

Similarity Dyes Removal from Wastewater by Coral Reef Waste as a Low-Cost

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Dyes Removal from Wastewater by Coral Reef Waste as a Low-Cost Adsorbent

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Abstract

Despite the positive impact of the rapid industrial growth in Indonesia, it has caused several problems. The non-biodegradable pollutant, such as reactive dyes that result from the textile industry, is harmful to the environment and human health. This contaminating agent should be removed from the waste before being disposed to the surrounding ecosystem. Adsorption is one of the simple and low-cost techniques to eliminate dye from the effluent. Waste from coral reefs is interesting to be explored as a dye-removing adsorbent because it is abundant in nature, cheap, and reusable. Therefore, this study aims to determine the adsorption performance of coral reef waste in removing several dyes, i.e., methylene blue (MB), remazol brilliant blue (RBB), disperse orange (DO), and vinyl sulfone (VS) from wastewater. The adsorption capacity was determined to evaluate the effectiveness of coral reef waste in removing the dyes at the isotherm model. Adsorption capacity and isotherm model were used to evaluate the effectiveness of this natural adsorbent. Based on the percentage removal and coefficient distribution value, the removal selectivity of RBB was the best, followed by DO, VS, and MB, respectively. In conclusion, coral reef waste is promising to be developed as a low-cost adsorbent for removing dyes from wastewater.

Keywords: adsorption, coral reef, dyes, low-cost adsorbent, wastewater.

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1. INTRODUCTION

The textile industry in Indonesia has been growing rapidly. In general, this industry uses dyes as an agent of coloring (Khodaie et al., 2013). About 10-15% of the dyes are missed from the fabric during the dyeing process and could pollute the water (Murugesan et al., 2007). Very small quantities of dyes (less than 1 ppm) in water are highly evident and unwanted (Robinson et al., 2001). Dyes are inert and challenging to biodegrade and decolorize when thrown into the waste stream. Hence, dyes in streams and rivers denote water pollution (Dulman & Cucu-Man, 2009).

Reactive dyes are widely used worldwide in industry, especially in textile, and could bind covalently with the fiber. These dyes are carcinogenic, harmful, and toxic to the organism (Ahmad et al., 2014). They also

carry a danger to aquatic organisms (O'Neill et al., 1999). One of the most commonly used dye water, methylene blue, is used for cotton and wood (El-Ashtoukhy & Fouad, 2015). It has some negative effects on the human being, such as respiratory disorders, and can cause diarrhea, gastritis, vomiting, and nausea (Chen et al., 1999). Remazol brilliant blue (RBB) is also widely used in the textile industry. It is one of the examples of reactive dyes which has the characteristics of light color, low energy consumption dyeing process, and high solubility in water (Baskaralingam et al., 2007). This reactive dye is highly carcinogenic and possesses toxicity to the organism. In vitro study of anthraquinone from RBB indicated its toxicity by inhibiting the mitochondrial adenine nucleotide translocation and oxidative phosphorylation. In addition, most of the dyes inhibited mitochondrial [14C] adenosine

diphosphate uptake in a partially competitive-noncompetitive manner (Chen et al., 1999). Therefore, the removal of such dyes from wastewater is needed to keep the environment.

Several treatments have successfully been conducted, such as precipitation, adsorption, flocculation, coagulation, reverse osmosis, sedimentation, electrochemical techniques, ozonization, membrane filtration, fungal degradation, and photodegradation to remove the dyes (Bhatt et al., 2012). Treatment with adsorption was found to be simple and cost-effective compared to other treatments. Some advantages of using this method are that it is relatively easy to degrade, can be used to remove different types of coloring, and can use natural material as an alternative low-cost adsorbent (Crini, 2006; Ho & McKay, 2003).

Lately, the development of cheaper and more effective adsorbents has been studied. Some materials, including natural materials, biosorbents, and waste materials from industry and agriculture, have been proposed by several workers as low-cost adsorbents (Crini, 2006). Indonesia has a lot of natural sources, especially from the sea. More than 70% of the area is filled by sea and can be explored. The coral reef is abundant and has the potential to be used as an adsorbent (Giyanto et al., 2017). Zubir et al. reported the chemical properties of coral from Bidong island and discovered that all corals contain calcium carbonate (CaCO_3) in the crystal form of aragonite structure (Ahmad Zubir et al., 2015).

