Synthesis of submicronic α-alumina from local aluminum slags

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Abstract

In this study, a high valued product submicronic α-alumina is successfully extracted from aluminum slags generated by the local aluminum industry. The extraction technique is based on the leaching of slags by H₂SO₄ followed by precipitation. The coarser aluminum-rich fractions of the slags are used in this study instead of the finer oxide-rich fractions that were commonly used in previous studies. The precipitation of the leached slags by NH₄OH is controlled by zetameter in order to determine the optimal precipitation pH. Then, the obtained gel showing the higher precipitation rate and the finer particle size is calcined at 1200 °C and characterized by XRF, XRD, FTIR, SEM, EDS and laser granulometry. Even without any pretreatment of slags, the XRF analysis reveals that a high purity and high extraction efficiency of 99.2% and 93.75% respectively can be achieved just at a leaching acid concentration of 15%. XRD spectrum shows that the produced alumina is a pure α-corundum, which is confirmed by FTIR spectrum showing only the Al-O bonds. The laser granulometry shows that the recovered powder exhibit a wide particle size distribution. It is between 50 nm and 20 µm while the average particle size (d50) is about 400 nm. SEM observations reveal that the grains are in the form of submicronic whiskers. The above characteristics allow the obtained alumina powder in this study to be used in the usual applications of alumina such as refractory, ceramic fibers, abrasive, etc. The obtained powders may assume also applications
as a thermally stable substitute for the commonly used transition alumina powders, which need further investigations in future studies.

**Keywords:** $\alpha$-alumina, aluminum slags, acid leaching, precipitation, calcination.

1. **Introduction**

Alumina is a compound that exhibits several phases like $\alpha$, $\gamma$, $\beta$ and $\theta$, etc [1]. While the most stable phase is $\alpha$-alumina with very interesting properties such as high hardness, high melting temperature, chemical stability and high elastic modulus [1–4]. Moreover, the combination of a high thermal coefficient, low thermal expansion and high compressive strength provides $\alpha$-alumina a good resistance to thermal shock [1, 5]. All these important properties make $\alpha$-alumina a material in demand in a wide variety of fields. It is used to produce refractory ceramics, high-strength ceramics, abrasives and other ceramic materials [2, 4]. Submicronic and Nano-sized $\alpha$-alumina powders are also in demand in the field of catalysis as a thermally stable alternative to the widely used transition alumina powders, that undergo phase transformations when used at high temperatures [6].

Submicronic $\alpha$-alumina powders can be produced by a wide variety of synthesis processes, including sol-gel [7], chemical synthesis [8], laser ablation [9], hydrolysis [10], thermal decomposition [11] and chemical vapor deposition [12], etc. However, these methods involve the use of relatively expensive raw materials such as aluminum sulfates [13]. It is therefore of great importance to find inexpensive alternative raw materials. One of the most important inexpensive sources from which we can obtain raw materials to produce $\alpha$-alumina powder is the treatment of aluminum industries wastes. As aluminum transformation industries can produce large quantities of aluminum slags, which is a waste material formed on the surface of molten aluminum and exposed to the atmosphere during the melting process. These slags contain many important components such as aluminum (12–20%), sodium chloride (20–25%), potassium chloride (20–25%), aluminum oxides (20–50%), and other compounds (2–5%) [14]. On the other hand, as these slags have been classified as toxic waste its disposal as a garbage dump is prohibited in many countries [15]. For this reason, the subject of recycling these wastes to eliminate their toxicity is of great interest. Accordingly, many studies have addressed the issue of the recycling and valorization of aluminum slags. Direct applications as raw materials in the refractory brick [16] or in the production of inert filling for construction, road paving, mortar components, polymer composites, adsorbents, and mineral wood have been reported [17]. While other researchers have aimed to use them to
manufacture other raw materials such as hydrogen gas [18], X-zeolites [19], aluminum sulfate (Al_2(SO_4)_{3.18}H_2O) [20] and alumina with different properties for many applications like catalysis [21,22], cement [23], biomaterials [24] and refractory [16].

