

PAPER • OPEN ACCESS

Synthesis of α -Alumina Developed from Waste Aluminium using Precipitation Technique

To cite this article: Aiman A. Bin Mokaizh and Jun Haslinda Binti Haji Shariffuddin 2021 *J. Phys.: Conf. Ser.* **1900** 012002

View the [article online](#) for updates and enhancements.

You may also like

- [MAPPING THE SHORES OF THE BROWN DWARF DESERT. III. YOUNG MOVING GROUPS](#)

T. M. Evans, M. J. Ireland, A. L. Kraus et al.

- [Thermodynamic Analysis of Alumina Refractory Corrosion by Sodium or Potassium Hydroxide in Glass Melting Furnaces](#)

Karl E. Spear and Mark D. Allendorf

- [Codeposition of Alumina Particles from Acid Copper Sulfate Bath](#)

Hidetaka Hayashi, Shinya Izumi and Isao Tari



ECS The Electrochemical Society
Advancing solid state & electrochemical science & technology

242nd ECS Meeting

Oct 9 – 13, 2022 • Atlanta, GA, US

Presenting more than 2,400 technical abstracts in 50 symposia

 **ECS Plenary Lecture featuring M. Stanley Whittingham,** Binghamton University Nobel Laureate – 2019 Nobel Prize in Chemistry

 Register now!



Synthesis of α -Alumina Developed from Waste Aluminium using Precipitation Technique

Aiman A. Bin Mokaizh^{1,2*}, Jun Haslinda Binti Haji Shariffuddin¹

¹ Faculty of Chemical and Natural Resources Engineering, Universiti Malaysia Pahang, 26300, Gambang, Pahang, Malaysia

² Department of Chemical Engineering, College of Engineering and Petroleum, Hadhramout University, Al-Mukalla, Yemen

*Corresponding author:: Ay.stmy@gmail.com

Abstract. The heated aluminium containers were added to a solution of 8.0 M H₂SO₄ solution, which eventually yielded a solution of Al₂(SO₄)₃ after a series of stepwise precipitation reactions. Al₂(SO₄)₃ was presented in large quantities of H₂SO₄ in the white semi-fluid solution; there were some unreacted aluminium parts. The solution was subjected to filtration and then mixed with anion in a ratio of 2:3, this resulted in the formation of a white layer Al₂(SO₄)₃•18H₂O. Thereafter, Al₂(SO₄)₃•18H₂O was calcined in an electric oven for 2 h at various calcination temperatures (500, 700, 900, 1100, and 1300°C). The mixtures were heated and cooled at a rate of 10°C/min. XRD was employed to investigate variations in temperature and determination of elemental accumulation of alumina produced. Al₂(SO₄)₃•18H₂O was due to a series of aluminium compositions produced from dehydration. All transitions from low temperatures to aluminium phases were converted to high-temperature α -Al₂O₃. The results obtained from X-ray disintegration showed that the α -Al₂O₃ phase was obtained at a reaction temperature of about 1150°C and above.

Keywords: α -Alumina, calcination, Phase-transformation, Precipitation

1. Introduction

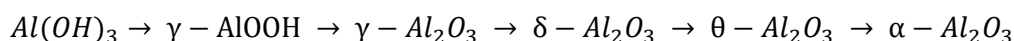
Alumina is an important ceramic acid, it possesses numerous industrial applications including, but not limited to high temperatures and micro-electronics. Alumina nano-powders are used for a range of uses in specialized fields but are not limited to telecommunications, material science, optoelectronics and fine ceramic composite materials. [1]. Alumina is represented as Al₂O₃ with 101.96 gmol⁻¹ molar mass and 3.95 to 4.1 gcm⁻³ density. Generally, alumina has a high melting point and hot spots in 2072 and 2977 °C. Other defining characteristics are an open and low neutron cross-sectional area for microwave radio frequencies with high-pressure strength, and high resistivity to abrasion, high chemicals, heat shock resistance, and a high refractory, and dielectric strength [2]. In recent years, α -Al₂O₃ nano-powders have been used for high-grade cleaning of gas-phosphatic compound, hydrothermal synthesis [2], plasma synthesis, left-gel method [3] sulphate solutions [4], hydrolysis of metal oxide, breakdown of organogenic-metal compounds in supercritical liquids, and aerosols [5].

