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Synthesis of silica-rich zeolite using quaternary ammonium-based templates

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Abstract. In this study, silica-rich zeolite was made at a ratio Si/Al= 30. The template used was cationic surfactant from quaternary ammonium, tetrapropylammonium bromide (TPAB) and cetyltrimethylammonium bromide (CTAB). The CMC for TPABr $(n - C_3H_7)4N$ is 10^{-3} M, and spherical micelle of CTAB is 8.9×10^{-4} M. The concentration of the TPAB and CTAB which used in this study were 0.0325; 0.125; 0.25 and 0.5 M. The results show that the type of synthesized zeolites are silica-rich sodalite with the crystal size 216 A°. The use of quaternary ammonium-based templates can increase crystallinity and reduce the size of silica-rich sodalite crystal grains but remain in the mesopore material size range. The concentration of the tetrapropylammonium bromide (TPAB) template is very influential on surface area and pore volume, whereas for cetyltrimethylammonium bromide (CTAB) it affects surface area but not on pore volume. The largest surface area and pore volume of 96 m^2/g and 3.4×10^{-2} obtained for silica-rich sodalite using a landfill template at small concentrations.

1. Introduction

Zeolites is a three-dimensional crystal structure with an aluminosilicate framework composed of SiO_4^{4-} and AlO_4^{5-} tetrahedral units that are connected by oxygen atoms. The ratio of Si^{4+} and Al^{3+} in the framework often reffered as the ratio of silica-to-alumina is generally written as either Si/Al mole ratio or SiO_2/Al_2O_3 mole ratio. The type of framework defines the structural properties of zeolites, including cage, pore opening, and channel structure [1]. The specific shape and size of the pores and channel in the zeolites make it have a specific selectivity so that it is often used as a catalyst. Besides, zeolites have heat-stable properties and have a large surface area. Zeolites consist of two types, namely natural zeolites and synthetic zeolites. Natural zeolite is an inorganic unrenewable natural material that has limitations because it cannot be renewed so that it will run out longer [2].

Generally, natural zeolites have a low Si/Al ratio and pore size of less than 10 A°, while for synthetic zeolites, the Si/Al ratio could be made in a range from 1 until up to 100. The ratio of Si/Al is used to describe the hydrophobicity of the zeolite surface. Synthetic zeolites always developed on various studies of the manufacturing process that adjusted to the purpose and interests of the application. Many parameters study the synthesis process (i) Si/Al and Si/Na ratios, (ii) reaction time,

and (iii) stirring in the reaction medium. The Si/Al and Si/Na ratios have a significant influence on the zeolite phase, long diffusion time, and the presence of stirring increases the zeolite crystallinity [3]. In the research was stated that zeolites formed at 100°C hydrothermal temperatures for three days, the ratio of Si/Al (v/v) = 1 yields sodalite (SOD) whereas at the ratio of Si/Al (v/v) = 15 and 25 produces NaP1[4]. The research also knew that the use of cetyltrimethylammonium bromide increases the crystallinity of the product because the peak of the XRD diffractogram is sharper and leaner. The sodalite zeolite with a Si/Al ratio of 1: 1 has a β-cages building unit consisting of six rings, and four rings joined to form a cuboctahedron. Six rings owned by this zeolite have small pores. Applications of sodalite zeolites include optical materials, hydrogen adsorption, and hydrogen storage [5].

It should be noted that zeolites with low Si/Al ratios or alumina-rich have some disadvantages; namely, the synthesis process uses low temperatures and is hydrophilic. High or silica-rich Si/Al zeolite ratio shows the surface area, large pore volume due to the density of the skeleton, and more hydrophobic, which causes higher adsorption capacity. Silica-rich zeolite is widely applied as adsorption, catalyst, and membrane [1]. Zeolites were synthesized in 400 W microwaves, at 180°C for 12 hours, resulted in ZSM-22 (TON) [6], in different types of solvents affecting their crystallinity, while the higher number of solvents improved the crystalline shape that appears in morphology ZSM-22. 1,2-dimethyl-3- (4-methyl benzyl) imidazolium and tetramethylammonium ions as organic structure-directing agents have been used in the synthesis of high silica LTA (Si/Al-17) [7]. Research [8] synthesized ZSM-5, which is a zeolite with a high Si/Al ratio with ultrasound time variation. The optimum conditions produced zeolite ZSM-5 at a temperature of 190°C within 20 minutes. Silica-rich zeolite has also been synthesized at hydrothermal temperature variations of 150, 160, 170, 180, 190°C Si/Al Ratios of 30, 35, and 40, the best results are obtained at 190°C within 18 hours [9].