In this study, a novel process was developed to recover alumina from aluminum slags. This process is based on acid leaching of the slags followed by alkaline precipitation. The coarse fraction of the slags, which is rich in aluminum, is used directly without any pretreatment instead of the finer oxide-rich fraction commonly used in other studies. The followed process involves several steps: selective crushing and sieving in order to recover the maximum aluminum compounds present in the slags, leaching in different concentrations of sulphuric acid (H_2SO_4), purification, precipitation by the addition of different amounts of ammonia varying the pH of the suspension between 4 and 10, filtration, drying and calcination. The study is assisted by XRF, XRD, FTIR, SEM, EDS and laser granulometry characterizations in order to optimize the extraction conditions namely, the leaching acid concentration and the precipitation pH for recover the maximum amount of alumina with the maximum purity and with well-known morphology and particle size distribution. The main objectives of the present study are to eliminate the harmful effect of aluminum slags on the environment and to provide the local ceramic industry with a very high value-added raw material that is submicronic α-alumina powder.

2. Experimental procedure

2.1. Raw materials

The aluminum slags used in this work were collected from a local aluminum transformer “El Ashir Min Ramadan”. The crushed slags have inherent heterogeneity. Where aluminum compounds concentrate in the coarser fractions, while oxide and non-metallic compounds accumulate in the finer fractions. This is because aluminum behaves plastically and remains unfragmented, while oxides and salts are brittle and fragment easily [17, 25]. A sieving of the above-mentioned slags was undertaken to separate the two fractions then just the fraction with particle size > 750 μm was taken in this study (Fig.1). The chemical analysis of the two fractions by XRF showed that the coarser fraction is rich in metallic aluminum (45.82%), aluminum oxide (33.18%) and aluminum hydroxide (17%) (Fig.1 b). The finer fraction is mainly aluminum oxide with a high content of impurities (MgAl_2O_4 (14%), CaCO_3 (16%) and SiO_2 (6%)). The taken fraction was characterized also by XRD and the major constituents
were found to be the aluminum compounds (Al, Al₂O₃ and Al(OH)₃) with a small amount of magnesium aluminate (MgAl₂O₄) and calcite (CaCO₃) (see section 3.3).

![Image](image.png)

**Fig. 1.** Shape of the used aluminum slags.

### 2.2. Methods

First, aluminum oxide was recovered as Al³⁺ ions by dissolving aluminum slags at various concentrations of H₂SO₄ (3, 5, 10, 15, 20 and 25%). The dissolution was carried out by putting 10 g of slag in a vial containing 100 ml of H₂SO₄. The vial was placed on a hot plate to maintain the temperature of the mixture at 90±2 °C. Then, the mixture was stirred continuously for 3 hours using a magnetic stirring bar. This allowed the three aluminum compounds (Al, Al(OH)₃ and Al₂O₃) contained in the aluminum slag rock to react and dissolve in H₂SO₄ giving aluminum sulphates according to the following reactions [26]:

$$\text{Al}_2\text{O}_3 + 3\text{H}_2\text{SO}_4 = \text{Al}_2(\text{SO}_4)_3 + 3\text{H}_2\text{O} \quad (1)$$

$$2\text{Al} + 3\text{H}_2\text{SO}_4 = \text{Al}_2(\text{SO}_4)_3 + 3\text{H}_2 \quad (2)$$

$$2\text{Al(OH)}_3 + 6\text{H}_2\text{SO}_4 = \text{Al}_2(\text{SO}_4)_3 + 3\text{SO}_2 + 6\text{H}_2\text{O} \quad (3)$$

The obtained aluminum sulfates solution was separated from the solid residue by filtration using filter paper with a diameter of 0.2 µm. The obtained solution has an acidic pH between 1.5 and 2.5. A chemical precipitation was then carried out at 40 °C by adding a solution of NH₄OH (10%) drop wise [26, 27], increasing the solution pH up to 10 in order to obtain the optimal pH which corresponds to the maximum precipitation rate. The operation was followed by measuring the zeta potential of the different solutions to control the particle size.
and the precipitation rate as a function of pH. The gel that showed the maximum precipitation rate was filtered and washed with distilled water to remove the maximum impurities. This gel was then dried in an oven at 80 °C for 24 hours. Finally, the obtained powder was calcined in Nabertherm furnace at 1200 °C for 2 hours with a heating rate of 5 °C/Min. Fig.2 is a summary diagram of the followed protocol.

