

# isotherm

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## ISOTHERM ADSORPTION OF 3000- $\mu\text{m}$ NATURAL ZEOLITE

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### Abstract

This study aims to develop an isotherm model of curcumin adsorption with natural zeolite with a size of 3000  $\mu\text{m}$ . The test was carried out by inserting zeolite into a solution of curcumin (10, 30, 40, 70, and 90 ppm) for 1 min, decanting for 10 minutes, and observing the changes in concentration that occurred. The isotherm model used is the Freundlich, Jovanovic, Dubinin-Radushkevich, Langmuir, Fowler-Guggenheim, and Temkin isotherm model. The results showed that natural zeolite adsorption on curcumin solution is suitable for the to Fowler-Guggenheim isotherm model. This is because the regression value of natural zeolite 3000  $\mu\text{m}$  is almost in accordance with the Fowler-Guggenheim adsorption isotherm model compared to other models. Based on this research, it is expected to provide new information to determine the adsorption isotherm model of zeolite with curcumin solution, especially at particle sizes 3000  $\mu\text{m}$ , and it is hoped that the natural zeolite adsorption process can help prospects in the treatment of liquid waste in the future.

Keywords: 3000  $\mu\text{m}$ , Isotherm adsorption, Natural zeolite.

## 1. Introduction

There are still many industries that dispose of waste, both radioactive waste, heavy metal waste, and dye waste which have dangerous impacts on the environment and human health [1]. One of the processes for treating wastewater is the adsorption process [2]. Adsorption is a method for removing wastewater contaminants [3]. There have been many studies on materials for adsorption, such as carbon [4], water hyacinth biomass [5], rice straw carbon [6], activated charcoal [7], Iron Oxide bound to polyacrylic acid [8], Biomass of *Rhizopus oryzae* [9], Chitosan Gel [10], And Zeolite [11].

Natural zeolite minerals, which are aluminosilicate compounds that are found in many areas in Indonesia, are scattered in various regions such as the city of Lampung [12] and Banten [13]. Lampung has a zeolite mineral potential of 18,945,000 m<sup>3</sup> [14]. One of the most relevant adsorbents and catalysts is zeolite, this is because zeolite has a large surface area and high adsorption capacity [15].

The phenomenon in which the adhesion of atoms to a solute on the surface of a substance occurs is called adsorption. Adsorption isotherm is one of the factors that affect the adsorption process. The relationship in the adsorption isotherm describes the phenomena and interactions between adsorbate and adsorbent [16]. Adsorption is divided into two types: physical adsorption (physisorption) and chemical adsorption (chemisorption), this adsorption classification is based on the interaction between the adsorbate and the adsorbent [17]. The illustration of the interaction between adsorbate and adsorbent is shown in Fig. 1.

Therefore, this study aims to develop an isotherm model of curcumin adsorption with natural zeolite with a size of 3000  $\mu\text{m}$  and to utilize natural zeolite, especially in Bayah, Banten, Indonesia. The novelty of this research is to describe a natural zeolite isotherm model measuring 3000  $\mu\text{m}$ . Analysis adsorption isotherm model to facilitate observation of the phenomenon and the interaction of natural zeolite (adsorbent) with curcumin solution (adsorbate). The materials used are natural zeolite (adsorbent) and curcumin solution (as a model of adsorbate). The materials used are natural zeolite (adsorbent) and curcumin solution (as a model of adsorbate). The curcumin solution was chosen because it has an ideal size (<1.4 nm) so that it can observe the adsorbent-adsorbate interaction so that it is possible to predict the characteristics of the adsorption isotherm experienced by zeolite [18]. The results showed that zeolite with a particle size of 3000  $\mu\text{m}$  can affect the adsorption process, and the adsorption isotherm on natural zeolites 3000  $\mu\text{m}$  represents the Fowler-Guggenheim model. This study provides new information to determine the adsorption isotherm model of zeolite with curcumin solution, especially at particle sizes 3000  $\mu\text{m}$ , and it is hoped that the natural zeolite adsorption process can help prospects in the treatment of liquid waste in the future.

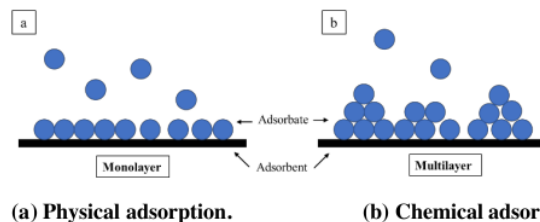


Fig. 1. Illustration of the interaction between adsorbate and adsorbent.

