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**Effect of CTA-Br Templete on Synthesis of NaX Zeolite from Fly Ashes  
and Applikation of NaX zeolite Obtained as an Adsorbent of Heavy Metals  
Cd and Cu**

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**FAKULTAS MATEMATIKA DAN ILMU PENGETAHUAN ALAM  
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# Effect of CTA-Br Templete on Synthesis of NaX Zeolite from Fly Ashes and Applikation of NaX zeolite Obtained as an Adsorbent of Heavy Metals Cd and Cu

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## Abstract

NaX Zeolites were prepared from fly ashes by hydrothermal method with and without CTA-Br templete at pH 14, temperature 60°C and sea water as a solvent. The NaX Zeolite obtained was characterized by X-ray diffraction (XRD), Fourier transformed infrared (FT-IR) spectroscopy, Scanning electron microscopy (SEM), and has charateristics of NaX Zeolite. N<sub>2</sub> adsorption desorption results showed that NaX zeolites prepared with and without CTA-Br templete have a specific surface area 5.130 m<sup>2</sup>g<sup>-1</sup> and 4.889 m<sup>2</sup>g<sup>-1</sup>, average pore size 30.21 nm and 25.84 nm, and total pore volume 0.078 cc g<sup>-1</sup> and 0.064 cc g<sup>-1</sup> respectively. The application of NaX zeolite is as an adsorbent to heavy metal Cd and Cu. The maximum adsorption capacities of NaX zeolites that were prepared with and without CTA-Br templete against heavy metal ions were 333.3 mg g<sup>-1</sup> and 303.0 mg g<sup>-1</sup> for Cd<sup>+2</sup> ion and 322.6 mg g<sup>-1</sup> and 285.7 mg g<sup>-1</sup> for Cu<sup>+2</sup> ion respectively. These results indicated that synthetic NaX zeolites have potential as adsorbent of heavy metals Cd and Cu. The results also suggested that the adsorption capacity was affected by specific surface area, average pore size and total pore volume.

**Keywords** : fly ash, NaX zeolite, adsorption, heavy metal

## 1. Introduction

The rapidly growing energy deman in developing world in general and Indonesia in particular demands that high energy generation should be achieved. Though there are different ways to generate energy; historically, coal-based power generation has been predominant in Indonesia. Thermal powaer plant plays an important role in fulfilling the growing energy demand of the people. Currently, coal-base thermal pwer insallations in Indonesia contribute about 65% of the total installed capacity for electricity generation. Indonesia coals have very high ash content (25 and 45%) and typically, the coal with an ash content of 40% is widely used in the power plants of Indonesia. As consequence, a huge amount of fly ash is generated in thermal power plants, causing several disposal-related problems. As far as the disposal of fly ash is concerned, presently, the fly ash utilization for different purposes is less than 10% in Indonesia, which is significantly less than that has been achieved in other countries. The

major fly ash utilization area is the construction industry, which utilization approximately 50% of the generated fly ash (due to its proven availability) for several applications as admixture in cement/concrete, lime pozzolanic mixture (bricks/blocks etc.). The other usage of fly ash are low lying area fill (17%), roads and embankments (15%), dyke raising (4%), brick manufacturing (2%) and zeolite etc.

Zeolite are porous crystalline aluminosilicates of  $\text{SiO}_4^{4-}$  and  $\text{AlO}_4^{5-}$  tetrahedra connected by oxygen bridges. Among various types of zeolite, NaX zeolite offers a large pore diameter (0.74 nm). The framework is composed by linking sodalite cavity through double six-ring which create a large supercage cavity accessible by a three-dimensional 12-ring pore system [1,2]. Because of its well-defined structure, large pore volume, and appropriate ion exchange capability, NaX zeolite is widely used as ion-exchanger [3], absorbent [4] and catalyst [5]. From literature, NaX zeolite can be synthesized from various precursor material and by various methods. Fly ash has the potential to be converted into zeolites.

## **2. Experiments**

### **2.1. Materials and reagents**

The materials used in this study is fly ash coal power plant Ombilin Tanah Datar, West Sumatra, NaOH (Merck), sea water beach of Padang, glacial acetic acid (Merck), template CTA-Br (Merck), aquadest, and wastewater Inlet House Achmad Mochtar Hospital Bukittingi. Glassware used as a beaker, test tube, measuring cup, pipette, funnel. Other equipment such as mortars, analytical balance, stirrer, furnaces, autoclaves, centrifuges, ovens, and filter paper. For characterization of the sample used XRD (X'Pert Powder Type Philip PW4030 / 60), FT-IR, SEM-EDX (JOEL JSM-6390LA type), and types Spectronik visible spectrophotometer 20.

### **2.2. Zeolite synthesis**

NaX zeolite synthesized from natural materials, namely fly ash and processed with alkali hydrothermal method using seawater. The sea water used is sea water taken Padang beach waterfront section, and then screened to separate impurities and stored in a sealed container.

For NaX zeolite synthesis process is done by taking 10 grams of fly ash are melted with 12 grams of NaOH using a mortar and then difurnace at a temperature of  $550^{\circ}\text{C}$  for 1 hour. The result of mixing reconstituted with 46 mL of sea water and the pH adjusted to pH

variations 12,13, and 14 by adding glacial acetic acid. Do two variations of the synthesis is by using templates CTA-Br as much as 3.14 grams for each process of synthesis and without templates and followed with alkaline hydrothermal processes in the autoclave for 4 days at 60° C, after which it is filtered to separate the precipitate and filtrate. The precipitate was dried in an oven at a temperature of 80° C for 24 hours. After calcination was carried out at a temperature of 550°C for 5 hours.

### **2.3. Characterization**

Zeolite obtained were characterized using FT-IR to see the chemical structure of the functional groups on the zeolite NaX 600 cm<sup>-1</sup> wave number 4000 cm<sup>-1</sup>, after it was done too characterization by XRD to determine the structure, type, and size of the zeolite crystals, subsequent to know the morphology of the surface shape, uniform particle size, and content of the elements contained in the zeolite was analyzed using SEM-EDX.

## **3. Results and Discussion**

### **3.1. Zeolite synthesis**

#### **a. Synthesized of NaX Zeolite NaX from Fly Ash Without Template**

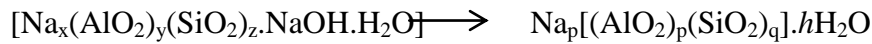
Fly ash is melted with NaOH to decompose components of silica and alumina which is mostly in the form of mullite and quartz to sodium silicate and sodium aluminate were instrumental in the formation of NaX zeolite framework. Cations Na<sup>+</sup> also will play a role in the process of ion exchange on the absorption of ammonia.

