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OPTIMIZING OF COMPETITIVE ADSORPTION METHYLENE BLUE AND METHYL ORANGE USING NATURAL ZEOLITE FROM ENDE-FLORES

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ABSTRACT

Waste from the textile industry is considered a potential source of environmental pollution, especially water, because it contains dangerous dyes. In this research, natural zeolite is an effective and efficient alternative to overcome pollution caused by methylene blue and methyl orange dyes. Activation of natural zeolite was carried out with 3 M HCl solution and was characterized using X-Ray Diffraction (XRD) and Scanning Electron Microscope (SEM). Simultaneously, the methylene blue and methyl orange adsorption processes were studied in variations in the weight of the adsorbent, contact time, and pH. The concentration of dyes left in the solution was observed with UV-Vis Spectrophotometer. The results showed that the adsorption capacity of natural zeolite in absorbing methylene blue was 21.189 mg / g and methyl orange was 18.208 mg / g. The optimum conditions of dyes adsorption are achieved with successive adsorbent weights 0.3 g and 0,4 g, following contact times 60 minutes and 90 minutes, and successive pH 6 and 2. The adsorbent weight factor, contact time, and pH affect the adsorption of dyes by natural zeolite from Ende.

Keywords: *natural zeolite, activation, adsorption, methylene blue, methyl orange*

INTRODUCTION

One of the leading industries being developed in Indonesia is the textile and garment industry. This industry has been able to reduce unemployment in Indonesia because it is a labour-intensive industry that absorbs many workers. Also, the textile industry plays a role as a contributor to foreign exchange [1]. However, the advancement of the textile industry can harm the environment if it is not balanced with the treatment of dye waste during the production process.

Liquid dye waste from the textile industry will make a significant contribution to environmental pollution [2].

The use of synthetic dyes in this industry is an alternative strategy because it is more stable than natural dyes, is easy to obtain, and is relatively inexpensive [3]. Organic dyes such as methylene blue and methyl orange are the most widely used dyes. The absorption ability of the fabric to dye is around 80-85%, and the rest will be lost in the washing process. Synthetic dye waste released into water will be a great danger to humans and the environment because of its carcinogenicity and even toxicity and is difficult to degrade by microorganisms [4].

Several methods have been developed during the last few years to control and remove dye waste present in waters, such as ultrafiltration, coagulation, biological treatment, electrochemical, and adsorption [5-8]. Among the developed methods, adsorption was chosen as the most potential method because of its easy operation, high efficiency and low cost, and it does not cause harmful side effects [9].

Adsorption is the process of absorption of a molecule (adsorbate) on the surface of another substance (adsorbent) due to the attractive force between the two substances [10]. The adsorbent innovation being developed is a natural mineral resource with high absorption capacity, is easily available at low cost, and has abundant availability. These criteria exist for natural zeolite [11].

Zeolite is an aluminosilicate mineral in the form of tetrahedral TO_4 (T = Al, Si), which is hydrated in alkaline and alkaline earth metals [12]. Natural zeolites scattered in Ende district - NTT are around 20 million tonnes. This large enough reserve can be maximally utilized for environmental purposes [13].

Utilization of natural zeolite Ende as a waste adsorbent in liquid has been carried out, and the results show that the quality of natural zeolite Ende as adsorbent is not different from the zeolite commonly used in research originating from the Bayah and Cikalong areas [14]. Anion dye adsorption study (methyl orange) using zeolite NaA / CuO showed that the adsorption efficiency of zeolite against waste reached 98% [15].

To further optimize the utilization of natural zeolite in Ende, an in-depth study of its potential and characterization as an adsorbent is needed. Natural zeolite has a large Si / Al ratio but still contains impurities in the form of metal oxides so that the surface area is low. An activation treatment is needed in order to increase its adsorption ability.

Based on the above background, it is necessary to research to obtain data on the characterization of natural zeolites in Ende. Besides, Ende's natural zeolite adsorption ability needs to be known not only for cation dyes (methylene blue) but also for its adsorption ability against anion dyes (methyl orange). This research is also an effort to overcome pollution caused by textile dye waste.