Based on the background description above, it can be said that the character of zeolites can be engineered according to their goals and interests. The parameters of the synthesis process highly determine the zeolite characters. When the Si/Al ratio is low or alumina-rich, the product zeolite is hydrophilic while the high Si/Al or silica-rich ratio produces hydrophobic zeolites, so in this study, a silica-rich zeolite can be applied as both an adsorbent and catalyst for compounds. The character is hydrophobic.

2. Experimental

2.1. Materials
Sodium silicate sol, NaOH pellets, Al(OH)$_3$ powder, cetyltrimethylammonium bromide (CTAB) powder, tetrapropylammonium bromide (TPA) powder, HF p.a. and HCl p.a. and aquadest.

2.2. Instruments
The instruments needed for analysis were FTIR Spectrometer (Shimadzu), X-ray diffractometer (XRD) (Bruker D2 Phaset 2nd Gen), Gas surface analyzer/GSA (Quantachrome NovaWin)

2.3. Procedure
Sodium aluminate is prepared by dissolving NaOH in distilled water and heated. After all the NaOH has dissolved, Al(OH)$_3$ was added slowly until the clear solution was formed.

Synthesis of zeolite was done by adding a drop of Na$_2$SiO$_3$ solution into the Sodium Aluminate solution; the mole ratio of Si/Al was 30. The mixing was carried out with a magnetic stirrer at room temperature. The mixture was transferred to stainless steel and then put into an autoclave and heated at 200°C for 24 hours. Samples from the autoclave were filtered and washed with distilled water until it reaches pH 7. The residue was dried in an oven at 110°C for 2 hours. Furthermore, the product was characterized using X-ray Diffraction (XRD), Fourier Transform InfraRed (FTIR), and Gas surface analyzer (GSA).

The next step was to carry out the same procedures by adding an ammonium surfactant. The types of surfactants used are cetyltrimethylammonium bromide and tetrapropylammonium bromide. Each

surfactant was assessed at concentrations of 0.0325 M, 0.125 M, 0.25 M, and 0.5 M. After the hydrothermal process at 200°C for 24 hours, the product was neutralized with distilled water, then dried. The next important step when the synthesis using a template is to eliminate it by calcining at a temperature above its decomposition temperature. The calcination temperature in this experiment was 550°C for 3 hours. Product characterization also using FTIR, XRD, and GSA.

3. Result and discussion

Zeolite was synthesized by reacting sodium aluminate and sodium silicate. In this research, the formation of sodium aluminate is obtained by dissolving aluminum hydroxide with sodium hydroxide, which is slowly heated. Mixing sodium aluminate and sodium silicate accompanied by stirring using a magnetic stirrer for 2 hours to form a homogeneous solution. When sodium silicate is mixed into the sodium aluminate solution, the solution turns white. That is because silanol derived from sodium silicate reacts with sodium aluminate through a polymerization reaction to form zeolites [10]. The formation of the gel indicates a strong interaction that is the start of the silicate and aluminate polymerization process. The formation of zeolite crystals occurs when condensation is accompanied by polymerization of aluminate species and silicates in a saturated solution. In mixing Na_2SiO_3 and $NaAlO_2$, two phases are formed, namely the solid phase as an amorphous gel and the solution phase as a saturated solution. Both phases are at equilibrium. Amorphous gels will dissolve and reorganize to form species that are seedings of the crystal nucleus and are the nucleation stage. At that stage, the magnitude of the Si/Al ratio affects the process of change from sol to gel. After the hydrothermal process at 200°C for 24 hours, a white crystalline solid was obtained. The hydrothermal process aims to uniform the crystals formed and perfect the growth of zeolite crystals [11]. The hydrothermal process involves water and heat, where the mixture is heated at high temperatures in tightly closed containers [12]. This situation is intended to create a balance between water vapor and solution so that there is no water vapor that evaporates, and the composition of the solution remains. At this hydrothermal stage, a condensation reaction occurs, which allows the formation of new bonds such as Al-O-Si, Si-O-S, (T-O-T), where T is the Si or Al atom [13].
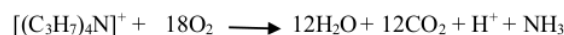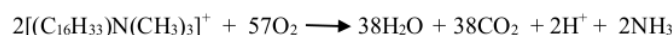
In this research, zeolite was synthesized at Si/Al ratio = 30. The resulting zeolite is called silica-rich sodalite zeolite. They have a high Si/Al ratio of more hydrophobic character. Hydrophobicity is properties related to interaction characteristic of polar compounds, especially water. Damjanovic [14] and de Ridder [15] stated that water uptake could be prevented by highly hydrophobic zeolites. Therefore, blocking pores by water molecules can be avoided. As a result, there will be more organic molecules such as compounds in coconut oil to diffuse and adsorb on pores [15] [16].

