MAREDAN NATURAL CLAY AS RAW MATERIAL FOR ZEOLITE SYNTHESIS

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Abstract: The Maredan Riau clay contains alumina silica components, which can serve as raw material for zeolite synthesis. This research has been conducted on the effects of aging time and crystallization temperature during the zeolite synthesis process, which is the result of converting Maredan clay raw materials. The synthesis of zeolite follows a series of steps, including activation of natural clay using 1M H_2SO_4 and calcination at 650ºC, curing of the post-calcination clay in 5 M NaOH, crystallization at different temperatures (80, 90, and 100ºC) for 8 hours, setting the final pH to up to 8 and drained. The solid obtained in zeolite was characterized to determine the ratio of Si/Al, type of zeolite and crystallinity, functional groups, and zeolite surface morphology. The analysis results with XRF at a crystallization temperature of 100°C showed that the Si/Al zeolite ratio was 1.75 which corresponds to the Si/Al ratio of sodalite-type zeolite. Accordingly, analysis with XRD also supports the formation of sodalite with a crystallinity of 51% and a crystal size of 30.03 nm. The FTIR and SEM characterization respectively showed the presence of D4R or D6R double ring external vibration of the sodalite at wave number 558.42 cm^{-1} and its morphology was round with petals which is a typical morphology of sodalite. It was concluded that the increase in crystallization temperature caused the crystallinity of the resulting sodalite to increase as well.
Keywords: crystallization; natural clay; maredan; reflux; sodalite

Abstrak: Lempung Maredan Riau mengandung komponen alumina silika, yang dapat berperan sebagai bahan baku untuk sintesis zeolit. Penelitian ini bertujuan mengamati efek waktu aging dan suhu kristalisasi selama proses sintesis zeolit, yang merupakan hasil konversi bahan baku lempung Maredan. Sintesis zeolit mengikuti serangkaian tahap, di antaranya aktivasi lempung alam menggunakan 1M H_2SO_4 dan kalsinasi pada suhu 650ºC, pemeraman lempung pasca kalsinasi di dalam 5 M NaOH, kristalisasi pada suhu yang berbeda (80, 90 dan 100ºC) selama 8 jam, pengaturan pH akhir hingga mencapai 8 dan dikeringkan. Padatan yang diperoleh berupa zeolit dikarakterisasi untuk menetapkan rasio Si/Al, jenis zeolit dan kristalinitas, gugus fungsi dan morfologi permukaan zeolit. Hasil analisis dengan XRF pada suhu kristalisasi 100°C menunjukkan bahwa rasio Si/Al zeolit adalah 1,75 yang sesuai dengan rasio Si/Al zeolit jenis sodalit. Sejalan dengan itu, analisis dengan XRD juga mendukung terbentuknya sodalit dengan kristalinitas 51% dan ukuran kristal 30,03 nm. Karakterisasi FTIR dan SEM masing-masing menunjukkan adanya vibrasi eksternal cincin ganda D4R atau D6R dari sodalit pada bilangan gelombang 558,42 cm^{-1} dan morfologinya berbentuk bulat berkelopak

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yang merupakan morfologi khas dari sodalit. Disimpulkan bahwa peningkatan suhu kristalisasi menyebabkan kristalinitas sodalit yang dihasilkan meningkat.
Kata kunci: kristalisasi; lempung alam; Maredan; refluks; sodalit

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Introduction

Zeolites are microporous crystalline hydrated aluminosilicates, which are found in various applications because of their unique physico chemical characteristics such as ion exchange and adsorption-desorption properties (Ghasemi *et al*., 2018). Raw materials for the synthesis of zeolite as silica and alumina have been reported to be found in many natural clays. Clay contains elements of silica and alumina which are important components in the synthesis of zeolite. The natural clay in Maredan Village, Tualang District, Siak Regency is around 625,000 tons which has not been used optimally. In general, clay is used as a basic material for making bricks and building materials. The results of XRD analysis stated that Maredan clay consists of quartz, kaolinite and illite minerals (Malik *et al*., 2020). The results of the XRF analysis that have been studied show that Maredan clay contains 48.762% SiO_2 and 26.248% Al_2O_3. The large amount of silica and alumina in Maredan clay causes these minerals to be used as raw materials in the synthesis of zeolites and this topic is discussed in this study.