Fig. 2. Summary diagram of the followed protocol.

2.3. Composition, microstructural and morphological characterizations

The chemical composition of the synthesized alumina powder has been determined using ZSX Primus IV wavelength dispersive X-Ray fluorescence spectrometer (WD-XRF), equipped with a 3/4 kW sealed X-ray tube, allows the vacuum analysis from Be to U. The crystallographic characterization of the phases existing in the used aluminum slags and in the synthesized alumina powder was done by Philips X’PERT Pro diffractometer equipped with a copper anti cathode (λ = 1.54 Å). The scanning angle 2θ was between 5 and 80° with a step interval of 0.017°.

For FTIR analysis, the synthesized powder was compressed into cylindrical pellets, and then characterized by Perkin Elmer Fourier transform infrared spectrometer in the range of 400-4000 cm⁻¹ at a resolution of 4 cm⁻¹.
The particle size, the zeta potential and the dispersion stability of particles inside the solution were measured at room temperature using Nano Partica SZ-100 nanoparticle analyzer. The electrophoretic light scattering (ELS) was the technique used for ZP analysis. The particle size of the synthesized alumina powder was also estimated in distilled water using HORIBA Partica LA-960 Laser Particle Size Analyzer. The morphology of the synthesized powders was observed by the use of JEOL JSM-7001F scanning electron microscope (SEM) operating in high vacuum mode at an accelerating voltage of 15 kV.

3. Results and discussions

3.1. Effect of leaching acid concentration

In the present study, the concentration of leaching H$_2$SO$_4$ acid used to dissolve the aluminum slag rocks was varied between 3 and 40%. Fig. 3 shows the percentage of alumina and sulphur trioxide in the recovered powder as a function of the concentration of leaching H$_2$SO$_4$ acid. As can be seen, the purity of the recovered alumina increased proportionally with the increase of the H$_2$SO$_4$ concentration to reach its maximum value (99.2%) at an optimal H$_2$SO$_4$ concentration of 15%. Beyond this acid concentration, the percentage of recovered alumina decreased with increasing acid concentration to reach a purity of 77.1% at an H$_2$SO$_4$ concentration of 40%. The slight increase in alumina percentage in the first stage was due to the increase of aluminum compounds solubility existing in the slag with increasing H$_2$SO$_4$ acid content (see equations 1-3). However, this solubility is limitable at a concentration of acid solution [26], which is 15% in this study. Therefore, above a concentration of 15% H$_2$SO$_4$, the solubility of aluminum in the solution has decreased. This resulted in an excess of sulfate anions (SO$_4^{2-}$) that did not find enough Al$^{3+}$ cations to completely transform into Al$_2$(SO$_4$)$_3$. These sulfate anions could not be removed in the precipitation phase. They remained in the precipitated gel and were converted to (SO$_3$) after calcination, resulting in a gradual decrease in the purity of the recovered powder with increasing H$_2$SO$_4$ concentration beyond 15%. It should be mentioned that the study of the purity of recovered alumina as a function of leaching acid concentration has not been addressed in other studies. However, the higher purity obtained in this study without any pretreatment of slags is comparable to the purity (99.28%) obtained by David et al [28] using pretreated aluminum slags.
In the present study, the efficiency of alumina extraction process $Q(\%)$ was calculated according to the following relationship:

$$Q(\%) = \frac{Q_{\text{Al extracted}}}{Q_{\text{Al initial}}} \times 100\% \quad (4)$$

Where $Q_{\text{Al extracted}}$ and $Q_{\text{Al initial}}$ are the quantity by gram of alumina in the extracted powder and the aluminum compounds in the slags, respectively. Fig. 4 represents the alumina extraction efficiency as a function of the leaching acid concentration. The efficiency of the extraction process increased from 74.12 to 93.75% as the leaching acid concentration was increased from 3 to 15%, and then decreased slightly to 87% when the acid concentration was increased from 15 to 40%. This confirmed the above results, indicating that the maximum dissolution of the aluminum compounds takes place at an optimum acid concentration of 15%.

