

# Utilization of Natural Zeolites as Cu (Ii) and Zn (Ii) Adsorbent

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## ABSTRACT

This research was aimed to determine the differences of physical and chemical characteristics from three natural zeolites from Tasikmalaya, Bayah and Lampung areas and to examine the characteristics of the natural zeolite adsorption capability to the micro-nutrients. The zeolite characterization analysis included some variables *i.e.* mineral types, morphology and pH, CEC (Cation Exchange Capacity), BSP (Base Saturation Point), Cu and Zn content of zeolite. Analysis of natural zeolite adsorption characteristics were done by Batch System. Data interpretation were done by using the isothermal adsorption equation of Langmuir, Freundlich and Brunauer-Emmett-Teller (BET). The results showed that zeolite from Bayah and Tasikmalaya had a similar morphology and it was categorized as mordenite types and platy structure, while zeolite from Lampung was a clinoptilolite type and tabular structure. The CEC of Tasikmalaya zeolite ( $137.58 \text{ cmol}(-)\text{kg}^{-1}$ ) was higher than Bayah ( $96.75 \text{ cmol}(-)\text{kg}^{-1}$ ) and Lampung ( $87.72 \text{ cmol}(-)\text{kg}^{-1}$ ). Adsorption capability at the Langmuir equation of Cu by Bayah zeolite ( $47.619 \text{ mg g}^{-1}$ ) was higher than Lampung ( $33.333 \text{ mg g}^{-1}$ ) and Tasikmalaya ( $28.571 \text{ mg g}^{-1}$ ). Effective adsorption of Zn by Bayah zeolite  $19.417 \text{ mg g}^{-1}$  was higher than Lampung ( $16.949 \text{ mg g}^{-1}$ ) and Tasikmalaya ( $1.395 \text{ mg g}^{-1}$ ).

**Keyword:** Adsorption, micro nutrient, zeolite

## ABSTRAK

Penelitian ini bertujuan untuk menentukan perbedaan sifat fisika dan kimia tiga zeolite alami yang berasal dari daerah Tasikmalaya, Bayah dan Lampung, dan untuk memperelajari daya jerap zeolite alami terhadap unsur hara mikro. Analisis terhadap sifat-sifat zeolit alami meliputi beberapa variabel, yaitu tipe mineral, morfologi, pH, KTK (Kapasitas Tukar Kation), kejenuhan basa, dan kandungan Cu dan Zn di dalam zeolit. Analisis terhadap daya jerap zeolite alami dilakukan dengan *Batch System*. Interpretasi data dilakukan dengan menggunakan persamaan adsorpsi isotherm Langmuir, Freundlich dan Brunauer-Emmett-Teller (BET). Hasil penelitian menunjukkan bahwa zeolit yang berasal dari Bayah dan Tasikmalaya memiliki kesamaan morfologi dan dikategorikan sebagai tipe *mordenite* dan berstruktur lempeng, sedangkan zeolite yang berasal dari Lampung merupakan tipe *clinoptilolite* dan berstruktur tiang (*columnar*). Kapasitas Tukar Kation (KTK) zeolit yang berasal dari Tasikmalaya ( $137,58 \text{ cmol}(+) \text{ kg}^{-1}$ ) lebih tinggi dari KTK zeolite yang berasal dari Bayah ( $96,75 \text{ cmol}(+) \text{ kg}^{-1}$ ) dan Lampung ( $87,72 \text{ cmol}(+) \text{ kg}^{-1}$ ). Berdasarkan persamaan adsorpsi isotherm Langmuir, daya jerap Cu dari zeolit yang berasal dari Bayah ( $47,619 \text{ mg g}^{-1}$ ) lebih tinggi dibandingkan dengan zeolit yang berasal dari Lampung ( $33,333 \text{ mg g}^{-1}$ ) dan Tasikmalaya ( $28,571 \text{ mg g}^{-1}$ ). Daya jerap Zn dari zeolite yang berasal dari Bayah ( $19,417 \text{ mg g}^{-1}$ ) lebih tinggi dibandingkan dengan zeolit yang berasal dari Lampung ( $16,949 \text{ mg g}^{-1}$ ) dan Tasikmalaya ( $1,395 \text{ mg g}^{-1}$ ).

**Kata kunci:** Penjerapan, unsur hara mikro, zeolit

## INTRODUCTION

One of the new developments to overcome the problem of fertilizer efficiency is making a slow release fertilizer using the biosorb method. The natural zeolite is a widely utilized mineral as an adsorbent. Zeolite is an aluminosilicate mineral with a cage structure ("Framework") in the three

dimensions. Zeolite has a very organized crystal with interconnected cavities to the all directions which causes very large surface area of zeolite (Sutarti and Rachmawati 1994) so that effectively used as adsorbent. This mineral is abundant in nature, cheap and has been widely studied as a metal adsorbent. According to Kundari *et al.* (2010), the natural zeolite effectively adsorbs Fe and Mn in the liquid waste. Zeolite can adsorb ammonium ions from urea as it has high CEC value which causes the number of lattice exchange and cavities in large quantities. So that the greater number of ammonium ions from