## 2. Isotherm Adsorption

### 2.1. Langmuir

The Langmuir adsorption model assumes that the adsorption only occurs in a monolayer, with a homogeneous surface and the adsorbent and adsorbate molecules don't interact [19]. The Langmuir equation is calculated using:

$$\frac{1}{Q_e} = \frac{1}{Q_{max}k_L} \frac{1}{C_e} + \frac{1}{Q_{max}} \quad (1)$$

where  $k_L$  is a constant in the Langmuir model,  $Q_e$  is the number of molecules adsorbed at equilibrium (mg/g), and  $Q_{max}$  is the adsorption capacity of the monolayer (mg/g).  $RL$  is an adsorption factor, explaining:

- (i)  $RL > 1$ , the adsorption process is not profitable
- (ii)  $RL = 1$ , the adsorption isotherm depends on the amount and concentration of adsorbed (linear adsorption process)
- (iii)  $RL = 0$ , the adsorption is too strong (irreversible)
- (iv)  $0 < RL < 1$  is the preferred adsorption process (adsorption can be controlled under certain conditions).

### 2.2. Freundlich

Freundlich's adsorption isotherm model informs multilayer adsorption, which occurs on heterogeneous surfaces [20]. The Freundlich equation is calculated using:

$$\text{Log } Q_e = K_f + \frac{1}{n} \log C_e \quad (2)$$

where  $K_f$  is Freundlich's constant and  $n$  is the adsorption intensity, which can predict the adsorption process which explains:

- (i)  $n < 1$ , the chemical adsorption
- (ii)  $n = 1$ , adsorption depends on the amount and concentration adsorbed combination of chemical and physical adsorption processes (the linear adsorption)
- (iii)  $n > 1$  is physical adsorption
- (iv)  $1/n < 1$  is normal adsorption
- (v)  $1/n > 1$  is cooperative adsorption
- (vi)  $1/n < 0$  is the preferred adsorption process (inhibits the desorption process)
- (vii)  $0 < 1/n < 1$  is normal adsorption (controllable under certain conditions)
- (viii)  $1/n = 0$ , the adsorbent surface is more heterogeneous

### 2.3. Temkin

The Temkin adsorption isotherm model describes types or factors that influence the adsorbent-adsorbate interaction [21, 22]. The Temkin equation is calculated using:

$$q_e = \beta_T \ln A_T + \beta_T \ln C_e \quad (3)$$

where  $A_T$  is the Temkin bond equilibrium constant (1.075 L/g),  $R$  is the gas constant (8.314 J/mol.K),  $T$  is the temperature, dan  $\beta$  is the adsorption heat constant (25.34 J/mol),  $\beta_T$  is a Temkin constant which describes:

- (i)  $\beta_T < 8$  kJ shows the physical adsorption.
- (ii)  $\beta_T > 8$  kJ shows the chemical adsorption.

#### 2.4. Dubinin-Radushkevich

The Dubinin-Radushkevich isotherm model describes the adsorption mechanism using the energy distribution on the heterogeneous surface of the adsorbent [23, 24]. The Dubinin-Radushkevich equation is calculated using:

$$\ln q_e = \ln q_s - (\beta \varepsilon^2) \quad (4)$$

where  $q_e$  is the number of adsorbates adsorbed at equilibrium (mg/g),  $q_s$  is the theoretical isotherm capacity (mg/g),  $\beta$  is the Dubinin-Radushkevich isotherm (mol/kJ<sup>2</sup>) dan  $\varepsilon$  is the constant for Dubinin-Radushkevich.

#### 2.5. Fowler-Guggenheim

The Fowler-Guggenheim isotherm model describes the lateral interactions of adsorbed molecules. The Fowler-Guggenheim isotherm model is one of the simplest correlations which allows prediction of the lateral interactions between adsorbates. The Fowler-Guggenheim equation is calculated using:

$$\ln \left( \frac{C_e e^{(1-\theta)}}{\theta} \right) - \frac{\theta}{1-\theta} = -\ln K_{FH} + \frac{2W\theta}{RT} \quad (5)$$

where  $K_{FH}$  is the Fowler-Guggenheim equilibrium constant (L/mg).  $W$  is the energy of the interaction between adsorbed molecules (kJ/mol),  $W$  is the heat of adsorption.

