Journal of Engineering Science and Technology
Vol. 17, No. 2 (2022) 1265 - 1275

ISOTHERM ADSORPTION OF 40-μm ZEOLITE PARTICLES FOR TREATMENT OF DYE WASTEWATER

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Abstract

This study aimed to determine the characteristics of the adsorption isotherm of curcumin dye solution by 40-μm zeolite particles. The 40-μm zeolite particles measuring were put in a certain concentration of curcumin dye solution and then the change in the concentration of the curcumin dye solution was analysed. Curcumin solution has an ideal size (less than 1.4 nm), so it is ideal to be used as a model for dye wastewater. To support our analysis, we use 4 isotherm models, namely Langmuir, Freundlich, Temkin, and Dubinin-Radushkevich The results showed that the 40-μm zeolite particles were able to adsorb curcumin solution as a dye wastewater model. The adsorption model that is suitable in this study, that is the Langmuir (R^2= 0.868) which explains the favourable monolayer adsorption. The most suitable results are followed by the Dubinin-Radushkevich model (R^2=0.938). The Dubinin-Radushkevich model describes the adsorption process following the pore-filling mechanism, this is following the characteristics of zeolites which have many pores. However, the adsorption process in this study is also compatible with the Freundlich and the Temkin adsorption models, in which the regression values respectively are 0.819; and 0.764. Freundlich's model shows the value of n=0.515<1 and $1/n$=1.940>1 explaining that the adsorption process occurs in a multilayer with chemical interactions and cooperative type. The Temkin model also shows a B_T value of 22.801 which explains the adsorption that occurred in the chemical interaction. This research provides new information regarding the use of zeolite particles of 40-μm size for the adsorption process, especially in dye wastewater.

Keywords: Curcumin dye solution, Isotherm adsorption, Zeolite.

1. Introduction

Currently, many industries use dyes in their operational activities. For example, the dye industry, textiles, and plastics industry. Water was widely used to dissolve dyes so that it produces a lot of coloured liquid wastewater [1]. Dyes can become pollutants in water. If the wastewater is discharged into the environment, it will pollute the water, cause poisoning of agricultural land, block the sun and interfere with the life of aquatic organisms, reduce the diversity of aquatic biota, cause disruption of ecosystem balance, cause carcinogens, to become mutagens in organisms [2, 3]. Therefore, it is necessary to manage dye wastewater. There are many techniques for managing wastewater including biological, physical, and chemical methods [4]. Chemical wastewater treatment can be done cheaply, and environmentally friendly, one of which is by using the adsorption process on wastewater molecules using adsorbents. Adsorption is a tertiary waste treatment technique [5].

Adsorption is the phenomenon of the adhesion of ions, atoms, or molecules from dissolved solids, liquids, or gases, which occur on the surface of a substance. The substance which the ion, atom, or molecule attaches is called an adsorbent, while the ion, atom, or dissolved molecule that is attached is called an adsorbate [6]. Adsorption isotherm is an important factor in the adsorption process because, in the adsorption isotherm, the interaction phenomenon between adsorbent and adsorbate is explained. Based on its molecular interactions, adsorption is divided into chemical and physical adsorption [7]. If the attractive force between the adsorbent and the adsorbate is very weak with the Vander Waal Tensile force, the adsorption process is physical adsorption [8]. If the tensile force between the adsorbent and the adsorbate is very strong which is almost the same as the strength of the chemical bond, the adsorption process is included in chemical adsorption, chemical adsorption is not easily reversed due to the strong tensile force [9].

Natural zeolite particle can be used as an adsorbent [10]. Zeolite is an alumino-silicate compound that has surface absorption properties. Zeolite particles have a microporous framework with a very large surface area because of all directions' interconnected cavities [11, 12]. This pore architecture will be very useful in the adsorption process. Zeolite is abundant and easy to find in Indonesia, especially in the Bayah, Lampung, and Tasikmalaya areas. Zeolite from the Bayah area has a good capability for adsorption because it has a modernite type and a platy structure [12].