Most of these technologies are used to structure nanometer particulates in an amorphous or gamma state. In many methods adopted, alumina loses its calcination, which repeats the nanocrystalline nature, since the γ -alumina turns into α -alumina, which quickly multiplies grain yield [6]. The growth of this grain eliminates the useful properties of the initial nanoscale alumina salts. Apart from the



aforementioned methods, the acids, which include sulfuric acid, hydrochloric acid and nitric acid, may react with the aluminium to yield aluminium salt. Hydrochloric and sulfuric acids were utilized within 3 to 12 m for the preparation of high-molecular α -alumina [6-7]. The implication of using a low concentration of acids is that it results in a long time and an incomplete reaction with aluminium due to the lower amount of heat produced and a low reaction rate. The reaction of aluminium with high concentrations of nitric acid produces nano-grids aluminium nitrate $\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$. At increased temperature, the nano-hydrate aluminium nitrate salt produces aluminium oxide, calcium and emits nitrogen dioxide gas, with red-brown gaseous gas [8]. Additionally, the reaction of aluminium with high concentrations of sulfuric acid results in the formation of octa-decahydrate aluminium sulfate $\text{Al}_2(\text{SO}_4)_3 \cdot 18\text{H}_2\text{O}$.

The calcination salts at an increased temperature of 350°C , initiate decay and the aluminium process begins. At this stage, the sulfur trioxide gas is removed from the colourless and acute smoke gas. At 900°C , the whole part of aluminium sulfate is aluminium. Octa-decahydrate aluminium sulfate of $667.42 \text{ gmols}^{-1}$ molar mass, 1.62 g cm^{-3} density and 86.5°C melting point is then formed. At 770°C , the water is converted to anhydrous aluminium sulphate. Besides, its little dissolution in alcohol suggests the presence of salt in the amorphous form at 800°C . After this, it is converted to $\epsilon\text{-Al}_2\text{O}_3$ at 900°C and $\alpha\text{-Al}_2\text{O}_3$ at $1100\text{-}1200^\circ\text{C}$ [9]. $\text{Alk}_{11}_3 \cdot 6\text{H}_2\text{O}$ alkoxide trichloride with Al-chloride is formed in the presence of chloride acid alumina and separated by alumina at 300°C [10]. Alumina occurs in a number of crystalline metastases structure, these include η -, γ -, δ -, θ -, β -, κ -, χ -, and α -alumina. Based on thermodynamics, the more stable phase of the rectangular α -alumina is $A = 4.758 \text{ \AA}$ and $c = 12.991 \text{ \AA}$ [11]. The separation of $\text{Al}(\text{OH})_3$ as aluminium compounds corresponds to a more stable phase at high temperatures in the aluminium structure. After the calcination temperature rises, the conversion sequence is presented in the equation below.



Several variables, including particle size, heating intensity, emission and ambient impact the transition of the aluminium process due to the impact on transition kinetics [11]. Based on a previous study, $\gamma\text{-Al}_2\text{O}_3$ is calcified at 800°C , transforming to Al-O_2 [12]. When $\delta\text{-Al}_2\text{O}_3$ reaches calcium at 1000°C , it is converted to $\theta\text{-Al}_2\text{O}_3$; finally, $\theta\text{-Al}_2\text{O}_3$ is converted to $\alpha\text{-Al}_2\text{O}_3$. Despite the aforementioned attributes, a small concentration of contaminants can change the barrier to alter the transformations. For instance, the calcination of $\gamma\text{-Al}_2\text{O}_3$ with a 3% platinum mixture can be lower than 1100°C in $\alpha\text{-Al}_2\text{O}_3$ [6]. In general, the aluminium cans were not converted to alumina in the processing of ceramic items. Thus, this present study was carried out to produce α -alumina by utilizing aluminium from waste collection sites. Specifically, aluminium drink boxes were selected for this conversion process due to their availability in large numbers as solid wastes in many countries. Primarily, they comprise aluminium with magnesium, manganese, iron, silicon, copper, and other metals.