#### 2.6. Jovanovic

The Jovanovic isotherm model has the same assumptions as of the Langmuir model [25]. The Jovanovic equation is calculated using:

$$\ln q_e = \ln q_{max} - K_J C_e \quad (5)$$

where  $q_e$  is the amount of adsorbate in the adsorbent at equilibrium (mg/g),  $q_{max}$  is the maximum absorption of the adsorbate, and  $K_J$  is the Jovanovic constant.

Table 1 shows the calculations used to investigate the adsorption model.

**Table 1. Test model specifications and test conditions.**

Isotherm Adsorption Model	Equations	Plot
Langmuir	$\frac{1}{Q_e} = \frac{1}{Q_{max} k_L} \frac{1}{C_e} + \frac{1}{Q_{max}}$	$\frac{1}{q_e}$ vs $\frac{1}{C_e}$
Freundlich	$\log Q_e = K_f + \frac{1}{n} \log C_e$	$\ln q_e$ vs $\ln C_e$
Temkin	$q_e = \beta_T \ln A_T + \beta_T \ln C_e$ $\Delta Q = -\Delta H$	$Q_e$ vs $\ln C_e$ or $\theta$ vs $\ln(C_e)$
Dubinin-Radushkevich	$\ln q_e = \ln q_s - (\beta \varepsilon^2)$	$\ln Q_e$ vs $\varepsilon^2$
Fowler-Guggenheim	$\ln \left( \frac{C_e e^{(1-\theta)}}{\theta} \right) - \frac{\theta}{1-\theta} = -\ln K_{FG} + \frac{2W\theta}{RT}$	$\theta$ vs $\ln \left( \frac{C_e e^{(1-\theta)}}{\theta} \right)$
Jovanovic	$\ln q_e = \ln q_{max} - K_J C_e$	$C_e$ vs $\ln Q_e$

### 3. Method

The material used as an adsorbent is natural zeolite originating from Banten, Indonesia, while the material used as an adsorbate model is a curcumin solution. In general, the adsorption process is carried out through the following steps: (1) curcumin solids are dissolved in water to form several concentrations (10, 30, 50, 70, and 90 ppm), (2) 3000  $\mu\text{m}$  zeolite particles are added to each variation of the concentration of curcumin solution, (3) the zeolite particles that have been put into curcumin solution are stirred for 60 seconds and be quiet for 10 minutes, (4) the solution is filtered, (5) the filtrate was measured and analyzed using a TDS & EC meter (Model E-1 portable), and (6) changes in concentration were analyzed using the adsorption isotherm model. The adsorption isotherm model used is the Freundlich, Jovanovic, Dubinin-Radushkevich, Langmuir, Fowler-Guggenheim, and Temkin isotherm model. Meanwhile, physicochemical properties of natural zeolite particles were analyzed using a Digital Microscope (BXAW-AX-BC, China) and X-Ray Diffraction (XRD).

### 4. Results and Discussion

#### 4.1. Physicochemical properties of natural zeolites 3000 $\mu\text{m}$

Figure 2 shows the zeolite particle microscope results. The results show that the particle size is in the range of 3000  $\mu\text{m}$ . Figure 3 shows the XRD comparison results of natural zeolite with zeolite standards. The analysis using XRD was carried out with the same procedure as the previous study [26]. The XRD method is needed to determine the proportion of each zeolite phase. The XRD pattern of natural zeolite according to the International Zeolite Association shows that natural zeolite consists of a clinoptilolite (HEU) and zeolite mordenite (MOR) phase [27]. The XRD results showed that the peak intensity was relatively lower than that of synthetic zeolites [28]. The clinoptilolite phase is the dominant phase because almost all peaks are clearly detected. Meanwhile, the peak of the mordenite phase is not clearly visible, because the intensity is low and overlaps with the clinoptilolite peak. Most likely the second phase of zeolite grows during hydrothermal processes or natural sedimentation [29, 30].