The hydrophobicity and zeolite-water interactions are strongly influenced by the chemical composition of zeolites. The hydrophobicity could be influenced by a number of silanol species because this species adsorbed water. The Al sites in the framework of zeolite could interact with water molecules [17]. For the same type of zeolite framework, the decreasing of Al content would increase the hydrophobicity. It can be said that at a higher Si/Al ratio it is more hydrophobic; therefore it has a higher adsorption capacity [14,18,19, 20, 21, 22, 23]. When the Si/Al ratio is high enough, the hydrophobicity of zeolites is no longer of significant importance because the effect of water competition and water cluster hindering has become negligible. Gonzalez-Olmos [24] found that the increasing the Si/Al ratio from 800 to 1366, zeolite with MFI structure unable to improve their adsorption capacity of MTBE, and even a slight decrease was observed when the surface area of zeolites increased from 265 to 330 m^2g^{-1}. However, in this case, the adsorption capacity of zeolites became independent of the Si/Al ratio for ratios over 800 [20].

When the formation process of sodalite zeolite was added to the surfactants as a template from organic compounds, the condensation polymerization process coincides with the interaction between the cationic surfactant micelles and negatively charged of silicate and aluminate species [25]. Removal of surfactants that act as pore molds can be achieved through calcination. The surfactant molecule has decomposed; it would leave a form of a pore hole of a certain size. This pore is then used as one of the active sites of the catalyst material. The size of the pore formed depends on the size of

the surfactant micelle. In this study, two types of surfactants with quaternary ammonium structure were examined, namely cetyltrimethylammonium bromide (CTAB) and tetrapropylammonium bromide (TPAB). The surfactants were chosen as templates pores because they have different alkyl chain lengths, the CTAB containing the longest alkyl chain is 16 carbons and three methyl groups (1 carbon), whereas the nitrogen atoms in TPAB bind four propyl groups (3 carbons). For the CTAB having CMC 1 and 2, CMC 1 of CTAB is pre-micelle to spherical micelle, the value is $8.9.10^{-4}$ M, while the CMC 2 of CTAB (spherical micelle to rodlike micelle) is $2.0.10^{-2}$. M [26]. The transformation from CMC 1 to CMC 2 depends on the temperature, the degree of dissociation, and the length of the alkyl surfactant chain. As the temperature increases, the concentration needed for transformation from CMC 1 to CMC 2 will increase. If the concentration and amount of surfactant needed is more, it is expected that micelles formed to interact with the silica-alumina framework will also be larger so that the pore mold is expected to be larger.

Meanwhile, the CMC for TPABr $(n-C_3H_7)_4N$ is 10^{-3} M [27]. The micellar formation takes place above a specific surfactant concentration, below which surfactant molecules are present as monomers [28]. In this study, using surfactant concentrations both of CTAB and TPAB were 0.0325; 0.125; 0.25 and 0.5 M. It is expected that there are differences in the interaction of these surfactants with aluminate and silicate species so that different pore sizes will be obtained. Decomposition reaction of CTA and TPA surfactants molecular are as follow.

