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HIGH PURITY ALUMINA POWDERS EXTRACTED FROM ALUMINUM DROSS BY THE CALCINING—LEACHING PROCESS

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Abstract

A processing technology was developed for alumina extraction from Al dross with the calcining-leaching process. The aluminum dross was mixed with soda and sintered at 1173K to yield soluble aluminates. Subsequently the sintered dross was leached with sulfuric acid to produce a solution containing aluminum . The unwanted metal ions, including Fe^{3+} and Na^{+}, were removed by ethylene diamine tetraacetic acid (EDTA) and water washing. Then NH_4HCO_3, was added controlling the crystallization of $NH_4AlO(OH)HCO_3$, and the drying and calcining process was carried out, resulting in ultra fine Al_2O_3 powders with high purity. The characteristics of the Al_2O_3 powders were examined by means of XRD and SEM. The extraction efficiency of Al_2O_3 can surpass 98% by optimization of the calcination and lixiviation processes. Well-dispersed fibriform Al_2O_3 powders were obtained by calcining at 1000℃ and the purity of the ultra fine Al_2O_3 powders was more than 99.36% .

Introduction

Billions tonnes of Al dross produced during the casting or remelting of aluminum have accumulated over the years. The majority of this dross is disposed off in landfill sites, causing serious pollution of the environment[1]. Finding methods for producing useful materials from the dross is a very important task for society.

Aluminium dross contains mostly aluminium, silicon and other metal oxides and salts such as KCl and NaCl. Thus it has the potential to be a rich alumina source, and is a good raw material suitable for production of aluminum and its chemicals. Different types of dross have been employed as raw materials by several authors. Dross includes considerable amounts of aluminum alloys, and it has been utilized as a deoxidsing agent in steel making, as a refractory material, and as a cement material [2-5]. Attempts have been made to extract alumina from the alloy dross by adopting either pyro- or hydrometallurgical methods [6,7]. Amer carried out the dissolution of waste dross in two steps, in order to produce a highly pure aluminum sulfate[8]. However, these processes have found little practical application because of the highly corrosive nature of the concentrated acid and alkali involved, and because these processes also constitute an environmental hazard.

The objective of this work is to facilitate the recycling of Al dross and produce high purity Al_2O_3. In this paper, the calcining–leaching method was used to recover Al_2O_3 from Al dross. In the proposed process, Al dross is mixed with soda prior to sintering in order to decompose the mullite phase present in the Al dross. Due to the low cost and stability of sulfuric acid, this acid is used as a lixiviant during the recovery of Al_2O_3 from sintered mixed dross. The impurity ions in the leaching liquor were removed by means of EDTA complexation, and washing with distilled water. This is a low temperature process that does not consume too much energy, and high purity Al_2O_3 is obtained.

Experimental

The primary raw materials of the experiment were Al dross, soda and H_2SO_4. The compositions of the Al dross are listed in table 1.

Table1 Chemical Composition of the Al dross(wt%)

Al	Ca	Fe	K	Mg	Na	Si	Ti	O
39.77	1.33	0.56	1.59	4.98	4.57	0.55	0.40	41.59

The recovery of Al_2O_3 from Al dross is based on application of hydrometallurgical processes such as acid or base leaching, purification, precipitation and calcination. The procedures of the experiment are illustrated in Fig.1. The crystalline phase and microstructure were characterized by X-ray diffraction (XRD) and scanning electron microscope (SEM).