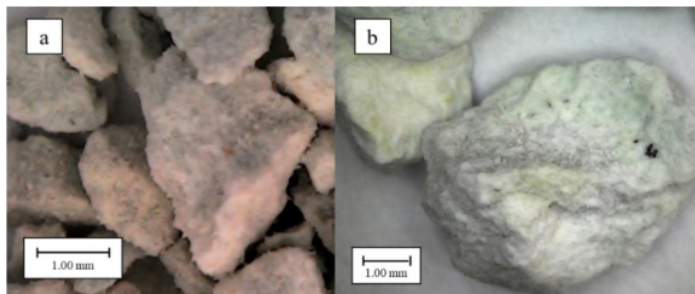


Fig. 2. Microscope image of zeolite particles 3000  $\mu\text{m}$ .

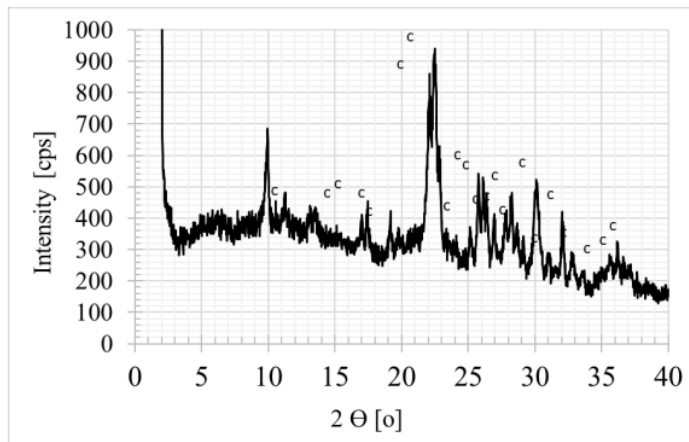


Fig. 3. The X-Ray diffraction (XRD) of natural zeolite.

#### 4.2. Isotherm adsorption models of natural zeolite

One of the important factors in adsorption is the adsorption isotherm. The relationship in the adsorption isotherm describes the phenomena and interactions between adsorbate and adsorbent [16, 31]. The adsorption isotherm model is analyzed by looking at the results of the regression data in accordance with the linearization of the mathematical model. Several parameters in the adsorption process can predict the phenomena that occur between the adsorbent and adsorbate during the adsorption process. The parameters of the adsorption isotherm model are presented in Table 2.

Adsorption isotherm is one of the factors that affect the adsorption process. The relationship in the adsorption isotherm describes the phenomena and interactions between adsorbate and adsorbent [16]. The adsorption isotherm model is analyzed by looking at the results of the regression data in accordance with the linearization of the mathematical model. Several parameters in the adsorption process can predict the phenomena that occur between the adsorbent and adsorbate during the adsorption process. The parameters of the adsorption isotherm model are presented in Table 2.

Langmuir model adsorption parameters using  $R^2$ ,  $q_{max}$ ,  $K_L$ , and  $R$ . Figure 4 depicts the Langmuir isotherm model. The results of the data calculation are shown in Table 2. The Langmuir isotherm model is a poor representation of the natural zeolite adsorption system. This can be seen from the regression value of  $R^2$  which is quite low, namely 0.2181. The correlation coefficient ( $R^2$ ) is calculated as an indicator of the suitability of experimental data with the proposed [29]. The results obtained  $0 < RL < 1$ , which indicates the preferred adsorption process (adsorption can be controlled under certain conditions). The Langmuir model assumes that the adsorption process occurs by forming a monolayer structure without any lateral interaction between the adsorbate and the adsorbent [25]. Thus, the adsorption system on natural zeolites is incompatible with the Langmuir isotherm model, which assumes that the adsorption process occurs on a homogeneous surface in the form of a monolayer. This is consistent with other studies [32].

Table 2. Parameters of the isotherm adsorption model.