METHODS

1. Materials

The materials used in this research were the natural zeolite from Ende - Flores, HCl p.a, HCl p.a Merck 38%, aquades, universal pH indicator, methylene blue Merck 115943, dan methyl orange C. I. 13025.

2. Instrumentation

The instruments used in this study were the Kokusan H-107 centrifuge, Titramax 101 shaker, Shimadzu AA-7000 SSA (Atomic Absorption Spectrophotometer), Shimadzu 1700 UV-Vis spectrophotometer, Scanning Electron Microscope (SEM) Carl-Zeiss

Bruker EVO MA10, X-Ray Diffraction (XRD) Bruker D4, analytical balances, furnaces, ovens and glassware.

3. Procedure

a. Preparation of Sample Natural Zeolite

Natural zeolite samples were resized into a fine powder with grain sizes passing a 200 mesh grain size, the pH neutralized with distilled water, then heated in an oven at 105 ^{o}C for 3 hours, and stored in a desiccator for further use.

b. Activation of Natural Zeolite

Natural zeolite samples are chemically activated. Chemical activation was carried out by mixing 50 grams of zeolite in HCl 3.0 M. The mixture was stirred with a magnetic stirrer for 3 hours, then rinsed with aquades until the pH was neutral and dried in a furnace at 300 ^{o}C for 3 hours.

c. Characterization of Natural Zeolit

Natural zeolite characterization was analyzed using X-Ray Diffraction (XRD) instrument, and the surface morphology of natural zeolite was analyzed by instrument Scanning Electron Microscope (SEM).

d. Preparation of Standard Solution Methylene Blue and Methyl Orange 1000 mg/L

0.5 g of methylene blue powder and 0.5 g of methyl orange were put into a 500 ml measuring flask and diluted to the limit mark.

e. Determination of Wavelength for Absorption of Methylene Blue and Methyl Orange

The determination of wavelength is done by measuring the absorbance of standard solutions in the range of 650 - 700 nm for the wavelength of methylene blue and 300 - 600 nm for the wavelength of methyl orange.

f. Preparation of Standard Curves for Methylene Blue and Methyl Orange

Standard series solution of methylene blue and methyl orange with a concentration of 1; 2; 3; 4; 5; and 6 mg/L made from the liquor through a dilution process. The absorbance was measured at the maximum wavelength.

g. Determination of the Optimum Weight

Adsorbent of zeolite with a weight variation of 0.1; 0.2; 0.3; 0.4; 0.5; 0.6 g is added in 20 mL of dyes with a concentration of 400 mg / L, the solution is shaken for 1 hour . The solution was centrifuged for 15 minutes at a speed of 5000 rpm. The absorbance of the dyes was determined using a UV-Vis spectrophotometer.

h. Determination of the Optimal Contact Time

The optimum weight obtained was added in 20 mL of methylene blue and methyl orange with a 400 mg / L concentration, then shaken with time variations of 30, 60, 90, 120, 150 minutes. The solution was centrifuged for 15 minutes at a speed of 5000 rpm. The absorbance of the dyes was determined using a UV-Vis spectrophotometer.

i. Determination of pH Optimum

The optimum weight obtained was added in 20 mL of methylene blue and methyl orange with a 400 mg / L concentration, then shaken at the optimum time. The solution was centrifuged for 15 minutes at a speed of 5000 rpm. The absorbance of the dyes was determined using a UV-Vis spectrophotometer.

j. Determination of Adsorption Isotherms

The optimum weight obtained was added in 20 mL of methylene blue and methyl orange with various concentrations of 100, 200, 400, 600, 800, 1000 mg/L, then shaken at the time, and the optimum pH was obtained. The solution was centrifuged for 15 minutes at a speed of 5000 rpm. The absorbance of the solution was measured using a UV-Vis spectrophotometer at the maximum wavelength obtained.