2. Experimentation

All the materials used were initially removed from aluminium boxes. Sulphuric acid (H_2SO_4), ethanol ($\text{CH}_2\text{CH}_3\text{OH}$) and purified water were easily accessible in the research lab. The aluminium boxes were subjected to heating at a temperature of 600°C in a carbonate furnace for 2 h; the coating and contamination were then removed. The heated aluminium cans were broken into smaller sizes for an increased reaction surface area using a sulfuric acid solution. Then, the boxes were distributed on the calculated stoichiometric numbers using $\pm 0.0001 \text{ g}$ accuracy of Digital Analytical Balance. In a previous study, sulphate acid from (3-12) M of alumina for high alumina was utilized [7]. In this research, 8.0 M plus sulphuric acid solution, H_2SO_4 was used due to its distinctive strength and excellent reactive capacity. The H_2SO_4 solution was gradually flushed away from the heating aluminium sheets because the reaction was strongly influenced by a large number of heat and gas smells. The cup stick was employed to adequately stir the mixture. The mixture was then filtered using the Buchner funnel to obtain a white solution of aluminium sulphate. Consequently, acidic aluminium sulphate solution was allowed to settle with a 2:3 ratio utilizing ethanol. The mixture was then left to cool, ultimately leading to a white precipitate that was further flushed with ethanol; the precipitate was

dried in an oven for 15 h at 110°C. The clean homogeneous substance was subjected to calcination in an electric oven at 500-1300°C for 2 h; heated and cooled at a rate of 10 min⁻¹.

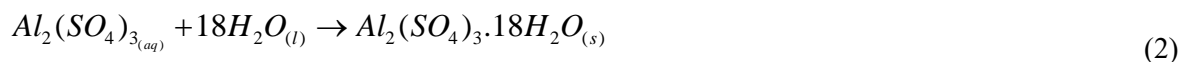
Pre-calcination and post-calcination mass percentage of weight loss were determined and recorded. Besides, alumina samples were subjected to the XRF test to determine the sample weights and elements such as aluminium and sulfur. XRF is a non-destructive analytical method employed for the identification of the contents of the sample elements. The EDX-720/800HS/900HS X-ray spectrometer and the XRF results were expressed in terms of the percentage of elements and potential oxide for the elements. Variations in the percentage of Al₂O₃ and SO₃ weighted percentages were determined to define the specific influence of the reaction of aluminium sulphate. Moreover, the characterization of the aluminium specimens was done using an X-ray diffractometer (Rigaku, AX-2500), 0.1542 nm CuK α was utilized to determine the phases present at various calcined heat range. The data mitigation procedures quickly detected peak status, relative density, and calculated the internal crystal d-range. Relatively unknown crystalline materials have been completely identified with full ICSD powder diffraction files.

3. Results and Discussion

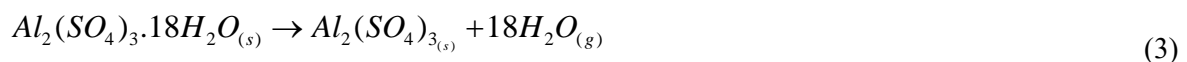
During the formation of aluminium sulfate, variables such as concentration, temperature and pressure were optimized. This process was converted to aluminium sulfate Al₂(SO₄)₃ after aluminium containers were added to 8 M H₂SO₄ to form aluminium sulphate. The chemical reaction of aluminium with H₂SO₄ is shown in Equation (1) for the formation of hydrogen gas and aluminium sulfate.



During the rain, a high quantity of ethanol was added to the white and semi-aluminium sulphate Al₂(SO₄)₃ to form a mixture. Thereafter, the resulting white precipitate octa-decahydrate was dissolved in aluminium sulfate, Al₂(SO₄)₃·18H₂O, salt, and ethanol. The white precipitate was thereafter subjected to filtration and rose with short ethanol to extract excess acid over salt. Subsequently, the filtrate was subjected to drying in a furnace for 15 h at 110°C and a small amount of powder was crushed. The XRD analysis of white sediment is presented in Figure 3. These findings were consistent with XRD results for Al₂(SO₄)₃·18H₂O, Al₂(SO₄)₃·17H₂O, and Al₂(SO₄)₃·16H₂O. The chemical reaction resulting in the formation of Al₂(SO₄)₃·18H₂O from Al₂(SO₄)₃ is detailed in Equation (2).