Nevertheless, only 5.2% of coral reefs in Indonesia are considered healthy (Zamani & Madduppa, 2011). Therefore, the unhealthy coral reef can be used to study its ability to absorb dyes. It has been recognized that coral reefs can precipitate material from seawater. The presence of calcium carbonate in coral reefs can be used to reduce environmental pollution (Smith & Kinsey, 1976). Hopefully, this material can be utilized as an effective adsorbent with high capacity. Hence, a small amount of adsorbent could adsorb dyes effectively and easy to be degraded. Some experiments reported that eggshells and coral powder could be used to immobilize heavy metals as an alternative to CaCO_3 (Ahmad et al., 2012). However, the use of coral reefs to remove dyes from wastewater is still unknown.

This preliminary study applied coral reef waste adsorbent from *Goniopora columna*

to remove several dyes from aqueous solutions. *G. columna* is local to the western and eastern Indian Ocean and the western Pacific Ocean, including southeast Asia and Indonesia (Sheppard et al., 2014). Methylene blue (MB), remazol brilliant blue (RBB), disperse orange (DO), and vinyl sulfone (VS) were used in this experiment. The adsorption was evaluated based on the Langmuir and Freundlich adsorption isotherm models, the removal percentage, and adsorption capacity. The interaction of coral reefs with dye was recognized by scanning electron microscopy (SEM). This study hoped that knowing the potency of the coral reef would provide novel insight into the alternative adsorbent.

2. MATERIALS AND METHODS

Materials

Coral reef waste of *Goniopora columna* (Figure 1.) was collected from Rancabuaya beach, Garut, West Java, Indonesia. Methylene blue (MB), remazol brilliant blue (RBB), disperse orange (DO), vinyl sulfone (VS), sodium hydroxide 2N (Merck), ethanol 96% (Merck), filter paper (Whatman 42), and aquadest.

The equipment used in this study were mortar and stamper, martel, furnace, spectrophotometer UV-Visible (Thermo Scientific® type Genesys 10S Uv-Vis), centrifugator (Hettich Zentrifugen® type EBA 20), oven (Hammet®), magnetic stirrer (Thermo Scientific®), digital scales(OHAUS®), SEM JEOL® (JSM-6510A), and other glassware (Pyrex).

Preparation and Characterization of Adsorbent

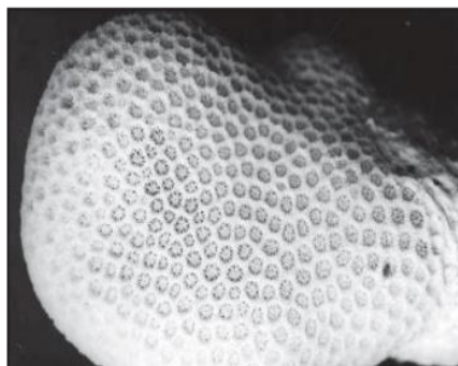


Figure 1. Coral reef of *Goniopora columna*.

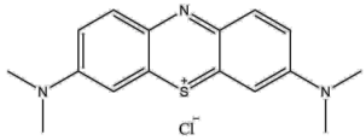
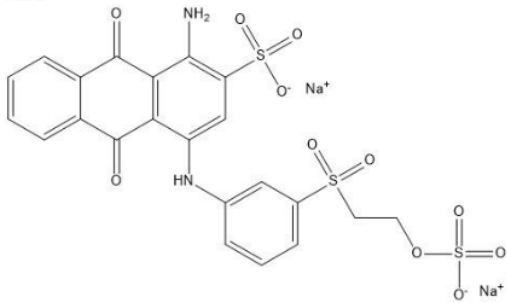
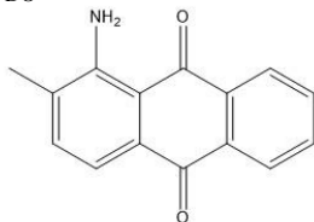
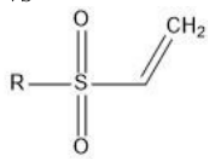
The impurities of coral reef waste were removed by washing the coral reef several times with distilled water, and then this coral reef was crushed mechanically using a grinder. This material was then passed through a mesh no. 100 to get a homogeneous particle size. The coral reef powder was activated using chemical and physical methods. First, the chemical method, the powder was treated with NaOH 2N at 100 °C for 120 minutes, and the pellets were neutralized to reach the neutral pH. After that, the physical method was dried at 60 °C and then calcined at 700 °C for 6 hours. SEM (JEOL® JSM-6510A) analysis

monitored the surface structure before and after sorption.