nitrogen fertilizer have been hydrolyzed and sequestered by zeolite. The entrapment of ammonium ions in the lattice exchange are temporary and can be easily released to the plant at the time required (Suwardi 2002). Therefore, zeolite basically is used as a carrier of metal cations so that the metal can be adsorbed and released slowly. In Indonesia, there are many types of natural zeolite, the zeolite deposit in Indonesia almost reach 250 million tons (Suwardi 2009). Generally, the type of zeolite minerals are clinoptilolite and mordenite. The minerals diversity are causing the differences of zeolites characteristics and adsorption capability. Zeolite has a potential to adsorb several elements. Erdem *et al.* (2004) described natural zeolite can be used effectively as an adsorbent for removing metal cations ( $\text{Co}^{2+}$ ,  $\text{Cu}^{2+}$ ,  $\text{Zn}^{2+}$ ,  $\text{Mn}^{2+}$ ) from the wastewater. Chiban *et al.* (2012) studied the adsorption and the removal of heavy metal from drinking water by natural zeolite. Wang and Peng (2010) reviewed utilization of natural zeolites as metal adsorbent in water and wastewater treatment. On the other hand, micro nutrients like Cu and Zn are essential nutrients needed by plants for growth and development. These elements are also deficient in plantation crops. Therefore, this research used natural zeolites as a carrier for slow release of micro nutrients fertilizer (Cu and Zn). This study was aimed to 1) characterize and differentiate the physical and chemical properties of the three natural zeolites, and 2) determine their effects on the capacity of the natural zeolites to adsorb micronutrients.

## MATERIALS AND METHODS

The experimental materials used in this study were natural zeolites taken from three source locations which were Lampung (ZA), Bayah (ZB), and Tasikmalaya (ZC), as well as microelements (Cu and Zn).

### Experimental Procedures

#### Characterization of Natural Zeolites

Physical characterization of three natural zeolites included some variables *i.e.* structure and morphology by *scanning electron microscope* (SEM), crystal structure by *X-Ray diffractometer* (XRD). The analysis of zeolites chemical characteristics involved the determination of cation exchange capacity (CEC), pH, Cu and Zn content of zeolite. Analysis of Cu content was measured from salt of  $\text{CuSO}_4 \cdot 5\text{H}_2\text{O}$ , while that of Zn content was from salt of  $\text{ZnSO}_4 \cdot 7\text{H}_2\text{O}$ .

#### Adsorption of Microelements (Cu and Zn) using Natural Zeolites

Adsorption experiments were carried out with Batch System using 0.02 g of zeolite from Lampung, Bayah and Tasikmalaya in 20 ml of Cu and Zn solutions. Variation in the concentrations of Cu and Zn tested were 20 ppm, 40 ppm, 60 ppm, 80 ppm, and 100 ppm each with a shaking time of 45 min. After shaking, the resulting extract was then sieved into a glass container with the use of Whatmann filter paper No. 42.

The measurement of Cu and Zn solution concentration before and after contact with zeolite was done with the use of the *atomic absorption spectrophotometer* (AAS). In order to obtain the sorption capacity, the amount of ions adsorbed per unit mass of adsorbent (in milligrams of ions per gram of adsorbent) was evaluated using the following expression.

$$\text{Adsorption Capacity (mg g}^{-1}\text{)} = \frac{[\text{C}_i] - [\text{C}_f]}{W} \times V$$

where:

$\text{C}_i$  = initial ion concentration (ppm),

$\text{C}_f$  = ion concentration after adsorption (ppm),

$V$  = volume of reacting solution (L),

$W$  = total zeolite (g)

The resulting adsorption values were then analyzed using the most commonly used isotherm theories which have been adopted for interpreted adsorption data namely, the Freundlich and Langmuir equilibrium isotherm theories, *Brunauer-Emmett-Teller* (BET) equation.

$$\begin{aligned} \text{Langmuir} & : q = \frac{q_{\text{sat}} \cdot K \cdot C_e}{1 + K \cdot C_e} \\ \text{Linear form} & : \frac{1}{q} = \frac{1}{q_{\text{sat}}} + \frac{1}{q_{\text{sat}} \cdot K \cdot C_e} \end{aligned}$$

where:

$q$  is total adsorbate ( $\text{mg g}^{-1}$ )

$q_{\text{sat}}$  is total adsorbate and the theoretical maximum, respectively ( $\text{mg g}^{-1}$ )

$K$  is a constant, and

$C_e$  is concentration (ppm)

$$\text{Freundlich} : q = K C_e^{\frac{1}{n}}$$

$$\text{Linear form} : \log(q) = \log(K) + \frac{1}{n} \log(C_e)$$

Where:

$C_e$  and  $q$  are similar definition with Langmuir while  $k$  and  $n$  are adsorption constant.