The raw materials for zeolite synthesis can come from natural materials containing silica aluminas such as pumice (Uzun *et al*., 2021), Coal Fly Ash (Ren *et al*., 2020), and Coal Bottom Ash (Gollakota *et al*., 2021). Raw materials containing silica and alumina such as fly ash are classified as dangerous (Xu & Van Deventer, 2000). Therefore, the use of clay (Damayanti, 2022) as a raw material for making zeolite is quite effective because clay is an easy material to find. Therefore, using clay as a primary material for zeolite is seen as a very efficient alternative because it uses a low-cost material. Iftitahiyah *et al*., (2018) have synthesized zeolite from kaolin clay to adsorb cationic dyes and as an alternative precursor. The use of clay has been widely used to synthesize zeolite by several researchers. The research of Bahgaat *et al*., (2020) has succeeded in synthesizing zeolite from natural clay Wadi Hagul which was synthesized with a calcination temperature of 800°C for 6 hours and refluxed using 1 M NaOH without a catalyst. Based on this research, it was found that the crystallinity was excellent, namely 100%. Based on these studies, it can be said that clay can be a promising raw material for synthesizing zeolite.

The amount of impurities such as quartz and iron (Fe) in Maredan clay is still a problem in zeolite synthesis because it can affect the framework and purity of the zeolite produced. One way to remove impurities in clay is by activation. Activation there are 2 ways, chemical and physical. In chemical activation using acid as an activator, minerals can dissolve impurities in clay such as K, Na, Ca, Mg and Fe. In this study, 1 M Sulfuric Acid was used as an activator to dissolve impurities in the clay to increase the purity of the zeolite produced. The physical activation process was carried out by calcining at a temperature of 650°C for 3 hours. According to Kassa *et al*., (2022), the calcination process at a temperature of 650°C was carried out to change the structure of the kaolin mineral into an amorphous metakaolin which would facilitate the reaction in the synthesis of zeolite.

Another way that can be used to increase the purity of the zeolite produced can also be done with the zeolite aging process. Aging is a step in zeolite nucleation that can affect the crystallinity and yield of the final product. This is because the increasing aging time will increase the speed of the zeolite crystallization process, accelerate the crystallization rate, and increase the number of crystal nuclei formed. The crystallization temperature also affects the type of mineral and the purity of the zeolite result (Ginting *et al*., 2019). According to Yang *et al*., (2018), the crystallinity of zeolite 4A gradually increased when the crystallization temperature was increased from 40°C to 90°C. The crystallinity of sodalite zeolite increases with increasing crystallization temperature. At the zeolite synthesis stage in this study, the crystallization temperatures of 80, 90 and 100°C were used.

Material and Methods

Activation of Maredan Natural Clay

Clay samples were taken in Maredan Village, Tualang District, Siak Regency, Riau Province. The clay samples were washed with distilled water and dried in the sun. Then the clay was ground and sieved using a $100 \geq x \geq 200$ mesh sieve (Endecotts LTD Sieve) (x is the particle size). The clay was dried in an oven (Stericell 222) at 105°C for 24 hours. Then the clay was stored in a desiccator.

It weighed as much as 10 grams of clay then a suspension was made in 100 mL of 1 M H_2SO_4 (Merck). The mixture was refluxed at 80°C for 3 hours and then filtered using the Whatman 42 filter paper. The clay was washed with aqua DM until the pH of the filtrate reached 4. Then the clay was dried in an oven at 105°C for 24 hours. The clay solids were ground and calcined at 650°C for 3 hours, then cooled in a desiccator.

Zeolite Synthesis

Maredan clay that has been activated as much as 10 g and 50 mL of 5 M NaOH (Merck) solution was put into a three neck flask. In the mixture, the aging process was carried out by stirring using a magnetic stirrer at a speed of 300 rpm at a temperature of 80°C for 5 hours. The resulting mixture was refluxed with temperature variations of 80, 90 and 100°C for 8 hours without stirring. The results obtained were filtered and washed with aqua DM until the pH of the filtrate approached 8. The residue obtained was dried at 105ºC for 24 hours.

Zeolite Characterization

Several stages of characterization were carried out to determine the characteristics of Zeolite. XRF (PANanalytical Epsilon 3) analysis was used to determine the elemental composition contained in the zeolite sample. XRD (PANanalytical Empyrean) analysis was used to determine the type of mineral and the purity of the zeolite. FTIR (Shimadzu IR Prestige-21) analysis aims to determine the type of bond vibration and functional groups contained in the zeolite. Surface morphology analysis of clay and zeolite samples was carried out using Scanning Electron Microscopy (SEM JOEL 6510- LA).

