# Preparation of nano α-Al$_2$O$_3$ from kaolin belitung by liquid polish milling

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**Nano α-Al$_2$O$_3$ powders were successfully prepared from Indonesian local kaolin, kaolin Belitung. Nano α-Al$_2$O$_3$ powders were prepared by alumina extraction processes through liquid polish milling of the calcined kaolin with a H$_2$SO$_4$ solution, thus producing kaolin derived aluminum sulfate. Calcination treatments were conducted at 600 $^o$C and 800 $^o$C. The greatest alumina extracted was about 70.90% for the local kaolin calcined at 800 $^o$C. The aluminum sulfate was neutralized with a NH$_4$OH solution until pH 7-8 to form white sols of Al(OH)$_3$. Sugar was added into the mixture to mask the sols, thus producing gels. The gel was dried at 200 $^o$C and then was calcined at 900 $^o$C for 5 hours, which resulted in the formation of alumina. The calcined alumina was characterized by X-ray diffraction (XRD) and scanning electron microscopy (SEM). The alumina phases consisted of a γ-Al$_2$O$_3$ phase of about 47.6% having crystallite sizes of about 4-5 nm and an α-Al$_2$O$_3$ phase of about 52.4% having crystallite sizes of about 19-26 nm. The SEM result shows that the average grain sizes of the alumina powder were less than 250 nm.**

**Key words: Nano α-Al$_2$O$_3$, Kaolin belitung, Liquid polish milling, H$_2$SO$_4$, Sugar, Calcination.**

## Introduction

Alumina-based ceramics are one of the most important oxide materials in industry because of their excellent physical and chemical properties, such as high bending strength, high fracture toughness, high hardness, excellent abrasion resistance, high dielectric strength at high voltage, are thermally stable (high melting point at 2050 $^o$C), and have a high resistance to chemical attack. These characteristics enable application of alumina as a material for structural, optical, biomaterial, and electrical purposes [1, 2, 3].

Bauxite is the common raw material for producing alumina via the Bayer process. On the other hand, other available resources of nonbauxitic materials that contain high alumina and low iron oxide contents, also have been processed in order to develop alternative technologies for producing alumina [4, 5, 6, 7]. Some examples of nonbauxitic raw materials are aluminosilicate compounds, such as sillimanite, kaolin, and mica. Kaolin, one of the aluminosilicates, has been used as a combined source of SiO$_2$ and Al$_2$O$_3$. Significant advances in alumina purity have been achieved using materials such as sulfates, nitrates, and chlorides as alumina precursors, to obtain high purity alumina [4, 8, 9, 10].

Kaolin is a soft, lightweight, often chalk-like sedimentary rock that has an earthy odor. Besides kaolinite, kaolin usually contains quartz and mica.

Kaolinite, the main constituent of kaolin, is white, greyish-white, or slightly colored. Its chemical structure is Al$_2$Si$_2$O$_5$(OH)$_4$ (39.8% alumina, 46.3% silica, 13.9% water) which represents a two-layer crystal (silicon-oxygen tetrahedral layer joined to alumina octahedral layer exist alternately) [10, 11]. The theoretical formula for kaolinite is Si$_2$Al$_2$O$_5$(OH)$_4$ (other formulaes are Al$_2$O$_3 \cdot$ 2SiO$_2 \cdot$ 2H$_2$O and Al$_2$O$_7$Si$_2 \cdot$ 2H$_2$O), which has a molecular weight of 258.071 g/mol [4, 11]. Kaolin contains a high percentage of aluminum oxide of about 39.8% and is a suitable substitute for bauxite from which alumina of high purity can be obtained. Therefore, it can be a suitable material for the production of nano a-Al$_2$O$_3$ because of the considerable content of alumina in the kaolin structure.

Several sintering and acid-extraction processes have been investigated for the production of alumina from kaolin [7, 12]. As an example, alumina can be synthesized from kaolin-derived aluminum sulfate or aluminum ammonium sulfate or aluminum chloride solutions via a conventional liquid-solid extraction process [4, 7, 8, 13]. The isolated alumina in the form of aluminum salts would be alumina precursors for the preparation of nano α-Al$_2$O$_3$. Many synthesis methods have been proposed for nano alumina preparation by chemical methods [14, 15], such as emulsion techniques that are accompanied by calcining a precursor [16, 17, 18, 19], templating techniques such as using a natural cellulosic material [20], pyrolysis processing [21, 22], and solution combustion synthesis using an organic compound as a fuel [23]. Nano α-Al$_2$O$_3$ has been prepared from aluminum salts at a temperature of 900 $^o$C by a sugar process [1].

