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Synthesis of Sodalite from Indonesian Kaolin with Conventional and Alkali Fusion Method

S N Khalifah[1,*], M Cahyawati[1], D K D Cahyani[1], A Arifah[1] and A Prasetyo[1]
[1]Department of Chemistry, Faculty Science and Technology, Universitas Islam Negeri Maulana Malik Ibrahim, Jalan Gajayana 50, 65144, Malang, Indonesia.

[*]E-mail: susikhalifah@gmail.com

Abstract. Sodalite has been synthesized from Indonesian kaolin. Kaolin was converted into more reactive metakaolin using conventional and alkali fusion methods. In the conventional method, kaolin is calcined at a temperature of 700 °C for 3 hours. Meanwhile, in the alkali fusion method, the mixture of kaolin and NaOH was calcined at a temperature of 600 °C for 2 hours. The effect of synthesis parameters such as aging time and crystallization temperature was investigated in conventional methods. Besides that, the crystallization time was investigated in the alkali fusion method. Transformation of kaolin into metakaolin and synthetic products was characterized using X-ray diffraction (XRD), X-ray Fluorescence (XRF), Fourier Transform Infrared (FTIR) and Scanning Electron Microscopy (SEM). The results showed that the kaolin structure contained a lot of quartz. In the conventional method, the kaolin structure cannot be converted into metakaolin. The synthesis product obtained from this method is a mixture of sodalite and quartz. Whereas, in the alkali fusion method, kaolin can be converted into metakaolin and sodium silicate. The synthesis product obtained is a pure sodalite.

1. Introduction

Sodalite is a type of zeolite which composed of β-cage that form micropore structure. β-cage (sodalite cage) consist of silica and alumina through association of simple 4 ring (S4R) and simple 6 ring (S6R) [1]. Sodalite has high stability compared with others zeolite that also have low Si/Al ratio such as NaA zeolite, NaX zeolite and NaY zeolite. High stability of sodalite was proposed by β-cage that establish sodalite framework with large density. Large density framework can decrease lattice energy. Therefore, it can increase stability of sodalite. Sodalite have much attention because it can be used in many fields such as ion exchange [2], optical material [3], hydrogen storage [1], nuclear waste isolation [3], infiltrating material for alumina and zirconia toughened alumina (ZTA) [4] and semiconductor [5].

Sodalite can be Synthesized using chemical precursor such as alumina, gamma alumina, aluminosilicate, silica gel, colloidal silica, sodium silicate and tetraethyl ortho silicate (TEOS) [5]. Synthesis using chemical precursor have been resulted pure product but also high cost synthesis. So, required low cost material that have large amount such as kaolin. One of kaolin that have abundant amount was Blitar kaolin. Blitar kaolin contains high alumina and silica. Therefore, it can be used as starting material for synthesis sodalite. Synthesis of sodalite from kaolin had been less reported, one of them produce pure sodalite [3] and with impurities nepheline [5].

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Route for sodalite synthesis from kaolin begin with conversion kaolin to metastability phase such as metakaolin using conventional method or calcination at high temperature. Synthesis of metakaolin using conventional method was reported resulting metakolin with impurities Quartz [6]. Another method for synthesis metakolin is alkali fusion. Alkali fusion is a method which use NaOH to dissolve impurities such as Quartz and followed by calcination. Synthesis of metakaolin using alkali fusion method revealed pure metakaolin and few impurities.

This research will compare between conventional and alkali fusion methods to formed metakaolin, then followed by hydrothermal method for synthesis of sodalite. Variation of aging time and crystallization temperature was investigated in the conventional method. Furthermore, variation of crystallization time was investigated in the alkali fusion method.

2. Experimental

2.1. Material

Kaolin that supplied from Blitar (76.1% Si, 9.6% Al, 6.44% K, 1.84% Ca, 3.19% Ti, 2.27% Fe) East Java, Indonesia was ground to 200-300 mess as starting material, sodium hydroxide (Merck, >99%); Al_2O_3 (Merck, 99%).

