

Preparing of Alumina from Aluminum Waste

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Abstract:- This research aims to take advantage of Aluminum Foil Waste (AFW) through using it as a raw material for preparing alumina (Al_2O_3), by the precipitation method. The effect of Hydrochloric Acid (HCl) concentration, on the digestion of AFW, was studied by varying it from 1 to 6 M, Sodium Hydroxide (NaOH) was used as precipitation agent, to precipitate the aluminum hydroxide from AlCl_3 obtained from previous step by raising the pH to different values (from 3 to 13) to study the effect of pH on precipitation process. It was found that the best acid concentration to digest the AFW was 5M, also the best pH to precipitate the $\text{Al}(\text{OH})_3$ was 9. Then the $\text{Al}(\text{OH})_3$ was washed, crushed, and dried at 105°C for 24 hr., and then it was converted to alumina by calcination process using different calcination temperatures (550, 675, and 800°C) to study the effect of calcination temperature on the resulting alumina. The purity of the prepared alumina investigated by XRF test, and it was more than 97% for the three samples which calcined at three different temperatures. The study of the evolution of crystalline phases of obtained powders was accomplished through XRD studies. XRD data showed that the samples calcined at 550, 675, and 800°C has $\gamma\text{-Al}_2\text{O}_3$ phase. Also the surface area was tested by using BET method, high surface area was obtained at 550°C about $246\text{m}^2/\text{g}$ and it was decreased with increasing the calcination temperature.

Keywords:- Alumina, Aluminum Waste, Aluminum Foil Waste, XRD, Surface Area.

I. INTRODUCTION

Aluminum is one of the most valuable component materials of municipal refuse in terms of (cigarette and candy casings, cans, wrapping foil, doors, siding, car machine and body portions, icy food dishes, pie plates and other various domestic waste) [1]. Aluminum foil (Al-foil) is one of the largest sources of aluminum waste that is difficult to recycle; so they are either buried or burned for disposal [2].

Aluminum in Municipal Solid Waste (MSW) landfills come from industrial waste, untreated raw curbside trash MSW, and aluminum wastes differently called dross, salt cake, baghouse fines, and other terms. Reactions associated with the presence of Aluminum in MSW landfill can be arise and become problematic issue for landfill processes by producing undesirable liquid leachate, heat, and gases, for example carbon monoxide, ammonia, hydrogen sulfide, and hydrogen [3].

Now MSW recycling is renowned as the most environmentally sound approach that makes a factor of benefits at every level: environmental, social and financial through decreasing greenhouse gas releases and turning materials that would then become waste into a worthy resource [4].

Aluminum waste recycling is important to the environment; actually, recycling 1 kg of aluminum saves 8 kg of bauxite, 14 kW of electricity, 4 kg of chemical products [5]. Aluminum waste can be recycled to yield numerous beneficial products (such as alumina) [6]. Therefore a many studies have been focused on the usage of waste for several uses and applications [7].

Alumina (Al_2O_3) or Aluminum Oxide is the only oxide formed by the metal aluminum and occurs in nature as the minerals corundum (Al_2O_3); diaspore ($\text{Al}_2\text{O}_3 \cdot \text{H}_2\text{O}$); gibbsite ($\text{Al}_2\text{O}_3 \cdot 3\text{H}_2\text{O}$); and most commonly as bauxite, which is an impure form of gibbsite [8]. Although the naturally occurring corundum, ruby and sapphire are also sometimes referred as alumina contains colors due to some impurities, but the usage of the term alumina is limited to the material which is produced from bauxite and used in aluminum production, chemical treating, and industrial ceramics, [9].

Several types of metastable alumina are found beside $\alpha\text{-Al}_2\text{O}_3$, for instance, ρ , γ , η , θ , δ , χ and $\kappa\text{-Al}_2\text{O}_3$. Those types of transition alumina can be formed from treating the aluminum hydroxides or aluminum salts thermally [10].

Alumina powders can be prepared by numerous well recognized methods, these methods can be divided into four main categories: (I) conventional method (Bayer process). (II) Chemical methods (III) thermos physical method and (IV) Mechanochemical Synthesis [11].