Generally, the *Brunauer–Emmett–Teller* (BET'equation) is written as follows:

$$\text{BET} : \frac{q}{q_{\text{mono}}} = \frac{K}{\left(1 - \frac{C_e}{C_s}\right) \left[1 + \frac{C_e}{C_s} (K - 1)\right]} \frac{C_e}{C_s}$$

$$\text{Linear form} : \frac{1}{a \left[\left(\frac{C_s}{C_e}\right) - 1\right]} = \frac{K - 1}{q_{\text{mono}} K} \left(\frac{C_e}{C_s}\right) + \frac{1}{q_{\text{mono}} K}$$

Where  $q$  is the amount of adsorption capacity ( $\text{mg g}^{-1}$ ),  $C_s$  is concentration of the solution used and added to the adsorbent (ppm),  $C_e$  is concentration of the solution when equilibrium (ppm).  $K$  is constant and  $q_{\text{mono}}$  is total adsorbent that adsorbed on monolayer ( $\text{mg g}^{-1}$ ).

## RESULTS AND DISCUSSION

### Physical and Chemical Properties of Zeolite

The physical and chemical characteristics of the natural zeolite from Lampung (ZA), Bayah (ZB) and Tasikmalaya (ZC) sources were analyzed in order to describe their respective initial condition (before treatment), as well as to differentiate the three zeolites used in this study. The *Scanning*

*Electron Microscope* (SEM) analysis showed the structure and morphology of the zeolite crystals.

Figure 1 illustrates the structural variation among the three natural zeolite crystals. All three natural zeolites exhibited sheet-like structure, but the Lampung zeolite was in a tabular form, while the Bayah and Tasikmalaya zeolites were in platy form. Results of the SEM analysis of the zeolite crystal structures indicated that there might be a need to refine the zeolite crystals in order to obtain better and homogenous forms.

The above findings were supported by the results of the *X-Ray diffractometer*(XRD) on the other physical characteristic, that is crystal intensity of the zeolites as depicted by Figures 2. On the other hand, Figure 3 shows the different patterns of diffractogram peaks of the three natural zeolite crystals, particularly at diffraction angle  $2\theta$ . As shown in Figure 3, the Lampung (ZA) zeolite is the clinoptilolite mineral type, while both Bayah (ZB) and Tasikmalaya (ZC) zeolites are mordenite type. It can be seen that in the Lampung (ZA) zeolite, there were three peaks, namely: at  $2\theta = 9.794^\circ$ ,  $22.727^\circ$ , and  $30.031^\circ$  with the highest score of 60 that is clinoptilolite-Ca (Figure 2). In comparison, the Bayah (ZB) zeolite possessed three peaks at  $2\theta = 22.237^\circ$ ,  $25.560^\circ$ , and  $28.039^\circ$  with the highest score of 37, that is mordenite (Figure 2). On the other

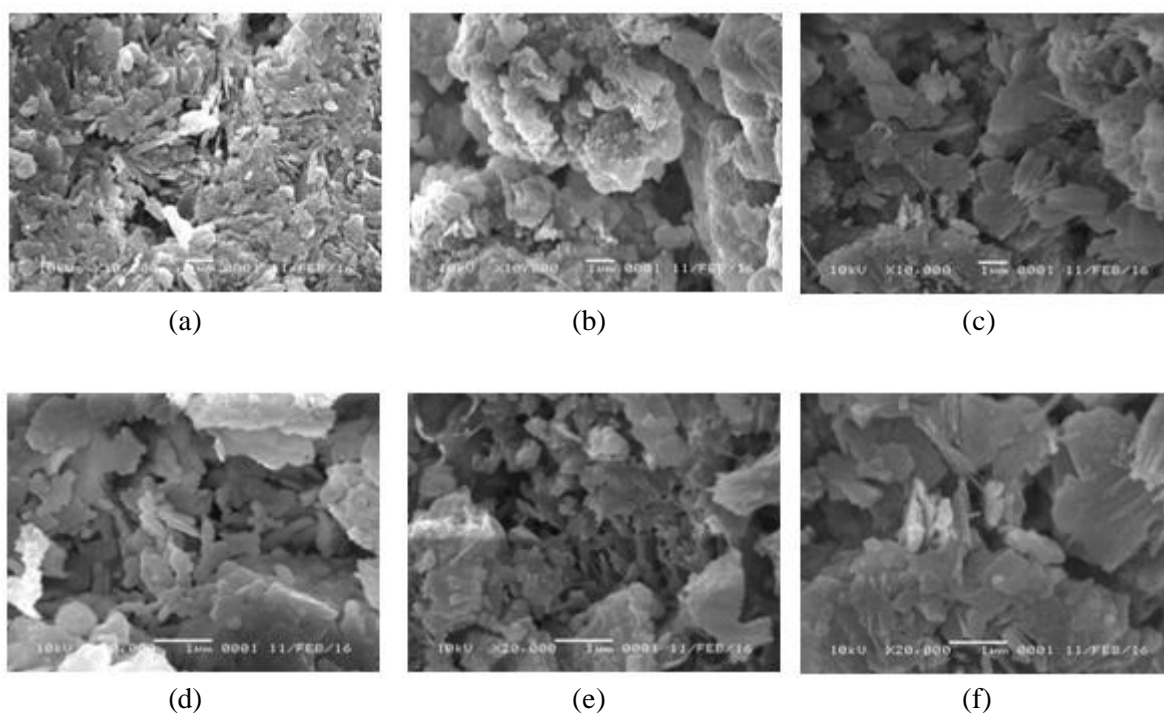
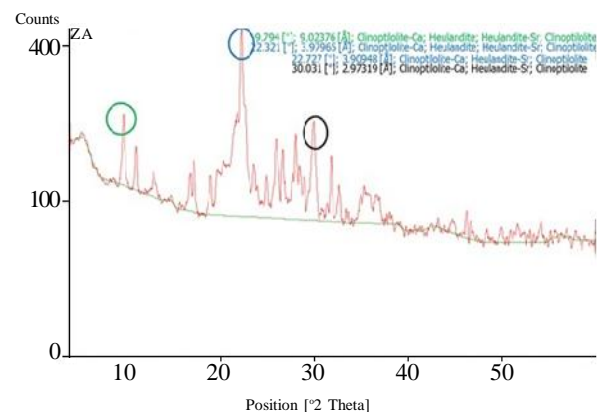
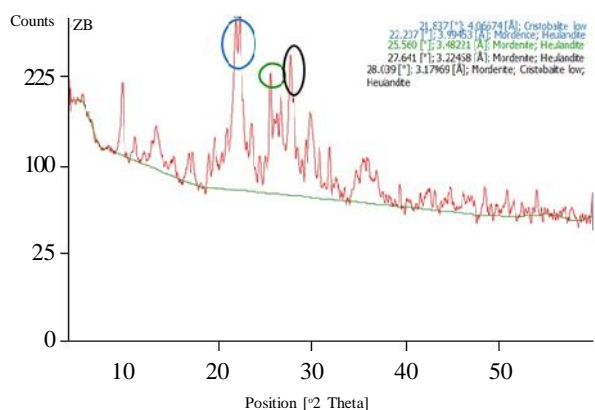