Furnace process aims to enhance the formation of the sodium silicate and sodium aluminate and eliminate other compounds that are not needed in the later crystallization process so that the zeolite synthesis has a high purity and absorption of ammonia for the better. Results furnace was added 46 mL of seawater to hydrothermal process that serves as mineralizer for the formation of minerals. The content of Na<sup>+</sup> is high enough in sea water can improve the process of forming sodium silicate and sodium aluminate, thus the building blocks of NaX zeolite framework more then the synthesis process will be more rapid progress.

Mixture of Fly Ash, NaOH and sea water has a pH of 14. To adjust the pH to 13 and 12 added glacial acetic acid. Acetic acid increases the concentration of H<sup>+</sup> ions dissolved in the mixture so as to increase the pH. The hydrothermal process is the process by which changes from amorphous fly ash into zeolite crystals. In this process of interaction between [SiO<sub>4</sub>]<sup>4-</sup> and [AlO<sub>4</sub>]<sup>5-</sup> tetrahedral bonding which will be arranged into a major component of the

formation of three-dimensional framework zeolite is hollow, the cavity is filled by cations  $\text{Na}^+$  derived from  $\text{NaOH}$  and possibly also of  $\text{NaCl}$  zeolite as a load balancer.

The following reactions occur during the process of alkaline hydrothermal synthesis:



### b. Synthesized NaX zeolite from fly ash with template CTA-Br

Template serves as a wake-up rectifier unit of primary tetrahedral framework formation of the zeolite framework, so that the distance between the tetrahedral-tetrahedral bonding to be the same. When the aluminate solution and silicate solution is mixed with the template, the template will form a micelle to reduce surface tension, hydrophobic groups will gather and hydrophilic head hydrophobic cluster will be away from each other so that formation of a circular cylinder.

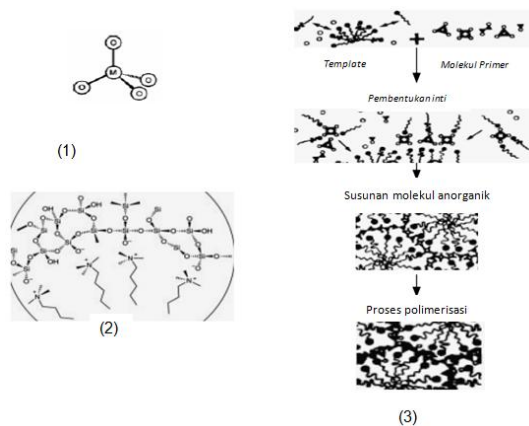


Figure 3.1. Schematic formation of zeolite as a template molecule steering (1) primary building units, (2) The process of formation of zeolites (zhao, 1996), (3) interaction CTA-Br Electrostatically hydrophilic group (head template) will interact with the primary building units (primary building units) of  $\text{TO}_4$ . Unit primer wake zeolite is a tetrahedral structure consisting of a central atom Si or Al surrounded by four oxygen atoms, namely  $[\text{SiO}_4]^{4-}$  or  $[\text{AlO}_4]^{5-}$ . Further interaction between  $\text{TO}_4$  and templates is the beginning of nucleation of zeolites, then will take place the growth of the zeolite crystal. The initial process of formation of the zeolites occurs continuously forming bonds Si-O-Al. Spontaneously template combined with inorganic molecules to form a structure which is then followed by solidification structure with heat treatment.

### 3.2. FT-IR Analisis of NaX Zeolite

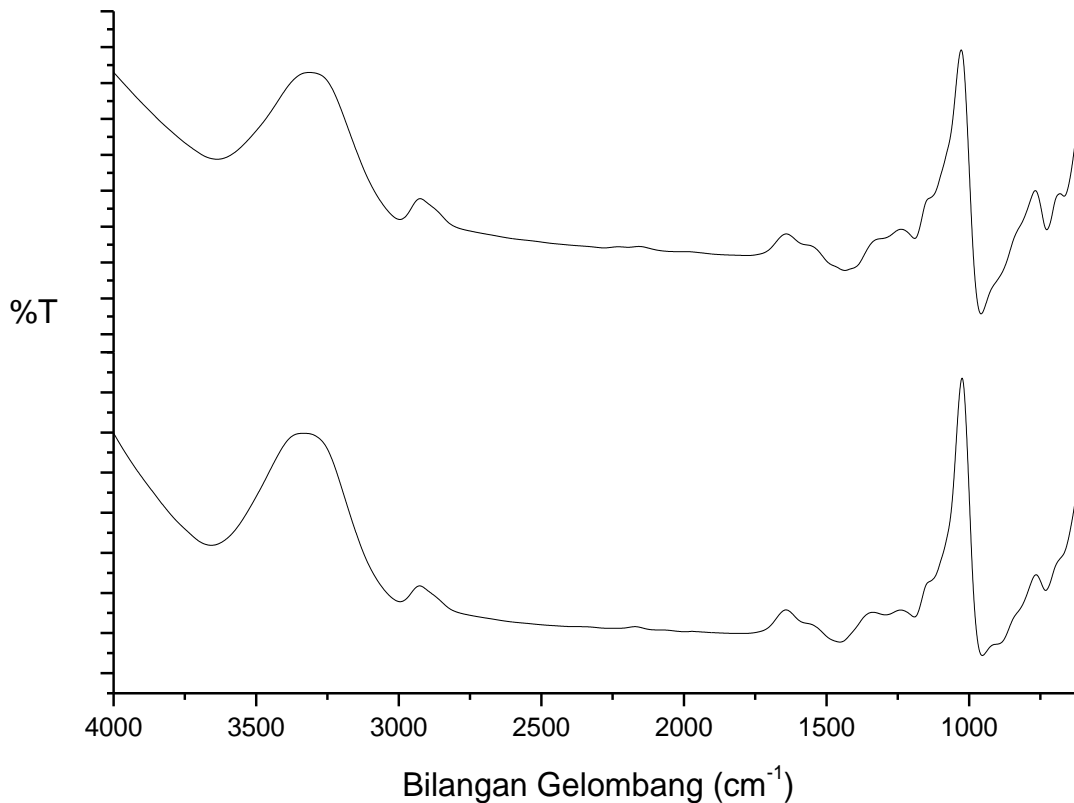


figure 3.2. FT-IR spectrograms synthesized NaX zeolite

Spectrograms of the picture above can be seen peaks in the following areas:

The area between  $3200-2000\text{cm}^{-1}$

The highlight in this area appear on any zeolite synthesized both with and without a template. This area shows the absorption bands associated with the hydroxyl group (OH) bonded, the cluster is estimated from water hydrates that bind the crystals. Then visible absorption bands in the region from  $2300-2000\text{ cm}^{-1}$ , where the area showed the presence of Si-H bond silanes. Of spectrograms above shows that these bonds appear on all zeolite synthesis.