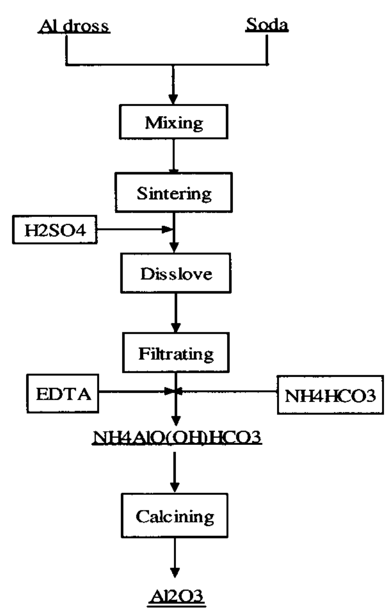


Fig.1 Flow chart of extraction of alumina from Al dross

Results and discussion

Sintering the mixture of Al dross and soda

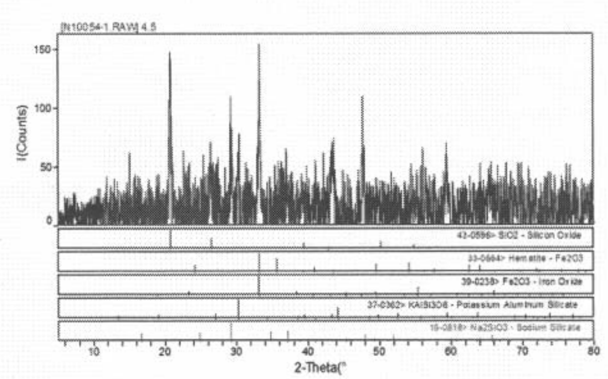
The main objective of sintering the mixture of Aluminum dross and soda was to break the crystalline mullite phase ($3Al_2O_3{\cdot}2SiO_2$) and the mesh framework of the glass phase, rendering free aluminate for leaching [9]. The following reactions occur during the calcining of fly ash and soda[10]:

$$Na_2CO_3=Na_2O+CO_2\uparrow \quad (1)$$

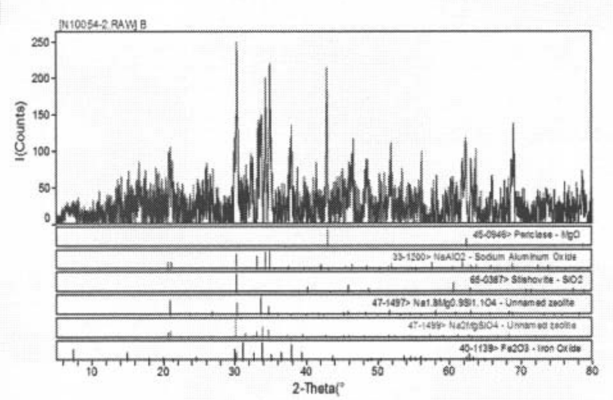
$$Na_2O+SiO_2=Na_2SiO_3 \quad (2)$$

$$3Na_2O+4SiO_2+3Al_2O_3\cdot 2SiO_2=3(Na_2O\cdot Al_2O_3\cdot 2SiO_2) \quad (3)$$

Figure 2 shows the XRD patterns of Al dross and the sintered mixture of Al dross and soda. From this figure we can see there are characteristic peaks of mullite in the raw material, while after sintering, these peaks disappeared and many new characteristic peaks were found. So the stable mullite crystalline phase was successfully broken-up by the sintering. $NaAlO_2$ is the major component of the sintered mix. $2CaO{\cdot}SiO_2$ and $Na_2O{\cdot}Fe_2O_3$ are the minor components. The soluble aluminum materials are dissolved. The process also produced insoluble residues called red mud.



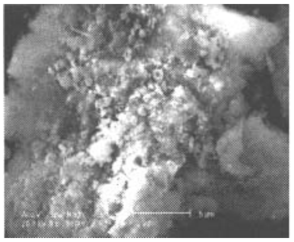
a.Raw material



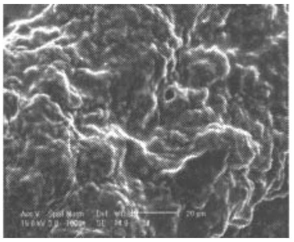
b. Sintered mix powders

Fig.2 XRD patterns of raw fly ash and calcined ash

SEM analysis of the sintered raw Al dross and sintered dross also gave similar results, as shown in Fig.3. SEM image of sintered dross exhibited mainly as clumps. They were formed by reactions of sodium hydroxide, aluminum, and silicon oxide in the sintering process. These clumps were irregular in shape. However, Glassy material was found in the SEM image of raw Al dross, as shown in Fig.3(a). This was the evidence of existence of a mullite phase in the raw material.



a.Raw material



b. Sintered mix powders

Fig.3 SEM images of raw Al dross and sintered ash

The sintering parameters such as sintering temperature and sintering time were considered to find the optimal activation efficiencies (rendering the maximum free aluminates). As shown in Fig.4, 1173K was the optimum temperature. At this temperature, the sintered material had the highest alumina extraction efficiency. Sintering below or above this temperature would produce abnormal sintered products, which led to lower alumina extraction efficiency.