Result and Discussion

Complex Compound Synthesis

Maredan clay before and after activation was characterized using XRF to determine the chemical composition and Si/Al ratio. The results of XRF characterization are shown in **Table 1**. Based on these data, it is known that Maredan clay contains 48.762% Si, 26.248% Al, 18.144% Fe and small amounts of other metals. The high content of Si and Al in Maredan clay causes Maredan clay to be used as a raw material for zeolite synthesis. However, before Maredan clay is used for zeolite synthesis, it is first activated using H_2SO_4 to reduce metal impurities, one of which is ferrous metal (Fe) from 18.144% to 14.623%.

Table 1. Chemical composition of Maredan clay before and after activation

Elemental Composition, %	LM	LM:4K
Si	48.76	53.36
Al	26.25	24.25
Fe	18.14	14.62
K	2.93	3.25
Ca	0.29	0.26
Mg	0.30	0.99

Table 2. Maredan clay Si/Al before and after activation

Clay Sample	Si (moles)	Al (moles)	Si/Al
LM	0.174	0.097	1.79
LM:4K	0.189	0.089	2.12

Information :

LM : Maredan clay without activation

LM:4K : Maredan clay which was activated using H_2SO_4 1 M, pH after washing was 4 and calcined at a temperature of 650°C/3 hours

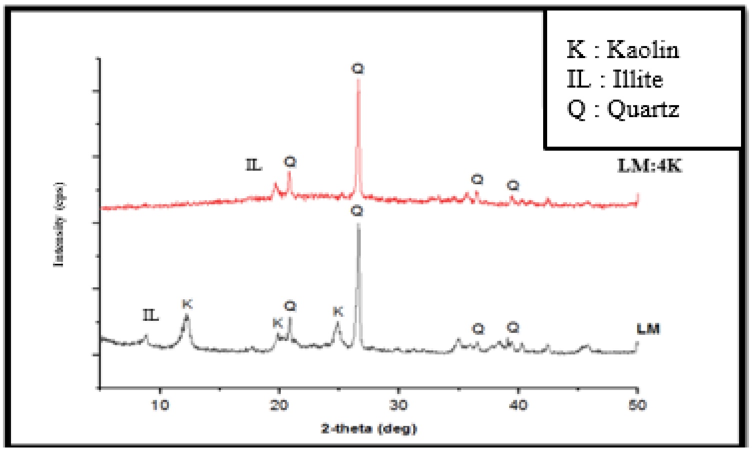

Figure 1. Maredan clay diffractogram pattern

The results of the Maredan natural clay diffractogram before and after activation are shown in **Figure 1**. showed that the unactivated Maredan clay contained illite, kaolin and quartz minerals. The illite peak appears at 2θ = 8.86°, the kaolin peak appears at 2θ = 12.18°, 19.84° and 24.82°. Quartz peaks can be seen at angles of 2θ = 20.92°, 26.64°, 36.5° and 39.04°. This is in accordance with the research of Malik *et al.*, (2020) which states that Maredan clay consists of quartz, kaolinite and illite minerals. Maredan clay diffractogram after activation showed the loss of sharp peaks of kaolin 2θ = 12.18°, 19.84° and 24.82° which indicated that kaolin had turned into amorphous metakaolin. According to Rahman *et al.*, (2019) after the calcination process at 750°C for 3 hours, the XRD pattern showed significant expected changes compared to the initial kaolin pattern, which was characterized by loss of the kaolin diffraction peak, accompanied by the appearance of amorphous aluminosilicates. Metakaolin is an amorphous material and the highest diffraction peak corresponds to the presence of quartz (SiO_2), which is the crystalline phase, in metakaolin. The transition of kaolin to metakaolin occurs in the temperature range of 400-800°C due to the OH structure dehydroxylation process of kaolin and metakaolin formation (Rahman *et al.*, 2019).

In the LM:4K sample, illite peaks can be seen at an angle of 2θ = 19.65° while quartz peaks can be seen at an angle of 2θ = 20.81°, 26.58°, 39.44° and 42.41°. The Si/Al ratio was determined from the ratio of silica and alumina content in the clay. The Si/Al ratio of Maredan clay before and after activation can be seen in **Table 2**. Based on these data, after activation using acid will increase the Si/Al ratio of Maredan clay from 1.79 to 2.12. The result is better than the research conducted by Panda et al., (2010) from 0.65 to 0.81. This ratio increased due to the loss of some impurities and an increase in the percentage of Si and Al dealumination.

