Aluminum Resource Recovery and High Value Aluminum Oxide Preparation from Secondary Aluminum Ash of Hazardous Solid Waste

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Abstract: Secondary aluminum ash, a typical hazardous solid waste in China's aluminum industry, generates over 4 million tons annually while posing environmental risks and holding significant resource potential. Using secondary aluminum ash from a specific enterprise as raw material, this study employed an alkaline roasting-water leaching method to extract aluminum resources. The effects of key parameters—including alkali-to-material ratio, roasting temperature, roasting duration, leaching temperature, liquid-to-solid ratio, and leaching time—on aluminum extraction efficiency were systematically investigated. Subsequently, α-Al_2O_3 ceramic materials were prepared from the aluminum leaching solution. The optimal process conditions were identified as: alkali-to-material ratio 1.1, roasting temperature 800°C, roasting time 120 minutes, leaching temperature 70°C, liquid-to-solid ratio 14:1 mL/g, and leaching time 80 minutes, achieving an aluminum extraction rate of 91.83%. XRD analysis of the roasting residue confirmed successful conversion of metallic Al, AlN, and Al_2O_3 into soluble $NaAlO_2$. After purification and calcination at 1200°C, the aluminum leaching solution yielded α-Al_2O_3 ceramic materials with purity exceeding 99%. Through process optimization, this study demonstrates efficient extraction and high-value conversion of aluminum resources from secondary aluminum ash, providing a comprehensive technical solution for solid waste utilization in the aluminum industry.

Keywords: secondary aluminum ash; alkaline roasting; aluminum leaching; α-Al_2O_3; resource utilization

1 Introduction

Secondary aluminum ash,a hazardous byproduct generated during aluminum extraction from primary aluminum ash[1],is produced in China at over 4 million tons annually.With the continuous expansion of the recycled aluminum industry,its emissions have shown a steady annual increase[2].The chemical composition primarily includes aluminum oxide,aluminum nitride(AlN),metallic aluminum,and fluorides/chlorides.Notably,AlN releases ammonia when exposed to water,while soluble fluorides and chlorides endow it with reactivity and leaching toxicity,posing potential ecological threats[3].Since 2021,secondary aluminum ash has been officially classified under the National Hazardous Waste Catalog(HW48 category),making its safe disposal and resource recovery critical for the sustainable development of the aluminum industry[1].

Current secondary aluminum ash treatment technologies primarily focus on two core aspects:deep detoxification and efficient aluminum extraction[3].In terms of deep detoxification,pyrometallurgical roasting faces challenges such as high energy consumption,while hydrometallurgical hydrolysis is prone to incomplete reactions and aluminum resource loss.For efficient aluminum extraction,the alkaline roasting-hydroleaching method involves mixing secondary aluminum ash w

ith NaOH for roasting,converting Al,AlN,and Al_2O_3into soluble $NaAlO_2$,followed by aluminum separation through hydroleaching[2].This process offers advantages like relatively simple workflow and high aluminum leaching efficiency.However,it was initially underdeveloped due to technical costs and lagging environmental policies.In recent years,with growing demand for resource recycling,this method has emerged as a research hotspot[8][11].

While existing research has explored this process,systematic optimization of process parameters affecting aluminum leaching efficiency remains inadequate,and effective integration between aluminum extraction and high-value-added product manufacturing is still lacking[5].To address this,this study investigates the impact of key parameters—including alkali-to-material ratio,roasting temperature,roasting duration,leaching temperature,liquid-to-solid ratio,and leaching time—on aluminum leaching efficiency using secondary aluminum ash from a specific enterprise.The optimal process conditions are determined,and the resulting leaching solution is utilized to produce high-purityα-Al_2O_3ceramic materials,thereby providing a comprehensive technical solution for the resource utilization of secondary aluminum ash.

2 Materials and Methods

2.1 Experimental Materials

The secondary aluminum ash used in the experiment was obtained from the aluminum ash and slag treatment workshop of a recycled aluminum enterprise.The original samples were crushed and ball-milled,then sieved through a standard sieve to obtain powder with particle sizes less than 150μm.The sieved samples were dried in a 105°C drying oven until a constant weight was achieved,cooled,and sealed for storage and subsequent use.

2.2 Raw Material Characterization Methods

The main chemical components of secondary aluminum ash were analyzed by X-ray fluorescence spectrometer(XRF).The phase analysis was carried out by X-ray diffraction(XRD)under the following conditions:Cu Kαradiation,tube voltage 40 kV,tube current 40 mA,scanning range 2θ=10~80,scanning speed 2/min.