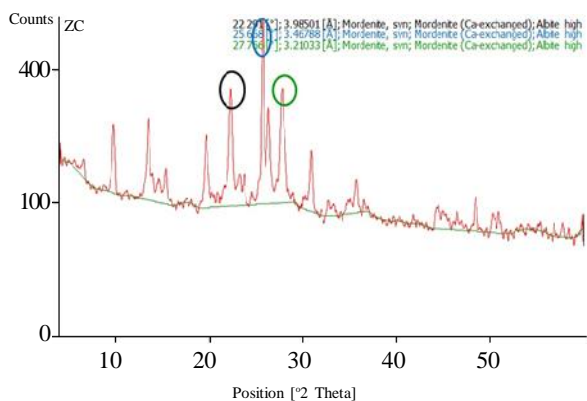
Figure 1. Images of formed crystals (SEM). Magnification:10000x for Lampung zeolite (a), Bayah zeolite (b), and Tasikmalaya zeolite (c). Magnification: 20000x for Lampung zeolite (d), Bayah zeolite (e), and Tasikmalaya zeolite (f).



A. Lampung Zeolit



B. Bayah Zeolit



C. Tasikmalaya Zeolit

Figure 2. Graphic diffraction of Lampung Zeolite (A), Bayah Zeolit (B) and Tasikmalaya Zeolit (C).

hand, the Tasikmalaya (ZC) zeolite displayed three peaks, namely: at  $2\theta = 22.291^\circ$ ,  $25.668^\circ$ , and  $27.756^\circ$  with the highest score of 58 that is mordenite (Figure 2). This results are similar with previous research. Razzak *et al.* (2013) revealed that Lampung zeolite have 3 strongest peaks at  $2\theta = 27.9583^\circ$ ,  $22.3963^\circ$ , and  $9.8631^\circ$ . According to ICDD 47-1870 data library, the existence of the strongest peaks may indicate that the mineral was appropriate with

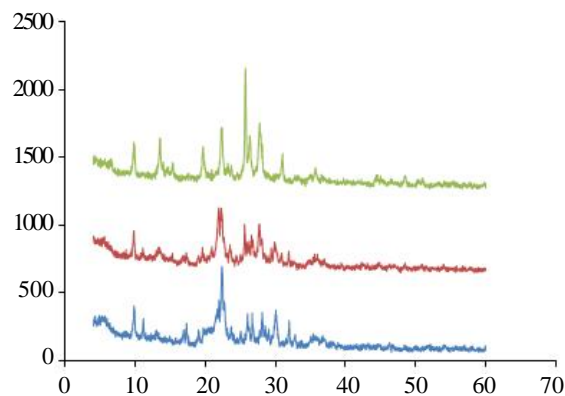


Figure 3. Graphic diffraction of Lampung, Bayah, and Tasikmalaya zeolites at diffraction angle  $2\theta$ . — : lobes [cts] ZA, — : lobes [cts] ZB, — : lobes [cts] ZC

potassium sodium calcium aluminum silicate hydrate of clinoptilolite-Na. While Bayah Zeolite have three strongest peaks available *i.e.* peak at  $2\theta$  of  $25.6897^\circ$ ,  $27.3619^\circ$  and  $26.6704^\circ$ , it can be assumed that Bayah zeolite belong to be a mineral mordenite group. Heraldry *et al.* (2003) said zeolite mineral type is determined by major cation exchangeability.