Table 3. Chemical composition of zeolite synthesized by Maredan clay

Zeolite Sample	**Elemental Composition (%)**					
	Si	Al	Fe	K	Ca	Mg
Z-A5-T80	50.443	26.396	15.780	2.533	0.472	0.560
Z-A5-T90	52.034	24.812	14.795	2.573	0.551	1.258
Z-A5-T100	49.613	27.147	16.159	2.434	0.441	0.155

Table 4. Si/Al of zeolite synthesized by Maredan clay

Zeolite Sample	Si (moles)	Al (moles)	Si/Al
Z-A5-T80	0.179	0.098	1.83
Z-A5-T90	0.189	0.092	2.01
Z-A5-T100	0.177	0.101	1.75

Information :

Z-A5-T80 : Zeolite with an aging time of 5 hours and a crystallization temperature of 80°C
Z-A5-T90 : Zeolite with an aging time of 5 hours and a crystallization temperature of 90°C
Z-A5-T100 : Zeolite with an aging time of 5 hours and a crystallization temperature of 100°C

The results of the XRF analysis of the synthesized zeolite are listed in **Table 3.** showed that the synthesized zeolite contained large amounts of Si and Al where other metals such as Fe, K, Ca and Mg were present as impurities. The chemical composition of the synthesized zeolite was compared with the chemical composition of Maredan clay after activation (**Table 1**). Based on the data in **Table 3**. showed that the Si content slightly decreased from 53.361% to 49.613% with the addition of NaOH. Previous research also showed a decrease in Si content such as Saputra's (2020) research from 55.889% to 49.613% at pH 4 conditions and an aging time of 5 hours and Poniran's (2020) research also showed the same results, from 52.360% to 51.064% at pH after leaching from 2 to 4. The percentage of Si, Al and Fe in the synthesized zeolite tends to be unstable with increasing crystallization temperature. This is because zeolite is in the form of a heterogeneous solid so the distribution of elements is uneven.

The Si/Al ratio of the synthesized zeolite is shown in **Table 4**. Based on these data, the Si/Al ratio of synthesized zeolite is between 1.75 - 2.01 which corresponds to the Si/Al ratio of sodalite zeolite. The research of Wahyuni *et al.*, (2019) showed that the Si/Al ratio for sodalite zeolite was between 1.5-3. The Si/Al ratio of the synthesized zeolite tends to decrease compared to the Si/Al ratio of the clay after activation. The decrease in the Si/Al ratio was due to a decrease in Si levels in the synthesized zeolite.

The crystallinity of the sodalite zeolite can be seen in **Figure 2**, the influence of the crystallization temperature will increase the crystallinity of the sodalite zeolite. According to Hamid *et al.*, (2020) the higher the crystallization temperature will increase the peak intensity. It is also appropriate in this study that the crystallization temperature with the reflux method increased the intensity of the zeolite peak obtained. Phenomena that arise due to an increase in temperature will cause the intensity of the crystalline phase to increase and the amorphous phase to decrease. This indicates an increase in product crystallinity caused by an increase in temperature. The percentage of sodalite crystallinity in all zeolite synthesis ranges from 31.12% - 51% with the highest crystallinity obtained by zeolite with a crystallization temperature of 100°C, which is 51%. This result is better than research conducted by Saputra (2020), which is 48.42 at pH 4 and has an aging time of 5 hours.