Alumina has many appealing properties which makes the material interesting for applications in many different areas. For example it is a very hard substance and its hardness is exceeded only by diamond and a little artificial materials. This property makes alumina suitable for use as an abrasive substance. Additional beneficial properties of the material are high melting point exceeding 2000°C (3632°F), chemical inertness, non-volatility and resistance to oxidation and corrosion. Al_2O_3 is an electrical insulator having high thermal conductivity [12]; it is also used in Glass industry, Catalysts, and Fillers [8]. Alumina adsorbents are widely used as desiccants in both heated (temperature swing adsorption) and heatless (pressure swing adsorption) dryers. They exhibit a large surface area and high strength against crushing and stable physical and chemical characteristics, even in high temperature and corrosive environments [13]. In addition, Alumina, in

general, is widely used as adsorbent to remove dissolved pollutants from contaminated water. Various chemical species, especially ions, are known to be adsorbed onto alumina [14].

Accordingly, in the present work, we report the green synthesis of alumina powder by the simple and cost-effective co-precipitation method using NaOH as precipitant and aluminum foil waste as raw material in order to get rid of those wastes by turning it into a useful product. In addition the effect of acid concentration, pH value, and calcination temperature were studied during preparing process.

II. EXPERIMENTAL SECTION

A. Alumina Preparation

➤ First: Preparing the AFW

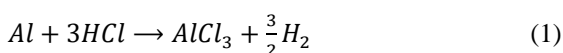
The raw material used in this study, which was AFW, obtained via domestic use from different sources, which was cut to small shredded chips of nearly 10×10 mm as shown in figure 1, and characterized by XRD and XRF tests.



Fig 1:- Shredded AFW

➤ Second: Digestion of AFW

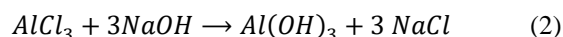
The prepared shredded chips of AFW was treated with digestive substance which was hydrochloric acid of five concentrations (1-6 M) each one alone by the following procedure. Five grams of prepared AFW was weighted and then it was added gradually to exactly 125 ml of each hydrochloric acid concentration that poured in 500 ml glass beaker. Because the reaction strongly occurs and may release large amounts of hydrogen gas and heat, the reaction was done in the outside the laboratory. When the release of the hydrogen gas was ended, a magnetic stirrer (LMS-1003/Korea) was used for 60 min to digest more amount of AFW which was not digested outside the lab. When the digestion process of AFW was completed, the aluminum chloride ($AlCl_3$) was produced according to the following balanced equation:



The produced aluminum chloride contained some impurities due to the fats and dirt stuck on the raw AFW. Therefore, the aluminum chloride was filtered by using vacuum filtration (VP115/American) to remove the impurities. After filtration, the pure solution of aluminum chloride ($AlCl_3$) was obtained. This solution was stored for the next step.

➤ Third: Precipitation Reaction

The precipitation agent in the alumina synthesis process according to this procedure was sodium hydroxide solution (NaOH). NaOH of 5 M concentration was prepared by dissolving 205 g of 97.5% NaOH in one liter distilled water. The precipitation reaction was achieved by adding the precipitation agent to the filtered $AlCl_3$ produced from the previous step. The addition was continuing at room temperature until the pH of the solution reached the desired values, which was measured by using pH meter (WTW- 3110/Germany). To select the best pH value, the pH of precipitation reaction was ranged from 3 to 13. $AlCl_3$ was converted to aluminum hydroxide which was similar to the white gel in appearance. Equation (2) describes the balance reaction in this step:



After maturation, the aluminum hydroxide (white gel) was washed several times with distilled water until the concentration of NaCl salt, the side product, was decreased for obtaining pure aluminum hydroxide product. After washing, the aluminum hydroxide was dried in an electrical oven (Mettler/Germany), at 105°C for 12 hr and then it was crushed using ceramic grinder until it became in the form of fine powder, which was white in color. The $Al(OH)_3$, which was obtained from five different acid concentration and various base pH values, was weighted to select the best acid concentration and the best pH. The best acid concentration which was dissolved all the 5 grams of AFW. The best pH of base gave additional quantity of $Al(OH)_3$, hence, produced more amount and best quality of alumina.