2.2. Synthesis sodalite

Kaolin was converted to metakaolin using conventional method by calcination at a temperature 700 ºC for 3 hours. The calcined product was ground and used as material for synthesis of zeolite. The alkali fusion for synthesis of metakaolin was done by crush 4.29 g kaolin with 8.58 g NaOH for 30 minutes. The mixture was fused at 600 ºC for 1 hour. The fused product grounded in a mortar and used to synthesis of zeolite. The molar composition for synthesis sodalite of both method was: $10SiO_2$: Al_2O_3: 0.96 Na_2O: 180 H_2O.

The mixture then age at room temperature (~30 ºC) for 10 days. Synthesis sodalite from Conventional product was investigated on crystallization temperature and aging time. Hydrothermal method for conventional product use crystallization temperature 90 ºC, 100 ºC, 110 ºC and aging time 24 hours, 36 hours, and 46 hours. The alkali fusion product was investigated on Crystallization time at 6 hours, 12 hours and 24 hours. The results of synthesis filtered and washed with aquades to pH 9 or 10. Sodalite obtained then dried at 100 ºC for 12 hours. The dried sample was kept on plastic bags for characterization using XRD, FTIR, and SEM.

2.3. Characterization

Chemical compositions of kaolin were determined using XRF (Philips Analytical's, PW2404, Almelo, The Netherlands). The phase identification was determined by XRD a Philips diffractometer with Cu-Kα radiation (Philips XRG 3100, Philips Analytical Inc) at 2 theta 5-60º. Infrared (IR) spectra were determined using Perkin Elmer Spectrum GX FTIR System spectrometer. Morphology of starting material and the synthesis product were observed by Hitachi S-570 Scanning Electron Microscopy (SEM) that operated at 15 kV.

3. Results and Discussion

3.1 Synthesis of metakaolin

The synthesis of metakaolin using conventional methods has been reported in previous studies where the results of XRD pattern indicated that the synthesis material still contain quartz as an impurities. The crystalline phase of material was dominated by quartz which reaches 90%. Quartz was stable at high temperatures hence calcination does not breakdown the quartz structure and no significant different on diffractogram pattern [7]. Therefore, Blitar kaolin includes low grade material. Furthermore, synthesis of metakaolin using alkali fusion method revealed sodium silicate that appeared in 2 theta 17º, 25º, 29.05º, 35.05º, 38º, and 49.05º (figure 1). Sodium silicate was formed by reaction of quartz on kaolin with NaOH at high temperatures.

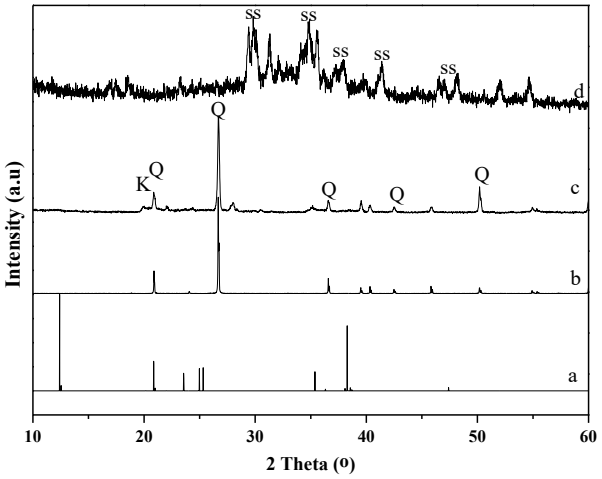

Figure 1. XRD pattern of (a) kaolin standard, (b) Quartz standard, (c) Blitar kaolin and (d) result of alkali fusion (Q = Quartz, K = kaolin, SS = sodium silicate).

The infared pattern of kaolin (figure 2) was shown vibration of Quartz as free Quartz at 785 cm^{-1}, while kaolin have vibration of Al-OH stretching at 3699 and 3450 cm^{-1}, H_2O bending at 1633 cm^{-1}, T-O-T asymmetric tetrahedral stretching at 1037 cm^{-1}, Al-OH octahedral stretching at 912 cm^{-1}, T-O-T (T= Si or Al) symmetric bending at 694 and 528 cm^{-1}, T-O-T asymmetric bending at 493 cm^{-1}.