**Fig. 3.** Alumina purity and SO$_3$ content vs. leaching acid concentration.
Fig. 4. Efficiency of alumina extraction process vs. leaching acid concentration.

Table 1 provides a comparison of the extraction efficiency obtained in this study using the coarser fractions of aluminum slags with the results obtained from other studies using the finer fractions, under almost the same leaching experimental conditions. It can be seen that 15% H₂SO₄ acid concentration was also reported as the optimal concentration for the maximum alumina recovery in the studies provided by David et al [26], Dash et al [27] and Das et al [28], in the case of non-pretreated slags. On the other hand, it was found that the aluminum slags fraction used in this work exhibited a higher extraction efficiency than that obtained in the mentioned studies. Only one study [27] has found a higher efficiency than ours, but with the pretreatment of the slags and with the use of a relatively high leaching acid concentration of 50%. This indicates that the metallic aluminum-rich coarse fractions selected in the present study were more suitable in terms of efficiency for the alumina extraction than the oxide-rich fine fractions selected in the studies [26], [27] and [28].

Table 1. Comparison of the obtained extraction efficiency with other studies.

| Raw material  | Particle size | Optimal H₂SO₄ leaching acid concentration | Efficiency of extraction | Temperature | Time | References       |
|---------------|---------------|------------------------------------------|--------------------------|-------------|------|-----------------|
| White coarser slags | >750 μm    | 15%                                      | 93.75% without pretreatment | 90 °C | 3 h | The present study |
| White finer slags | <850 μm <850 μm | 30% and 50%                              | 85% without pretreatment and 95% with pretreatment | 90 °C | 1 h | [27] |
| White finer slags | <850 μm    | 30%                                      | 88% with pretreatment     | 90 °C | 3 h | [26] |
| White finer slags | 75 μm      | 15%                                      | 83.49% with pretreatment | 90 °C | 5 h | [28] |
3.2. Effect of the precipitation pH

The zeta potential is a measure that can provide us with information about the surface functionality, the stability of the dispersed particles as well as interaction of dissolved compounds with the solid dispersed particles (catalysis). In this study, the zeta potential was monitored as a function of the precipitate pH during the drop wise addition of (NH₄OH) in order to control the particle size and stability and to test the functionality and interactivity of the precipitated alumina surface. Fig. 5 shows the zeta potential of the prepared gel versus the pH value of the medium at ambient temperature. According to the figure, the zeta potential decreased meaningfully with increasing pH and took its negative values when the pH was higher than 8. While the isoelectric point, at which the surface acquired a net zero charge (pZ = 0 mV) was found at a pH_{zpc} (pH of zero potential charge) of about 8. According to [22,29], the amphoteric equilibria can be represented by the following equation:

\[
\text{Al} - \text{OH}_2^+ \overset{<\text{pH}_{zpc}}{\rightleftharpoons} \text{Al} - \text{OH} \overset{>\text{pH}_{zpc}}{\rightleftharpoons} \text{AlO}^- \quad (5)
\]

Thus, the precipitated alumina particles acquired a net negative charge in an aqueous medium at pH > pH_{zpc} and a net positive charge at pH < pH_{zpc}. Furthermore, the pH_{zpc} value of about 8 for the recovered alumina was consistent with the range of 8 to 9.2 reported by [22, 29] for pure catalytic Al₂O₃ particles. This means that the alumina powders recovered from aluminum slags in this study may assume good catalytic-grade properties, which should be further verified in future studies by the measurement of particles surface area and porosity.
Fig. 5. Zeta potential of alumina precipitates at different pHs.