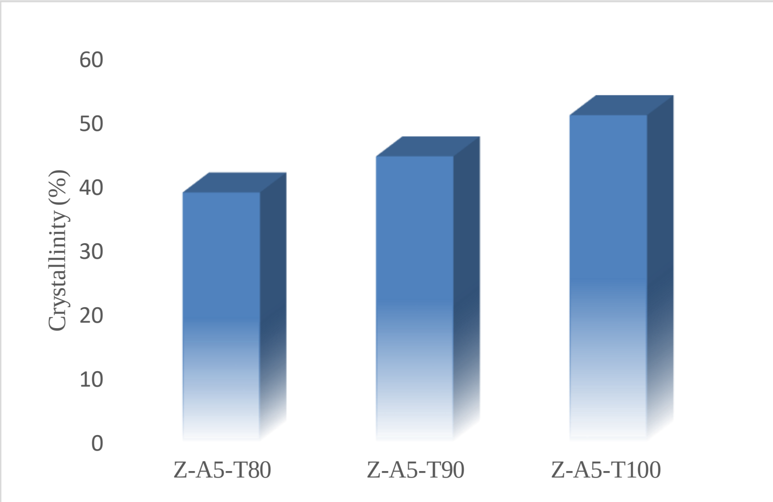

Figure 2. Crystallinity of synthesized sodalite zeolite

Based on these results can be seen in **Figure 3**, it is also known that the crystal size of the synthesized zeolite is in the range of 30.03 - 32.30 nm with the smallest crystal size having a crystallization temperature of 100°C, which is 30.03 nm. This research is comparable to research conducted by Malik (2020), 46.40; 41.24 and 45.28 nm at aging conditions of 3 hours, 5 hours and 7 hours. The higher the crystallization temperature, the more crystal nuclei are formed so that the crystal growth takes longer which causes the crystal size to get smaller.

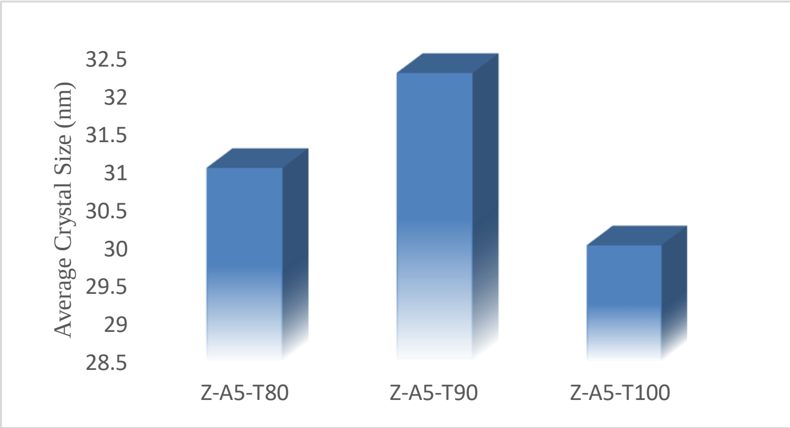

Figure 3. Average crystal size of synthesized sodalite zeolite

Characterization using FTIR was carried out in the wave number range of 400-1200 cm^{-1}, in the wave number range there were functional groups possessed by zeolite. The results of the IR spectrum of the zeolite are shown in **Figure 4.**

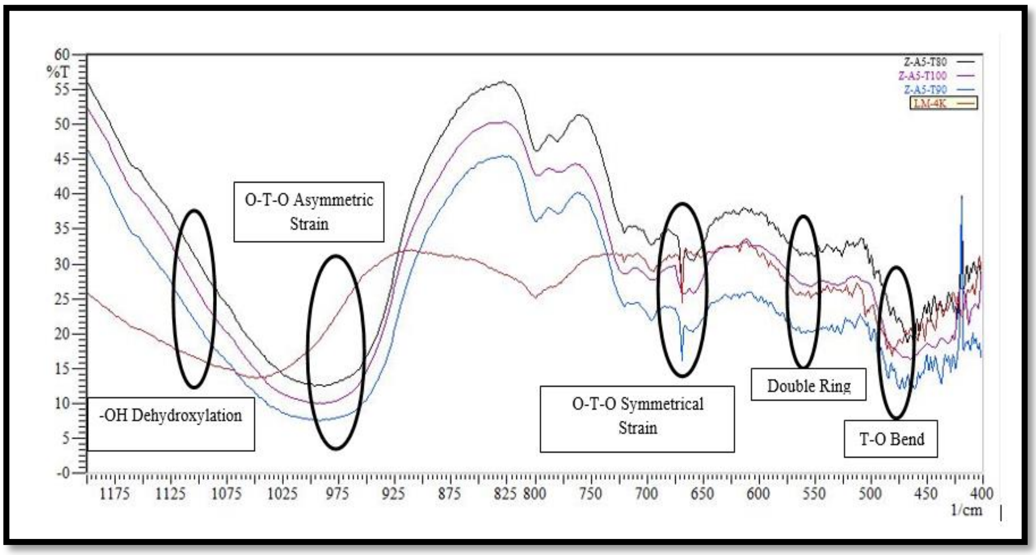

Figure 4. Result of IR spectrum with variation of crystallization temperature

The interpretation of the peaks formed has the same criteria because they contain the O-T-O functional group (T can be replaced with Si or Al) which is common in zeolites. The absorption band in the area between 420-500 cm^{-1} is related to the T-O bending vibration absorption. The absorption band in the area between 1250-950 cm^{-1} shows asymmetrical internal (O-T-O) and external (T-O-T) strain vibration absorption. The absorption band in the 850-650 cm^{-1} region is a symmetrical strain vibration.