The adsorption capacity can be determined using the formula:

$$qe = \frac{V\,(C_o - C_e)}{m} \qquad (1)$$

Where:

qe	=	adsorption capacity (mg/g)
V	=	volume of solution (L)
C_o	=	initial dye concentration (mg/L)
C_e	=	finish dye concentration (mg/L)
m	=	mass of zeolite (gram)

RESULTS AND DISCUSSION

1. Preparation of Sample Natural Zeolite

The initial preparation is done by refining the zeolite to produce a fine powder measuring 150 mesh. This process aims to uniform the granules and enlarge the surface to optimise the zeolite's adsorption capacity. Washing and heating are carried out in the preparation stage to remove impurities and evaporate the water present to increase the pore size [16].

2. Activation of Natural Zeolite

Natural zeolite still contains water vapour and metal oxides, causing low adsorption and ion exchange capabilities. The quality of natural zeolites can be improved through the activation process. This process aims to remove absorbed metal oxides and cover the zeolite surface so that the contact area becomes the largest. The addition of the contact area can increase the ability of zeolite as an adsorbent [17].

3. Characterization of Natural Zeolite

XRD is a qualitative and quantitative analysis method that functions to analyze the structure of zeolite powder. When analyzed by XRD, all materials containing certain crystals will produce distinctive peaks. The diffractogram of the results of natural zeolite analysis is shown in Figure 1

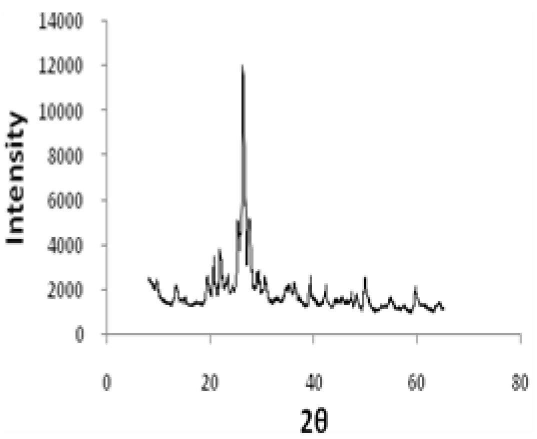

Fig 1. Diffractogram of Natural Zeolite

XRD analysis showed that the natural zeolite of Ende was a mixture of clinoptilolite and mordenite. The characteristic peaks evidence this in the JCPDS (Joint Committee on Powder Diffraction Standard) data with high intensity appearing at 25.60°, 26.25°, and 27.67° for mordenite and clinoptilolite intensity appearing at angles of 9.74°, 13.38°. And 29.07°. The peaks with the highest intensity are owned by mordenite, and this indicates that mordenite is a type of natural zeolite with a large abundance scattered in Ende.

The morphology of the crystal particles was observed by SEM (Scanning Electron Microscope) at a magnification of 3000 times. Figure 2 shows the surface morphology of the natural zeolite in Ende. The material shows the natural zeolite indicator based on SEM observations in the form of flat sheets shaped like bars with stacked and random arrangements [18].

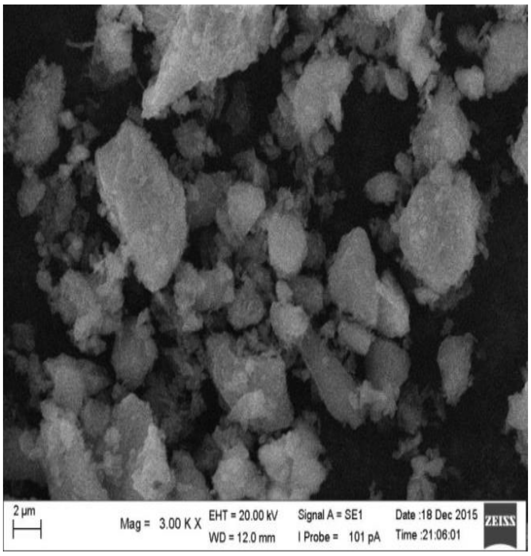

Fig 2. Surface morphology of natural zeolite

4. Determination of the Maximum Wave-length for Dyes Absorption

The concentrations of dyes can be qualitatively compared from the intensity of the color that will fade when the adsorption process ends. Quantitatively, the determination is measured by a UV-Vis spectrophotometer in the wavelength range of 600 - 700 nm for methylene blue and 400 - 550 nm for methyl orange. The maximum wavelengths of methylene blue and methyl orange were obtained at 664 nm and 463 nm.