Fig. 6 shows the appearance of the Al$_2$(SO$_4$)$_3$ solution recovered after precipitation by NH$_4$OH at different pH values. It was clear that the pH value had a strong influence on the opacity of the solution and the rate of precipitation, where the solution became increasingly opaque as the pH has increased. The increase in the opacity of the solution was due to the increased rate of alumina precipitation as (Al$_6$(OH)$_{10}$(SO)$_{4.5}$H$_2$O) with the addition of NH$_3$OH according to the following reaction [18]:

$$10 \text{NH}_4\text{OH} + 3 \text{Al}_2(\text{SO}_4)_4 \rightarrow \text{Al}_6(\text{OH})_{10}(\text{SO})_{4.5} \text{H}_2\text{O} + 5 (\text{NH})_4\text{SO}$$ (6)

Therefore, it was important to add the maximum possible amount of NH$_3$OH to achieve the maximum precipitation rate of alumina according to the equation (6). However, the precipitation of alumina reached its maximum rate when all Al$^{3+}$ ions in the solution precipitated as (Al$_6$(OH)$_{10}$(SO)$_{4.5}$H$_2$O), that is what happened at a pH > the neutralization pH between Al$_2$(SO$_4$)$_3$ acid and NH$_4$OH base. While a large increase in pH above the neutralization pH by the addition of NH$_3$OH did not result in further precipitation. It just gave an excess of NH$_3$OH and the solution was therefore diluted. This is why we preferred to stop the addition of ammoniac at a pH of 9.5 slightly higher than the neutralization pH. This pH was considered to be optimal because it gave the higher opacity of the solution correspond to the higher precipitation rate of alumina (Fig. 6).
The variation in the precipitation pH by adding NH₄OH lead always to the formation of the same aluminum hydroxide phase, but with different morphologies (size and shape) [30, 31]. Fig. 7 shows the granulometric distribution of the precipitated alumina particles at different pH levels measured by zetameter. It can be seen that there was an inversely relation between the precipitated alumina particle size and the medium pH, where the particles size decreased from 23 to 0.45 nm by raising of pH from 4.5 to 9.5. The obtaining of fine particle size in the suspension can lead to fine particles of the final recovered alumina powders. Therefore, it was necessary to choose the higher possible precipitation pH to obtain fine powders, but our process stopped at pH of 9.5, because as it was shown previously the large raising of the pH above the neutralization pH did not lead to any more alumina precipitation.

3.3. Microstructural and morphological characteristics of the recovered alumina

The aluminum hydroxide powder obtained after the drying of the precipitated gel at 80 °C presents an amorphous structure [13, 26]. This powder was calcined at 1200°C for dehydroxylation and desulfurization. During this calcination, a crystallization and some phase transformations were also performed. The XRD spectrum of the alumina powder obtained after the calcination is shown in Fig. 8. The intense and sharp diffraction peaks indicated that the obtained powder was of high crystallinity. Moreover, in comparison with the spectrum of aluminum slags, the peaks of aluminum (Al), aluminum hydroxide (Al(OH)₃), magnesium
aluminate (Al₂MgO₄) and calcite (CaCO₃) were disappeared. The disappearance of these peaks indicated that the extracted alumina was of high purity. It also indicated that the chemical process used in this work was effective in extracting and recovering almost all of the aluminum (Al) contained in the slags in the form of alumina (Al₂O₃). On the other hand, the comparison of the XRD pattern of the synthetized alumina with the international database (ICDD) showed that this latter was a single corundum phase (α-Al₂O₃) (ICSD 01-081-1667 [32]). Similar observations were also made in previous studies by the calcination of aluminum hydroxide powders at the same temperature of 1200 °C [13, 26, 31]. However, the use of lower calcination temperatures in some other studies has led to the recovery of other alumina phases from aluminum slags, such as γ-alumina at 600 °C [22], η-alumina at 900 °C [26], α+η alumina at 1000 °C [13] and α+γ alumina at 1050 °C [28]. This means that the appropriate structure can be obtained for any application by simply changing the calcination temperature. In this study, we have focused only on the most stable polymorph α-alumina, which can be used in a wide range of applications especially at high temperatures.

Fig. 8. XRD patterns of the used aluminum slags and the recovered alumina powder.