The formation of sodalite zeolite is characterized by the presence of a double ring vibration absorption band, which is a characteristic feature of the structure of sodalite zeolite. Each zeolite has a different double ring structure and the double ring of sodalite zeolite is a D4R or D6R double ring. The specific character of the double ring is shown in the absorption region of 500-650 cm^{-1} which is shown in all samples. According to Kianfar, (2019) absorbance bands around 1250-950, 820-650 and 500-420 cm^{-1} are peaks that are not sensitive to changes in structure. While in the 650-500 cm^{-1} area is a peak that is sensitive to changes in the structure and composition of the framework.

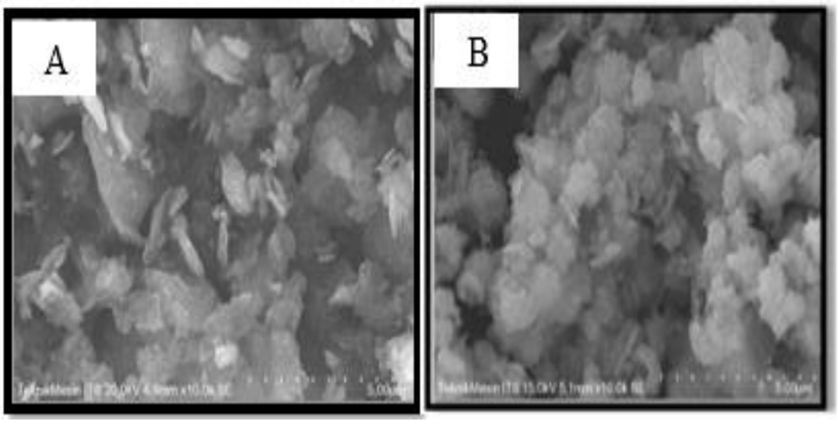

Figure 5. A. SEM analysis results from LM:4K sample (10,000x magnification) and **B**. SEM analysis results from Z-A5-T90 zeolite (10,000x magnification)

Analysis using SEM was carried out to determine the surface morphology of the clay and zeolite samples. The results of SEM characterization of clay and zeolite samples are shown in **Figure 5.** Based on **Figure 5. A**. The morphology of LM:4K clay can be seen in the form of thin plates that are spread unevenly. The morphology of this sample is also in accordance with the research of Yu *et al*., (2022) which states that the surface morphology of kaolin is in the form of thin plates.

The micrograph in **Figure 5.B.** with a magnification of 10,000x shows that the synthesized zeolite with an aging time of 5 hours and a crystallization temperature of 90°C produces almost the same morphology in the form of spherical crystals. The effect of aging time on the morphology of the zeolite produced is related to the homogeneity of the zeolite. The longer the aging time is carried out, the more clearly the spherical shape with petals will be seen with a smoother surface as a characteristic of sodalite zeolite and more homogeneous. Increasing the temperature causes the particle shape and crystal morphology to become clearer. This proves that the better particle shape results in increased aggregation.

Conclusion

A purity of 51% sodalite and a crystal size of 30.03 were obtained at a crystallization temperature of 100 °C. The result of XRF characterization showed that the ratio of Si/Al zeolite synthesized was between 1.75 which corresponded to the ratio of Si/Al zeolite sodalite. Based on XRD data, the type of mineral that composes synthetic zeolite is a mixture of sodalite zeolite, and LTA zeolite, with the remaining components in the form of muscovite clay and quartz as non-clay minerals. From the FTIR analysis, the formation of the sodalite framework was indicated by the external vibration of the D4R or D6R double rings at a wave number of 558.42 cm^{-1}. The results of SEM characterization showed that the morphology of activated Maredan clay (LM:4K) was in the form of thin plates, while the morphology of the synthesized zeolite showed spherical, petal-shaped crystals which were the typical morphology of sodalite zeolite.

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