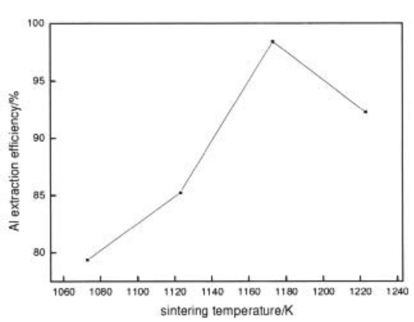


Fig.4 Effect of sintering temperature of alumina extraction efficiency

Dissolving the sintered mixture

The purpose of the leaching process is to leach the soluble Al ions (in the form of sodium aluminates). The possible reactions that take place in the leaching of the raw Al dross are as follows :

$$Na_2\cdot Al_2O_3\cdot 2SiO_2+4H_2SO_4+(2n-4)H_2O =2Al^{3+}+2Na^{+}+4SO_4^{2+}+2(SiO_2\cdot nH_2O) \quad (4)$$

$$Na_2SiO_3+H_2SO_4=Na_2SO_4+H_2SiO_3\downarrow \quad (5)$$

The leaching parameters such as the concentration of sulfuric acid, leaching time and leaching temperature were considered to find the optimal extraction efficiency of Al_2O_3.

The results presented in Fig.5 and Fig.6, indicate that the efficiency of aluminum species extraction increases with an increase in both acid concentration and leaching time. Increasing the acid concentration can accelerate the leaching reaction and achieve high extraction efficiency. However, when acid concentration reached a certain value, the extraction efficiency may not improve. The optimum acid concentration was 0.9mol/l at which the alumina extraction efficiency reached the maximum. To some degree, prolonging the leaching time can increase the aluminum extraction efficiency,as prolonging the leaching time will allow the completion of the leaching reactions.

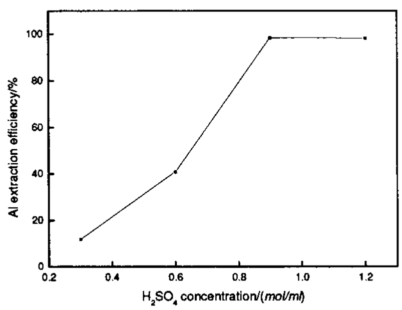


Fig.5 Effect of H_2SO_4 concentration on alumina extraction efficiency

The dissolution time was defined as the period that started at the time when the sintered mixture was introduced into the acid and ended when the liquid was separated from the red mud. This was the time period that the dissolution solution actually contacted the sintered mixture product. The longer the dissolution process is run, the greater the loss of the alumina extraction efficiency. This was due to the dissolution of $Na_2O{\cdot}Fe_2O_3$ as well as the side reactions. Since the particle size of the sintered mixture was very small, a short dissolution period actually gave the best result. The exact relationship between the dissolution time and aluminum extraction efficiency is shown in Fig.6.

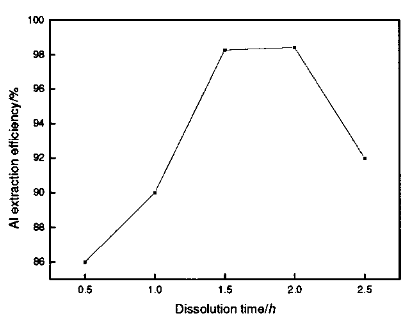


Fig.6 Effect of dissolution time on alumina extraction efficiency

Temperature was the most important factor affecting alumina dissolution from the sintered mixture. From the results presented in Fig.7, it can be seen that when the temperature is lower than 80°C the extraction efficiency is low. Increasing the leaching temperature can accelerate the progress of the leaching reactions and improve the aluminum extraction efficiency.