➤ Fourth: Calcination

The calcination process of aluminum hydroxide produced was carried out in different calcination temperatures. The goal was to know the effect of calcination temperature on the quality of the formed alumina. The calcination process was carried out in an electrical furnace (Carbolite CWF1200/ UK), at 550, 675 and 800 °C for 4 hrs. The prepared samples were named A550, A675, and A800. The appearance of the calcined samples was white in color as shown in fig. 2. Equation (3) describes the balance reaction in this step:

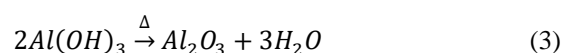




Fig 2:- Prepared Alumina Powder

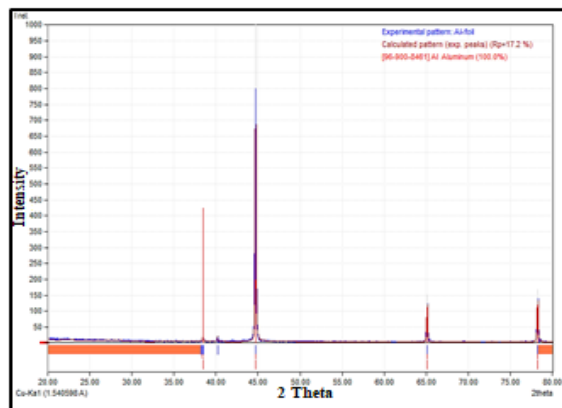


Fig 3:- XRD pattern for AFW

B. Characterization of the Prepared Alumina

The purity of prepared alumina was investigated by X-Ray Florescence (XRF), (Spectro 2011/Germany) according to TurboQuant-Powders method.

Samples crystalline phases were identified by X-ray diffraction (XRD). During the performance of the experiments in Bruker (D2) 2010/Germany, samples were exposed to Cu-K α radiation which was obtained from a copper X-ray tube and operated at 40 kV and 30 mA ($\lambda=1.5406 \text{ \AA}$). The 2 theta scan was from 20 to 80 $^\circ$.

Specific surface area analysis was carried out using the Brunauer-Emmett-Teller (BET) method by N $_2$ adsorption, Quantachrome/ Qsurf 9600 Thermo Finnegan Co. USA.

The results of AFW, shows that AFW contains a high percent of aluminum; about 98.9% also it contains some impurities are listed in table 3. Peaks were detected at $2\theta = 38.4712^\circ, 44.743$ and 65.1176° in XRD pattern, it has very good match with the peaks positions of Al from PCPDFWIN software no.(040787) where its three peaks appeared at $2\theta= 38.472^\circ, 44.738^\circ$ and 65.133° .

III. RESULTS AND DISCUSSION

A. Aluminum Foil Waste Analysis

At first, the foil waste was elementally analyzed using the XRD and XRF instrument and the results are shown in table 1 and figure 3.

B. Results of Prepared Alumina

➤ The Effect of Acid Concentration

The effect of HCl concentration, which is ranging from (1-6 molar) on the amount of alumina yield is shown in figure 4. The amount of aluminum foil waste, pH of the precipitation reaction and calcination temperature, were fixed on 5 g, 9 pH values and 550 $^\circ\text{C}$ respectively.

Compound	AFW (%)
Al	98.9
SiO $_2$	0.53
P $_2$ O $_5$	0.202
SO $_3$	0.007
Fe $_2$ O $_3$	0.255
CaO	0.061
Cl	0.0128
Other elements	0.0322

Table 1:- XRF results for AFW (w/w%)

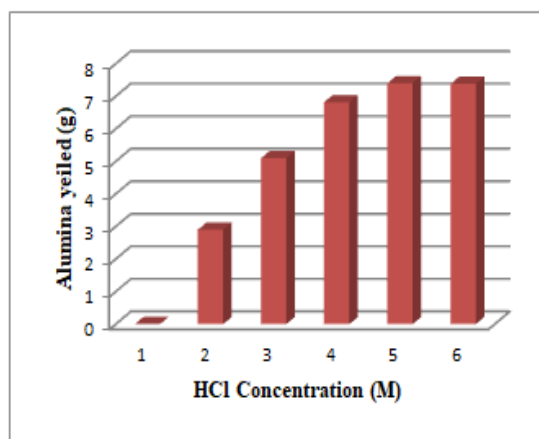


Fig 4:- the Effect of Acid Concentration on Alumina Yield

The results shows that the amount of alumina produced increase with increasing the acid concentration, where it was zero when the acid concentration was equal to 1 M, because the aluminum foil wastes were not digest with the low concentration of hydrochloric acid. At 5 and 6 M the higher amount of alumina was obtained, it was 7.4 and 7.38 grams 5 and 6 M acid concentration respectively. Thus, the concentration of 5 M was chosen as the best acid concentration which was digests all the 5 grams of aluminum foil waste, and convert it into aluminum chloride (AlCl $_3$). The 5 M concentration was used in the further experiment.