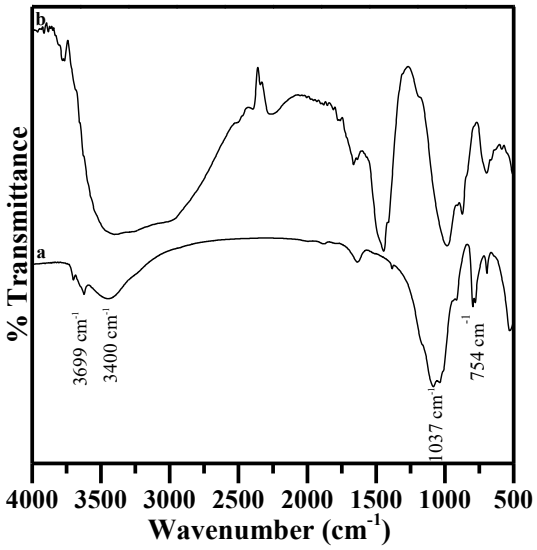

Figure 2. FTIR spectra of (a) kaolin and (b) result of alkali fusion.

Analysis using FTIR showed that metakaolin has similiar spectrum with kaolin (figure 2) but loses vibration of Al-OH octahedral at 3665 cm^{-1} as a result of convertion alumina octahedral to tetrahedral. Analysis using FTIR showed that alkali fusion process cause shifting of asymmetric T-O-T vibration at 1037 cm-1 to 984 cm^{-1} and loss vibration at 3699 cm^{-1}, 912 cm^{-1}, 754 cm^{-1} and the appearance vibration at 872 cm^{-1}. The shifting of vibration indicated transformation structure of octahedral alumina in kaolin to tetrahedral alumina in metakaolin. Alkali fusion process also cause demerge absorption from OH vibration at wavenumber 3400 cm^{-1} which is uptake of sodium silicate, the vibration enlarged because sodium silicate was hygroscopic, then vibration of asymmetric T-O-T strain at 984 cm^{-1}, T-O-T symmetric bending at 697 cm^{-1} and 520 cm^{-1}, asymmetric bending T-O-T at 461 cm^{-1} and asymmetric CO_3^{2-} strain of Na_2CO_3 at 1445 cm^{-1}. Asymmetric CO_3^{2-} vibration (Na_2CO_3) arises due to reaction of sodium silicate with carbon dioxide in the air. The FTIR spectra indicates alkali fusion process was not only produce sodium silicate but also metakaolin.

Morphology of kaolin (figure 3) was layered plate as a result of octahedral and tetrahedral sheet. Based on the research before, micrograph results from metakaolin that preparated from conventional method showed that the material was partially change to amorphous material. Moreover, based on the micrograph of alkali fusion product, layered kaolin was fully transformed to amorphous material.

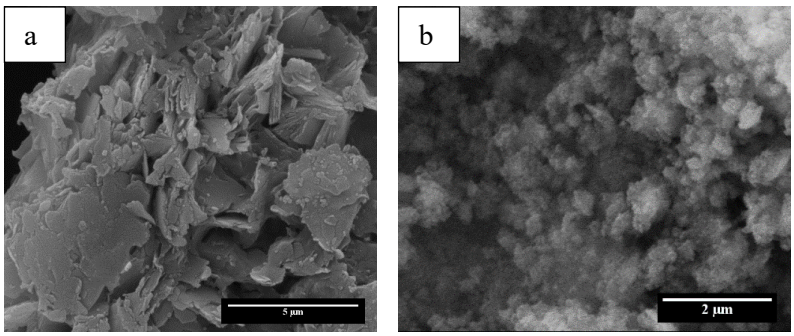

Figure 3. Morphology of (a) kaolin and (b) alkali fusion product.

3.2. Synthesis of sodalite

3.2.1. Synthesis of sodalite from kaolin prepared using conventional method

Metakaolin obtained from conventional methods was converted to sodalite and investigated in aging time. XRD pattern of product revealed sodalite denoted at 2 theta 14.5; 24.98; 32.5; 35.01; 43.9° and quartz as impurities (figure 4). Investigation on aging time does not show significant changes in crystallinity of product obtained.