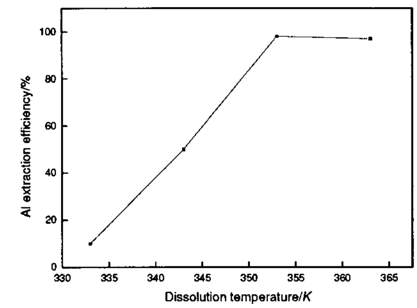


Fig.7 Effect of dissolution temperature on alumina extraction efficiency

2.3 Precipitated the aluminum

The leaching liquor obtained from the previous processes contained Al^{3+}, Na^{+}, Fe^{3+}, Ca^{2+}, Mg^{2+}, and other impurities. In order to obtain high purity Al_2O_3, the impurity ions had to be removed. The selective precipitation of ions by pH value adjustment was found to be unsuccessful in separating Fe^{3+} and Al^{3+}[11]. We chose ethylene diamine tetraacetic acid (EDTA), a complexing agent, to remove the Fe^{3+} and other impurity ions. Selectivity arises because the complexing ability of each metal in the leaching liquor with EDTA is different [12,13].

The pH value of the leaching liquor was adjusted to 3.0 before the EDTA was added, and then the solution was continuously stirred for 30 minutes. The color of the solution turned from dull yellow to bright yellow. NH_4HCO_3 was subsequently added into the solution to precipitate the aluminum. At the same time the solution was quickly stirred. After filtration, white precipitates and yellow solution were obtained. These results show that the Fe^{3+} contained in the leaching liquor was effectively removed. The other soluble impurities absorbed in the precipitates could be removed by three cycles of washing. A white gelatinous precipitate was obtained and then dried in a microwave oven for 5 min and the powder calcined at 1000°C for 2 h. Active γ-Al_2O_3 powder was obtained. The XRD pattern and SEM images of the Al_2O_3 product are shown in Figs.8 and 9. The purity of the Al_2O_3 product was as follows: Al_2O_3 99.36%, MgO 0.51%, CaO 0.06%, Fe_2O_3 0.04%, and SiO_2 0.03%. The result indicated that further investigation of strategies for Mg removal is necessary to achieve higher purity Al_2O_3.

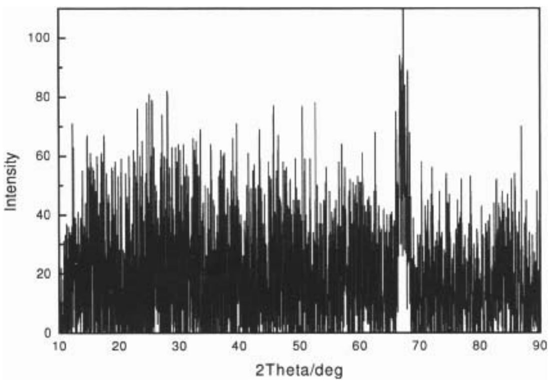


Fig.8 XRD pattern of Al_2O_3 product

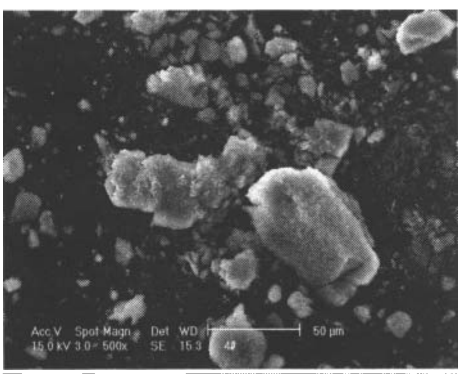


Fig.9 SEM images of α-Al_2O_3

Conclusions

(1) Al dross can be used can be used as a bauxite substitute in the soda sintering process for alumina production. Optimum conditions for a desilicated ash sintering were 1173K and 60 min.

(2) Optimum conditions for sintered for Al dross dissolution were an H_2SO_4 concentration of 0.9mol/l at 358K for 1.5h.

(3) The Al_2O_3 purity was improved by means of EDTA complexation of other impurity metals and washing with distilled water. Finally, High purity (Al_2O_3>99%) active α-Al_2O_3 was obtained.

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