Isotherm adsorption models	Parameters	Value	Informations
Langmuir	$q_{max}$ (mg/g)	181.8180	The maximum monolayer adsorption capacity
	$K_L$ (L/mg)	0.9999	Langmuir adsorption constant
	$R_L$	0.0834	$0 < R_L < 1$ , indicating favorable adsorption.
	$R^2$	0.2181	The correlation coefficient.
Freundlich	$n$	2.5753	$n < 1$ implies adsorption with a chemical process. $n > 1$ defines adsorption with a physical process.
	$1/n$	0.3883	$1/n < 1$ indicating a normal adsorption. $1/n > 1$ implies cooperative adsorption
	$k_f$ (mg/g)	612.3475	The Freundlich constant
	$R^2$	0.305	The correlation coefficient.
Temkin	$A_T$ (L/g)	216.913	Equilibrium binding constant
	$\beta_T$ (J/mol)	28.4631	$\beta_T < 8$ kJ indicates physical adsorption $\beta_T > 8$ kJ indicates chemical adsorption
	$R^2$	0.2354	The correlation coefficient.
Dubinin	$q_s$ (mg/g)	229.0178	The maximum adsorption capacity of the adsorbent.
	$\beta$ (mol <sup>2</sup> /kJ <sup>2</sup> )	0.0931	the Dubinin-Radushkevich isotherm saturation capacity
	$E$ (kJ/mol)	2.31740	Energy $E < 8$ kJ/mol is a physical adsorption process
	$R^2$	0.0916	The correlation coefficient.
Fowler-Guggenheim	$W$ (kJ/mol)	34.456	The attraction between adsorbed molecules, and the presence of an exothermic process.
	$K_{FG}$ (L/mg)	$1.5614 \times 10^{-14}$	The Fowler-Guggenheim constant.
	$R^2$	0.8971	The correlation coefficient
Jovanovic	$K_j$ (L/mg)	0.0462	Jobanovic adsorption constant
	$q_{max}$ (mg/g)	421.491	The maximum monolayer adsorption capacity
	$R^2$	0.4325	The correlation coefficient



Freundlich model adsorption parameters using  $R^2$ ,  $n$ , and  $1/n$ . Figure 5 depicts Freundlich's isotherm model. The Freundlich isotherm model is a poor representation of the natural zeolite adsorption system. The regression value of  $R^2$  which is owned by this model is quite low, namely 0.305. Meanwhile, other parameters such as  $n$  (preferential adsorption from one adsorbate to another) are 2.57 ( $n > 1$ ), which indicates the physical adsorption process. The value of  $n$  in the range 2-10 has good adsorption characteristics [33]. Meanwhile, the value of  $1/n$  which ranges between less than 1 and greater than zero indicates a normal adsorption process (controllable under certain conditions). Thus, the adsorption system on natural zeolites does not fit the Freundlich isotherm model. This is consistent with other studies [34].

The Temkin model adsorption parameters using  $R^2$ ,  $A_T$ , and  $\beta_T$ . Figure 6 depicts Temkin's model isotherm. The positive value of the constant B (slope) corresponds to the positive value of the variation in the adsorption energy parameter  $\beta_T$  which indicates that the adsorption is exothermic. The value of  $\beta_T < 8$  kJ indicates the physical adsorption process. High  $A_T$  shows an interesting interaction between adsorbate and adsorbent. The  $A_T$  values for all adsorption systems show relatively small values, meaning that there is less affinity between the adsorbent and adsorbate molecules because there is a dominant physical interaction confirmed by  $\beta_T$  parameters. Physical interactions involve only the weaker interactions. This is supported by low  $\beta_T$  results indicating a weak interaction between adsorbent and adsorbate [35]. Thus, the adsorption system on zeolite does not fit the Temkin isotherm model.

The adsorption parameters of the Dubinin-Radushkevich model using  $R^2$ ,  $q_e$ ,  $\beta$ , and  $E$ . Figure 7 illustrates the Dubinin-Radushkevich isotherm model. The  $E$  value estimates the type of absorption reaction. The Dubinin-Radushkevich isotherm model is the most unfavorable representation of the natural zeolite adsorption system. The regression value for  $R^2$  of this model is very low, namely 0.0916. The  $E$  value obtained is around 2.3174 kJ/mol, which indicates that the value  $E < 8$  kJ/mol indicates that the adsorption takes place through a process of physical interaction [16]. Physical interactions involve only the weaker interactions. Thus, the adsorption system on natural zeolites does not fit the Dubinin-Radushkevich isotherm model.

The adsorption parameters of the Fowler-Guggenheim model using  $R^2$ ,  $W$ , and  $K_{FG}$ . The Fowler-Guggenheim isotherm model is the best representation of the natural zeolite adsorption system. The regression value of  $R^2$  which is owned by this model is the best among the other five models, namely 0.8971. Figure 8 depicts the Fowler-Guggenheim isotherm model. The Fowler-Guggenheim equation is one of the most probable equations for lateral interactions. The results show that  $W$  is positive, which means that the interactions between the adsorbed molecules are attractive. When the  $W$  value is positive, the adsorption will increase, and this is due to an increase in the interaction between adsorbed molecules with increasing load. Small identical  $K_{FG}$  means that the adsorbent-adsorbate interaction on the surface is less efficient in adsorbing adsorbate molecules because of the dominance of physical interactions [16]. This is because the Fowler-Guggenheim isotherm shows that there is a lateral interaction at a set of localized sites with weak interactions due to the Van der Waals interaction effect between the adsorbed species on neighboring sites [35].