2.3 Alkaline roasting-water leaching test

Weigh 20.0 g of pretreated secondary aluminum ash,mix it with sodium hydroxide according to the preset alkali material ratio(mass ratio of NaOH to secondary aluminum ash),and place it in a corundum crucible.Transfer the crucible into a box-type resistance furnace and heat it to the preset temperature at a rate of 5°C/min for constant temperature calcination.After the calcined product cools naturally,grind it and set aside.

The effects of alkali-to-bulk ratio(0.5:1,0.8:1,1.0:1,1.1:1,1.2:1,1.5:1),calcination temperature(600,700,800,850,900°C),and calcination time(60,90,120,150,180 min)on aluminum leaching rate were investigated.Based on optimized calcination conditions,the influence of leaching temperature (40,50,60,70,80,90°C),liquid-to-solid ratio(8:1,10:1,12:1,14:1,16:1,18:1 mL/g),and leaching time(30, 60,80,100,120 min)on aluminum leaching rate was further examined.

The aqueous leaching experiment was conducted as follows:5.0 g of calcined residue was weighed and placed in a conical flask.Deionized water was added according to the preset liquid-to-solid ratio.The flask was then placed in a constant-temperature water bath with an agitator

and leached at 200 rpm under constant temperature.After leaching,the suspension was separate d by vacuum filtration.The filtrate was diluted to the desired volume and the aluminum concen tration was determined using the EDTA titration method.The aluminum leaching rate was calcul ated according to Equation(1).Each experiment was repeated three times,and the results were av eraged.

Calculation formula for aluminum leaching rate:

$$\eta=\frac{C\times V}{m\times\omega}\times100\%$$

In the formula:ηis the aluminum leaching rate,%;C is the aluminum concentration in the fi ltrate,g/L;V is the filtrate volume,L;m is the mass of calcined slag,g;ωis the mass fraction of a luminum in the calcined slag,%.

2.4 Preparation of Regenerated Alumina

The leachate obtained under optimal conditions was purified and calcined.Ammonia water was added to adjust the pH to 8.5,followed by NaOH to reach 13,and most impurities were re moved through precipitation filtration.The purified filtrate was placed in a 75°C water bath,whe re 2 g/L polyethylene glycol 2000 was added as a dispersant according to the leachate volume and thoroughly dissolved.Hydrochloric acid was then slowly added to adjust the pH to 9.5,and the mixture was aged for 10 hours before filtration to obtain the precursor.The precursor was calcined in a muffle furnace at a heating rate of 5°C/min until reaching 700°C for 2 hours t o produceγ-Al_2O_3.The temperature was further increased to 1200°C for constant-time calcination of 2 hours to obtainα-Al_2O_3ceramic raw material.The product phase was analyzed by XRD,and the purity was determined according to the standard"Alumina"(GB/T 24487-2022)[9].

3 Results and Discussion

3.1 Characteristics of Secondary Aluminum Ash Raw Materials

The chemical composition analysis of secondary aluminum ash is presented in Table 1.As shown in the table,the sample contains 60%-70%Al_2O_3,5%-15%metallic Al,and 5%-12%AlN,alon g with additional components such as SiO_2,MgO,Na_2O,K_2O,and fluorine and chlorine.This comp osition aligns closely with the typical range of secondary aluminum ash,indicating that the raw materials used in this study are representative[6].

表 1 二次铝灰主要化学成分（质量分数，%）

Table 1 Chemical composition of secondary aluminum dross (mass fraction, %)

组分	Al_2O_3	Al	AlN	SiO_2	MgO	Na_2O	K_2O	F	Cl
含量	60-70	5-15	5-12	5-10	2-5	1-3	1-2	0.5-2	1-3

Figure 1 presents the XRD spectrum of secondary aluminum ash.The analysis reveals the primary phases:α-Al_2O_3,γ-Al_2O_3,metallic Al,AlN,$MgAl_2O_4$spinel,Na_3AlF_6cryolite,NaCl,and KCl.The prominent AlN diffraction peak confirms the presence of aluminum nitride,while the diffraction peaks of soluble chlorides indicate the presence of salt impurities in the raw material.

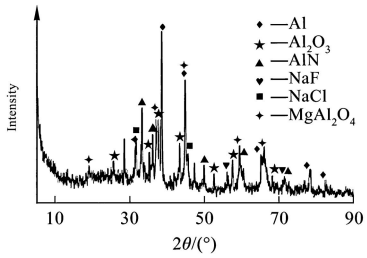

Fig. 1 XRD pattern of secondary aluminum dross

3.2 Optimization of Alkalization Roasting Process

3.2.1 Effect of Alkali Material Ratio on Aluminum Leaching Rate

Under fixed conditions of 800°C roasting temperature,120-minute roasting time,70°C leaching temperature,14:1 mL/g liquid-to-solid ratio,and 80-minute leaching time,the effect of alkali-to-material ratio on aluminum leaching rate was investigated,as shown in Figure 2a.The aluminum leaching rate increased from 70.0%to 91.0%when the alkali-to-material ratio rose from 0.8 to 1.1.A slight increase to 92.0%occurred at 1.2,but the rate declined to 91.0%at 1.3,indicating that the optimal range for alkali-to-material ratio is 1.1–1.2[7].Insufficient alkali amounts prevent complete conversion of aluminum-containing phases into soluble aluminates,while excessive alkali may lead to side reactions or higher reagent costs.Considering both aluminum leaching efficiency and reagent expenses[12],an alkali-to-material ratio of 1.1 was selected as the optimal condition.