In this study, preparation of nano α-Al$_2$O$_3$ from Indonesian local kaolin (kaolin Belitung) is reported

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using a simple and commercial method. Most of the materials used in these methods are technical and local compounds and are found on an industrially scale. The purposes of this paper are to investigate the effect of the calcination temperature on the process of alumina extraction from kaolin Belitung by liquid polish milling with a $H_2SO_4$ solution and to prepare nano $\alpha$-$Al_2O_3$ from the kaolin-derived aluminum salts.

## Experimental Procedure

### Materials and instruments

Materials used in this research were Indonesian local kaolin (kaolin Belitung), a 25% ammonia solution, 95-97% sulfuric acid, technical grade sugar, and kerosene. All materials were obtained from a local market and Merck Inc.; whereas instruments used were a mortar, a pot mill, a Heraus electrical furnace, a Philips PANalytical X-ray diffractometer, and a JEOL JSM-35C scanning electron microscope (SEM).

### Preparation of aluminum salts from Kaolin belitung

Kaolin Belitung was initially prepared and calcined at three different temperatures (room temperature/no calcination (A), 600 °C (B), and 800 °C (C)).

About 251.85 gram of kaolin Belitung (A) was dispersed in 315.78 gram of a 95-97% $H_2SO_4$ solution. The mixture of kaolin and the acid was milled in an alumina pot mill for 24 hours. Then, the mixture was filtered to separate the residue and the filtrate. The filtrate was used as an alumina precursor for nano $\alpha$-$Al_2O_3$ preparation, whereas the residue was prepared for further analysis.

As a comparison, two other batches were prepared from different calcined kaolin samples. The second batch consisted of the kaolin Belitung calcined at 600 °C and sulfuric acid (B), whereas the third batch of nano alumina was prepared from the kaolin Belitung calcined at 800 °C (C).

### Preparation of nano $\alpha$-$Al_2O_3$ from the kaolin derived aluminum salts

About 300 ml of filtrate A was prepared in a 500 ml beaker. A 25% ammonia solution was added to the filtrate A until pH 7-8, and stirred constantly to give a white sol of $Al(OH)_3$. About 10% (w/v) sugar was added appropriately into the mixture and stirred until evenly mixed with the mixture until it produced a white gel. The gel was dried in the oven at 200 °C to produce black charcoal precursors. The precursors were milled in a non polar solvent such as kerosene for 12 hours and were successfully calcined at 900 °C for 5 hours in a Heraus electric furnace.

The procedure was also conducted for the filtrates B and C.

### Characterization of the residues and alumina powder

The residues of samples A, B, and C produced from the preparation of kaolin-derived aluminum salts were analyzed and characterized using chemical analysis,

XRD, and SEM. On the other hand, the calcined alumina powder was characterized using XRD and SEM.

## Results and Discussion

### Chemical characteristics of the kaolin Belitung before and after extraction by liquid polish milling

The results of chemical analysis on the kaolin residue before and after extraction by liquid polish milling using a 95-97% $H_2SO_4$ solution are listed at Table 1. According to Table 1, it seems that the addition of 315.78 gram of 95-97% $H_2SO_4$ solution had optimally extracted alumina from kaolin Belitung, because 70.90% of the total alumina content had been extracted from the kaolin. Therefore, by quantitative analysis, the alumina content in the kaolin residue was about 71.75%, 34.38%, and 29.09% for samples A, B, and C, respectively.

The initial calcination significantly influenced the level of extraction of alumina from the kaolin by the liquid polish milling process with the $H_2SO_4$ solution (Table 1). The higher the initial calcination temperature used, the higher the level of extraction of alumina, and a higher alumina content was obtained. According to Chambers & Holliday[24], when an aluminosilicate is heated at at 550-900 °C, the bonds in the alumino-silicate will be destroyed and broken, giving an amorphous structure which is more reactive, thus the $Al_2O_3$ compounds from calcined kaolin are more easily extracted, compared with non-calcined kaolin. However, secondary minerals such as spinel and mullite will be produced at a higher calcination temperature.