The Fourier transform infrared analysis (FTIR) of the synthetized alumina powder in the region of wave numbers 400–4000 cm⁻¹ is shown in Fig. 9. A peak observed at around 450 cm⁻¹ may be due to Al–O bending vibrations in the AlO₆ octahedral units [1, 13]. An intense
and broad absorption peak due to the stretching Al-O modes was also found in the range of 550 to 850 cm\(^{-1}\) [13, 33–35]. The absorption band at 1635 cm\(^{-1}\) was due to the bending vibrations of H-O-H bonds in the hydration water [29], where the residual traces related to water were due to hygroscopic nature of the KBr pellets employed as mounting material [13]. The bands observed around 2845, 2926, 3428 and 3785 cm\(^{-1}\) were due to the stretching vibration of -O-H. It is quite possible that this effect is due to the moisture adsorbed on the surface of the synthesized powders [13,36]. A consolidated list of the wave numbers and the attribution of bands of the calcined powder was given in Table 2. The FTIR result may account for the approximated chemical composition [AlO\(_x\)(OH)\(_y\)zH\(_2\)O] for the surface of the synthetized \(\alpha\)-Al\(_2\)O\(_3\). This surface composition was similar to those suggested for partially dehydroxylated Al\(_2\)O\(_3\) surfaces [29]. Hence, the FTIR spectrum of the extracted \(\alpha\)-Al\(_2\)O\(_3\) indicated that they expose largely clean surfaces constituting aluminum oxide and hydroxide, as well as H\(_2\)O molecules. Moreover, no clear peaks of sulfur or carbon contaminants found to be on their surface.

Table 2. Attribution of the bands of the calcined alumina powder vs. the wave number.

| Wave number (cm\(^{-1}\)) | Attribution     |
|---------------------------|-----------------|
| 2850, 2926, 3428 and 3785 | \(\nu\)(O-H)    |
| 1635                      | \(\delta\)(H-O-H) |
| 550-850                   | \(\nu\)(Al-O)   |
| 450                       | \(\delta\)(Al-O) |

![Fig. 9. FTIR spectra of the alumina powder calcined at 1200 °C.](image)

Fig. 9. FTIR spectra of the alumina powder calcined at 1200 °C.

Fig. 10 (a) shows the particle size distribution of the synthesized alumina powder measured by laser diffraction particle sizing method. As it can be seen, the recovered alumina showed a
wide particles size distribution that ranged from 40 nm to 20 µm. The bi-modal character of the particle size distribution was very clear in the size distribution curve. Two peaks were found corresponding to two populations, the first one lower than 0.6 µm and the second one higher than 0.6 µm. The observation of the synthesized powder by SEM (Fig. 11) showed that the coarse fractions (≥0.6 µm) were due to the agglomeration of the small particles (≤0.6 µm), which was due to the high energy of α-Al₂O₃ nanoparticles surfaces thermodynamically unstable at room temperature [37]. This agglomeration was mainly insured by the weak hydrogen-type bonds between the small particles due to the presence of (OH) groups on the particles surfaces as shown in the FTIR results (Fig. 9). By the use of the cumulative presentation of the particle size (Fig. 10 (b)), it can be seen clearly that about 51% of the particles were lower than 0.6 µm and the remaining 49% were higher than 0.6 µm, while the average particle size (d50) of the grains was found to be in the order of 400 nm. This large particle size distribution tends to maximize particle packing density. Where the large particles can form a rigid structure, and the fine active particles form the matrix and bind the large particles after sintering, which is very suitable for certain applications such as refractory materials [17] and electrode-coated for lithium-ion batteries [38]. It is worth noting that some applications like catalysis, abrasives and nanocomposites need disperse fine α-alumina particles with narrow particle size distribution [37]. However, this alumina powder grade can be obtained easily using the recovered alumina through many methods like sieving [39], supersonic vibration [40] or by the selective corrosion and refined fractionated coagulation separation [37].

Fig. 10. The particle size distribution of the synthesized powder.
The morphology and the chemical composition of the recovered α-alumina powders are of great importance. Where they can have a considerable impact on the microstructure, catalytic sensitivity and mechanical behavior of the elaborated materials [6]. In agreement with laser diffraction particle sizing results, SEM observation of the synthesized α-alumina in Fig. 11 showed a wide particle size distribution, with particles sizes ranging from submicron to tens of micrometers. The individual α-Al₂O₃ particles have almost a morphology of short whiskers with irregular ends, each approximately 1μm in average length and 0.2 μm in average diameter. These submicronic whiskers tended to stick together to form micro-particles. As a result, the synthetized α-Al₂O₃ powder showed a bimodal distribution nature as shown in Fig. 11. It should be mentioned that other morphologies of α-Al₂O₃ extracted from aluminum slags can be obtained, such as the spherical morphology [41] or the irregular morphology [28]. These differences in the morphology were mainly due to the differences in the used products and experimental protocol (pH, temperature, type and concentration of leaching acids, etc). The atomic composition of the recovered α-Al₂O₃ powder was also analyzed by EDS, which revealed the presence of high amount of Al and O (99.59 at.%) with very small amounts of impurities confirming the high purity of the recovered alumina (Fig. 11 (d)).
**Fig. 11.** SEM micrograph of the recovered alumina powder with different magnifications (a) (b) (c) and the composition of the recovered powder (d).