Other materials have been widely used as adsorbents such as carbon soursop peel waste [13], activated carbon from almond bark [14], aloe vera waste biomass [15], iron oxide-graphene oxide nanocomposite [16], modified chitosan [17], tungsten oxide [18], methane [19], modified clays [20], MOFs material [21], zeolite particle [22], and many more. From these current studies, no one has specifically discussed the adsorption process using specific zeolite particle sizes to adsorb the curcumin solution which is used as a model for dye wastewater. Curcumin solution has an ideal size (less than 1.4 nm) [23]. The particle size of 1.4 nm is ideal for use as a wastewater dye model because it can be absorbed by the pores of zeolite particles that have a pore diameter of less than 2 nm [24].

Therefore, this study aimed to determine the characteristics of the adsorption isotherm of curcumin solution by 40 μm zeolite particles. The main novelty in this study was to analyse the isotherm adsorption of 40-μm zeolite particles to adsorb curcumin dye solution as a model of pollutant in wastewater. The zeolite particles measuring 40 μm were put in a certain concentration of curcumin solution and then the change in the concentration of the curcumin solution was analysed. The curcumin solution as a model can facilitate the observation of adsorbent-adsorbate interactions and make it easier to predict the adsorption isotherm model that occurs in the zeolite. This research provides new information regarding the use of zeolite particles of 40 μm size for the adsorption process, especially in dye wastewater.

2. Methods

2.1. Materials used in this study

The natural zeolite particles of 40 μm were used as an adsorbent in this study. The natural zeolite particles were obtained from Bayah Banten, Indonesia. The curcumin dye was used as an experimental adsorbate model. The curcumin dye was carried out from PT. Motasa Indonesia, Mojokerto, Indonesia. Test instruments used in this study include the XRD (XRD, PANalytical X'Pert PRO; Philips Corp., The Netherland), TDS meter (Model E-1 portable), and the digital microscope (BXAW-AX-BC, China; 1000x magnification).

2.2. Adsorption method

This research was conducted during the pandemic, the research was done at home uses existing equipment. The adsorption process was done by weighing 1 g of zeolite particles and mixing it with 50 mL of curcumin dye solution. The concentration of curcumin dye solutions was varied to 10, 30, 50, 70, 90, and 100 ppm. The mixture was then stirred for 1 minute with the speed of 300 rpm and settle for 10 minutes to occurs the adsorption process. Adsorption can occur within 10 minutes and changes in ppm can be detected, during the adsorption process the temperature was the room temperature. The mixed solution was then filtered. The filtrate was then analysed with a TDS meter. In this study, repetition is only done once. The adsorption process was analysed from changes in the concentration of curcumin solution (ppm). To analysis the characteristic of the zeolite particles more closely, we also saw zeolite particles using a Digital Microscope. The analysis phase of Bayah zeolite particles was also carried out in this study using X-Ray Diffraction (XRD).

The adsorption process occurs during the solution mixing procedure. When the solution was stirred there is an interaction between the adsorbate-adsorbent. The mixture settled for a while before measuring the ppm to occur the natural precipitation. The next step after the adsorption process was carried out is to evaluate the adsorption model that occurs. Several model adsorptions were used in this study for analysed of adsorption process including Langmuir, Freundlich, Temkin, and Dubinin-Radushkevich.

2.3. Isotherm adsorption models used in this study

2.3.1. Langmuir

Langmuir's Isotherm model explains that the adsorption process forms a single layer (monolayer) on the outside of the adsorbent surface. There are four assumptions of the adsorption isotherm model, namely:

(i) The molecules react on the adsorbent surface.
(ii) Each side can only bind one adsorbate molecule.
(iii) Each side has the same potential energy.

(iv) After one adsorbate molecule binds to the adsorbent surface, there are no other bonds between the adsorbed molecules and other adsorbates.