The silica content (silicate) in the residue increased as the alumina content decreased, because silica is insoluble or slightly soluble in acidic conditions. A reaction between silicate with acid would give insoluble compounds such as stable dispersed silica [24]:

$$SiO_3^{2-}{}_{(aq)} + 2H^+ \rightarrow H_2SiO_{3(aq)} \rightarrow SiO_2 \cdot xH_2O \qquad (1)$$
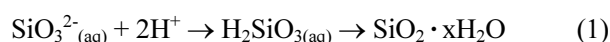
However, the content of impurities in the residue, such as $Fe_2O_3$, CaO, MgO, $K_2O$, $Na_2O$, and $TiO_2$, was decreased in the very acidic medium.

### Mineralogical characteristics for the kaolin Belitung and the residue structures, and the calcined alumina powder

The effect of the initial calcination on the kaolin Belitung transformation after extraction by liquid polish milling using 95-97% sulfuric acid could be observed in the XRD results, which are presented in Figure 1.

Figure 1 show typical XRD patterns of the pure kaolin Belitung before extraction; and samples A, B, C after extraction by liquid polish milling using 95-97% sulfuric acid. The XRD analysis of the pure kaolin Belitung in Figure 1, shows that the kaolin consists of kaolinite as the main component with strong peaks of kaolinite main crystal planes at diffraction angles of
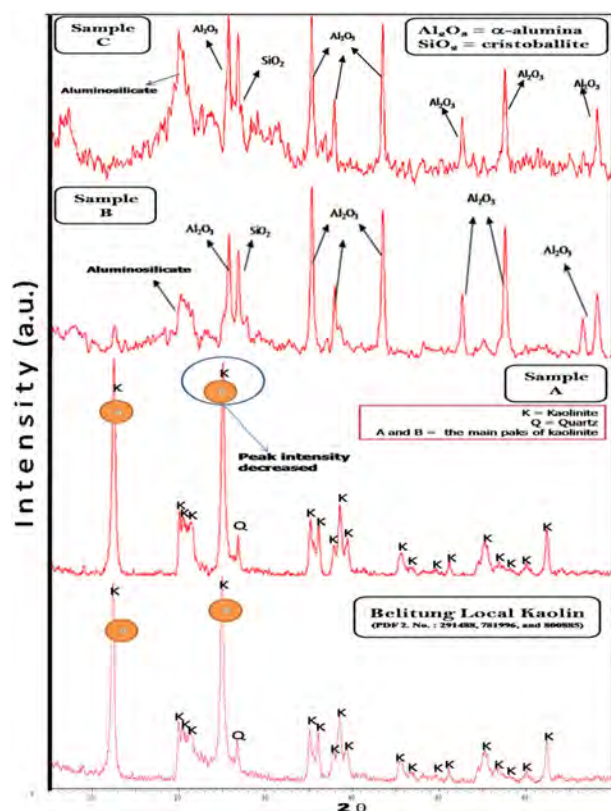
**Fig. 1.** XRD patterns of pure kaolin Belitung before extraction; and samples A, B, C after extraction by liquid polish milling using 95-97% sulfuric acid.
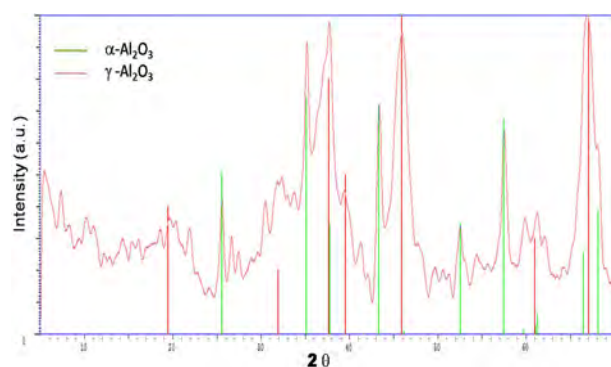


**Fig. 2.** XRD pattern of alumina powder calcined at 900 °C.

**Table 1.** Composition of kaolin Belitung before and after extraction by liquid polish milling using 95-97% sulfuric acid.

| Component | Belitung Local Kaolin | Component content (%) | | |
|---|---|---|---|---|
| | | Sample A (Room Temperature) | Sample B (600 °C) | Sample C (800 °C) |
| Aluminosilicate | - | 9.91 | 18.95 | 16.97 |
| SiO$_2$ | 46.73 | 49.14 | 57.79 | 61.60 |
| Al$_2$O$_3$ | 37.70 | 27.05 | 12.96 | 10.97 |
| Fe$_2$O$_3$ | 0.71 | - | - | - |
| CaO | 0.05 | - | - | - |
| MgO | 0.06 | - | - | - |
| Na$_2$O | 0.04 | - | - | - |
| K$_2$O | 0.49 | - | - | - |
| TiO$_2$ | 0.40 | - | - | - |
| $^a$L.O.I | 13.83 | 13.19 | 10.30 | 10.46 |