The results of the XRD analysis were consistent with those of the SEM tests, *i.e.* the crystal structure and mineral type of the Lampung (A) zeolite were different from those of the Bayah (B) and Tasikmalaya (C) zeolite crystals. These observations were also consistent with existing literature which stated that basically zeolite possesses three structural variations, namely: a) chain-like structure, with acicular and prismatic crystal forms, for example: natrolite, b) sheet-like structure, with platy form, or tabular form commonly with pronounced basal cleavage, for example: heulandites, and c) framework structure in which the crystals possess closely similar dimensions, for example: chabazite (Sapoetra 2006).

The results of the zeolite chemical analysis are summarized in Table 1, consisting of cation exchange capacity (CEC), bases, pH, and Cu and Zn content for each natural zeolite. As shown, the three zeolites exhibited varying CEC values. The Tasikmalaya (ZC) zeolite had the highest CEC ( $137.580 \text{ cmol}(-)\text{kg}^{-1}$ ), followed by the Bayah (ZB) zeolite ( $96.750 \text{ cmol}(-)\text{kg}^{-1}$ ), while the Lampung (ZA) zeolite produced the lowest CEC value ( $87.720 \text{ cmol}(-)\text{kg}^{-1}$ ). The higher was the resulting CEC value, the higher was the level of isomorphic substitution that occurred in the Tasikmalaya zeolite, and the greater was its negative charge. This resulted from the process of isomorphic substitution of the Si atoms by Al

Table 1. Chemical characterization of zeolite.

Natural Zeolite	CEC (cmol(-) kg <sup>-1</sup> )	Bases (cmol(+)kg <sup>-1</sup> )				BSP (%)	pH H <sub>2</sub> O	Cu content (ppm)	Zn content (ppm)
		K	Na	Ca	Mg				
ZA	87.720	6.670	2.450	54.390	10.120	83.960	6.730	0.0007	0.0138
ZB	96.750	3.210	3.690	94.890	10.560	116.130	8.920	0.0003	0.0044
ZC	137.580	3.310	5.150	106.030	10.510	90.870	7.280	0.0003	0.0021

Note: ZA: Lampung Zeolit; ZB: Bayah Zeolit; ZC: Tasikmalaya Zeolit

atoms in the zeolite structure, in which Al brought about higher negative charge. When Al<sup>3+</sup> substitute for Si<sup>4+</sup> in the central cation position of the tetrahedron, a net negative charge is generated. This negative charge is counterbalanced primarily by alkali and alkaline Earth cations like Na<sup>+</sup>, K<sup>+</sup>, Ca<sup>2+</sup>, and Mg<sup>2+</sup> (generally called “exchange cation”). The exchange cations are loosely held near the seat of charge within the zeolite structure and can be replaced by other cations in solution (Ming and Mumpton 1989).

Aside from this, both Tasikmalaya (ZC) and Bayah (ZB) zeolites showed a tendency of higher Ca cation exchange compared to the other cations (K, Na and Mg). This was because Tasikmalaya and Bayah zeolites are of mordenite mineral type, which is among those that typically possess a higher percentage of Ca and, consequently, more Ca cations (in the case of Tasikmalaya and Bayah zeolites) are exchanged in the contact solution.

Specifically for Tasikmalaya zeolite, its mineral mordenite tended to exhibit higher Ca cation exchange capacity (CEC), as shown in the result of the XRD analysis. The Bayah zeolite yielded a higher Base Saturation Point (116.13%) compared to that of Tasikmalaya (90.87%) and Lampung zeolite (83.96%). Base saturation point increased due to the increasing of exchangeable cations such as Ca-dd, Mg-dd, K-dd, Na-dd. BSP of Bayah and Tasikmalaya zeolite were high that can be caused by these zeolite were mordenit type. Mordenit type zeolite has high Ca-dd content (contributor zeolite of Ca-dd). Other factors affected the base saturation point of zeolite was level of natural zeolite purity. Zeolites have a low level of purity when there are impurities like other minerals. Other minerals *i.e.* heulandite, albite and cristobalite. The zeolitic tuffs of the WC area contain zeolite minerals in the range 50 < mass % < 20 with sepiolite, albite, quartz, muscovite and mordenite as accessory minerals (Diale 2011). According Ginting *et al.* (2007) generally the formation of natural zeolites is in alkaline environment, therefore it is suspected that an alkaline mineral can be

increase base saturation point of zeolite. Association with these minerals are influenced by environmental forming.