Adsorption forms a single layer (monolayer) as modelled in Fig. 1(a) [6, 25]. The Langmuir process is expressed by Eqs. (1) and (2):

$$\frac{1}{Q_e} = \frac{1}{Q_{\max} K_L} \frac{1}{C_e} + \frac{1}{Q_{\max}} \tag{1}$$

$$R_L = \frac{1}{1+K_L C_e} \tag{2}$$

where Q_e is the number of molecules adsorbed per gram adsorbent at equilibrium (mg/g), K_L is the constant in the Langmuir adsorption model, Q_{max} is the monolayer adsorption capacity (mg/g) and C_e is the equilibrium concentration of adsorbate (mg/L), while R_L is the constant of dimensionless of adsorption separator factor [26].

(i) $R_L > 1$, is unfavourable of adsorption process (occur the most desorption processes).
(ii) $R_L = 1$, adsorption process is linear (depending on the concentration absorbed and the amount absorbed).
(iii) $R_L = 0$, adsorption process is Irreversible (adsorption is too strong).
(iv) $0 < R_L < 1$ adsorption process is favourable (adsorption process can be controlled).

2.3.2. Freundlich

The Freundlich adsorption model describes adsorption in general. This model can describe the adsorption process that occurs chemically or physically. The adsorption occurs the multilayer adsorption and the bond are not too strong with the distribution of heterogeneous energetic from the active site of adsorbent. The multilayer adsorption is modelled in Fig. 1(b). The Freundlich isotherm formula is expressed by Eq. (3):

$$\ln Q_e = \ln K_f + \frac{1}{n} \ln C_e \tag{3}$$

where Q_e is the amount of adsorbate absorbed per unit of adsorbent (mg/g), K_f is the relative adsorption capacity, C_e is the concentration of adsorbate under equilibrium conditions (mg/L), and n is the intensity of adsorption indicating the linearity [6, 27]. The value of n can predict using:

(i) $n = 1$, is the linear adsorption.
(ii) $n < 1$, is the chemical interaction adsorption.
(iii) $n > 1$, is the physical interaction adsorption.
(iv) $0 < 1/n < 1$, is the Favourable adsorption process.
(v) $1/n > 1$ is a cooperative adsorption process.

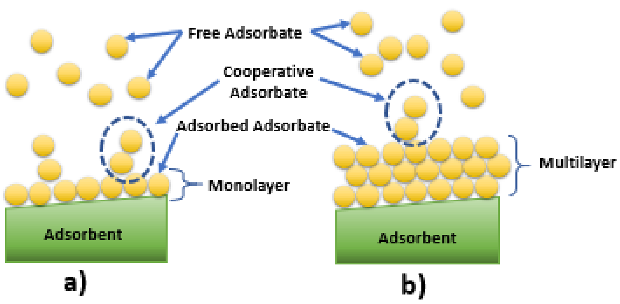

Fig. 1. Model of adsorption process (a) monolayer (b) multilayer.

2.3.3. Temkin

Temkin isotherm adsorption explains the correlation of isotherm adsorption and interaction indirect adsorbate. There are three postulates of the Temkin isotherm adsorption model:

(i) The increasing surface coverage of adsorbent affected the decreases in adsorption heat linearly,

(ii) On the adsorbent surface, the isotherm adsorption process shows uniform binding energy distribution,

(iii) The interaction of adsorption involves the interaction between adsorbent-adsorbate [6, 23, 28]. The Temkin isotherm adsorption model expressed in Eq. (4) :

$$Q_e = B_T lnA_T + B_T lnC_e \tag{4}$$

where B_T is the constant of adsorption heat (if the B_T < 8 kJ/mol, the adsorption process shows physical adsorption; and B_T > 8 kJ shows the chemical adsorption), A_T is the Temkin equilibrium constant, and the T expressed the absolute temperature.