The area between  $1600-850\text{ cm}^{-1}$

In this area showed an absorption band, which reported an Al and Si atom is substituted in the tetrahedral shape. In the span of this region showed the presence of vibration range of Si-O

and Al-O in  $\text{AlO}_4$  or  $\text{SiO}_4$  tetrahedral. Their wave numbers in this region can describe one of the characteristics of the formation of zeolites precisely on spectrograms range of  $1447.24$  to  $1435.06 \text{ cm}^{-1}$  and  $969.11$  to  $964.96 \text{ cm}^{-1}$  (Belviso C, 2009). From this data it can be concluded that the zeolite has been formed in all the synthesis results both with and without a template template for this area showed that all the samples in the test had a tetrahedral framework of Si-O and Al-O which is forming the zeolite framework.

The area between  $750\text{-}650 \text{ cm}^{-1}$

This area (the red box) is an area of fingerprints for NaX zeolite. For variation template synthesis using CTA-Br can be observed in the spectrograms [a] contained a single peak at  $715.73 \text{ cm}^{-1}$ , then the spectrograms [b] division occurs peaks at wave numbers  $716.73$  and  $701.46 \text{ cm}^{-1}$  and division more clearly visible on spectrograms [c] seen at  $723.25$  and  $669.81 \text{ cm}^{-1}$ . Cleavage is also well visible on spectrograms [e] and [f]. Of all the spectrogram above, for zeolites synthesized with template without template at all pH variations show a peak in the vicinity of  $746, 668, 690 \text{ cm}^{-1}$  which is a specific area for zeolite NaX (SM Holmes, 1998). Can be observed whereby the increase in pH, the division occurs specific peak on the zeolite NaX increasingly clear, this shows the amount of crystalline zeolite NaX more. At lower pH number of  $\text{H}^+$  ions contained in the mix more and more, so it will compete with the  $\text{Na}^+$  ions to bind to the main framework of the zeolite led to the formation of zeolites NaX slower and less marked by division peaks are less clear.



### 3.3. XRD analysis of NaX Zeolite

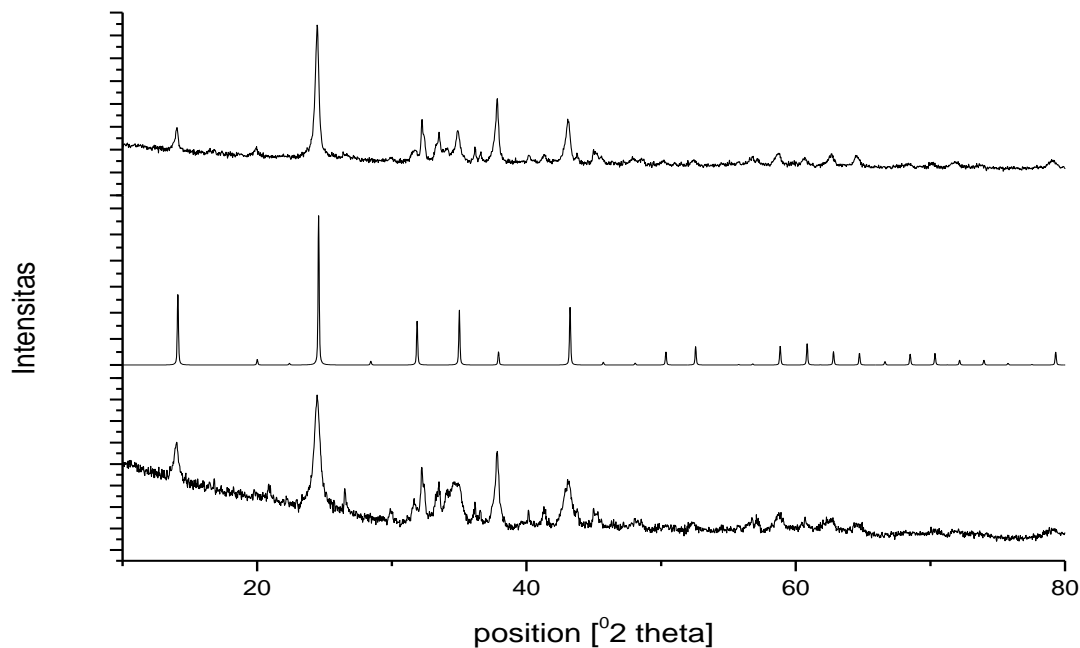


Figure 3.3. The XRD pattern synthesized zeolite NaX

[(X = peak NaX zeolite, Q = quarsa (SiO<sub>2</sub>), C = corundum (Al<sub>2</sub>O<sub>3</sub>)]

The diffraction pattern NaX zeolite synthesis results compared with the diffraction pattern database faujasite / NaX hydrate in Treacy and Higgins (J. Phys. Chem.7 (4), 2758-2764). For all zeolite synthesis using either a template or no template at all pH variation showed peaks at specific 2θ for zeolite NaX (Treacy, 2001). Can be seen from the diffraction pattern of the above, for the zeolite synthesized without using templates pH of 12 is still seen some peaks quarsa (Q) and corundum (C), but with increasing pH peak quarsa (Q) and corundum (C) becomes less and NaX zeolite peaks increased. This shows that the higher the pH, the basic compound (quarsa and corundum) more quickly transformed into zeolite crystals. The highest diffraction peaks contained in the diffraction pattern of zeolite template 14. The high pH and more regular diffraction peaks shows that in this variation amorphous compounds more quickly turned into NaX zeolite crystals.

XRD analysis for the sixth synthesized zeolites have in common with a database layout 2θ faujasite / NaX hydrate which is the reference compound for zeolit NaX. Zeolites are synthesized by the addition of template pH of 14 is a compound with diffraction pattern for the most regular, high intensity and has the highest similarity to the database with the highest

peak on  $2\theta = 24.5001$   $3258.62$  intensity can be said with this condition are the best conditions for synthesis NaX zeolite compared to other variations.