3.2.2 Effect of Calcination Temperature on Aluminum Leaching Rate

Under fixed conditions of an alkali-to-material ratio of 1.1,roasting time of 120 minutes,leaching temperature of 70°C,liquid-to-solid ratio of 14:1 mL/g,and leaching time of 80 minutes,the effect of roasting temperature on aluminum leaching rate was investigated,as shown in Figure 2b.When the roasting temperature increased from 600°C to 800°C,the aluminum leaching rate rose from 72.5%to 91.8%.At 900°C,the leaching rate slightly decreased to 89.6%.This phenomenon can be attributed to the following:higher temperatures accelerate solid-phase reaction rates,promoting $NaAlO_2$formation.However,excessively high temperatures(>800°C)may cause localized sintering of materials,hindering reactant diffusion and mass transfer,thereby adversely affecting aluminum leaching[2].Therefore,the optimal roasting temperature was determined to be 800°C.

Under the optimal alkali-to-aluminum ratio and calcination temperature conditions,the effect of calcination time on aluminum leaching rate was further investigated.Results showed that extending the calcination time from 60 minutes to 120 minutes increased the aluminum leaching rate from 86.5%to 91.8%.Further extension to 180 minutes showed no significant change in leaching efficiency[7].Considering both energy consumption and leaching efficiency,the optimal calcination time was determined to be 120 minutes.

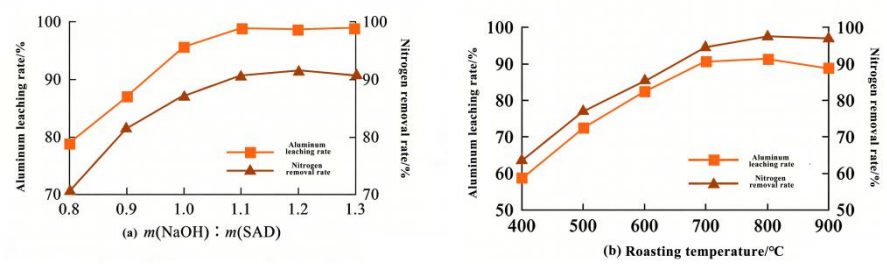

Fig. 2 Effects of alkali ratio (a) and roasting temperature (b) on aluminum leaching rate

3.3 Optimization of Water Immersion Process

The leaching process parameters were further optimized under the optimum conditions(alkali-to-aluminum ratio 1.1,800℃for 120 min).The effects of leaching temperature,liquid-solid ratio and leaching time on the leaching rate of aluminum were investigated.

The leaching temperature experiment showed that the leaching rate of aluminum increased from 85.6%to 91.8%when the leaching temperature was increased from 40℃to 70℃under the condition of liquid-solid ratio 14:1 mL/g and leaching time 80 min.When the temperature was increased to 90℃,the leaching rate of aluminum decreased slightly to 90.5%.Therefore,the optimum leaching temperature was 70℃.

Liquid-solid ratio experiments demonstrated that under leaching conditions of 70°C and 80 minutes,the aluminum leaching rate increased from 88.2%to 91.8%when the liquid-to-solid ratio was raised from 8:1 mL/g to 14:1 mL/g.No significant change in aluminum leaching rate was observed when the ratio was further increased to 18:1 mL/g[4].Therefore,the optimal liquid-solid ratio is 14:1 mL/g.

The leaching time experiment showed that the leaching rate of aluminum increased from 86.7%to 91.8%when the leaching time was extended from 30 min to 80 min at 70℃and 14:1 mL/g.The leaching rate of aluminum remained stable when the leaching time was extended to 120 min.Therefore,the optimal leaching time was 80 min.

The optimum conditions of the alkaline roasting and leaching process are as follows:the ratio of alkali to material is 1.1,the roasting temperature is 800℃,the roasting time is 120 min,the leaching temperature is 70℃,the ratio of liquid to solid is 14:1 mL/g,and the leaching time is 80 min.Under these conditions,the leaching rate of aluminum is 91.83%.