Preparation of Stock and Dye Solutions

Four dyes were used in this study, i.e., MB, RBB, DO, and VS. The chemical structures and their properties are shown in Table 1. The stock solution was prepared by dissolving 0.01 g of each dye powder into 100 mL distilled water to reach the final 100 mg/L dye solution concentration. Five different initial concentrations of each dye were prepared by dilution of stock solution.

Table 1. Chemical structures and the properties of dyes

Compound	Molecular weight	Melting point	Solubility in water
MB 	319.9 g/mol	100-110 °C	43.600 mg/L at 25 °C
RBB 	626.5 g/mol	305 °C	10 – 50 mg/mL at 21 °C
$C_{22}H_{16}N_2Na_2O_{11}S_3$ DO 	237.25 g/mol	205.5 °C	0.332 mg/L at 25 °C
$C_{15}H_{11}NO_2$ VS 	118.16 g/mol	-26 °C	100 mg/mL at 17 °C
$C_4H_6O_2S$			

Samples Analysis

UV-visible spectrophotometer (Thermo Scientific® type Genesys 10S UV-visible) was used to determine the concentration of the adsorbates. The maximum wavelength of MB, RBB, DO, and VS were 649, 590, 445, and 635 nm, respectively. To confirm the homogeneity and linearity over the concentration range used in this study, calibration curves for these dyes' concentrations were measured and produced.

Adsorption Experiment

The adsorption of MB, RBB, DO, and VS onto coral reef were studied at room temperature at different initial concentrations. The initial dye concentrations were set at 6, 5, 4, 3, and 2 mg/L for MB; 100, 90, 80, 70, and 60 mg/L for RBB; 60, 50, 40, 30, and 20 mg/L for DO; and 90, 80, 70, 60, and 50 mg/L for VS. For the adsorption experiments, 1.0 g of adsorbent and 100 mL of each initial dye solution was mixed in 250 mL Erlenmeyer flask on a thermal shaker at 25 °C with the velocity of 500 rpm. After 120 minutes, the suspensions were filtered, and the concentrations of the dye remaining in the supernatant were analyzed using a UV-visible spectrophotometer.

Adsorption Capacity

The adsorption capacity was calculated by measuring the amount of dye adsorbed per unit mass of adsorbent (q_e in milligrams of dyes per gram of adsorbent) using the equation 1.

$$q_e = \frac{C_o - C_e}{m} \times V \quad (1)$$

C_o is the initial dye concentration (mg/L), C_e is the equilibrium dye concentration (mg/L), m is the amount of the adsorbent used (g), and V is the volume of aqueous phase (L). The removal efficiency of dyes by adsorbent is examined in percentage as follow in equation (2), while the coefficient distribution is calculated using equation (3):

$$\text{Removal efficiency} = \frac{C_o - C_e}{C_o} \times 100 \quad (2)$$

$$K_d = \frac{q_e}{C_e} \quad (3)$$

Adsorption Isotherm Model

The Langmuir and Freundlich adsorption isotherm models were used to the experimental data for the uptake of MB, RBB, DO, and VS by the coral reef waste. The expression of the Langmuir isotherm model is:

$$q_e = \frac{kC_e b}{(1 + kC_e)} \quad (4)$$

C_e is the equilibrium solution phase concentration (mg/L), q_e is the equilibrium solid phase concentration (mg/g), k is the enthalpy-related constant, and b is the Langmuir isotherm sorption capacity (/mg). Plotting C_e/q vs C_e gives Langmuir isotherm, where $1/b$ is the slope and $1/kb$ is the intercept.

The Freundlich isotherm model is expressed in equations (5) and (6) in their non-linear and linear forms, respectively.

$$q_e = kfC_e^{1/n} \quad (5)$$

$$\text{Log } q_e = \text{Log } kf + \left(\frac{1}{n}\right) \text{Log } C_e \quad (6)$$

q_e and C_e were described previously, kf is the sorption capacity constant and n is the intensity constant. Plotting $\text{Log } q_e$ vs $\text{Log } C_e$ gives Freundlich isotherm, where the slope is the value of $1/n$ and intercept is equal to $\text{Log } kf$.