$$2[(C_{16}H_{33})N(CH_3)_3]^+ + 57O_2 \longrightarrow 38H_2O + 38CO_2 + 2H^+ + 2NH_3$$

$$[(C_3H_7)_4N]^+ + 18O_2 \longrightarrow 12H_2O + 12CO_2 + H^+ + NH_3$$

Illustration, the interaction of quaternary ammonium surfactant with aluminate silicate species, can be seen in Figure 1. In the calcination process at 550^oC, the CTAB and TPAB surfactant templates are decomposed into carbon dioxide gas, water vapor, and ammonia gas, so that the hole remains as a pore in this material.
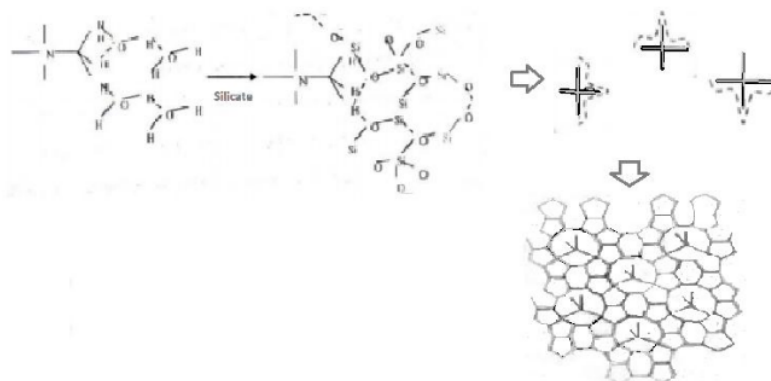

Figure 1. The formation scheme of zeolite framework uses quaternary ammonium-based surfactants as a pore template

To identify the zeolite results, characterization using XRD to determine the crystallization of minerals formed and FT-IR to identify functional groups contained in a compound, as well as Surface area analyzer for pore size, pore-volume, and surface area.

3.1. Characterization of zeolite synthesized using Infrared Spectrophotometer

This characterization aims to identify functional groups contained in a compound. Zeolites generally have a characteristic infrared absorption region around the wave number 300-1200 cm^{-1}. The zeolite structure has internal strands and external strands. Internal braid on the product zeolite appeared in the absorption area around 950-1250 cm^{-1}, indicating the existence of asymmetrical stretching vibrations from Si — O and Al — O from the aluminosilicate framework. According to the references [29], the sodalite wave numbers are 428-461 cm^{-1}, which is the S4R buckling vibration. S4R is a specific zeolite character which is shown by the appearance of absorption in the region. This S4R is an external link between the zeolite layers and the other. The absorption band at the wave number 661-714 cm^{-1} is symmetrical stretching vibration Si-O-Si or Si-O-Al, and ~ 990 cm^{-1} is asymmetrical stretching vibration. In addition, the purpose of FTIR analysis is also to determine the presence of directing molecules/surfactant templates. Figure 2 shows the spectra zeolite synthesized using the TPAB template, while Figure 3 uses the CTAB template.
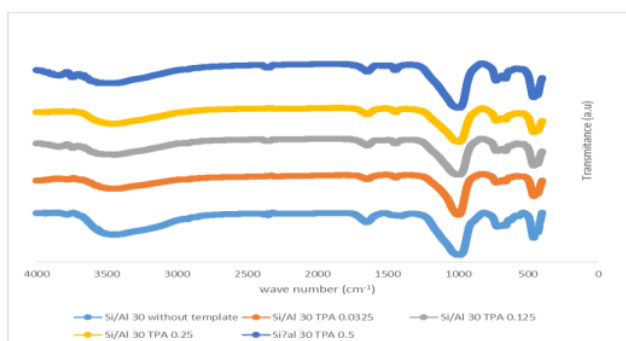

Figure 2. The spectra of zeolite synthesized using the TPAB (tetrapropylamonium bromide) template in various variations
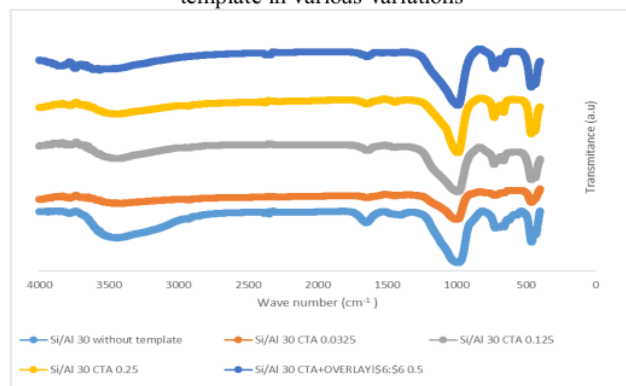