When the relatively complex molecule Al₂(SO₄)₃·18H₂O is converted at a high temperature, alumina salts are the products formed. Calibration at Al₂(SO₄)₃·18H₂O loses bonding in complex molecules and provides sufficient variations to form new compounds that are simple and stable for the elements. At a high calcined heat range, Al₂(SO₄)₃·18H₂O is converted to alumina via completely evaporating water vapour, which then loses sulfur trioxide gas; the corresponding reactions are presented in Equations (3) and (4) [9]. Some volatile foreign substances that are in the gaseous phase also fail. Calcinations produce thermal energy for alumina, and this is the most stable crystalline alumina; this is important for altering alumina.



Aluminium boxes for commercial products include aluminium boxes of more than 97% of aluminium. Approximately 3% of trace elements compose of 1% each of magnesium and manganese, and 1% of iron, silicon and copper. To evaluate the samples, XRF analysis was performed on Al₂(SO₄)₃·18H₂O before and after calcination at 500-1300°C for 2 at each stage. The weighted contents of elements in the sample after each XRF test are given in Table 1.

Table 1: Weight of the oxides available after calcium for 2 h at 500-1300 °C in $\text{Al}_2(\text{SO}_4)_3 \cdot 18\text{H}_2\text{O}$

Calcination Temperature (°C)	Al		S		Z	M	Fe	C
	$_2\text{O}_3$	O_4	iO_2	nO	nO	$_2\text{O}_3$	uO	
500	12 .40	8 7.23	0 .08	0 .03	0 16	0. 10	0. 10	-
700	23 .75	7 5.84	0 .05	0 .02	0 18	0. 16	0. 16	-
900	93 .74	4. 61	0 .07	0 .15	0 65	0. 74	0. .04	0
1100	94 .87	3. 32	0 .42	-	0 73	0. 66	0. 66	-
1300	96 .40	1. 40	0 .90	0 .13	0 65	0. 52	0. 52	-

Figure 1 displays weight changes (expressed in percentage) that were caused by $\text{Al}_2(\text{SO}_4)_3 \cdot 18\text{H}_2\text{O}$, both pre and post calcination with various thermal ranges, with traces of aluminium oxides and sulphate ions. Figure 2 indicates the rate of loss of weight for the calcified $\text{Al}_2(\text{SO}_4)_3 \cdot 18\text{H}_2\text{O}$. The removal of water from sulfur trioxide and calcareous $\text{Al}_2(\text{SO}_4)_3 \cdot 18\text{H}_2\text{O}$ salts resulted in a weight loss. At different temperatures, each mixture was extracted. The diffusion of water, sulphur and salt relies on the heat range of the calcination. The amount of diffusion increases as calcine temperature increases. The percentage change before Al_2O_3 calcium is 12.51% and 86.88% of SO_3 of $\text{Al}_2(\text{SO}_4)_3 \cdot 18\text{H}_2\text{O}$. At 500°C, weight content was almost content all along the way. $\text{Al}_2(\text{SO}_4)_3 \cdot 18\text{H}_2\text{O}$ calcined. Notwithstanding, a weight loss of 54.78% was observed, which was as a result of the removal of ethanol and water below 500°C. The elements that make up these compounds are not detectable using XRF due to their characteristic light nature. The procedure for dehydrating $\text{Al}_2(\text{SO}_4)_3 \cdot 18\text{H}_2\text{O}$ salt consists of numerous transformation stages, usually from high to low hydrating stage. Thereafter, it is subjected to full dehydration to produce $\text{Al}_2(\text{SO}_4)_3$. The traces of various hydrated salts such as $\text{Al}_2(\text{SO}_4)_3 \cdot 16\text{H}_2\text{O}$, $\text{Al}_2(\text{SO}_4)_3 \cdot 14\text{H}_2\text{O}$, $\text{Al}_2(\text{SO}_4)_3 \cdot 9\text{H}_2\text{O}$, and $\text{Al}_2(\text{SO}_4)_3 \cdot 5\text{H}_2\text{O}$ may be found. Below a temperature of 650°C, no significant variation was observed in the number of sulfate ions recorded; hence, the high rate of reduction in weight of $\text{Al}_2(\text{SO}_4)_3 \cdot 18\text{H}_2\text{O}$ below 650°C was conclusive as a result of the salt being dehydrated.