### 3.4. SEM-EDX analysis of NaX Zeolite

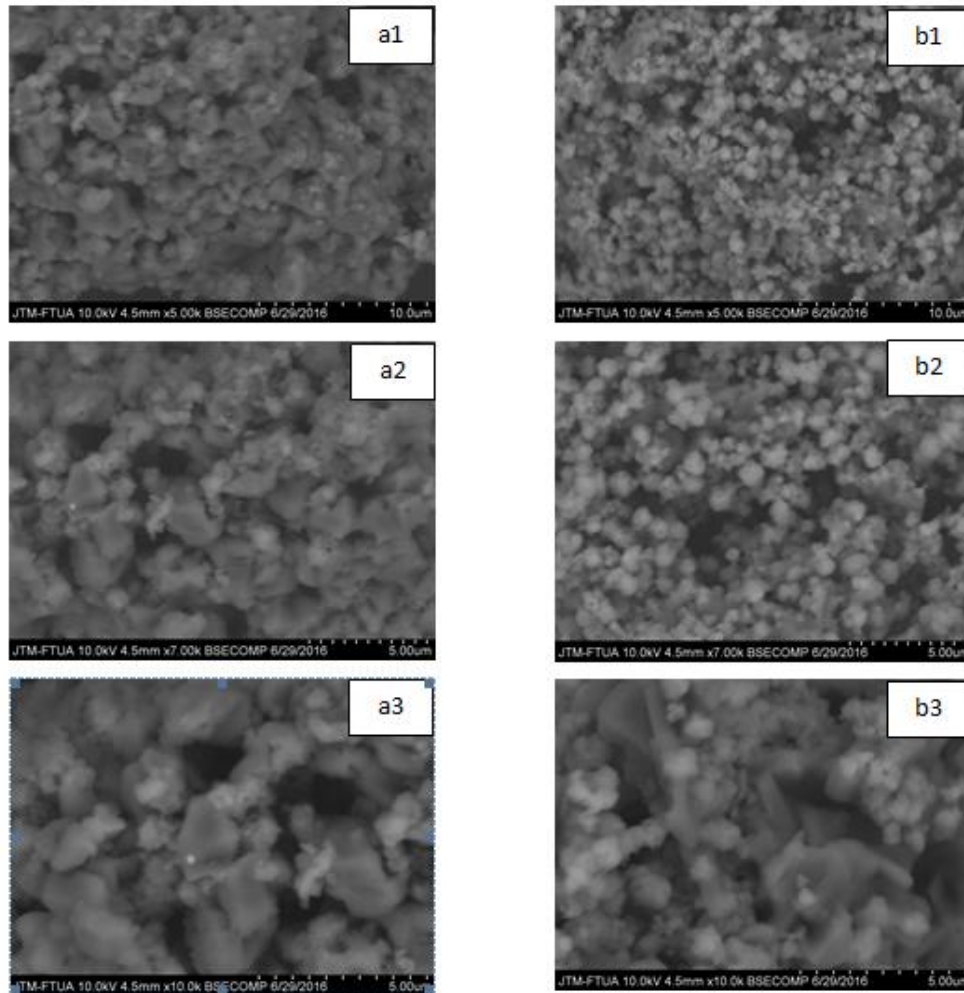


Figure 3.4. SEM photos NaX zeolite synthesis results, the series left image template for synthesis without magnification (a1). 5000x, (a2) 7000X, (a3) 10000x. right image sequence with a template for the synthesis of magnification (b1) 5000x, (b2) 7000X, (b3) 10000x

In the figure 3.5. synthesized zeolite looks good with or without a template template with variations in pH 14 has shaped the dominant morphology sperikel (ball) which are arranged to form agglomerates which have pores. Sperikel shape on these SEM images show that the types of minerals that become dominant constituent component of this compound is in accordance with the NaX zeolite NaX zeolite pure morphology (Belviso C, 2009). Seen in the picture zeolite with the addition of the template has a smaller particle size, high uniformity

and pore more and agglomeration (clumping) is less than the zeolite without a template. This is caused by the action template that form patterns in the direction of crystal formation and regular. The level of agglomeration (clumping) and a pore formed affect the zeolite surface area that would affect the ability adsorbs zeolite. Big blob provide a smaller surface area.

EDX analysis is used to determine the composition of compounds that exist in zeolite synthesis, where EDX paired with SEM to obtain a more accurate analysis of the product.

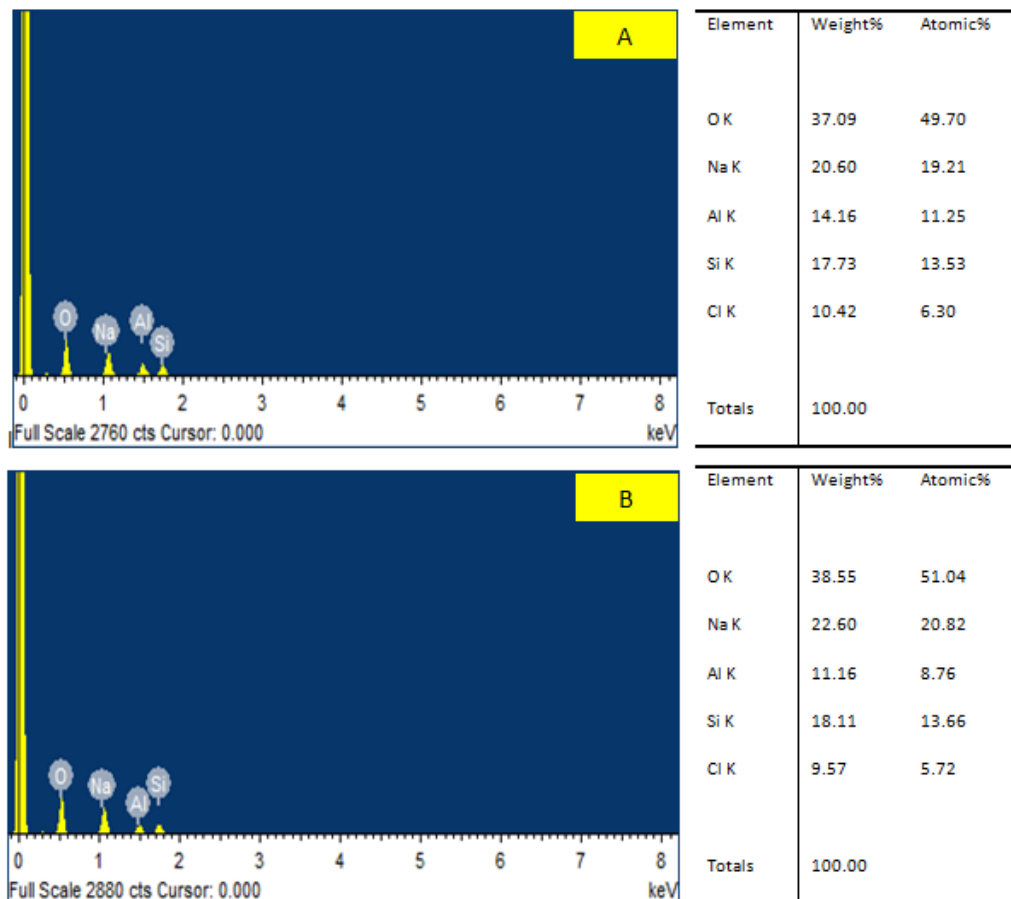


Figure 3.5. EDX spectrum for zeolite synthesis (A) without template, (B) with template