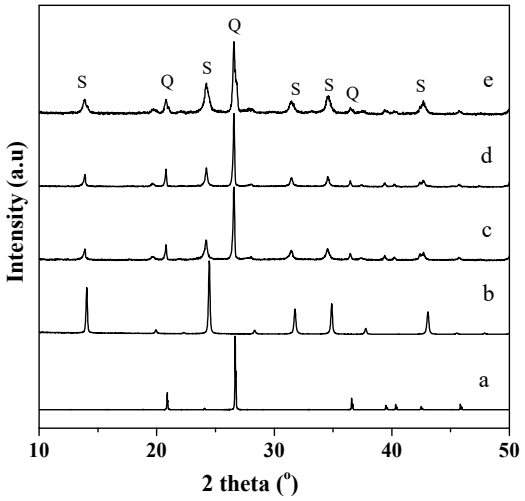

Figure 4. XRD pattern of (a) Quartz standard, (b) Sodalite standard, (c) aging for 24 hours, (d) 36 hours and (e) 48 hours (Q = Quartz, S = Sodalite) at crystallization temperature 100 ºC.

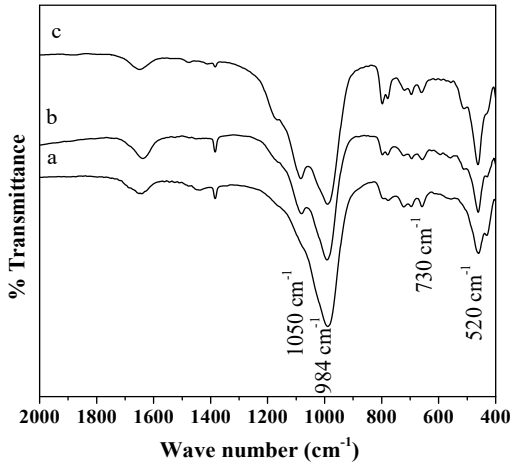

Figure 5. FTIR spectra of product with aging time (a) 24 hours, (b) 36 hours and (c) 48 hours at crystallization temperature 100 ºC.

FTIR spectra of product (figure 5) was showed vibration of Si-O bending at 520 cm^{-1}, Si-O-Si symmetric stretching at 730 cm^{-1}, and Si-O-Si asymmetric stretching at 1050 cm^{-1} which refer to Quartz present. While sodalite appear at 984 cm^{-1} that refer to T-O-T asymmetric stretching. Increasing of aging time elevate vibration at 520 cm^{-1}, 730 cm^{-1}, and 1050 cm^{-1} due to increasing Quartz based on XRD analysis. Based on micrograph of sodalite (figure 6) have shown that increasing particel size proportionate with aging time which the particel size approximately 1.5 μm at aging time 48 hours.

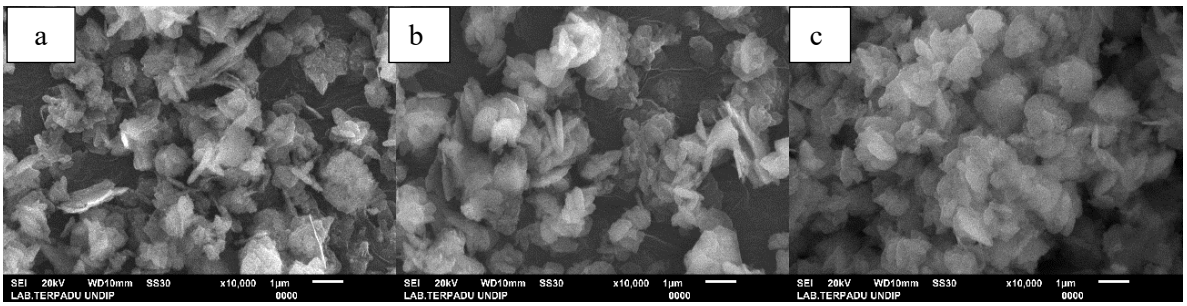

Figure 6. Micrograph of product with aging time (a) 24 hours, (b) 36 hours and (c) 48 hours at crystallization temperature 100 ºC.

The effect of crystallization temperature was also investigated on the conventional metakaolin method. The XRD pattern of resulting product was shown at figure 7. Based on the analysis using XRD the products which formed were sodalite and quartz. Crystallization temperature was not significantly influence the product obtained. Analysis using FTIR (figure 8) showed crystallization temperature decrease intensity of Quartz by decrease vibration at 520 cm^{-1}, 732 cm^{-1} and 1045 cm^{-1} denote vibration of Quartz even though XRD analysis was not showing different result. The micrograph of sodalite produced (figure 9) was shown decreasing of particel size by increasing crystallization temperature.