5. Determination of the Optimum Weight

The adsorbent weight used ranges from 0.1 to 0.6 g. The effect of adsorbent weight on the adsorption capacity of zeolite on methylene blue and methyl orange is shown in Table 1.

Table 1 Adsorption capacity of natural zeolite against methylene blue (MB) and methyl orange (MO) at various adsorbent weight

Adsorbent Weight (g)	Adsorption Capacity (mg/g)	
	MB	MO
0.1	4.817	3.935
0.2	6.913	4.093
0.3	12.454	7,178
0.4	11.761	10.514
0.5	9.483	9.205
0.6	8.013	8.184

Based on Table 1, the optimum adsorbent weight for methylene blue uptake is 0.3 g. The optimum adsorbent weight for methyl orange uptake is 0.4 g. The optimum point occurs because the more adsorbent is used, the more surface area (active site of the zeolite) is, so the greater the possibility of adsorption. However, after the optimum weight, there is a decrease in the dye content that is absorbed. This decrease is because not all active sites are filled with an adsorbate. Large numbers of active sites take longer to reach an equilibrium state [19].

The highest adsorption capacity is obtained when all active sites of the adsorbent have been filled with an adsorbate. In the methylene blue and methyl orange adsorption process, the highest adsorption capacity was obtained with a small adsorbent weight. The adsorbent weight can increase the active side, thus allowing the active side to not interact with the adsorbate and cause a decrease in adsorption capacity [20]. Therefore, in the results of this study, the sample with high adsorbent weight did not produce a high adsorption capacity.

6. Determination of the Optimal Contact Time

Determining the optimum contact time aims to determine the time required to achieve the adsorption equilibrium of methylene blue and methyl orange by natural zeolite adsorbent. Generally, the longer the contact time between the adsorbent and the adsorbate, the more adsorbed the adsorbate will be. The zeolite adsorbent in adsorbing methylene blue and methyl orange increased respectively at 60 minutes and 90 minutes (optimum contact time), then decreased slightly in the next minute. Adsorbate so that the addition of adsorption time will only have a slight increase in the adsorption capacity or tend to be constant [21]. The effect of contact time on the adsorption capacity of natural zeolites on methylene blue and methyl orange is presented in Table 2.

Table 2 Adsorption capacity of natural zeolite against the dyes at various contact times

Contact Time (minute)	Adsorption Capacity (mg/g)	
	MB	MO
30	11.825	9.467
60	12.978	9.713
90	11.891	10.815
120	11.675	10.014
150	11.228	9.624

In determining the optimum contact time, it is necessary to know that the longer the adsorption time, the electrical stability will be disturbed. The adsorption capacity after the optimum contact time tends to decrease [22].

7. Determination of Optimum pH

One of the important parameters that determine the uptake of dyes by natural zeolite is the degree of acidity (pH). Changes in pH can affect the chemical and surface properties of the adsorbent, the solubility of the adsorbate, and ion competition in the absorption process [23]. The absorption of natural zeolite to methylene blue increased at pH = 6, 13.298 mg/g. The increase in the adsorption capacity of methyl orange occurred at pH = 2 (optimum pH) of 11.427 mg/g. This indicates that the dye adsorption process is influenced by electrostatic interactions or ion exchange with H + ions on the active site of the zeolite. The decrease in adsorption capacity at higher pH is due to the condition that the active site of the zeolite is saturated [24]. The adsorption and absorption capacities of dyes at various pH are shown in Table 3.

Tabel 3 Adsorption capacity of natural zeolite against the dyes at various pH

pH	Adsorption Capacity (mg/g)	
	MB	MO
2	11.407	11.427
4	12.823	10.325
6	13.298	10.011
8	13.014	9.763
10	12.115	8.728

The bond between zeolite adsorbent and dye adsorbate in this study is the Van der Waals bond. Van der Waals bonds are defined as the attractions between molecules due to dipole-dipole attractions. Dipolar molecules combine with neighbouring molecules until the negative pole of one molecule approaches the positive pole of another molecule. Natural zeolites contain a certain amount of negative charge and positive charge. Adsorbate, which has a negative charge, will bind to the positive charge of natural zeolite, while adsorbate, which has a positive charge, will bind to the negative charge of natural zeolite.