These *Base Saturation Points* (BSP) are linearly related to their respective pH values. In general, the higher BSP, the higher the pH would also be. According to the statement Soepardi (1983), the percentage of base saturation and pH is a definite correlation. It caused base saturation point reflected the comparison alkaline cations with acid cations (H, Al and Fe). When concentration of alkaline cations are higher, therefore the acid cation concentration are lower (high pH). The higher BSP and pH values indicated lower H<sup>+</sup> ion concentration. When pH was high, the competition between metals and H<sup>+</sup> ion decreased so that the opportunity of metals to be adsorbed by zeolite increased. According to Wang *et al.* (2008) at this high pH, H<sup>+</sup> ion competition as competitors would be declined because the metal ion solution was alkaline. With the decreased of H<sup>+</sup> ion so that metal ion could be adsorbed optimally. The other factor that influences zeolite BSP value is its degree of purity. The degree of purity of a natural zeolite is lower when there is a contaminant, namely other minerals such as heulandite, albite, and cristobalite.

The other important parameters in determining zeolite adsorption capacity are the levels of Cu and Zn elements contained in the micronutrients used (Table 2). These data are useful in establishing the initial condition of the materials used in the study. The Cu content in CuSO<sub>4</sub>.5H<sub>2</sub>O and Zn content in ZnSO<sub>4</sub>.7H<sub>2</sub>O solutions using HCl as extractive yielded 25% higher as compared to using *aquadest* as extractive. This result meant that HCl produced 25% more solution than *aquadest*.

Table 2. Micronutrient Content

Extractant	Cu Content (ppm)		Zn Content (ppm)	
	CuSO <sub>4</sub> .5H <sub>2</sub> O		ZnSO <sub>4</sub> .7H <sub>2</sub> O	
Aquades	0.00609		0.02416	
HCl 25%	0.00927		0.24788	

Table 3. Values of parametric adsorption of Cu (II) based on Langmuir, Freundlich and BET isothermal equations.

Natural Zeolite	Langmuir			Freundlich			BET		
	R <sup>2</sup>	q <sub>e</sub> (mgg <sup>-1</sup> )	K	R <sup>2</sup>	K	1/n	R <sup>2</sup>	q <sub>mono</sub> (mgg <sup>-1</sup> )	K
ZA	0.990	33.333	-0.435	0.069	33.113	0.081	0.975	13.699	-18.250
ZB	0.998	47.619	7.000	0.094	32.810	0.111	0.963	25.707	-389.000
ZC	0.998	28.571	-0.289	0.045	32.359	0.061	0.952	10.000	-11.111

Note: ZA: Lampung Zeolit; ZB: Bayah Zeolit; ZC: Tasikmalaya Zeolit

Table 4. Values of parametric adsorption of Zn (II) based on Langmuir, Freundlich and BET isothermal equations.

Natural Zeolite	Langmuir			Freundlich			BET		
	R <sup>2</sup>	q <sub>e</sub> (mg g <sup>-1</sup> )	K	R <sup>2</sup>	K	1/n	R <sup>2</sup>	q <sub>mono</sub> (mg g <sup>-1</sup> )	K
ZA	0.841	16.949	-0.172	0.029	24.831	0.06	0.949	25.64	-55.71
ZB	0.854	19.417	-0.103	0.015	32.211	-0.06	0.954	34.60	32.11
ZC	0.687	1.395	-0.102	0.099	27.164	-0.22	0.998	31.65	158.00

Note: ZA: Lampung Zeolit; ZB: Bayah Zeolit; ZC: Tasikmalaya Zeolit

### Micronutrient Adsorption Capacity of Zeolite

The adsorption values for Cu (II) and Zn (II) at varying concentrations were analyzed further with the use of Langmuir, Freundlich, and BET (*Brunauer-Emmett-Teller*) isothermal adsorption equations (Tables 3 and 4). As shown, Langmuir and BET models gave different adsorption results. Langmuir's formula yielded a more effective adsorption estimate, which can be used as optimum adsorption capacity for a given adsorbent, whereas the BET model produced an adsorption value at the first layer. Tables 3 and 4 show that in almost all isothermal models used, the Bayah (ZB) zeolite demonstrated the highest Cu and Zn adsorption capacity compared to Lampung (ZA) and Tasikmalaya (ZC) zeolites. It can be seen that the effective Cu adsorption capacity of the Bayah zeolite was 47.619 mgg<sup>-1</sup>, with a coefficient of determination (R<sup>2</sup>) of 0.998, while the its adsorption capacity at the first layer was 25.707 mgg<sup>-1</sup> with an R<sup>2</sup> value of 0.963 (Figure 4). Bayah zeolite also possessed an effective Zn adsorption capacity at 19.417 mg g<sup>-1</sup> with a coefficient of determination (R<sup>2</sup>) of 0.854, while its adsorption capacity at the first layer was 34.600 mgg<sup>-1</sup> with R<sup>2</sup>= 0.954 (Figure 5). This indicates that the adsorption capacity of the Bayah zeolite at the crystal surface is far greater. The Bayah zeolite produced a negative charge particularly at the surface of the mineral crystal.