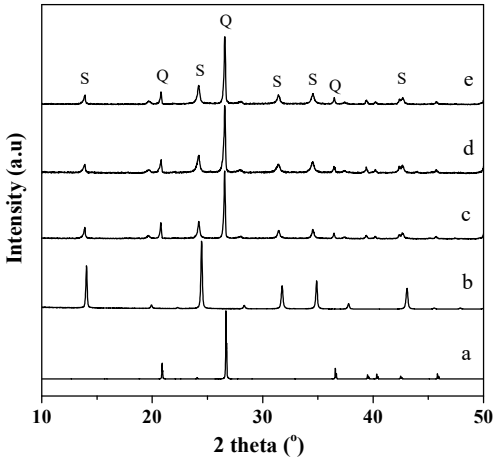

Figure 7. XRD pattern of (a) Quartz standard, (b) sodalite standard, crystallization temperature for (c) 90 °C, (d) 100 °C and (e) 110 °C at aging time 48 hours.

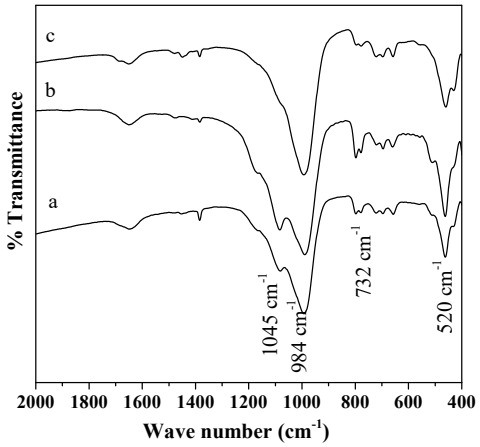

Figure 8. FTIR spectra of product with crystallization temperature for (a) 90 °C, (b) 100 °C and (c) 110 °C at aging time 48 hours.

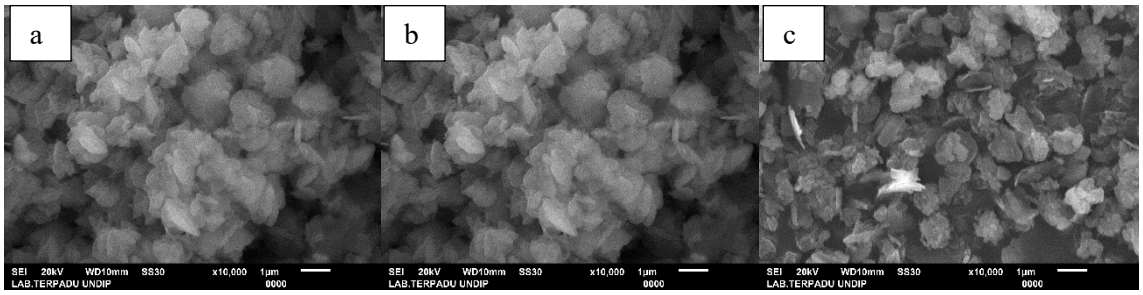

Figure 9. Micrograph of product with crystallization temperature for (a) 90 °C, (b) 100 °C and (c) 110 °C at aging time 48 hours.

3.2.2 Synthesis of sodalite from kaolin prepared using alkali fusion method

The effect of crystallization time have been investigated on the synthesis sodalite from kaolin prepared using alkali fusion method. The diffractogram as XRD result was shown in figure 10. Based on the analysis using XRD can be known that product formed was pure sodalite due to reaction of quartz with NaOH on alkali fusion. Investigation on crystallization time showed that crystallization time affected width of peak consequence of smaller crystal of sodalite.