12.385 ° (001) and 25.016 (002); and quartz as the minor component. When the kaolin was extracted by liquid polish milling using 95-97% sulfuric acid (Sample A), the XRD result showed the sample having the same mineral composition and an XRD pattern with pure kaolin, as shown in Figure 1. However, one of the main peak intensities from kaolinite in sample A decreased, especially at a diffraction angle of 25.016, corresponding to the (002) crystal plane of kaolinite (see Figure 1). The peak intensity of the sample was lower than the peak intensity from pure kaolin, this phenomenon indicated sample A had lost some components during liquid polish milling. This might probably be caused by the extraction of some alumina compounds of the

kaolin (sample A) with sulfuric acid during liquid polish milling, leading to the formation of aluminum sulphate in the solution. This is in accordance with the result of the chemical analysis on sample A, showing a decrease of the alumina content in the kaolin (see at Table 1). This result is significantly different from the XRD result on the samples B and C after extraction by liquid polish milling using 95-97% sulfuric acid, in which the samples used were kaolins calcined at 600 °C and 800 °C, as shown in Figure 1. The XRD identification showed that both samples B and C have the same mineral composition and XRD patterns in their residues (see Figure 1). The mineral phase identified in both residues of samples B and C were radical aluminosilicates, cristoballite, and α-Al$_2$O$_3$, the most stable alumina phase with strong peaks of α-Al$_2$O$_3$ main crystal planes at diffraction angles of 25.556 ° (012), 35.165 (104), 43.316 (113), and 57.459 (116). Nevertheless, by contrast, the profiles of peak intensity of a-Al$_2$O$_3$ for both samples B and C with the in initial calcination temperatures are mostly different to those indicated by Figure 1. The significant changes can be seen in the decrease of alumina peak intensities in the residue of sample B, as shown by the diffraction patterns in Figure 1. The effect of the initial calcination temperature on alumina extraction from the kaolin with sulfuric acid by liquid polish milling is significantly shown in the percentage of the alumina content in the sample residue. The content of alumina in the kaolin residue for both samples B and C decreased as the initial calcination temperature increased, which was in accord with the results of chemical and XRD analyses (see Table 1 and Figure 1).
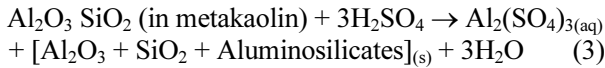
**Table 2.** Crystallite sizes of kaolin, cristoballite, and α-$Al_2O_3$ for samples A, B, and C

| Initial Calcination Temperature (°C) | Crystallite sizes (nm) | | |
|---|---|---|---|
| | α-$Al_2O_3$ | $SiO_2$ | Kaolinite |
| Room temperature (A) | - | - | 23 |
| 600 (B) | 25 | 6 | - |
| 800 (C) | 18 | 3 | - |

During the calcination the structure of kaolin was degraded and two molecules of water were released. This process is known as dehydroxylation, which is a reaction of the decomposition of kaolinite crystals to a partially disordered structure. The first presence of dehydroxylation was identified by changes of mechanical, electrical properties and also by weight loss. The results of an isothermal firing show that the dehydroxylation begins at ~ 420 °C [4, 11]. The chemical equation describing this process is:

$$Al_2O_3 \cdot 2SiO_2 \cdot 2H_2O \rightarrow Al_2Si_2O_7 + 2H_2O \text{ (g)} \qquad (2)$$
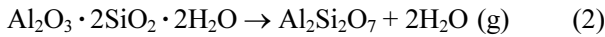
The result of dehydroxylation is a new phase called a meta kaolinite, which is an amorphous phase. During liquid polish milling of the kaolin (sample A) or calcined kaolins (samples B and C) in a sulfuric acid medium, the alumina in all kaolin samples is extracted and dissolves in the $H_2SO_4$ which leads to formation of aluminum sulfate (according to the XRD results):

$$Al_2O_3 \ SiO_2 \text{ (in metakaolin)} + 3H_2SO_4 \rightarrow Al_2(SO_4)_{3(aq)}$$
$$+ [Al_2O_3 + SiO_2 + \text{Aluminosilicates}]_{(s)} + 3H_2O \qquad (3)$$