4. Conclusions

The objective of this study was to treat aluminum slags in order to eliminate their environmental threat and to exploit them in the extraction of high value-added alumina. A controllable acid leaching technique followed by precipitation was used for the extraction. The following important conclusions can be drawn from this study:

1. The coarser aluminum-rich fractions of the slags used in this study were found to be technically more suitable for alumina extraction, compared to the finer oxides-rich fractions used in other studies.

2. A high purity of recovered alumina (99.2%) and a highest extraction efficiency (93.75%) were achieved at an optimum leaching H₂SO₄ concentration of 15%.

3. The XRD analysis revealed that the recovered alumina was a pure and high crystalline corundum (α-alumina).

4. FTIR spectrum of the extracted α-Al₂O₃ indicated that it exposed largely clean surfaces, with no clear signs about undesirable contaminants.

5. The granulometric analysis of the produced alumina showed a bi-modal character of the particle size distribution, ranging from 40 nm to 20 μm.

6. SEM observation of the synthesized α-alumina showed that the individual α-Al₂O₃ particles have almost a short submicron whiskers morphology.

7. The above properties of the recovered alumina powder allow it to be used in the most common applications of α-alumina.

REFERENCES:

[1] X. Su, J. Li, J. Mater. Sci. Technol., 27 (2011) 1011–1015.
[2] Y. Huang, Y. Xia, Q. Long, S. Liao, Y. Li, J. Liang, J. Cai, Ceram. Int., 41 (2015) 2607–2610.
[3] G. Feng, W. Jiang, J. Liu, C. Li, Q. Zhang, L. Miao, Q. Wu, Ceram. Int., 43 (2017) 13461–13468.
[4] L. Liu, X. Zhang, L. Zhu, Y. Wei, C. Guo, H. Li, Z. Für Naturforschung B, 74 (2019) 579–583.
[5] S.H. Lim, K.Y. Zeng, C.B. He, Mater. Sci. Eng. A, 527 (2010) 5670–5676.
[6] W.L. Suchanek, J.M. Garces, P.F. Fulvio, M. Jaroniec, Chem. Mater., 22 (2010) 6564–6574.
[7] G. Paglia, C.E. Buckley, A.L. Rohl, R.D. Hart, K. Winter, A.J. Studer, B.A. Hunter, J.V. Hanna, Chem. Mater., 16 (2004) 220–236.
[8] Y. Wang, J. Wang, M. Shen, W. Wang, J. Alloys Compd., 467 (2009) 405–412.
[9] G.P. Johnston, R. Muenchausen, D.M. Smith, W. Fahrenholtz, S. Foltyn, J. Am. Ceram. Soc., 75 (1992) 3293–3298.
[10] T. Ogihara, H. Nakajima, T. Yanagawa, N. Ogata, K. Yoshida, N. Matsushita, J. Am. Ceram. Soc., 74 (1991) 2263–2269.
[11] E. Kato, K. Daemon, M. Nanbu, J. Am. Ceram. Soc., 64 (1981) 436–443.
[12] H. Noda, K. Muramoto, H. Kim, J. Mater. Sci., 38 (2003) 2043–2047.
[13] I.N. Bhattacharya, P.K. Gochhayat, P.S. Mukherjee, S. Paul, P.K. Mitra, Mater. Chem. Phys., 88 (2004) 32–40.
[14] M.C. Shinzato, R. Hypolito, Waste Manag., 25 (2005) 37–46.