The above data can be used to calculate the ratio of Si / Al ratio in zeolites. For zeolite synthesis without a template at a pH of 14 has a ratio of Si / Al = 1.25 and for zeolite synthesis with a template at a pH of 14 has a ratio of Si / Al = 1.62. Of Si / Al ratio is further reaffirmed that the zeolite synthesis has a ratio of Si / Al < 2, which is the nature of the NaX zeolite.

### 3.5. Surface Area Analyzer of NaX zeolite obtained with and without CTA-Br templete in preparing (N<sub>2</sub> adsorption desorption )

Analisis *surface area analyzer* dilakukan dengan metode Bruenne Emmet Teller (BET) digunakan untuk mengetahui luas permukaan dan Barret Joyner Halenda (BJH) untuk menentukan distribusi ukuran pori zeolit hasil sintesis. Analisis SAA dilakukan pada suhu 77<sup>0</sup>K.

Zeolit hasil sintesis dengan *templete* memiliki luas permukaan yaitu 5,130 m<sup>2</sup>/g yang diperoleh dengan metode BET, rata-rata ukuran pori 30,21 nm dan volume total pori 0,07749 cm<sup>3</sup>/g yang diperoleh dengan motode BJH. Sedangkan untuk zeolit tanpa *templete* memiliki luas permukaan yaitu 4,889 m<sup>2</sup>/g yang diperoleh dengan metode BET, rata-rata ukuran pori 25,841 nm dan volume total pori 0,06371 cm<sup>3</sup>/g yang diperoleh dengan motode BJH. Ukuran pori yang diperoleh pada penelitian ini tergolong ke dalam zeolit mesopori karena memiliki ukuran pori dalam rentang zeolit mesopori yaitu 2 – 50 nm<sup>12</sup>. Zeolit hasil sintesis memiliki kurva isoterm tipe IV karena tekanan relatif mengalami kenaikan dari 0,25 sampai 0,45 yang menjelaskan pembentukan mesopori. Histersis *loop* yang diperoleh pada tekanan relatif 0,5 sampai 1 merupakan klasifikasi dari isoterm tipe IV<sup>36</sup>.

Tabel 4.1 Data N<sub>2</sub> Adsorpsi-Desorpsi

| Perlakuan              | Luas Permukaan          | Rata-Rata Ukuran Pori | Volume Total Pori          |
|------------------------|-------------------------|-----------------------|----------------------------|
| Dengan <i>Template</i> | 5,130 m <sup>2</sup> /g | 30,21 nm              | 0,07749 cm <sup>3</sup> /g |
| Tanpa <i>Template</i>  | 4.889 m <sup>2</sup> /g | 25.841 nm             | 0.06371 cm <sup>3</sup> /g |

### 3.6. The analysis of adsorption capacity of NaX zeolite against heavy metals (Cd and Cu)

Adsorpsi ion logam berat dilakukan dengan menggunakan ion logam Cd<sup>2+</sup> dan Cu<sup>2+</sup>. Adsorpsi dilakukan dengan variasi konsentrasi yaitu (50, 100, 150, 200 dan 250) mg/L dan juga dengan variasi waktu kontak (30, 60, 90, 120 dan 150) menit menggunakan zeolit NaX dengan dan tanpa *templete* 0,015 g pada 150 rpm. Konsentrasi larutan ion logam berat (Cd<sup>2+</sup> dan Cu<sup>2+</sup>) setelah adsorpsi oleh zeolit NaX hasil sintesis dapat dilihat pada Lampiran 11.

Hasil kapasitas adsorpsi ion logam berat oleh zeolit NaX hasil sintesis dapat dilihat pada Lampiran 12 dan grafik adsorpsi ion logam berat oleh zeolit NaX dapat dilihat pada gambar berikut:

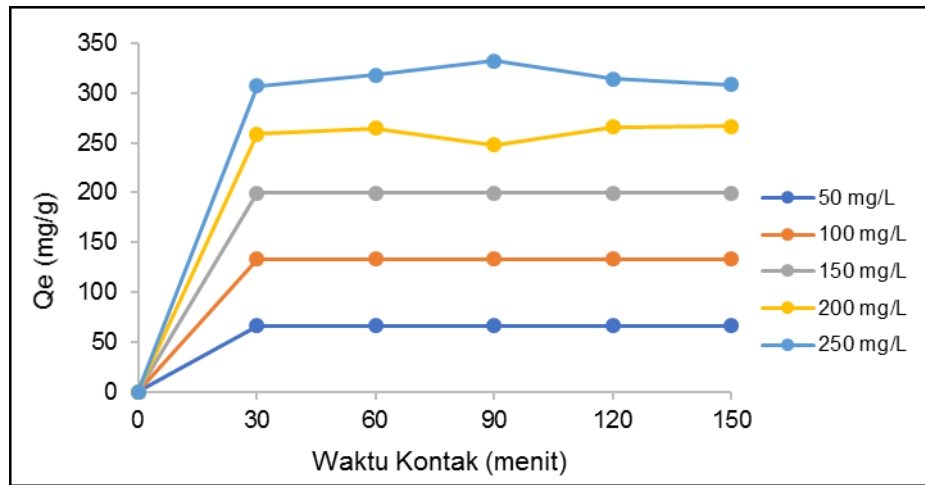


Figure 3.6. The adsorption capacity of NaX zeolite preparing with CTA-Br template against heavy metals Cu