Figure 1 reveals that the calculated sulphate ions that occurred at 600°C. The weight temperature of Al_2O_3 arose due to the corresponding increase in temperature. Sulfate ions are separated into sulfur trioxide and get eliminated through gas, this process aided the increased weight loss. Improving the calcination temperature causes weight loss. At this temperature, it began to produce sharp and colourless sulphur trioxide gas. Examples of weight loss of sulfate ions in the calcination process at 800°C, 900°C and 1000°C are provided in figures (1) and (2); these show a significant reduction in weight loss after being calcined. The aforementioned changes took place largely as a result of the destruction of a larger percentage of sulfur trioxide gas as the sulfate ions emitted at this temperature. When calcinating at about 1000°C, 86.41% rate of weight reduction was observed, which almost immediately did not change the sample at high-temperature calcium. Weights of aluminium oxide corresponded to 94.30% while the weight of sulfate ions corresponded to 4.58%. The figures showed that at 1000°C almost sulphate ions were disrupted. Alumina was therefore the calcareous residue. Figure (1) indicates that at heat above 1000°C calcenes had a small amount of isochron ion because small concentrations of sulphur trioxide molecules had been cut off from alumina. About 1.18% of sulfate ions were found at calcined samples at 1300°C. Thus, calcination or higher calcination temperature serves as the optimum condition for sulfur trioxide molecules to go out of the alum.

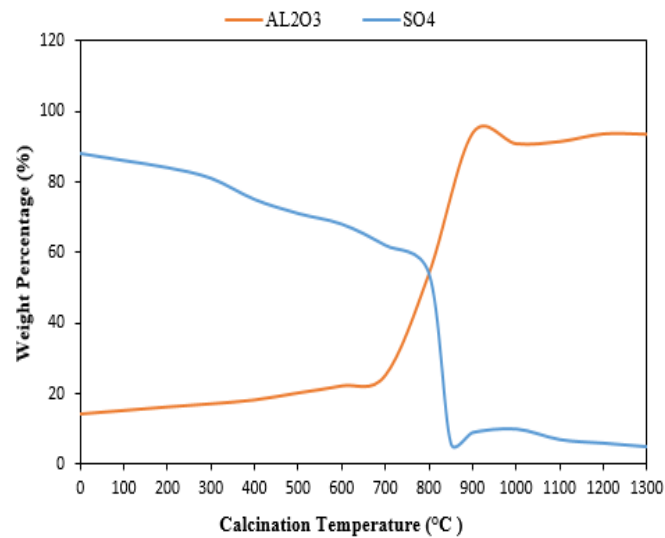


Figure 1. $\text{Al}_2(\text{SO}_4)_3 \cdot 18\text{H}_2\text{O}$ after calcination for 2 h at 500-1300°C with the weight of aluminium oxide and sulfate ions.

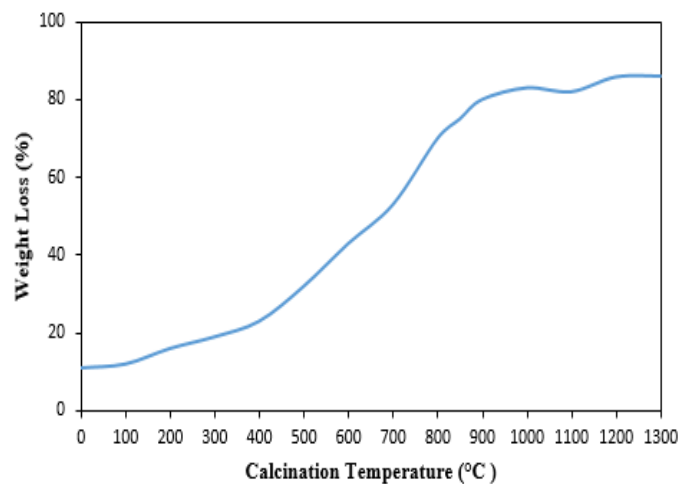


Figure 2. The percentage of $\text{Al}_2(\text{SO}_4)_3 \cdot 18\text{H}_2\text{O}$ weight loss after calcination for 2 h at 500-1300°C