Based on the main peaks of the XRD patterns from Figure 1, the crystallite sizes of kaolinite, cristoballite, and α-$Al_2O_3$ for samples A, B, and C can be calculated using the Scherrer equation [1]:
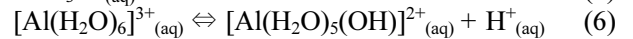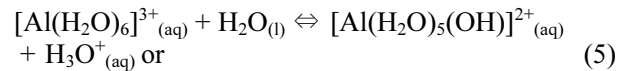
$$D = \frac{K\lambda}{\beta cos\theta} \qquad (4)$$

where $D$ is the crystal size, $K$ is a shape factor with a value of 0.9-1, λ is the wavelength of the X-rays (1.54056 Å), and β is the value of the *full width half maxima* (FWHM). Using the Scherrer equation, the crystallites sizes of the three alumina samples can be calculated and are presented in Table 2.

Figure 2 shows the XRD pattern of the isolated alumina from kaolin Belitung after thermal treatment at 900 °C. According to Figure 2, when sugar was used as a gelling agent in a water medium, the precursors of isolated alumina in the form of kaolin-derived aluminum sulfate which was neutralized with ammonia was found to transform and crystallize at a temperature of 900 °C, with the broadening and overlapping sharp peaks of α-$Al_2O_3$ and γ-$Al_2O_3$. Quantitative analyses of the diffractogram patterns, including the crystallite
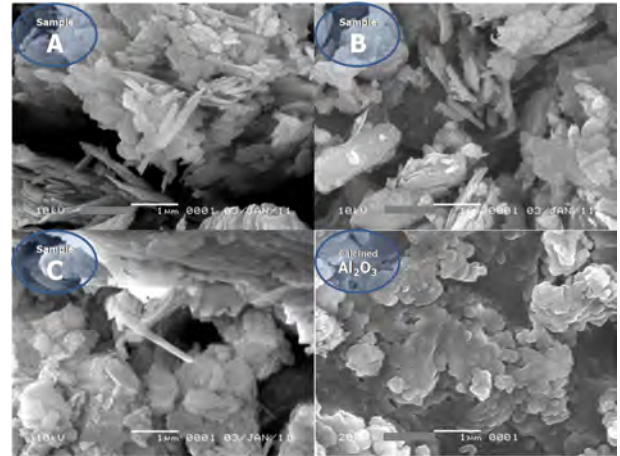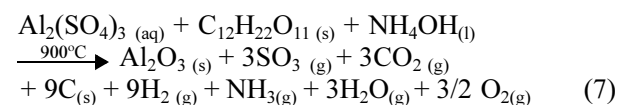


**Fig. 3.** Typical SEM images of samples A, B, C and alumina.

sizes and phase compositions of α-$Al_2O_3$ and γ-$Al_2O_3$, were performed for calcined alumina powder using XRD software. The quantitative results show that the alumina phases in the calcined alumina powder consisted of the γ-$Al_2O_3$ phase of about 47.6% having crystallite sizes of about 4-5 nm (PDF 2. 10-0425) and the α-$Al_2O_3$ phase of about 52.4% having crystallite sizes of about 19-26 nm (PDF 2. 83-2080).

When kaolin-derived aluminum sulfate was produced in a water medium, a hydrated aluminum ion would be produced from aqueous aluminum sulphate in the ion complex form of aluminum hexaaqua ion, where the reaction is as follows [25]:

$$[Al(H_2O)_6]^{3+}_{(aq)} + H_2O_{(l)} \Leftrightarrow [Al(H_2O)_5(OH)]^{2+}_{(aq)}$$
$$+ H_3O^+_{(aq)} \text{ or} \qquad (5)$$
$$[Al(H_2O)_6]^{3+}_{(aq)} \Leftrightarrow [Al(H_2O)_5(OH)]^{2+}_{(aq)} + H^+_{(aq)} \qquad (6)$$