2.3.4. Dubinin-Radushkevich

Dubinin-Radushkevich isotherm is the adsorption process on the heterogeneous surface of the adsorbent which has many pore structures. The adsorption occurs free energy. The adsorption process occurs by filling the micropore of the adsorbent with the adsorbate molecule [29]. Dubinin-Radushkevich isotherm is written in Eq. (5):

$$ln\, Q_e = ln\, Q_s - (\beta\varepsilon^2) \tag{5}$$

where Q_S is the saturation capacity of theoretical isotherms (mg/g), β is the constant of Dubinin-Radushkevich isotherm, and $\mathcal{E}$ is the Polanyi potential (J/mol). $\mathcal{E}$ can be calculated by the Eq. (6) [6]:

$$\varepsilon = RT\, ln\left[1 + \frac{1}{C_e}\right] \tag{6}$$

where C_e is the equilibrium concentration of solute. To calculate the free energy of the adsorption process per adsorbate molecule can use Eq. (7) [23]:

$$E = \frac{1}{\sqrt{2\beta}} \tag{7}$$

where E is the energy needed to remove the molecules from the surface of the adsorbent. If E < 8 kJ/mol, the adsorption is the physical adsorption. If 8 < E < 168 kJ/mol, the adsorption is the chemical adsorption.

3.Result and discussion

Figure 2 shows the photograph of zeolite particles. Fig. 2(a) is the agglomeration of zeolite particles. The particles were very fine with greenish. Fig. 2(b) is a photograph of zeolite particle grains. The particle sizes were 0.04 mm (40 µm).

Figure 3 shows the comparison of Bayah's natural zeolite XRD pattern compared to the standard zeolite XRD pattern according to the International of Zeolite Association [30]. using XRD analysis was done using the previous literature [31]. The results showed that Bayah's natural zeolite consisted of mordenite (MOR) and clinoptilolite (HEU) zeolites. The peak intensity of Bayah's natural zeolite is relatively lower than that of synthetic standard zeolites, which indicates that the sample also contains a nanocrystalline phase. The dominant phase of natural Bayah zeolite is the clinoptilolite phase because many peaks are detected. There are several mordenite peaks but not visible such as 2ϴ 6.51° and 22.2° which are very low in intensity and overlap with the clinoptilolite peaks. The results obtained are following the literature which reports that Bayah's natural zeolite has mordenite and clinoptilolite phases [32, 33].

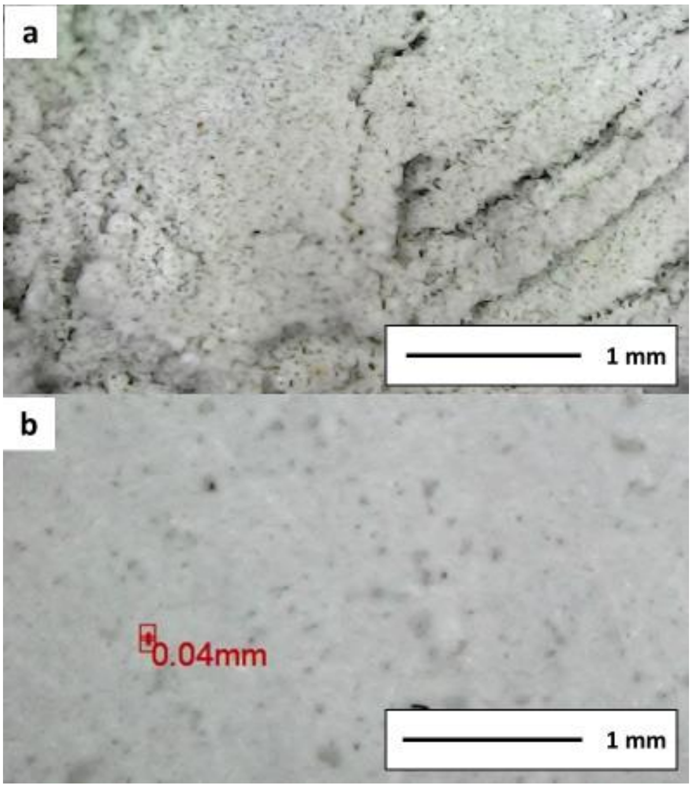

Fig. 2. The photograph of zeolite
(a) the agglomeration particles (b) the grain of particles.