Figure 3. The spectra of zeolite synthesized using the CTAB (cetyltrimethylammonium bromide) template in various variations

The addition of the surfactant template no change the fingerprint area of the FTIR spectra, both samples without surfactants and with surfactants CTAB as well as TPAB showed similar absorption pattern in the area. The wavenumber ~ 461 cm^{-1} indicates the presence of bending vibrations from 4 single rings (S4R). In addition, the wave number ~ 1000 cm-1 shows a peak which indicates the existence of asymmetrical stretching of Si-O-Si or Si-O-Al. This shows that the zeolite produced is sodalite zeolite. On the other side, there is no absorption of wave numbers that indicate the presence of

groups containing N, meaning that in the sample, the surfactant has decomposed. From both figures 2 and 3, it was observed that the addition of surfactants significantly affected the absorption of 3400 cm⁻¹ wavenumbers, indicating that there was a decrease in the number of O-H groups in the sample. It is predicted that the decrease in the number of OH groups is correlated with the formation of an increasingly larger Si-O-Si or Si-O-Al zeolite framework. The increase of surfactant concentration from at 0.0325, 0.125, 0.25, and 0.5 both on CTAB and TPAB did not significantly affect the overall spectra of zeolites synthesized with surfactants.

3.2. Characterization of zeolites synthesized by X-Ray Diffraction (XRD)

The XRD (X-Ray Diffraction) method is a qualitative analysis method that provides information about the crystallization of a mineral because each mineral has a characteristic diffractogram pattern. The crystallinity of the sample can be seen from the display of the diffractogram pattern. Diffractograms that have clear peaks separation patterns and high peak sharpness intensities have good crystallinity. The change in the relative intensity of the peaks on the diffractogram shows the number of measured crystal fields are not the same. For example, at a certain angle 2θ, which has a relatively high intensity, then at the angle 2θ, there are many areas of the same crystal. The tendency of the sharper peak and separation between the peaks clearly shows the better crystallinity.

The synthesized zeolite has a diffractogram pattern, as shown in Figures 4 and 5. Based on the figure, the zeolite synthesized has similar patterns and peaks with standard sodalite zeolites. Zeolite sodalite standard RRUFF ID R040141 has peaks at 2θ = 14.17°, 24.62°, 31.90°, 35.03°, and 43.25° or referring to JCPDS data the angles of 2θ sodalite standard are 14.16°, 24.66°, 31.99°, 35.13° and 43.39°. Zeolite sodalite has a cubic crystal system, usually in the plane hkl (111), forming a pseudohexagonal prism. The value of 2θ for each zeolite synthesized and standard sodalite can be seen in Table 1 for the TPAB template and Table 2 for the CTAB template.
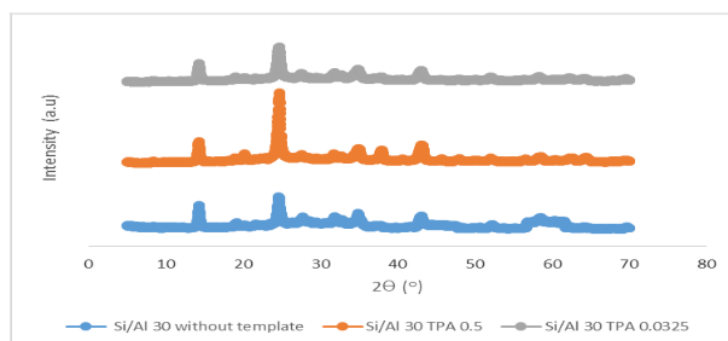