8. Determination of Adsorption Isotherms

The relationship between the adsorbate concentration and the absorption rate on the adsorbent surface can be determined through the adsorption isotherm. The results obtained indicate that the increase in the initial concentration of dyes is followed by an increase in the amount of these substances adsorbed (adsorption capacity). Table 4 shows the adsorption capacity of natural zeolite at various initial adsorbate concentrations.

Table 4 Adsorption capacity of natural zeolite against the dyes at various initial concentrations

Concentration adsorbate (mg/L)	Adsorption Capacity (mg/g)	
	MB	MO
100	8.655	6.409
200	11.751	8.132
400	14.218	11.427
600	16.013	14.503
800	21.017	17.113
1000	21.189	18.208

The adsorption capacity of natural zeolite to adsorbed dyes increased the initial concentration of the adsorbate. Because the higher the adsorbate concentration, the more the number of dyes in the adsorbed solution.

The nature of the zeolite influences the zeolite's ability to absorb an adsorbate. Zeolite with chemical activation provides quality values that meet the standards. Impurities such as water vapour and other minerals that can interfere with the absorption process tend to be less so that the zeolite can adsorb more the dyes molecules. The excellent quality of natural zeolite is proven by testing methylene blue and methyl orange adsorption, which results in large adsorption capacity values [25]. The adsorption capacity of dyes reached the highest values, namely 21.189 mg/g and 18.208 mg/g, respectively.

The determination of the adsorption isotherm of dyes by natural zeolites was analyzed using two adsorption isotherm models, namely Langmuir and Freundlich models. The Langmuir isotherm is based on the adsorption of the monolayer on the homogeneous active site of the adsorbent, while the Freundlich isotherm describes the adsorption on heterogeneous surfaces. The linear form of the Freundlich equation is expressed in the equation:

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

Where: Ce = the adsorbate equilibrium concentration (mg/L), qe = the adsorption capacity at equilibrium (mg/g), Kf and n = the constants and the adsorption intensity. The linear form of the Langmuir isotherm equation is:

$$\frac{C_e}{q_e} = \frac{C_e}{Q} + \frac{1}{Qb} \quad (3)$$

Q = the maximum adsorption on the monolayer (mg/g) and b = the Langmuir constant, which is related to the binding (mL/mg) affinity. Also, b = a measure of the adsorption energy [26].

In the adsorption of methylene blue and methyl orange with natural zeolites, the Langmuir isotherm correlation value is greater than that of Freundlich. This shows that the adsorption process that occurs is monolayer (homogeneous) adsorption. Isotherm parameter values qm, Kf, n, and R^2 are presented in Table 5.

Tabel 5 Natural zeolite adsorption isotherm parameters for methylene blue and methyl orange

Parameters		MB	MO
Langmuir	q (mg/g)	21.189	18.208
	b (L/mg)	0.387	0.148
	R^2	0.998	0.992
Freundlich	K_f (mg/g)	9.252	7.951
	n	0.142	0.738
	R^2	0.805	0.615

The Langmuir isotherm dominates the dyes adsorption process. This indicates that the adsorption only occurs on one active site. When the molecule occupies an active site, there will be no further absorption. If the active site of the adsorbent is saturated with adsorbate, the addition of adsorbate concentration does not increase the number of dyes adsorbed [27].

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CONCLUSION

Ende natural zeolite can act as dye waste adsorbent by chemical activation treatment using 3 M HCl. The adsorption capacity of natural zeolite absorbs methylene blue reaches 21.189 mg/g, and methyl orange reaches 18.208 mg/g. The optimum conditions for the absorption of methylene blue and methyl orange occurred at the adsorbent weight of 0.3 g and 0.4 g. The contact time was 60 minutes and 90 minutes, and the optimum pH occurred at pH 6 and 2. Weight factor adsorbent, contact time, and pH simultaneously influence both methylene blue and methyl orange absorption.

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