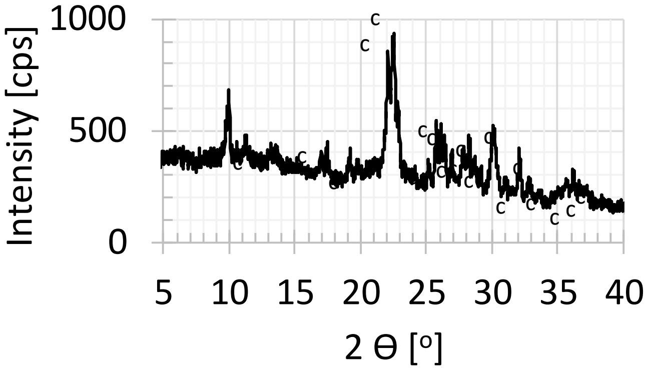

Fig. 3. XRD pattern of Bayah natural zeolites.

Figures 4(a), (b), (c), and (d) respectively show fitting data with the model adsorption isotherm Langmuir, Freundlich, Temkin, and Dubinin-Radushkevich. The most suitable adsorption isotherm model in this study is the Langmuir (has R^2= 0.868). Other parameters obtained in this work are R_L = 0.073. 0 < R_L < 1 which describe the adsorption have the favourable monolayer adsorption [34]. The most suitable results are followed by Dubinin-Radushkevich with a regression value of 0.8342.

The Dubinin-Radushkevich occur the adsorption process was filling the micropore of adsorbent with the adsorbate molecule [35]. The parameter of the Dubinin-Radushkevich model is shown in Table 1. The β as the Dubinin-Radushkevich constant is influenced by the pore volume of the adsorbent. The pore volume has an impact on the energy value (E). The β value indicates a high adsorption capacity; this has an impact on the maximum bond energy value [36].

In addition, the result shows that the adsorption process is also compatible with the Freundlich and the Temkin models. The regression values of all models were more than 0.700.

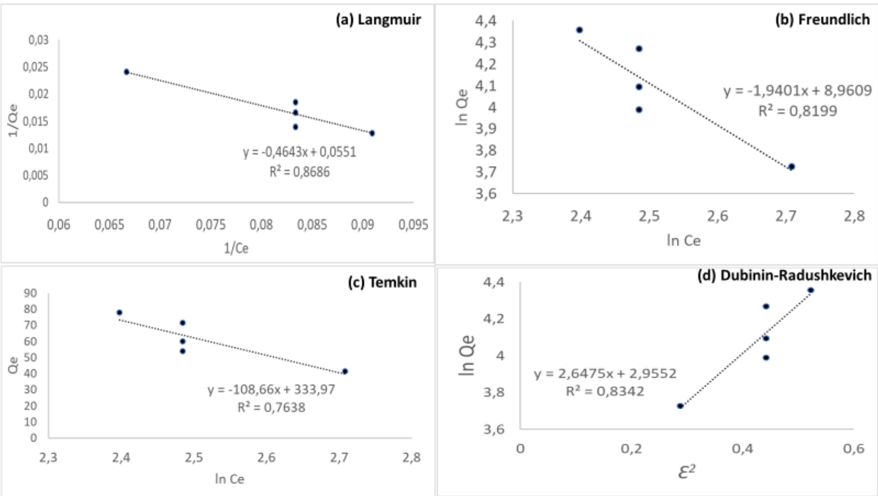

Fig. 4. (a), (b), (c), and (d) respectively show fitting data with the model adsorption isotherm Langmuir, Freundlich, Temkin, and Dubinin-Radushkevich.

Table 1 summarizes the adsorption phenomena described based on the model parameters used in this study. From the results of Table 1, we get several points.

(i) Langmuir stated the maximum monolayer adsorption capacity. From the data, we get R_L 0.073. 0 < R_L < 1 indicates favourable adsorption.

(ii) From Freundlich's data, the data show n < 1, indicating the chemical interaction adsorption. Next, the score is obtained 1/n > 1, indicating a cooperative adsorption process.

(iii) Temkin stated B_T > 8 kJ shows the chemical adsorption.

(iv) The Dubinin-Radushkevich isotherm model stated Energy (E) < 8 kJ/mol is a physical adsorption process.

Table 1. The adsorption parameter.