Figure 3 presents the findings of the white solid XRD analysis produced through precipitation before and after calcification at 500 to 1300°C. XRD results indicate that before calcification, the sample corresponds to XRD of $\text{Al}_2(\text{SO}_4)_3 \cdot 18\text{H}_2\text{O}$. At 400°C, the phase XRD results were found to be aluminium sulfate $\text{Al}_2(\text{SO}_4)_3$. The XRD contains some unknown columns. Thus, there may be other crystalline phases such as different hydrate aluminium sulphates or compounds in the sample. At this temperature, the production of alumina could not be achieved since the temperature was relatively low; so, it is likely that sulfate ions will be separated. After calcining at 400°C, the peaks of the aluminium sulfate phase were combined with the ICS code 073249. 2 hills (or Miller Index) showing the presence of aluminium sulfate 20.87° (104), 24.56° (11 (3), 31.76° (024), 34.74° (11-6), 41.53° (303), 43.98° (11-9), and 45.75° (306) [12,13]. It is worthy of mentioning that as the temperature rose from 400 to 500°C, there was a corresponding rise in density of the aluminium sulfate phase, with XRD results. With other moistening agents, the presence of an aluminium sulfate phase at 800°C was relatively low at 500°C, but that the water molecules and contaminants have already been eliminated at the aforementioned heat range. Structurally, aluminium sulphate appears crystalline and rhomboidal at 500 and 700°C. When calcined at 900 °C, the aluminium sulphate lost its crystalline structure and

small amorphous pellets were $2^\circ\text{C} = 43.98^\circ$. It appears at -47° and $65^\circ - 68^\circ$, these two sandstones depend on the interference of signals X-X, G, and B alumina. Previous studies had revealed that the γ , θ and δ -alumina peaks were in the range of 43.95 to 46.89 DEG and 64.79-67.80 DEG [14-16]. Within the range of the aforementioned heat, alumina sulfate phases lose contact and naturally amorphous.

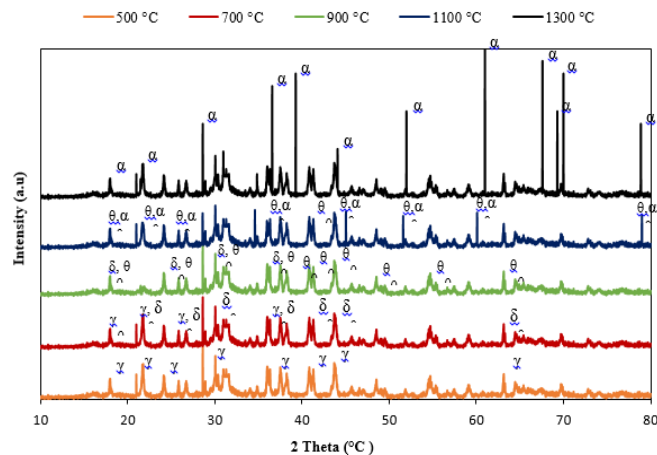


Figure 3. XRD results for 2 h from 500 to 1300°C for $\text{Al}_2(\text{SO}_4)_3 \cdot 18\text{H}_2\text{O}$, where $\gamma = \gamma$ -Alumina; $\theta = \theta$ -Alumina; $\delta = \delta$ -Alumina; $\alpha = \alpha$ -Alumina

This finding is in line with the observations of previous studies, as calcium samples lost contact and amorphous at 1000°C , with the exemption that the density of amorphous ribs has increased. Several small and sharp peaks with an amorphous groove in comparison to the calcination results at 900 and 1000°C emerged for calcined with 1100°C . The findings above show that some aluminium particles showed that in this heat range there was a structurally crystalline alumina process that was reorganised to create α -alumina [17]. α -, γ -, δ -, and θ -alumina were present in a single and reduced order. From structural analysis, it is revealed that the alumina crystal tends towards crystallinity, and the crystalline system is rhomboidal. The calcined samples at 1100°C were started with conversion to α -alumina with a quite high structurally crystalline appearance. The aforementioned result is based on the XRD reference database (ICSD assembling code 025778). The crystalline structure of this alumina is rhombic with 3.95 g cm^{-3} density and $a = b = 4.7510 \text{ \AA}$ and $c = 12.9700 \text{ \AA}$. XRD peaks showing the existence of traces of α -alumina 2θ (Shaft Index) = 25.94° (012), 36.17° (104), 38.84° (110), 44.81° (113), 51.38° (024), 58.26° (116), 60.12° (211), 62.65° (018), 68.17° (214), 70.54° , and 78.76° (119) [18]. Calcination at 1300°C was similar to that of a calcined sample at 1100°C with clear peaks of α -alumina. The intensities of the XRD hills are intensified and showed that more α -alumina