3.4 Phasic Analysis of Calcined Residue

To validate the phase transformation mechanism during the calcination process,XRD analysis was performed on the calcined slag obtained under optimal conditions(alkali-to-material ratio 1.1,calcination at 800°C for 120 minutes),as shown in Figure 3.Compared with the XRD patterns of the raw material(Figure 1),the diffraction peaks of Al,AlN,and $Al_2O_3$are almost completely absent,with $NaAlO_2$being the predominant phase.The characteristic diffraction peaks are located at 2θ=20.1°,23.5°,32.2°,34.8°,and 41.3°[7].This indicates that during the alkaline calcination process,Al,AlN,and Al_2O_3in the raw material were successfully converted into soluble $NaAlO_2$,laying the foundation for subsequent aluminum extraction by water leaching.Additionally,no significant fluorine-or chlorine-containing phases were detected in the patterns,suggesting that the calcination process promoted the transformation or volatilization of certain impurities[3].

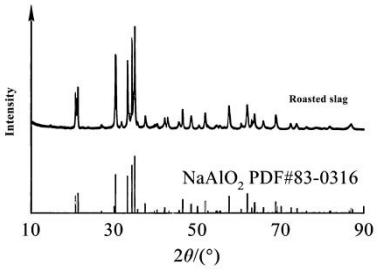

Fig. 3 XRD pattern of roasted residue under optimal conditions

3.5 Preparation and Characterization of Regenerated Alumina

The leachate obtained under optimal conditions was purified and calcined,followed by XRD analysis as shown in Figure 4.The 700°C calcined product exhibited characteristic diffraction peaks ofγ-Al_2O_3(2θ=37.6°,45.8°,67.0°),indicating the product existed in the metastableγ-Al_2O_3form at this temperature[9].After 1200°C calcination,the diffraction pattern showed only the characteristic peaks ofα-Al_2O_3(2θ=25.6°,35.2°,43.4°,57.5°),with theγ-Al_2O_3peaks completely disappeared, demonstrating complete conversion to the stableα-Al_2O_3phase and no other impurity phases detected[10].

The product purity analysis demonstrated that the finalα-Al_2O_3achieved 99.2%purity,meeting the technical requirements specified in the national standard"Alumina"(GB/T 24487-2022)and qualifying for use as a ceramic industrial raw material.This outcome confirms that the extraction of aluminum resources through alkaline roasting and hydrolysis,followed by purification and calcination,enables the successful production of high-purity alumina materials from secondary aluminum ash,thereby achieving its high-value conversion.

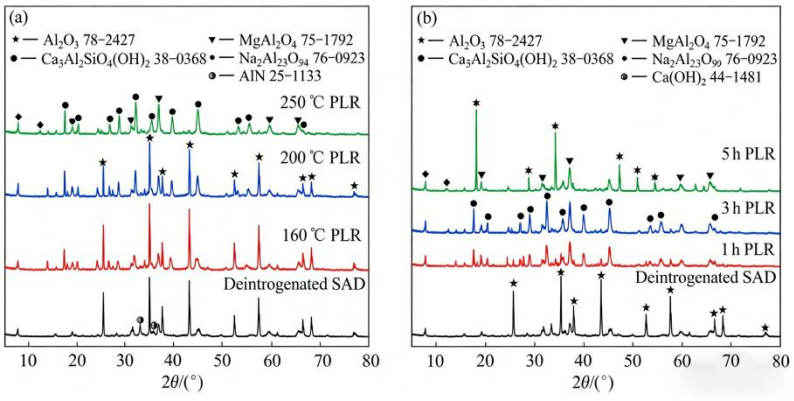

Fig. 4 XRD patterns of regenerated alumina at different calcination temperatures

4 Conclusion

This study optimized the process parameters for extracting aluminum from secondary aluminum ash using the alkaline roasting-water leaching method,successfully preparing high-purityα-Al_2O_3ceramic materials.The optimal conditions were:alkali-to-material ratio 1.1,roasting temperature 800°C,roasting time 120 min,leaching temperature 70°C,liquid-to-solid ratio 14:1 mL/g,and leaching time 80 min,achieving an aluminum leaching rate of 91.83%.XRD analysis demonstrated

that Al,AlN,and Al_2O_3in the raw materials were successfully converted into soluble $NaAlO_2$during roasting.After purification and calcination at 1200°C,theα-Al_2O_3obtained reached a purity of 99.2%,meeting relevant national standards.This study achieved efficient extraction and high-value-added conversion of aluminum resources from secondary aluminum ash,providing a feasible technical solution for the resource utilization of solid waste in the aluminum industry.

The following directions warrant further attention:evaluating the adaptability of secondary aluminum ash from different sources to the process;investigating the migration patterns of impurity elements during the roasting-leaching process and their impact mechanisms on product purity; optimizing the deep impurity removal process,and conducting scale-up experiments and economic assessments to facilitate the industrial application of the technology[8][12].

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