The Jovanovic model of adsorption parameters using  $R^2$ ,  $K_j$ , and  $q_{max}$ . Figure 9 depicts the Jovanovic isotherm model. The Jovanovic model assumes that there are no interactions in the adsorbed layer [36]. The adsorption surface model is in fact the same as that described by Langmuir. The results show that the adsorption of a monolayer or monolayer of the Jovanovic equation assumes a monolayer localized adsorption without lateral interaction [37].

Based on the  $R^2$  value for each adsorption model between natural zeolite and curcumin solution, the adsorption results were obtained that matched the Fowler-Guggenheim isotherm model. For the adsorption system with zeolite adsorbent, the other 5 models are not suitable. Adsorption system with natural zeolite adsorbent with curcumin solution at large particle sizes (3000  $\mu\text{m}$ ) following the order of the Fowler Guggenheim > Jovanovic > Freundlich > Temkin > Langmuir > Dubinin model. The adsorption system with natural Zeolite adsorbent has a good correlation with the Fowler-Guggenheim model which informs the multilayer adsorption process. This model is one of the simplest correlations which allows prediction in the lateral interactions between the adsorbates.

The Fowler-Guggenheim model takes the lateral interactions of adsorbed molecules during the adsorption process. The Fowler-Guggenheim isotherm shows that there is a localized lateral interaction with weak interactions (Van der Waals interaction effect) between adsorbed species in other layers [35]. The results of the empirical interaction energy data ( $W$ ) show that  $W > 0$ , which indicates an interesting interaction between adsorbed molecules. The adsorption interaction energy varies linearly based on loading. The energy of the adsorption interaction will increase with the addition of the load, this is due to the increased interaction between the adsorbed molecules. The plot of the Fowler-Guggenheim equation was carried out by calculating the surface area using the maximum adsorption capacity value. To verify the validity of the Fowler-Guggenheim model, it is necessary to recalculate the amount adsorbed using the equilibrium concentration value and the Fowler-Guggenheim parameter.

Based on the results of the 6 models that have been tested, only 1 model fits the characteristics of natural zeolite 3000  $\mu\text{m}$ . This is due to the large zeolite particle size which causes a small surface area so that the adsorption process is not optimal.

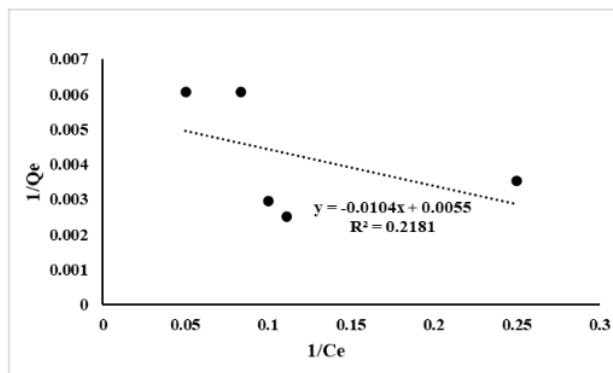


Fig. 4. Langmuir isotherm model of natural zeolite 3000  $\mu\text{m}$ .

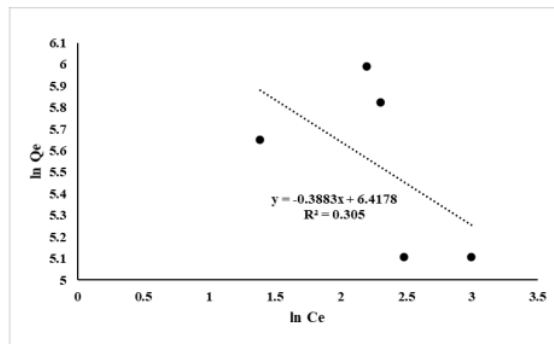


Fig. 5. Freundlich isotherm model of natural zeolite 3000  $\mu\text{m}$ .