Figure 4. The diffractogram pattern of zeolite synthesized using the TPAB (tetrapropylammonium bromide)
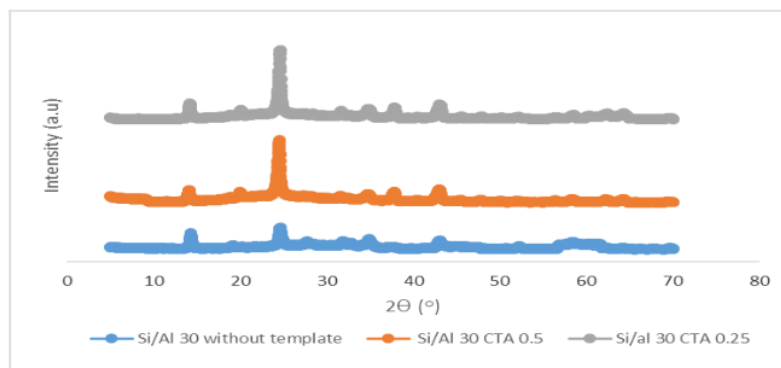
Figure 5. The diffractogram pattern of zeolite synthesized using the CTAB (cetyltrimethyamonium bromide)

Tabel 1. The value 2θ diffractogram of zeolite synthesized using TPAB template and standard

	2θ (°)		
Sodalite References RRUFF	Ratio Si/Al 30 without template	Ratio Si/Al 30 TPAB 0.0325	Ratio Si/Al 30 TPAB 0.5
14.17	14.31	14.274	14.26
24.62	24.65	24.644	24.71
31.90	31.87	31.831	31.79
35.03	34.89	34.899	34.94
37.96	37.80	37.851	-
43.25	43.02	43.002	43.15
45.00	-	-	-
52.51	52.23	52.110	-
60.82	61.51	-	-

Based on Figures 4 and 5 and tables 1 and 2, all zeolites synthesized using both the TPAB and CTAB templates have diffractogram peaks that are very similar to sodalite zeolites. Therefore, it can be said that all synthesis products contain sodalite zeolites. The figure also observed differences in peak intensity. The addition of TPAB template at a small concentration of 0.0325 M did not affect to the intensity of the peak when compared without the template, but when the concentration was increased almost 16 times to 0.5 M, there was a significant increase in the main peak that is at $2\theta = 24.66°$. This data shows that the crystalline fields that compose the sodalite zeolite are more regularly and numerously. It can be said the addition and increase in template concentration increase the crystallinity of sodalite better than without template.

The use of CTAB at a concentration of 0.25 M increased the intensity of the main peaks of sodalite when the concentration was increased to 0.5 M, the increase in peak intensity was not significant. However, when compared to sodalite without the CTAB template, it is clear that the intensity increases and the baseline is flatter. This shows an increase in crystallinity in the product**.**

Table 2. The value 2θ diffractogram of zeolite synthesized using CTAB template and standard

	2θ (°)		
Sodalite Reference RRUFF	Ratio Si/Al 30 without template	Ratio Si/Al 30 CTA 0.25	Ratio Si/Al 30 CTA 0.5
14.17	14.31	14.185	14.11
24.62	24.65	24.616	24.54
31.90	31.87	31.691	31.62
35.03	34.89	34.905	34.79
37.96	37.80	37.864	37.65
43.25	43.02	43.121	43.02
45.00	-	45.584	-
52.51	52.23	-	-
60.82	61.51	-	-

Table 3. The crystals size of the zeolite synthesized

Sample	Crystal size A°
Si/Al 30 without template	216.9
Si/Al 30 TPA 0.0325	204.81
Si/Al 30 TPA 0.5	209.27
Si/Al 30 CTA 0.25	234.73
Si/Al 30 CTA 0.5	206.07

Table 3 shows the size of the synthesized sodalite crystals. The size of the crystal grains is calculated using the Scherer equation. The use of templates at both low (0.0325 M) and high (0.5 M) concentrations has the effect of reducing the size of the crystal grains. While the presence of CTAB at moderate concentrations (0.25 M) increased the size from 216.9 A° to 234.73 A°, but using a CTAB template concentration of 0.5 M the size actually dropped to 206.07 A°. The size reduction will correlate with its surface area. Crystal size on each sample is influenced by the concentration and type of template used as a pore molding agent and directing its structure at once, therefore, each sample experiences a different crystal growth rate.