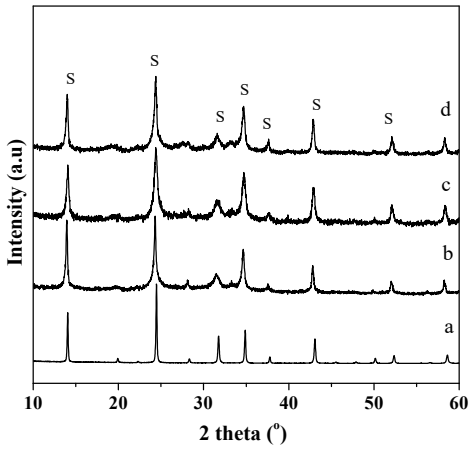

Figure 10. XRD pattern of sodalite standard (a), crystallization time for 6 hours (b), 12 hours (c) and 24 hours (d) (S = sodalite) at crystallization temperature 100 °C and aging time 48 hours.

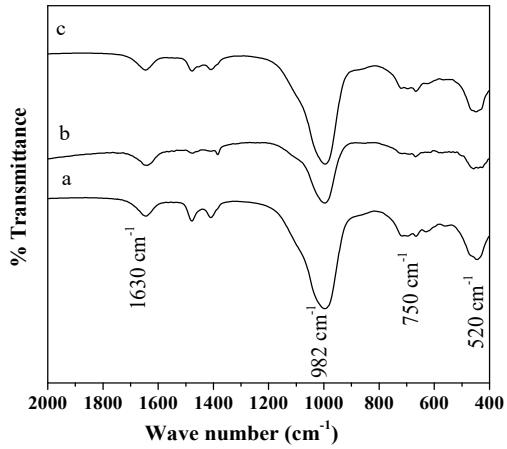

Figure 11. XRD pattern of sodalite with alkali fusion method at crystallization time (a) 6 hours, (b) 12 hours and (c) 24 hours in the crystallization temperature 100 °C and aging time 48 hours.

The FTIR spectra of sodalite (figure 11) showed that the product which formed was pure sodalite. Crystallization time also increases crystallinity of product referred by sharpening peak of O-H bending vibration at 1650-1600 cm^{-1} due to the presence of water absorbed physically. Other absorptions appear at wavenumbers 1250-950 cm^{-1} indicated the internal T-O-T asymmetric stretching, 720-650 cm^{-1} related to internal symmetric stretching of T-O-T, and 820-720 cm^{-1} referred to external symmetric stretching of T-O-T (Auerbach et al, 2003). Based on the micrograph results (figure 12) indicated that the product not only irregular particle, but also homogen. Crystallization time affect the size of particle compatible as XRD result, at 6 hours formed approximately 0.5 μm, and decline to 0,2 μm at 12 hours.

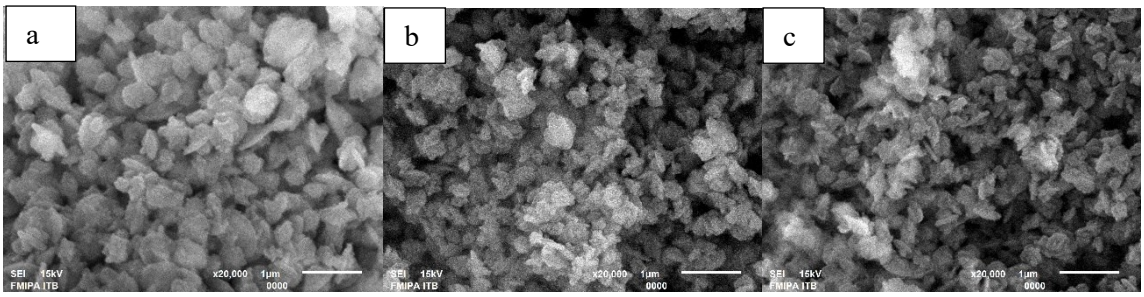

Figure 12. Micrograph of sodalite at crystallization time (a) 6 hours, (b) 12 hours and (c) 24 hours in the crystallization temperature 100 °C and aging time 48 hours.

4. Conclusion

This research reports that the transformation of Blitar kaolin using conventional methods has produced a product with quartz impurity. Whereas, when using the alkali fusion method, it produces metakaolin and sodium silicate. Sodalite synthesized from conventional methods has not been proven to be pure because there is still quartz impurities. Whereas, when using the alkali fusion method pure sodalite products have been obtained. Synthesis parameters such as the aging time, crystallization temperature, and crystallization time proven have effect on increasing the crystallinity and decreasing the particle size of the product obtained.

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