➤ *The Effect of pH in Precipitation Reaction*

This step was performed to determine the influence of the pH in precipitation reactions (which was ranging from 3 to 13) on the amount of alumina yield, which was controlled by adding NaOH solution to the AlCl₃ which was obtained from the previous step by using the best HCl concentration with fixed amount of aluminum foil waste and calcination temperature at 5 g and 550°C, respectively. The results are illustrated in figure 5.

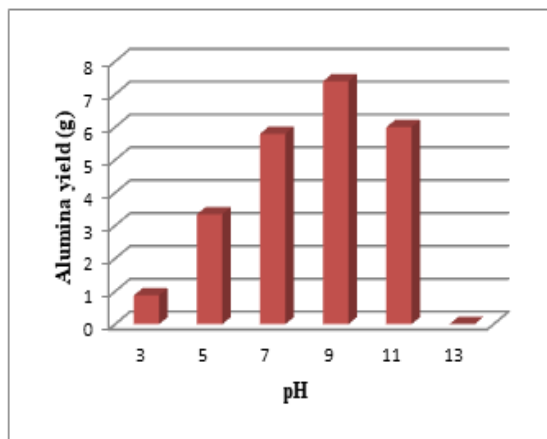


Fig 5:- the Effect of pH on Alumina Yield

From figure 5, note that the increase of alumina produced with increasing the pH value until it reached 9 then it was decrease, while there is no yield at pH 13, other Published studies show that increasing the pH of a solution containing aluminum will form dissolved species consisting of associated Al³⁺ and OH⁻, and when the supply of OH⁻ is sufficient, the aluminum hydroxide will be precipitate. If the pH increases more, this precipitate is re dissolved as an anion contains more than three groups of hydroxide per atom of aluminum [15], the Al(OH)₃ which in turn calcined to get the alumina. So the best pH to precipitate the Al(OH)₃ from AlCl₃ was 9 which gives higher amount of Al(OH)₃ which in turn gives higher amount of alumina (7.4 g).

➤ *The Effect of Calcination Temperature*

The effect of calcination temperature was studied on the purity of prepared alumina, the phase of the alumina produced, and the surface area.

➤ *XRF Results*

The chemical analysis of the prepared alumina at different calcination temperatures obtained by XRF and expressed as oxides wt%, taking into account the aluminum content in the AFW which is relatively high purity.

Figure 6 shows that there is no significant effect of calcination temperature on the purity of prepared alumina, where that all alumina prepared at different calcination temperature, almost have the same purity which was ranging from 97.6-97.7%.

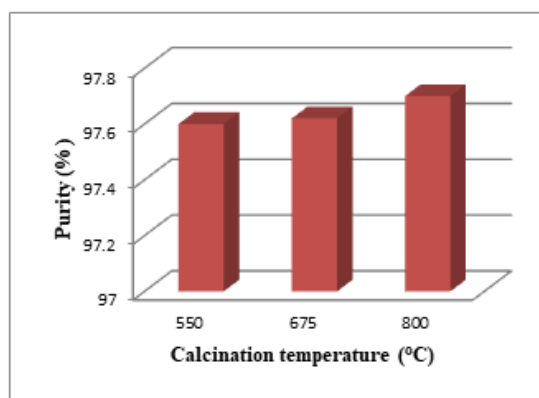


Fig 6:- The Effect of Calcination Temperature on the Purity of Prepared Alumina

The impurities for prepared alumina, shown in table 2 below, are attributed to the presence of impurities in aluminum foil waste, as the foil contains the ratios of iron and silicon, etc. Also the presence of chlorine was from the use of hydrochloric acid in the process of preparation, and some impurities present in NaOH used in precipitation reaction. This result is consistent with the XRF results of γ-Al₂O₃ in previous research [16].

Compound	A550 (%)	A675 (%)	A800 (%)
Al ₂ O ₃	97.6	97.62	97.7
SiO ₂	0.47	0.56	0.36
P ₂ O ₅	0.64	0.65	0.65
SO ₃	0.0006	0.001	0.0006
Fe ₂ O ₃	0.7	0.7	0.67
CaO	0.11	0.12	0.12
Cl	0.36	0.32	0.14
Other elements	0.1194	0.029	0.3594

Table 2:- XRF Results of Prepared Alumina (w/w, %)

➤ *XRD Results*

The phase determination of the alumina was done by using patterns of X-ray diffraction (XRD). Figure 7 show the X-ray diffraction patterns of prepared alumina which calcined at various temperatures (550, 675, and 800°C) for 4 hours.