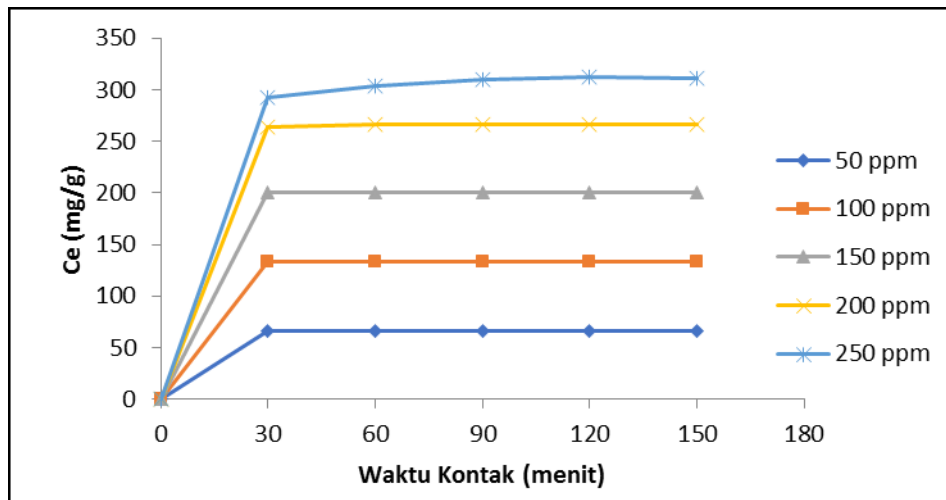


Figure 3.7. The adsorption capacity of NaX zeolite preparing without CTA-Br template against heavy metals Cu

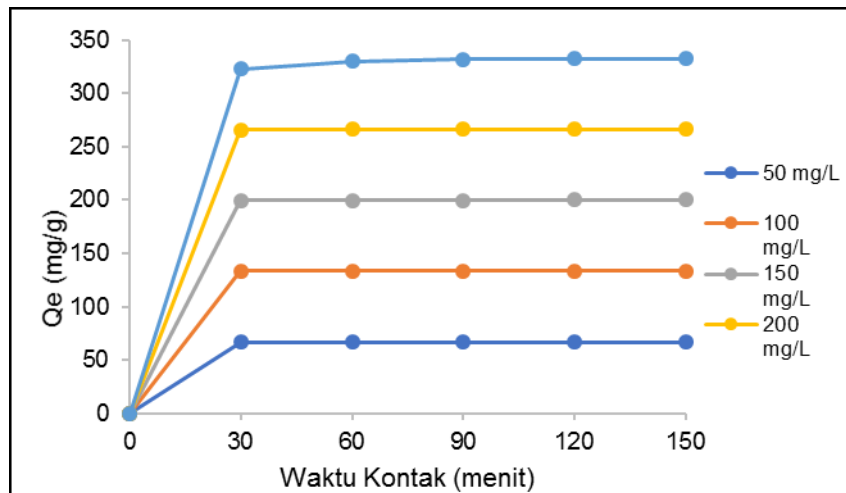


Figure 3.8. The adsorption capacity of NaX zeolite preparing with CTA-Br templete againt heavy metals Cd

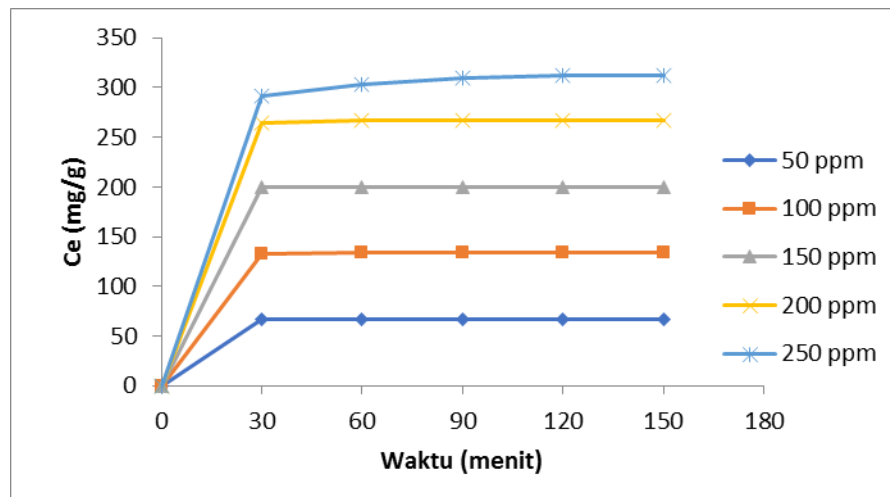


Figure 3.9. The adsorption capacity of NaX zeolite preparing without CTA-Br templete againt heavy metals Cd

Pengaruh waktu kontak terhadap kapasitas adsorpsi masing-masing ion logam berat oleh zeolit NaX hasil sintesis memiliki kecenderungan yang sama dari waktu kontak 30 menit sampai 150 menit. Hasil yang diperoleh terjadi karena zeolit NaX hasil sintesis dengan dan tanpa *template* mengalami tingkat jenuh dalam waktu yang singkat, sehingga walaupun waktu diperpanjang tidak akan mempengaruhi kapasitas adsorpsi dari masing-masing ion logam berat. Hal ini juga menunjukkan kemampuan adsorpsi dari zeolit NaX relatif cepat.

Pengaruh konsentrasi larutan ion logam berat terhadap kapasitas adsorpsi ion logam berat adalah semakin tinggi konsentrasi larutan ion logam berat pada waktu kontak yang