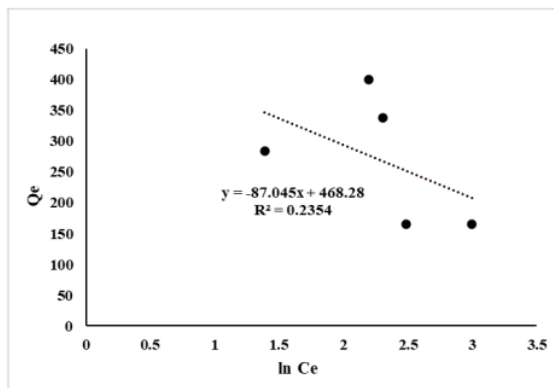


Fig. 6. Temkin isotherm model of natural zeolite 3000  $\mu\text{m}$ .

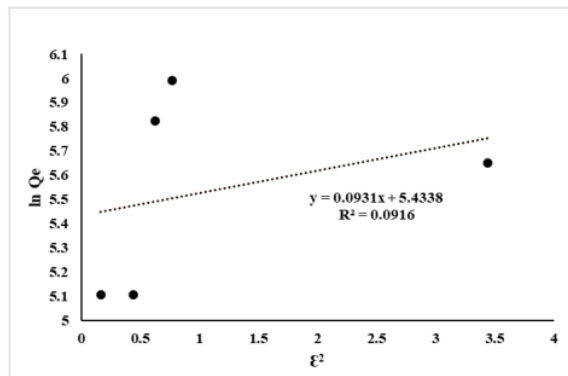


Fig. 7. Dubinin-Radushkevich isotherm model of natural zeolite 3000  $\mu\text{m}$ .

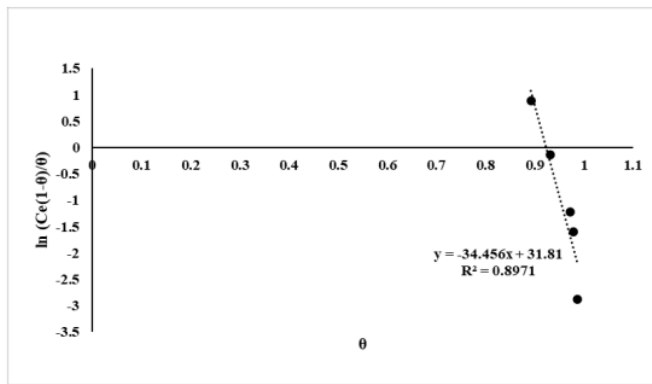


Fig. 8. Fowler-Guggenheim isotherm model of natural zeolite 3000  $\mu\text{m}$ .

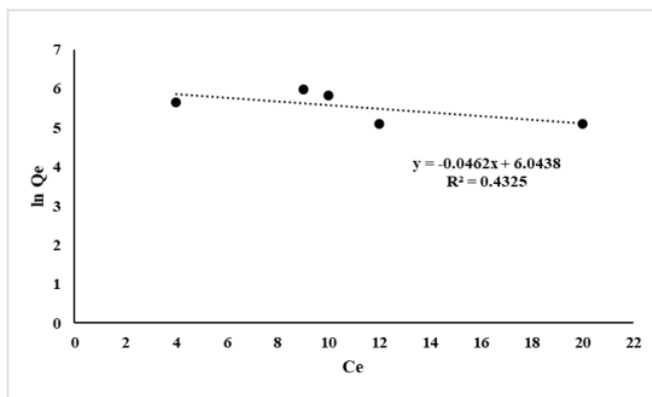


Fig. 9. Jovanovic isotherm model of natural zeolite 3000  $\mu\text{m}$ .

### 5. Conclusion

This study is to determine the natural zeolite adsorption isotherm model with a size 3000  $\mu\text{m}$  based on the characteristics of zeolite (adsorbent) with curcumin solution (adsorbate) and to utilize natural zeolite, especially in Indonesia. Based on testing through several isotherm models, namely: Langmuir, Freundlich, Temkin, Dubinin-Radushkevich, Fowler-Guggenheim, and Jovanovic isotherm models. The results show that the adsorption of natural zeolite on the curcumin solution matches the Fowler-Guggenheim isotherm model. This is because the results of the regression value on natural zeolite 3000  $\mu\text{m}$  are almost in accordance with the Fowler-Guggenheim adsorption isotherm model compared to other models. Based on the results of this study, it is expected to provide new information to determine the adsorption isotherm model of natural zeolite with curcumin solution, especially at particle sizes 3000  $\mu\text{m}$ .

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