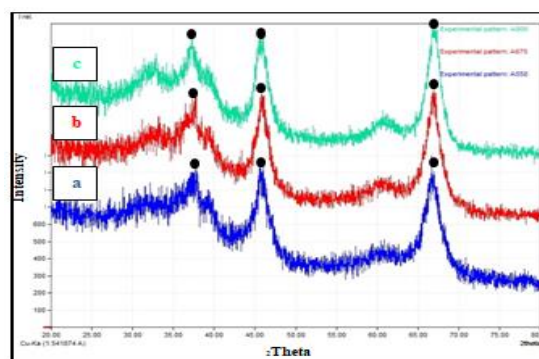


Fig 7:- XRD pattern of prepared alumina, (a) prepared alumina calcined at 550°C, (b) prepared alumina calcined at 675°C, (c) prepared alumina calcined at 800°C

It is clear from the above figure that all the prepared alumina powder at calcination temperatures from 550 to 800°C (a, b, and c) exhibits the same three broad diffraction peaks at 2θ angles 37.6°, 46° and 67°. These reflections are referred to 100% γ-alumina phase (JCPDS card number 10-0425) [4]. That means that in this range of calcination temperature from 550°C to 800°C, γ-alumina is the only identified phase [17]. These results are in similar with other many researchers [18, 19].

➤ *Surface Area Results*

Specific surface areas were calculated by Brunauer-Emmett-Teller (BET) method according to (ISO-9277), It was observed that all prepared alumina had high surface area values (206 to 246 m² g⁻¹), Higher surface area of alumina indicated that this can be used as a very good adsorbent and also as catalytic material [20].

From figure 8 it can be seen that when the calcination temperature increased from 550°C to 800°C, the surface area of the samples was decreased from 246 m² g⁻¹ to 206 m² g⁻¹. This tendency is in agreement with reports of other researchers for alumina [21].

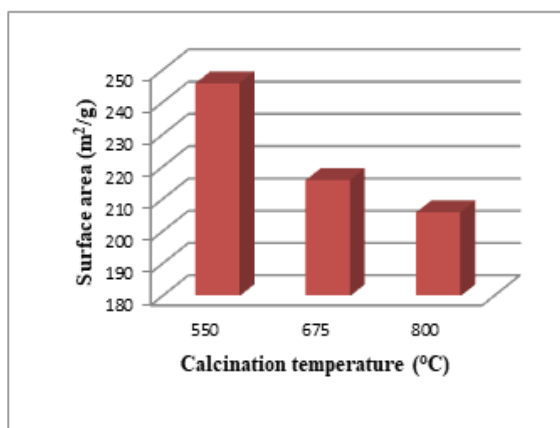


Fig 8:- The Effect of Calcination Temperature on the Surface Area of Prepared Alumina

This decrease in surface area may be due to the destruction of some small micro pores and rebuilding of large macro pores by the sintering process [22].

The specific surface area of calcined samples at 550, 675, and 800 °C are shown in Table 3.

Sample name	Calcination temperature (°C)	Surface area (m ² g ⁻¹)
A550	550	246
A675	675	216
A800	800	206

Table 3:- Specific Surface Area of the Calcined Samples at Different Temperatures

IV. CONCLUSION

Aluminum foil waste can be used as raw material in alumina preparation process, with the best concentration of hydrochloric acid of 5 M. Also the best pH value in precipitation reactions was at 9. XRF results shows High purity alumina was achieved (greater than 97. However XRD results showed that obtaining γ-alumina phase during calcination at temperatures 550, 675, and 800°C. The calcination temperature had a significant effect on the surface area of the resulting alumina. As the heat increased, the surface area was reduced; the highest surface area was 246m²/g at 550°C calcination temperature.