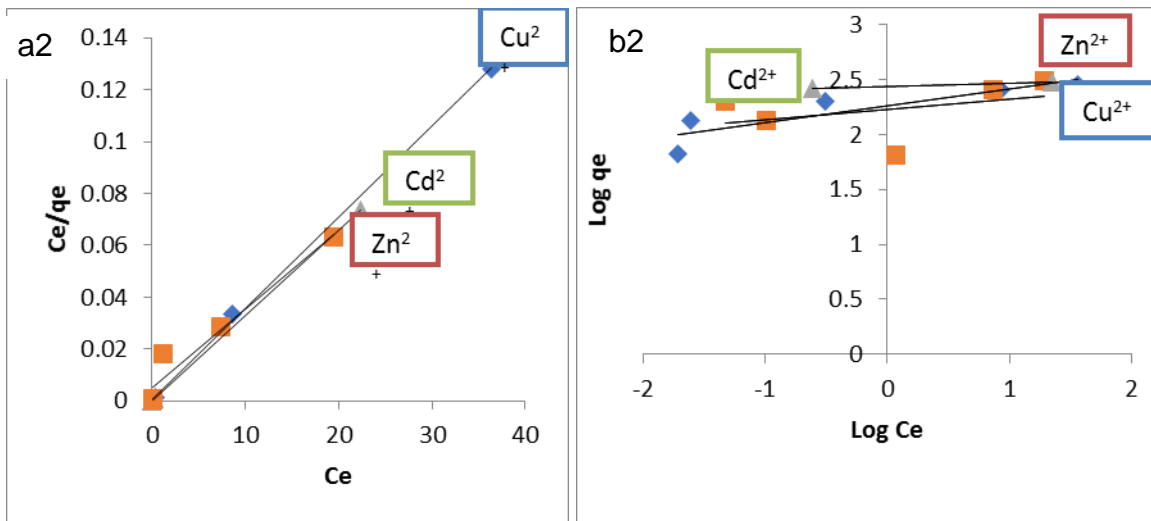
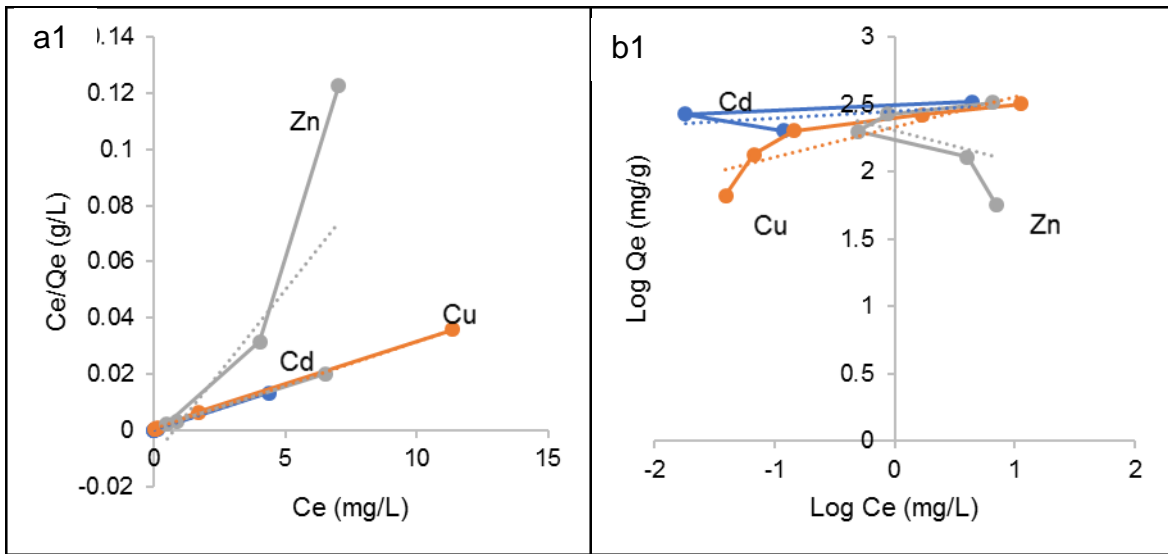
sama maka kapasitas adsorpsi juga meningkat. Hal ini sesuai dengan hasil penelitian Priyadi (2014) bahwa semakin tinggi konsentrasi larutan ion logam berat maka kekuatan transfer massa juga akan semakin besar, sehingga kemampuan adsorpsi ion logam berat oleh zeolit juga semakin besar. Ion logam berat akan berpindah dari permukaan luar ke dalam pori dari zeolit yang menyebabkan terjadinya penyerapan ion logam oleh zeolit. Semakin tinggi konsentrasi ion logam berat juga menyebabkan semakin banyaknya ion logam berat yang akan mengalami pergantian ion dengan  $\text{Na}^+$  yang ada pada zeolit ataupun berikatan dengan struktur zeolit<sup>28</sup>.

Zeolit dapat mengadsorpsi logam berat karena zeolit memiliki pori. Ion logam berat masuk ke dalam pori zeolit dan diikat oleh ion negatif yang terdapat pada zeolit. Adanya *template* pada zeolit NaX juga berperan untuk pembentuk pori dari zeolit sehingga penyerapan ion logam berat oleh zeolit menjadi lebih besar. Luas permukaan zeolit juga akan mempengaruhi penyerapan ion logam berat oleh zeolit. Semakin besar luas permukaan zeolit maka penyerapan ion logam berat juga akan semakin besar.

Muatan-muatan negatif zeolit terbentuk karena adanya struktur kerangka tiga dimensi tetrahedral silikat  $[\text{SiO}_4]^{4-}$  dan aluminat  $[\text{AlO}_4]^{5-}$  yang terikat melalui atom oksigen. Keberadaan aluminium pada kerangka zeolit menyebabkan zeolit bermuatan negatif dan muatan negatif tersebut diimbangi oleh kation  $\text{Na}^+$ . Kation  $\text{Na}^+$  dapat menyebabkan terjadinya pertukaran ion di dalam zeolit dengan ion logam berat yang diserap oleh zeolit. Ion logam berat akan masuk ke dalam zeolit sehingga konsentrasi dari larutan ion logam berat setelah adsorpsi oleh zeolit akan berkurang<sup>28</sup>.

### **3.7. Isoterm adsorption of heavy metals by NaX Zeolite**

Analisis isoterm adsorpsi ion logam berat ( $\text{Cd}^{2+}$  and  $\text{Cu}^{2+}$ ) oleh zeolit NaX hasil sintesis dilakukan untuk melihat model adsorpsi yang diikuti oleh penelitian ini. Pada penelitian ini digunakan model isoterm adsorpsi yang umum yaitu isoterm adsorpsi Langmuir dan Freundlich dan dilakukan pada waktu kontak yang telah dianggap stabil yaitu untuk ion logam  $\text{Cd}^{2+}$  and  $\text{Cu}^{2+}$  yaitu pada waktu kontak 60 menit.



Gambar 4.11 Isoterm Adsorpsi: a1. Langmuir dengan *template* b1. Freundlich dengan *template* a2. Langmuir tanpa *template* b2. Freundlich tanpa *template*

Parameter isoterm adsorpsi dapat dilihat pada tabel 4.3 Nilai koefisien korelasi ( $r$ ) menentukan isoterm adsorpsi yang sesuai dari kedua isoterm adsorpsi yang digunakan. Nilai korelasi yang lebih besar atau semakin mendekati satu yang dipilih sebagai isoterm adsorpsi yang sesuai untuk penelitian. Isoterm Langmuir dan Freundlich adalah isoterm adsorpsi yang sering digunakan untuk melihat karakteristik proses adsorpsi. Isoterm adsorpsi Langmuir berasumsi bahwa terjadi adsorpsi homogen adsorbat pada permukaan adsorben dan terjadi adsorpsi *monolayer*. Sedangkan isoterm adsorpsi Freundlich berasumsi bahwa adsorpsi *multilayer* adsorbat oleh adsorben. Isoterm adsorpsi Freundlich dapat digunakan apabila



memiliki nilai konstanta Freundlich ( $n_f$ )  $1 < n < 10$ <sup>37</sup>. Contoh perhitungan persamaan Langmuir dan Freundlich dapat dilihat pada Lampiran 13.