[15] P.E. Tsakiridis, J. Hazard. Mater., 217 (2012) 1–10.
[16] S.O. Adeosun, O.I. Sekunowo, O.O. Taiwo, W.A. Ayoola, A. Machado, Adv. Mater., 3 (2014) 6.
[17] A. Gil, S.A. Korili, Chem. Eng. J., 289 (2016) 74–84.
[18] A. Meshram, K.K. Singh, Resour. Conserv. Recycl., 130 (2018) 95–108.
[19] T. Hiraki, A. Nosaka, N. Okinaka, T. Akiyama, ISIJ Int., 49 (2009) 1644–1648.
[20] J. Hazard. Mater., 217 (2012) 1–10.
[21] I.N. Bhattacharya, P.K. Gochhayat, P.S. Mukherjee, S. Paul, P.K. Mitra, Mater. Chem. Phys., 88 (2004) 32–40.
[22] M.C. Shinzato, R. Hypolito, Waste Manag., 25 (2005) 37–46.
[23] F.H. Kamil, A. Salmiaton, R.M.H.R. Shahruzzaman, R. Omar, A.G. Alsultsan, Bull. Chem. React. Eng. Catal., 12 (2017) 81–88.
[24] A. Gil, S.A. Korili, Chem. Eng. J., 289 (2016) 74–84.
[25] M.S. Reddy, D. Neeraja, Sādhana, 43 (2018) 124.
[26] Md.S.R. Sarker, Md.Z. Alam, Md.R. Qadir, M.A. Gafur, M. Moniruzzaman, Int. J. Miner. Metall. Mater., 22 (2015) 429–436.
[27] T. Hiraki, T. Miki, K. Nakajima, K. Matsubae, S. Nakamura, T. Nagasaka, Materials, 7 (2014) 5548–5553.
[28] B.R. Das, B. Dash, B.C. Tripathy, I.N. Bhattacharya, S.C. Das, Miner. Eng., 20 (2007) 252–258.
[29] B. Dash, B.C. Tripathy, I.N. Bhattacharya, T. Subbaiah, Int J Met. Eng, 1 (2012) 78–82.
[30] E. David, J. Kopac, J. Hazard. Mater., 261 (2013) 316–324.
[31] M. Mahinroosta, A. Allahverdi, J. Clean. Prod., 179 (2018) 93–102.
[32] F. Karouia, Traitement Thermique de Boehmite de Taille et Forme de Particules Contrôlées: Vers l’optimisation Des Propriétés de l’alumine Gamma, PhD Thesis, Université de Toulouse, Université Toulouse III-Paul Sabatier, 2014.
[33] H. Zhang, D. Zhu, S. Grasso, C. Hu, Ceram. Int., 44 (2018) 14967–14973.
[34] A.S. Brown, M.A. Spackman, R.J. Hill, Acta Crystallogr. Sect. A, 49 (1993) 513–527.
[35] J. Shah, M. Ranjan, V. Davariya, S.K. Gupta, Y. Sonvane, Appl. Nanosci., 7 (2017) 803–813.
[36] J.K. Pradhan, I.N. Bhattacharya, S.C. Das, Mater. Sci. Eng. B, 77 (2000) 185–192.
[37] J.S. Lee, H.S. Kim, N.-K. Park, T.J. Lee, M. Kang, Chem. Eng. J., 230 (2013) 351–360.
[38] S. Pu, L. Li, J. Ma, F. Lu, J. Li, Sci. Rep. 5 (2015) 11575.
[39] K. Nakahira, T. Hotta, M. Naito, N. Shinohara, Y. Cho, S. Katori, H. Emoto, T. Yamada, T. Takahasashi, M. Okumiya, J. Eur. Ceram. Soc., 23 (2003) 1661–1666.
[40] C.J. Papini, W.K. Yoshiïto, D. Gouvêa, R.M.L. Neto, in: Mater. Sci. Forum, Trans Tech Publ, 2005, pp. 73–78.
[41] M. Mahinroosta, A. Allahverdi, J. Environ. Manage., 223 (2018) 452–468.
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Fig. 1. Shape (a) and composition (b) of the used aluminum slags.

Fig. 2. Summary diagram of the followed protocol.

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