Model	Parameter	Result	Notes
Langmuir	Q_{max} (mg/g)	18.149	The maximum monolayer adsorption capacity
	K_L (L/mg)	0.026	Langmuir adsorption constant
	R_L	0.073	$0 < R_L < 1$, indicating favourable adsorption.
	R^2	0.868	The correlation coefficient.
Freundlich	n	0.515	$n < 1$, is the chemical interaction adsorption
	$1/n$	1.940	$1/n > 1$ is a cooperative adsorption process
	K_f (mg/g)	7792.37	The relative adsorption capacity
	R^2	0.819	The correlation coefficient.
Temkin	A_T (L/g)	21.606	The Equilibrium binding constant
	B_T (J/mol)	22.801	$B_T > 8$ kJ shows the chemical adsorption
	R^2	0.764	The correlation coefficient.
Dubinin-Radushkevich	q_s (mg/g)	523.480	The maximum adsorption capacity of the adsorbent.
	β (mol^2/kJ^2)	4.110	The Dubinin-Radushkevich isotherm saturation capacity
	E (kJ/mol)	0.349	Energy $E < 8$ kJ/mol is a physical adsorption process
	R^2	0.8342	The correlation coefficient.

The adsorption process that occurs in this study is illustrated in Fig. 5. In this study, cooperative adsorption occurred. The adsorbate fills the pores of the zeolite by physical interaction. However, on the surface of the adsorbent occur monolayer and multilayer adsorption with chemical interactions. Based on the results obtained, 40-µm zeolite has a very good performance in the adsorption process, both of zeolite surface and the porous part are active interact with the adsorbate.

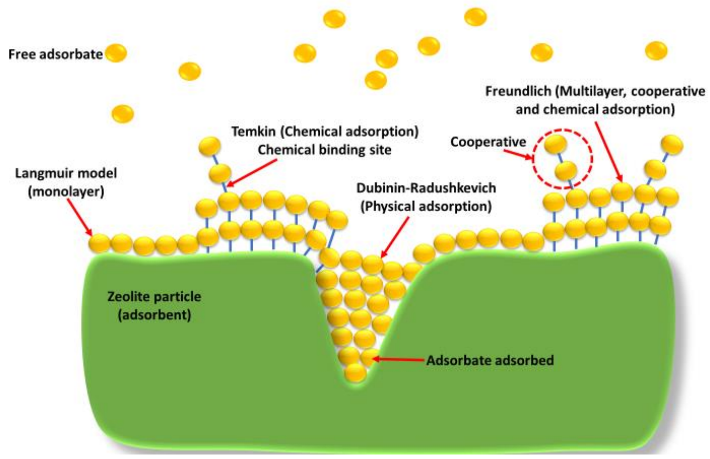

Fig. 5. The adsorption process that occurs in this study.

4. Conclusion

This study investigated the characteristics of the adsorption isotherm of curcumin dye solution by 40 µm zeolite particles. The results showed that the 40-µm zeolite particles were able to adsorb curcumin solution as a dye wastewater model. The adsorption model that is suitable in this study, that is the Langmuir (R^2= 0.868) which explains the favourable monolayer adsorption. The most suitable results are followed by the Dubinin-Radushkevich model (R^2=0.938). The Dubinin-Radushkevich model describes the adsorption process following the pore-filling mechanism, this is following the characteristics of zeolites which have many pores. However, the adsorption process in this study is also compatible with the Freundlich and the Temkin adsorption models, in which regression values respectively were 0.819; and 0.764. Freundlich's model shows the value of n=0.515<1 and 1/n=1.940>1 explaining that the adsorption process occurs in a multilayer with chemical interactions and cooperative type. The Temkin model also shows a B_T value of 22.801 which explains the adsorption that occurred in the chemical interaction. This research provides new information regarding the use of zeolite particles of 40-μm size for the adsorption process, especially in dye wastewater.

Acknowledgements

We acknowledged RISTEK BRIN, Grant Penelitian Terapan Unggulan Perguruan Tinggi and Bangdos, Universitas Pendidikan Indonesia.

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