3.3. Characterization of zeolites synthesized by Gas Surface area Analyzer (GSA)

The Gas Surface area Analyzer (GSA) method uses nitrogen adsorption to determine the total surface area, pore volume, and pore size of a porous solid. The BET equation can only be used for adsorption isotherms which have a P/Po value of around 0.05-0.3 [30]. The analysis results obtained isothermal of adsorption-desorption graphs of sodalite synthesized shown in Figure 6 and Figure 7.
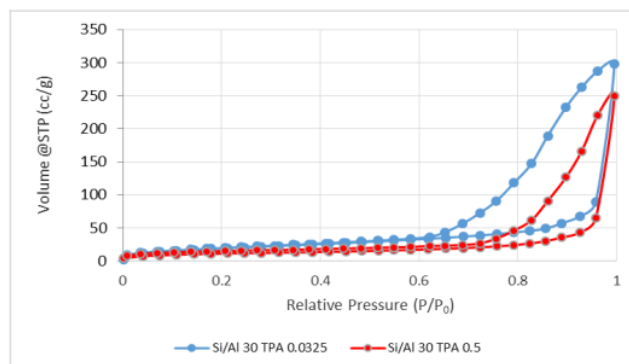
Figure 6. Isotherm adsorption of sodalite zeolite synthesized using TPAB template
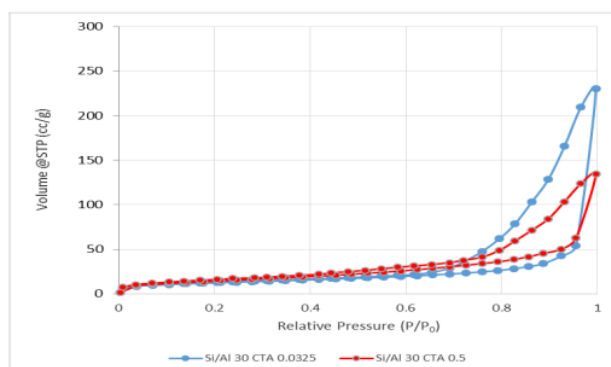

Figure 7. Isotherm adsorption of sodalite zeolite synthesized using CTAB template

The isothermal adsorption-desorption graphs of sodalite zeolite synthesized with TPAB templates (Figure 6) and CTAB (Figure 7) have hysterisis loops in the area of about 0.7. An isothermal curve is a type IV isothermal graph (based on IUPAC). This indicates that the synthesized sodalite zeolite is classified as mesoporous solid. The absorption ability of synthesized sodalite zeolites with TPAB is higher than CTAB.

Tabel 4. Surface area, pore diameter and volume synthesized sodalite zeolite

Code of sample	Surface area (m^2/g)	Average Half pore width (A^o)	Average of pore diameter (A^o)	Pore volume (cc/g)
Si/Al 30 TPA 0.0325	96.537	18.354	36.708	3.4×10^{-2}
Si/Al 30 TPA 0.5	52.241	14.653	29.306	1.9×10^{-2}
Si/Al 30 CTA 0.0325	64.525	19.93	39.86	2.3×10^{-2}
Si/Al 30 CTA 0.5	77.653	21.537	43.074	2.8×10^{-2}

From the data in Table 4, it can be seen that the concentration of the template influences the mesoporous characters. Both TPAB and CTAB templates are surfactants with quaternary ammonium hydrophilic portions. TPAB has a shorter alkyl chain, but the head binds to 4 propyl groups (C3), causing a pore size slightly smaller than CTAB, which has one alkyl group C16 but the head binds

only three methyl groups (C1). It is possible that the dominant role in determining the size of the pore mold is the group that is in the nitrogen atom**.** In this study, the pore surface area is proportional to the pore volume of the synthesized sodalite zeolite. From this data, the sodalite zeolite adsorption capacity can be correlated well for their surface area, and no depending on the size of the pore radius. The highest adsorption ability is TPA 0.0325, which is equal to 3.4×10^{-2} cc/g.

Conclusions

Based on the data obtained and discussion, it can be concluded that silica-rich sodalite has been successfully synthesized using a ratio of Si/Al = 30. The size of silica-rich sodalite crystals without templates is 216 A°. The use of quaternary ammonium-based templates can increase crystallinity and reduce the size of silica-rich sodalite crystal grains but remain in the mesopore material size range. The concentration of tetrapropylammonium bromide (TPAB) template is very influential on surface area and pore volume, whereas for cetyltrimethylammonium bromide (CTAB) it affects surface area but not on pore volume. The largest surface area and pore volume of 96 m^2/g and 3.4×10^{-2} were obtained for silica-rich sodalite using a landfill template at small concentrations.

Acknowledgment

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