REFERENCES

- [1]. Buryakovskaya O. A., Meshkov E. A., Vlaskin M. S., Shkolnikov E. I., and Zhuk A. Z., "Utilization of Aluminum Waste with Hydrogen and Heat Generation", Materials Science and Engineering 250, 012007, 2017.
- [2]. Bayus J., Ge C. , and Thorn B., "A preliminary environmental assessment of foil and metallized film centered laminates", Resources Conservation and Recycling, Volume 115, Pages 31-41, December 2016.
- [3]. Calder G. V., and Stark T. D., "Aluminum Reactions and Problems in Municipal Solid Waste Landfills", Practice Periodical of Hazardous, Toxic, and Radioactive Waste Management, Vol. 14, No. 4, October 1, 2010.
- [4]. El-Amir A. A. M., Ewais E. M. M. , Abdel-Aziem A. R., Ahmed A., and El-Anadouli B. E. H. , "Nano-alumina powders/ceramics derived from aluminum foil waste at low temperature for various industrial applications", Journal of Environmental Management, Volume 183, Part 1, Pages 121-125, 1 December 2016.
- [5]. Osman A. I., Abu-Dahrieh J. K., McLaren M., Laffir F., Nockemann P., and Rooney D., "A Facile Green Synthetic Route for the Preparation of Highly Active γ-Al₂O₃ from Aluminum Foil Waste", Scientific Reports | 7: 3593, 2017.
- [6]. Li G. , Weizhong W., Long T., Tian Z., Cao Z., Yang J., Gan S., Zhang K., and Huang R. , "A general and facile method to prepare uniform gamma-alumina hollow microspheres from waste oil shale ash", Materials Letters, Volume 133, Pages 143-146, 15 October 2014.
- [7]. López-Andrés S., Fillali L., Jiménez J.A., Tayibi H., Padilla I., and López-Delgado A., "Synthesis of Alumina Based on Industrial Waste Material", Wastes: Solutions, Treatments and Opportunities, (Spain), 1st International Con., 2011.
- [8]. Davis K., "Material Review: Alumina (Al₂O₃)", School of Doctoral Studies (European Union) Journal, Issue 2, p109-114. 6p, 2010.
- [9]. Kumar R., Prabhakar V., and Saini J., "Alumina", International Journal of Current Engineering and Technology, ISSN 2277 – 4106, 2013.

- [10]. Shirai T., Watanabe H., Fuji M., and Takahashi M., “Structural Properties and Surface Characteristics on Aluminum Oxide Powders”, Ceramic foundation engineering research center annual report, Review article, vol. 9, 23-31, 2009.
- [11]. Ghanizadeh S., “Synthesis and Processing of Nanostructured Alumina Ceramics”, Ph.D. Thesis, Loughborough University, England, 2013.
- [12]. Dey S., “Synthesis and Application of γ -Alumina Nanopowders”, M.Sc. Thesis, National Institute of Technology, Rourkela, Orissa, 2011.
- [13]. Serbezov A., Moore J. D., and Wu Y., “Adsorption Equilibrium of Water Vapor on Selexsorb-CDX Commercial Activated Alumina Adsorbent”, Journal of Chemical & Engineering Data, 56, 1762–1769, 2011.
- [14]. Miyazaki A., and Balint I., “Purification of Waste Water Using Alumina as Catalysts Support and as an Adsorbent”, Waste Water - Treatment and Reutilization, ISBN 978-953-307-249-4, 2011.
- [15]. Hem J. D. and Roberson C. E., “Form and Stability of Aluminum Hydroxide Complexes in Dilute Solution”, Geological Survey Water-Supply Paper 1827-A, United States Governments Printing Office, Washington, 1967.
- [16]. Ghoniem M. G., Sami T. M., El-Reefy S. A. and Mohamed S. A., “The production of high purity alumina from solid wastes obtained from aluminum factories”, WIT Transactions on Ecology and The Environment, Vol 180, ISSN 1743-3541, 2014.
- [17]. Martí'n-Ruiz M.M., Pe´rez-Maqueda L.A., Cordero T., Balek V., Subrt J., Murafa N., and Pascual-Cosp J., “High surface area α -alumina preparation by using urban waste”, Ceramics International 35 (2111–2117), 2009.
- [18]. Zhang Y., and Jia Y., “Preparation of porous alumina hollow spheres as an adsorbent for fluoride removal from water with low aluminum residual”, Ceramics International 42(17472–17481), 2016.
- [19]. [19] Liu L., Luo S., Wang B., and Guo Z., “Investigation of small molecular weight poly(acrylic acid) adsorption on γ -alumina”, Applied Surface Science 345 (116–121), 2015.
- [20]. Das B.R., Dash B., Tripathy B.C., Bhattacharya I. N., and Das S.C., “Production of eta-alumina from waste aluminium dross”, Minerals Engineering 20: 252–258, 2007.
- [21]. Suzuki N. and Yamauchi Y., “One-step synthesis of hierarchical porous γ -alumina with high surface area”, J. Sol-Gel Sci. Technol., 53, 428–433, 2010.
- [22]. Ravanchi M. T., Fard M. R., Rayeeni S. F., and Yaripour F., “Effect of Calcination Conditions on Crystalline Structure and Pore Size Distribution for a Mesoporous Alumina”, Journal Chemical Engineering Communications, Volume 202, Issue 4, 2014.