The hydrolysis and neutralization process of the aluminum salt solution was accelerated with the addition of a $NH_4OH$ solution until pH 7-8, and until the white sol of aluminium hydroxide ($Al(OH)_3$) formed. However, the presence of a disachcharide template, during the preparation, significantly reduces the tendency to agglomeration in the as-synthesized alumina particles. The template contains hydroxyl and ether groups in its structure, which hydrogen bonds with the hydroxyl groups from the particle surfaces. As a result, the surface hydroxyl groups get capped with the olygomer (disachcharide). The template coating on the alumina particle surfaces results in reduced aggregation of particles due to the steric hindrance provided by the olygomer. A total reaction is assumed as follows:

$$Al_2(SO_4)_3 \ _{(aq)} + C_{12}H_{22}O_{11 \ (s)} + NH_4OH_{(l)}$$
$$\xrightarrow{900°C} Al_2O_{3 \ (s)} + 3SO_{3 \ (g)} + 3CO_{2 \ (g)}$$
$$+ 9C_{(s)} + 9H_{2 \ (g)} + NH_{3(g)} + 3H_2O_{(g)} + 3/2 \ O_{2(g)} \qquad (7)$$

**Microstructures**

Figure 3 shows typical SEM images of samples A, B, C and alumina using a JEOL JSM-35C scanning

electron microscope.

Figure 3 show the results of SEM analysis of the local kaolin samples without and with thermal treatment at temperatures of 600 °C and 800 °C after liquid polish milling with 95-97% sulfuric acid (SEM images of samples A, B and C, respectively). The three SEM images did not show any significant differences in the morphology of all samples. SEM analysis of sample A, shows the morphology and the microstructure of kaolin Belitung without the thermal treatment after liquid polish milling with 95-97% sulfuric acid, having sheet and plate-like structures. These structures are similar with the morphology of kaolinite. This has a 1:1 sheet structure composed of $SiO_4$ tetrahedral sheets and $Al(O, OH)_6$ octahedral sheets with pseudo-hexagonal symmetry. The sheets are created from planes, which are occupied as follows: $O_6 - Si_4 - O_4 - (OH)_2 - Al_4 - (OH)_6$ [11]. However, SEM analysis of samples B and C show relatively similar morphologies and micro-structures with sample A, also having sheet and plate-like structures in most regions of the particle morphology. These structures probably correspond to the aluminosilicate structures, according to the XRD results in Figure 1. Nevertheless, some regions in the morphology of samples B and C are grain-like, which probably corresponds to alumina and silica particles. These particles might also be covered by alumino-silicate particles in samples B and C.

Figure 3 also shows a SEM image of the alumina powder in which an appreciable formation of agglomerates after thermal treatment at 900 °C can be seen. The average grain size of the alumina in the case of the sample calcined at 900 °C for 5 hours were less than 250 nm. The addition of a sugar template is important in the preparation of alumina. The crystal or particle size produced from the aluminum salt and hydroxide will be smaller with the sugar present. This is because the concentration of $Al^3$ ions in the precursor will be lower over the course of the phase transformation with a certain amount of sugar. As the calcination temperature is increased, the material is considered to be so deficient in $Al^{3+}$ content that it retards the crystallite grain growth. Furthermore, this masking compound will be distributed across the particle surfaces, such that the binding distance will increase because sugar restricts the grain contact among the nucleated $Al_2O_3$ and restrains crystallite growth [1, 25].

## Conclusions

Nano $\alpha$-$Al_2O_3$ powders were successfully prepared by alumina extraction processes through liquid polish milling of kaolin Belitung with a $H_2SO_4$ solution. The addition of sugar was an important step for the preparation of nano $\alpha$-$Al_2O_3$. It is concluded that the initial calcination significantly influenced the level of extraction of alumina from the kaolin by the liquid polish milling process with a $H_2SO_4$ solution. The higher the initial calcination temperature used, the higher the level of extraction of alumina, and a higher alumina content was obtained. The highest alumina extracted was about 70.90% for the local kaolin calcined at 800 °C. However, the kaolin residue consisted of radical aluminosilicates, silica, and alumina. Calcination of kaolin-derived aluminum sulfate at 900 °C with a $NH_4OH$ solution and the presence of sugar leads to the formation alumina. The alumina phases formed in calcined alumina powder consist of the $\gamma$-$Al_2O_3$ phase of about 47.6% having crystallite sizes of about 4-5 nm and the $\alpha$-$Al_2O_3$ phase of about 52.4% having crystallite sizes of about 19-26 nm. The average grain sizes of the alumina powder were less than 250 nm, as given by SEM. The study reports that kaolin Belitung could be a promising raw material for the preparation of nano $\alpha$-$Al_2O_3$.

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