Lampung zeolite has clinoptilolite mineral, that has a little more Al atom so that the negative charge generated for ion exchange was less. On the other hand, Bayah and Tasikmalaya zeolites are mordenite type of zeolite, zeolite of this type usually has the lower Si/Al ratio so that the adsorption power is better. This is related with the research of Zulfa (2011) which stated that the Lampung zeolite has 78% of clinoptilolite that has the lower Si/Al ratio (3.78) than mordenite zeolite (2.86). The amount of Al in the clinoptilolite zeolite is less so that the negative charge which neutralized is less. In addition, mordenite zeolite has good adsorption power because it has a surface area and relatively large pore size when compared to other natural zeolites. Here are the cavity contents of mordenite zeolite of 0.5 m<sup>3</sup>g<sup>-1</sup> and clinoptilolite of 0.4 m<sup>3</sup>g<sup>-1</sup>, while the surface area of the mordenite and clinoptilolite zeolite were consecutively 183.78 m<sup>2</sup>g<sup>-1</sup> and 170.81 m<sup>2</sup>g<sup>-1</sup>.

From the data and the correlation coefficient (R<sup>2</sup>) are known that the adsorption of Cu and Zn using Bayah zeolite follow the model of Langmuir and BET equation compared to Freundlich. This is consistent with the research which stated that the adsorption of molecules or ions on the solid surface were generally limited to a single molecular layer (monolayer). Thus the adsorption usually follows the Langmuir adsorption equation (Emelda *et al.* 2013) and the research of Chunfeng (2009) about the synthesis zeolite adsorption of fly ash to the Zn

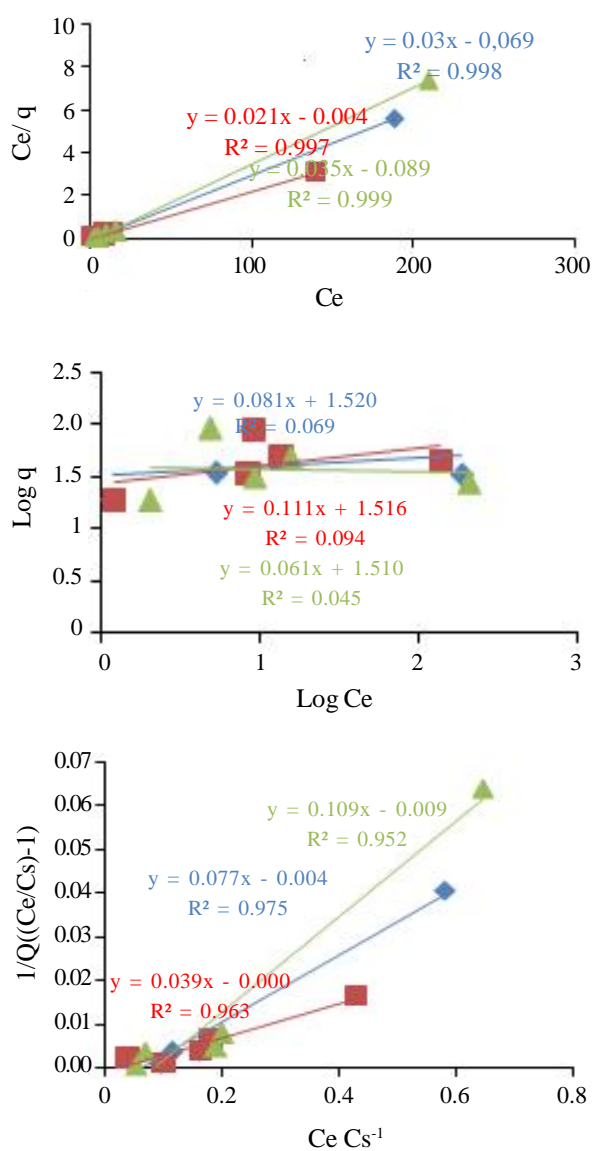


Figure 4. Langmuir, freundlich BET linearized equation of Cu (II). ♦ : ZA, ■ : ZB, ▲ : ZC.

metal. In this study, the result adsorption of Cu according to Langmuir isotherm model is larger than the BET model, while the Zn adsorption is opposite. This may be related to the formed bond. Langmuir isotherm adsorption occurs chemically because of their strong chemical bond between the adsorbate and adsorbent surface. While the BET isotherm adsorption usually occurs because of the physical adsorption of which adsorbate molecules can form more than one layer of adsorbate on the surface. At Cu adsorption may almost of the adsorption process occurs, there is no interaction between molecules absorbed or it is only formed a single layer when maximum adsorption occurred. While the Zn adsorption process occurred largely in the pores or formed many layers as a result of the occurred inter-

