Aluminum Smelter Baghouse Dust

A kinetic study of the NaOH leaching experiments of aluminum smelter baghouse dust is conducted with the shrinking core model. For the fluid particle reactions of spherical particles, two simple idealized models have been proposed: the progressive conversion model and the

Fig. 2 Effect of bath temperature and solids content on aluminum smelter baghouse dust leaching with time (1 M NaOH, 250 rpm). **a** 10 g/L solids content, **b** 50 g/L solids content

shrinking core model. For particles that have a high porosity and a low reaction rate, the solid phase is converted continuously and progressively throughout the particle. This reaction mechanism is modeled by the progressive conversion model. For particles which have limited porosity, the reaction occurs only at the solid product–unreacted core interface. The unreacted core of material shrinks in size during the reaction and this reaction mechanism is modeled by the shrinking core model [7]. As shown in Fig. 3, the agglomeration of aluminum smelter baghouse dust forms a porous structure. For this reason, a kinetic study of the NaOH leaching of baghouse dust is conducted based on the shrinking core model.

A schematic diagram of the rate-determining steps and the equations of the shrinking core model are shown in Fig. 4 [8–10]. As shown in Fig. 4, when the diffusion is fast and the chemical reaction is slow, the overall reaction rate is determined by the chemical reaction. On the other hand, when the diffusion is slow and the chemical reaction is fast, the overall reaction rate is determined by the

diffusion. When there is no significant difference of rates between the reaction steps, the overall reaction rate is determined by the combined chemical reaction as well as the diffusion.

Based on the equations of the shrinking core model, the reaction rate, k , can be obtained with different bath temperatures, and an Arrhenius plot is obtained based on the temperatures and rate constants. A schematic diagram of an Arrhenius plot and the Arrhenius equations are shown in Fig. 5 [11]. From the slope of the Arrhenius plots, the activation energy can be obtained. In general, reactions with high activation energies are highly temperature-dependent. In this case, temperature can influence both the production rate and economics of the process. The activation energy from the experiments can be used to distinguish a chemical reaction from a physical process. Physical processes (i.e. diffusion control) generally have low activation energy values and the value is typically smaller than 5 kcal/mol. Chemical reactions normally have activation energy values between 10 and 25 kcal/mol [7]. For this

Fig. 3 SEM images of aluminum smelter baghouse dust with particle size smaller than 45 μm

Fig. 4 Schematic diagram of rate-determining steps and equations of the shrinking core model [8–10]

Fig. 5 Arrhenius plot and equation [11] **Fig. 6** Application of the shrinking core model and Arrhenius plot with 10 g/L solids content. **a** pore diffusion control, **b** chemical reaction control, **c** fluid film diffusion, **d** Arrhenius plot based on pore diffusion control

Fig. 7 Application of the shrinking core model and Arrhenius plot with 50 g/L solids content. **a** pore diffusion control, **b** chemical reaction control, **c** fluid film diffusion, **d** Arrhenius plot based on pore diffusion control

research, the three different reaction equations of the shrinking core model (i.e. chemical reaction, pore diffusion, and fluid film diffusion), and the Arrhenius plots are drawn for the 1 M NaOH leaching data in Figs. 6 and 7.

In Fig. 6, the application of the shrinking core model and Arrhenius plot are drawn based on the leaching data with 10 g/L solids content. For a kinetic study, three different reaction equations of the shrinking core model are used to compare the linearity of those equations. In general, when an R^2 value is close to 1, it indicates that the regression line fits the data. In Fig. 6, the R^2 value of the pore diffusion control model shows numbers between 0.912 and 0.947. This R^2 value is relatively close to 1 when compared to the R^2 values of other reaction equations as shown in Fig. 6. Therefore, reaction equations for pore diffusion control could explain the empirical data more accurately than other reaction equations. Thus, the

activation energy of aluminum smelter baghouse dust leaching with 10 g/L solids content is calculated based on the pore diffusion control model. It can be seen in Fig. 6d that the calculated activation energy is about 4.8 kcal/mol and this value corresponds to the typical activation energies of the physical processes (i.e. diffusion control). When the solids content is increased to 50 g/L, the calculated activation energy is about 8.1 kcal/mol. This value corresponds to the typical activation energies of the mixed control of physical processes (i.e. diffusion) and chemical processes. In this case, there is no significant difference of rates between the chemical reaction and the physical processes at the 50 g/L leaching condition. The activation energy for leaching with 50 g/L solids content is higher than for leaching with 10 g/L solids content. As shown in Fig. 2, the higher activation energy corresponds to a decrease in the aluminum leaching rate. Additionally, the

Fig. 8 XRD analysis of the aluminum smelter baghouse dust

higher activation energy indicates that other factors may affect the overall reaction kinetics, such as agglomeration of particles during leaching.