Tabel 4.3 Isoterm Adsorpsi Langmuir dan Freundlich dengan zeolit dengan *template*

| Ion       | Langmuir |                 |       | Freundlich |        |       |
|-----------|----------|-----------------|-------|------------|--------|-------|
|           | $K_L$    | $Q_m$<br>(mg/g) | r     | Kf         | N      | r     |
| $Cu^{2+}$ | 6,199    | 322,581         | 0,999 | 214,583    | 4,413  | 0,568 |
| $Cd^{2+}$ | 50,251   | 333.333         | 0,715 | 280,543    | 19,801 | 0,871 |

Tabel 4.4 Isoterm Adsorpsi Langmuir dan Freundlich zeolit tanpa *template*

| Ion       | Langmuir |                 |       | Freundlich |        |       |
|-----------|----------|-----------------|-------|------------|--------|-------|
|           | $K_L$    | $Q_m$<br>(mg/g) | r     | Kf         | N      | r     |
| $Cu^{2+}$ | 3.003    | 285.714         | 0.998 | 183.2314   | 6.5789 | 0.881 |
| $Cd^{2+}$ | 110.01   | 303.030         | 1     | 277.332    | 35.714 | 0.998 |

Dari tabel 4.3 diatas diperoleh nilai r dari isoterm adsorpsi Langmuir dan Freundlich. Berdasarkan hasil yang diperoleh, didapatkan nilai r dengan isoterm adsorpsi Langmuir untuk ion logam  $Cd^{2+}$  and  $Cu^{2+}$  yaitu berturut-turut 0,999 dan 0,715 dan dengan isoterm adsorpsi Freundlich yaitu berturut-turut 0,871, 0,568 dan 0,415. Berdasarkan hasil tersebut menunjukkan bahwa penelitian ini lebih mengikuti isoterm adsorpsi Langmuir. Isoterm adsorpsi Langmuir berarti bahwa terjadi adsorpsi terjadi pada permukaan yang homogen dan terjadi adsorpsi *monolayer*. nilai kapasitas adsorpsi ion logam berat ( $Cd^{2+}$ ,  $Cu^{2+}$  dan  $Zn^{2+}$ ) masing-masingnya 333,3333 mg/g, 322,5806 mg/g dan 86,2069 mg/g. berdasarkan nilai tersebut kapasitas ion logam  $Cd^{2+} > Cu^{2+} > Zn^{2+}$ . Kapasitas adsorpsi ion logam  $Cd^{2+} > Cu^{2+} > Zn^{2+}$  disebabkan oleh selektivitas ion logam  $Cd^{2+} > Cu^{2+} > Zn^{2+}$ <sup>38</sup>.

Berdasarkan tabel 4.4 diperoleh nilai koefisien kolerasi (r) untuk ion logam berat  $Cu^{2+}$  0.998, ion  $Zn^{2+}$  0.973, dan ion  $Cd^{2+}$  1 pada isotherm Langmuir sedangkan nilai koefisien kolerasi (r) untuk ion logam berat  $Cu^{2+}$  0.881, ion  $Zn^{2+}$  0.382, dan ion  $Cd^{2+}$  0.998 pada isotherm Freundlich. Hal ini menunjukkan bahwa adsorpsi logam berat  $Cu^{2+}$ ,  $Zn^{2+}$ , dan  $Cd^{2+}$

oleh zeolit NaX hasil sintesis cenderung mengikuti model isotherm Langmuir karena memiliki nilai koefisien kolerasi ( $r$ ) mendekati nilai 1. Isotherm Langmuir menggambarkan bahwa adsorpsi cenderung pada permukaan monolayer dan mengasumsikan bahwa penyerapan terjadi pada adsorben yang homogen<sup>14</sup>. Dari isotherm Langmuir diperoleh kapasitas adsorpsi maksimum untuk ion logam  $\text{Cu}^{2+}$ ,  $\text{Zn}^{2+}$ , dan  $\text{Cd}^{2+}$  masing-masingnya 285,174 mg/g, 333,333 mg/g dan 303,030 mg/g.

Zeolit dengan template memiliki kapasitas adsorpsi lebih tinggi dibandingkan tanpa template. Hal ini disebabkan template mempengaruhi pori dari zeolit yang dihasilkan. Template akan meningkatkan kemampuan penyerapan oleh zeolit NaX.

#### 4. Conclusions

NaX Zeolites were prepared from fly ashes by hydrothermal method with and without CTA-Br templete at pH 14, temperature 60°C and sea water as a solvent. The NaX Zeolite obtained was characterized by X-ray diffraction (XRD), Fourier transformed infrared (FT-IR) spectroscopy, Scanning electron microscopy (SEM), and has charateristics of NaX Zeolite.  $\text{N}_2$  adsorption desorption results showed that NaX zeolites prepared with and without CTA-Br templete have a specific surface area 5.130  $\text{m}^2\text{g}^{-1}$  and 4.889  $\text{m}^2\text{g}^{-1}$ , average pore size 30.21 nm and 25.84 nm, and total pore volume 0.078  $\text{cc g}^{-1}$  and 0.064  $\text{cc g}^{-1}$  repectively. The application of NaX zeolite as an adsorbent to heavy metal ions  $\text{Cd}^{+2}$  and  $\text{Cu}^{+2}$ . The maximum adsorption capacities of NaX zeolites that were prepared with and without CTA-Br templete against heavy metal ions were 333.3  $\text{mg g}^{-1}$  and 303.0  $\text{mg g}^{-1}$  for  $\text{Cd}^{+2}$  ion and 322.6  $\text{mg g}^{-1}$  and 285.7  $\text{mg g}^{-1}$  for  $\text{Cu}^{+2}$  ion respectively. These results indicated that synthetic NaX zeolites have potential as adsorbent of heavy metals ions  $\text{Cd}^{+2}$  and  $\text{Cu}^{+2}$ . The results also suggested that the adsorption capacity was affected by specific surface area, average pore size and total pore volume.

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