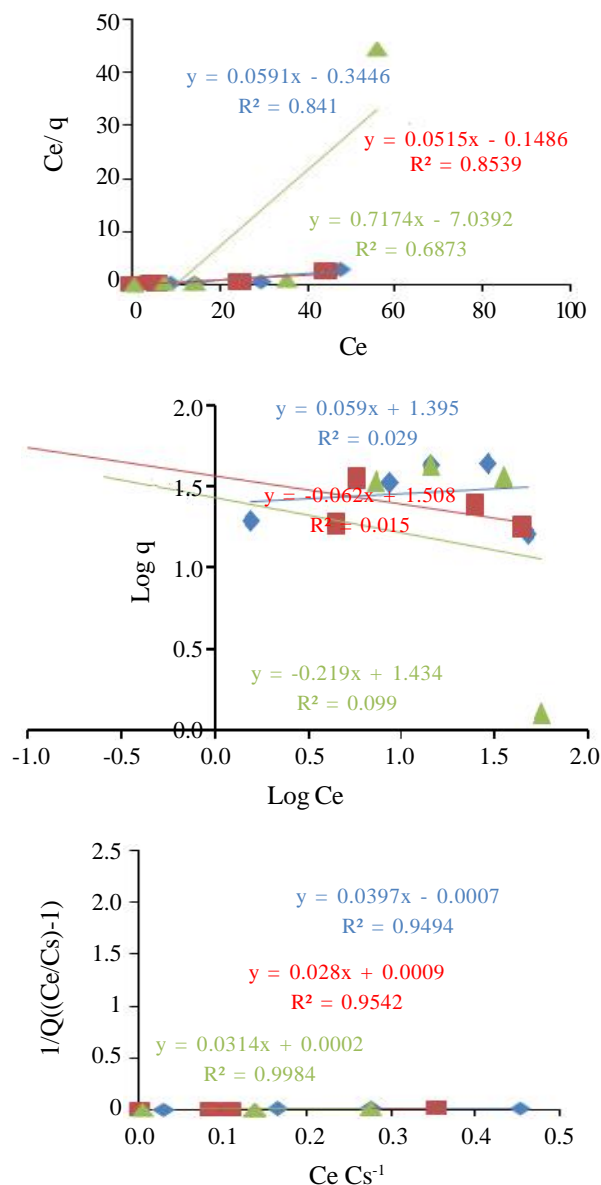


Figure 5. Langmuir, freundlich BET linearized equation of Zn (II). ♦ : ZA, ■ : ZB, ▲ : ZC.

metal bonding. This shows that Bayah zeolite adsorption on crystal surfaces is much greater.

If the CEC value of zeolite (Table 5) compared with adsorption capacity according to the Langmuir and BET equation, adsorption ability of zeolite tended

Table 5. Comparison – CEC with Adsorption Rating.

Natural Zeolite	CEC mg g <sup>-1</sup>	Value of Parametric Adsorption of Cu (II) Based on Formula		Value of Parametric Adsorption of Zn (II) Based on Formula	
		Langmuir	BET	Langmuir	BET
		q <sub>e</sub> (mgg <sup>-1</sup> )	q <sub>mono</sub> (mgg <sup>-1</sup> )	q <sub>e</sub> (mgg <sup>-1</sup> )	q <sub>mono</sub> (mgg <sup>-1</sup> )
ZA	87.72	33.333	13.699	16.949	25.641
ZB	96.75	47.619	25.707	19.417	34.602
ZC	137.58	28.571	10.000	1.395	31.646

to follow the pattern of its CEC value. Generally, zeolite that has a high CEC value can be seen having a high adsorption capacity through Cu and Zn. This is because the zeolite has a high CEC, it means zeolite has the number of lattice exchange and cavities in large numbers so that more number of metals are sequestered by zeolite. The active site as isomorphic substitution result of Si by Al caused the growing of ion exchange by zeolite. According to Said and Nurul (2008), a zeolite synthesized from coal bottom ash has a high CEC so that the adsorption power for Cu is also high.

## CONCLUSIONS

Bayah and Tasikmalaya had similar morphology structure and mineral types. Bayah and Tasikmalaya zeolites were mordenite mineral types and platy structured morphology, while Lampung zeolite was a clinoptilolite mineral type and tabular structured morphology.

Observed differences in CEC values implied variation in the adsorption capacity based on Langmuir and BET formulas. The capacity of the Bayah zeolite to adsorb Cu (II) is greater than those of the Tasikmalaya and Lampung zeolites. The study effective adsorption capacity of the Bayah zeolite was  $47.619 \text{ mg g}^{-1}$  [1 g Bayah zeolite adsorbed 47.619 mg Cu (II)], while that of the Lampung zeolite was only  $33.333 \text{ mg g}^{-1}$ , and that of the Tasikmalaya zeolite was only  $28.571 \text{ mg g}^{-1}$ . Further, the effective adsorption capacity of the Bayah zeolite with regard to Zn (II) amounted to  $19.417 \text{ mg g}^{-1}$ , which was greater than that of the Lampung zeolite ( $16.949 \text{ mg g}^{-1}$ ), and Tasikmalaya zeolite at  $1.395 \text{ mg g}^{-1}$ .

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