Thermodynamic Study of the NaOH Leaching

The thermodynamic modeling of aluminum smelter baghouse dust leaching is studied for compatibility with the Bayer process. The XRD analysis of the aluminum smelter baghouse dust shown in Fig. 8 indicates that the major phase of the aluminum smelter baghouse dust is the aluminum metal. Thus, the following reaction equations of aluminum leaching with sodium hydroxide are proposed for the equilibrium composition diagram:

Table 3 Reaction equations of aluminum during NaOH leaching

Reaction equations	ΔG° (kcal)	
	100 °C	1000 °C
$\text{Al} + 3\text{H}_2\text{O}(\text{l}) + \text{NaOH}(\text{l}) = \text{Na}[\text{Al}(\text{OH})_4] + 1.5\text{H}_2(\text{g})$	−116.350	−89.673
$2\text{Al} + 2\text{H}_2\text{O}(\text{l}) + 2\text{NaOH}(\text{l}) = 2\text{NaAlO}_2 + 3\text{H}_2(\text{g})$	−221.893	−229.821
$2\text{Al} + 3\text{H}_2\text{O}(\text{l}) = \text{Al}_2\text{O}_3 + 3\text{H}_2(\text{g})$	−211.080	−217.644
$\text{Al} + 3\text{H}_2\text{O}(\text{l}) = \text{Al}(\text{OH})_3 + 1.5\text{H}_2(\text{g})$	−102.415	−83.343

Fig. 9 Equilibrium composition diagram of aluminum smelter baghouse dust leaching using the HSC chemistry

The amount of the aluminum in the dust is about 13 wt.%. Thus, for 100 % aluminum recovery, the total aluminum content in leachate should be 0.1 mol for 20 g/100 mL solids content. Based on this condition, the solution consists of 5.5 mol H_2O and 1 mol NaOH . This amount of solution is enough to promote the reactions forward (Eqs. 1–4) based on Le Chatelier's principle. In Table 3, proposed reaction equations, as well as the corresponding Gibbs free energy at standard state are listed. For the same molarity of aluminum at 100 °C, the formation of $\text{Na}[\text{Al}(\text{OH})_4]$ has the largest negative value when compared to the other reactions. Therefore, $\text{Na}[\text{Al}(\text{OH})_4]$ is the most stable form at 100 °C. When the temperature increases, the formation of $\text{Na}[\text{Al}(\text{OH})_4]$ decreases, and the formation of sodium aluminate (NaAlO_2) increases, as shown in Fig. 9 based on the HSC chemistry. Above 700 °C, sodium aluminate (NaAlO_2) is the most stable form after dissociation of water from $\text{Na}[\text{Al}(\text{OH})_4]$.

The Bayer process, which is the primary production method of aluminum, produces alumina from the bauxite ore. In the Bayer process, sodium hydroxide is used to dissolve aluminum from the bauxite in autoclaves. The main leach reactions are as follows [7]:

Gibbsite: $\text{Al}(\text{OH})_3 + \text{NaOH} = \text{Na}[\text{Al}(\text{OH})_4]$

Boehmite and diasporite: $\text{AlOOH} + \text{NaOH} + \text{H}_2\text{O} = \text{Na}[\text{Al}(\text{OH})_4]$

Based on the main leach reactions for the bauxite ore, the final product after leaching is sodium aluminate, which is the same final product obtained after leaching of aluminum smelter baghouse dust. Thus, the leached compound could be reverted back to produce high-quality alumina making aluminum smelter baghouse dust a viable secondary source for the aluminum industry. Its feasibility, however, still needs to be further examined since the generation of hydrogen gas during leaching should be handled properly due to its flammable or potentially explosive nature [12–15].

Conclusions

In this research, alkaline leaching of aluminum smelter baghouse dust is studied at different temperatures and solids content. With 1 M of NaOH, aluminum leaching from the dust is effective when the bath temperature is high and the solids content is low. Based on the kinetic study, the rate-determining step of aluminum smelter baghouse dust leaching is the pore diffusion, and the calculated activation energy is about 4.8 kcal/mol at 10 g/L solids content. However, at 50 g/L solids content, the overall reaction is controlled by the diffusion as well as the chemical reaction and the calculated activation energy is about 8.1 kcal/mol. A thermodynamic study is performed to determine the final product of the aluminum after leaching. The results indicated that sodium aluminate should be present in the leachate. This product is the same for the Bayer process. Therefore, aluminum smelter baghouse dust could be utilized as a secondary source for the Bayer process for alumina recovery. In this research, aluminum smelter baghouse dust leaching is mainly focused on small particle size since a larger surface area can cause some problems during the separating process when using an electrostatic separator, and the smaller particle size guarantees that the larger surface area will be attacked by the reagents. However, further study about utilizing a larger particle size as well as handling the hydrogen gas evolution should also be investigated.

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