4. Conclusion

Alumina (Al_2O_3) was produced from the reaction between the sulfuric acid from the waste aluminium, and the deposition of calcium. The samples were characterized using X-Ray Fluorescence Spectrometers and X-ray powder diffraction. The X-Ray Fluorescence Spectrometers test result indicated that the increase in calcined temperature resulted in a corresponding increase in the amount of alumina, and the sulfate ions were reduced. The highest weight of the alumina reached using calcium was 96.79 % at 1300°C . The aforementioned technique of producing alumina from aluminium wastes α - Al_2O_3 has industrial application due to its reproducibility; hence, many crystalline α - Al_2O_3 are completely reproducible. Furthermore, the cost of producing them are relatively low since the needed raw materials for their production are abandoned aluminium boxes, sulfuric acid and ethanol [19]. Aluminum sulphate $\text{Al}_2(\text{SO}_4)_3 \cdot 18\text{H}_2\text{O}$ calcined vapour, sulphur trioxide and unstable exhaust were removed at high temperatures. Calcined $\text{Al}_2(\text{SO}_4)_3 \cdot 18\text{H}_2\text{O}$ is hydrogen which is a forming alpha alumina extracted from sulphate ions at different heat peaks. The water molecules were reduced to about 600°C and the SO_3 gas was extracted from the salt at 600 to 1000°C . After calcification at

1200°C, the alumina was converted to α -alumina. The findings from XRD analyses indicated that the α -alumina phase (α -Al₂O₃) samples were found after calcination at varying temperatures which were higher than 1200°C. A rise in the calcined temperature ultimately resulted in weight loss. The weight gain had increased after calcinating temperature, but weight loss had increased by around 1000°C.

References

- [1] Ribeiro M J, Abrantes J C, Ferreira J M and Labrincha J A 2002 *Ceram. Int.* **28** 319-26.
- [2] Suchanek W L 2010 *J. Am. Ceram. Soc.* **93** 399-412
- [3] Nguefack M, Popa A F, Rossignol S and Kappenstein C 2003 *Phys. Chem. Chem. Phys.* **5** 4279-89
- [4] Nieto M I and Tallon C 2006 *Adv. Sci. Technol.* **45** 223-30.
- [5] Janbey A, Pati R K, Tahir S and Pramanik 2001 *J. Eur. Ceram. Soc.* **21** 2285-89
- [6] Mishra P 2002 *Mater. Lett.* **55** 425-29
- [7] Gonczy S T and Mitsche R.T. U.S. *Patent* 4615875 7 October 1986.
- [8] Pacewska B and Keshr, M 2002 *Thermoc. Acta* **385** 73-80.
- [9] Bhattacharya I N, Gochhayat K, Mukherjee P S, Paul S and Mitra P K 2004 *Mater. Chem. Phys.* **88** 32-40
- [10] Smith R L, Rohrer G S and Perrotta A J 2001 *J. Am. Ceram. Soc.* **84** 1896-1902
- [11] Shackelford J F and Doremus R H 2008 *Structure, Prop. & Process (Springer:New York USA)*.
- [12] Kloprogge J T, Geus J W, Jansen J B H and Seyken D 1992 *Thermoc. Acta* **209** 265-76
- [13] Arena U 2012 *Waste Manag.* **3** 625-39
- [14] Wood T E and Wilson D M 1990 *U.S. Patent* 4954462
- [15] Young D J 2008 *Elsevier, New York, NY, USA*.
- [16] Yang R J, Yen F S, Lin S M and Chen C C 2007 *J. Cryst. Growth* **299** 429-35
- [17] Ogihara T, Nakajima H, Yanagawa T, Ogata N and Yoshida K 1991 *J. Am. Ceram. Soc.* **74** 2263-69
- [18] Sarkis J, Helms M M and Hervani A A 2010 *Corporate Social Responsib. & Environ. Manage.* **17** 337-54
- [19] Ijburg I I M, Bruin H D, Elberse P A and Geus J W 1991, *J. Mater. Sci.* **26** 5945-49