3. RESULTS AND DISCUSSION

It was known that the coral reef comprises calcium carbonate and can produce about 4 kilograms of CaCO_3 per square meter per year, especially in a shallow area that leads to the sea. The ability of the coral reef to precipitate material from seawater is high. It keeps pace with a rising sea (Giyanto et al., 2017; Smith & Kinsey, 1976). In addition, Ahmad et al. (2012) also found from FT-IR analysis that coral spectra appear centered between 1450 and 1500 cm^{-1} , and these bands match the ν_3 (asymmetric stretching of carbonate) mode (Ho & McKay, 2003). Coral reef produces CaCO_3 from the polyp part as forming the skeleton. This compound is required to protect the ecosystem (Ahmad et al., 2012). Calcium carbonate is amorphous and has three polymorphs, namely calcite, vaterite, and aragonite. The calcite is thermodynamically the most stable under ambient conditions (Smith & Kinsey, 1976). The ion of Ca^{2+} and CO_3^{2-} play a crucial agent in adsorbing the dyes. The charge of dyes

gives an ionic interaction on the surface of CaCO_3 . Zhao and Gao (2010) also reported that CaCO_3 could be used to remove acidic pink res B (APRB) dye solution (Shaw et al., 2015).

Percentage Removal of Dyes

This analysis was used to know the percentage of removal of different dyes from the adsorbent. Five initial and final concentrations were measured to evaluate the adsorption capacity. The list is shown in Table 2. For MB, the percentage removal decreased as the initial concentration decreased, and the average removal was the lowest of the other dyes. VS also had similar results with MB, the percentage removal decreased as the initial concentration decreased, but it has a better average percentage removal value.

In contrast, RBB and DO had different results. The percentage removal increased as the initial concentration decreased. Although for DO at an initial concentration of 20 mg/L, the removal percentage decreased. It was

suggested because the optimum concentration was between 30-40 mg/L. The most interesting thing is for RBB. The removal percentage was the highest and had a similar value in all concentrations. It was indicated that this adsorbent is good for RBB treatment. RBB is an anionic dye since it has an anthraquinone group (Lazim et al., 2015). The chemical structure of RBB consists of more reactive atoms than the others. It is suspected to be the reason that RBB could bind to the surface of the adsorbent.

The distribution coefficient (K_d) values were analyzed by comparing the dyes efficiencies. The distribution coefficient is used to compare the adsorption capacities of different dyes under the same experimental conditions. The ratio of dye concentration in the solid phase to that in the equilibrium solution after a specified reaction time was stated in the K_d value (Ahmad et al., 2012). A high value of K_d shows high solid-phase retention through adsorption and chemical reactions.

Table 2. The amount of removal percentage and the coefficient distribution for each added dye concentration.

	Initial conc. (mg L ⁻¹)	q _e (mg/g)	Removal (%)	K _d (L g ⁻¹)
Methylene blue	6	0.30	49.50	0.10
	5	0.26	52.60	0.11
	4	0.20	49.75	0.10
	3	0.11	37.67	0.06
	2	0.03	16.50	0.02
Remazol brilliant blue	100	9.11	91.05	1.02
	90	8.23	91.41	1.06
	80	7.34	91.80	1.12
	70	6.50	92.84	1.30
	60	5.58	92.95	1.32
Disperse orange	60	3.94	65.67	0.19
	50	3.67	73.46	0.28
	40	3.27	81.83	0.45
	30	2.41	80.23	0.41
	20	1.53	76.35	0.32
Vinyl sulfone	90	6.71	74.6	0.29
	80	5.73	71.61	0.25
	70	4.86	69.39	0.23
	60	4.04	67.38	0.21
	50	3.13	62.58	0.17

Nevertheless, a small value of K_d denotes that a high number of dyes remained in the solution. As shown in Table 2, the K_d value is comparable to removal efficiency. The highest value comes to RBB, with an average value of 1.16. Based on the percentage removal and coefficient distribution value, the selectivity of dye sorption onto coral reefs was $RBB > DO > VS > MB$. RBB has a large molecular weight and is also bulky based on the chemical structure. The interaction between $CaCO_3$ and the dyes might be through ionic interaction, and RBB contributed the most interaction due to its structure.

Equilibrium Adsorption Isotherm

The Langmuir and Freundlich adsorption isotherms model was used to

analyze the dye concentrations for the interpretation of equilibrium isotherm data. Generally, the interaction of adsorbents and adsorbate is described in the adsorption isotherm and thus is critical in optimizing the use of adsorbents. The Langmuir adsorption has been used successfully for many monolayer adsorption processes that assume the adsorption occurs at specific homogeneous sites within the adsorbent (Bulut & Aydin, 2006). In the Langmuir equation, see equation (4), the b value is the Langmuir constant related to the affinity of the binding site. At the same time, K_b represents a partial limiting adsorption capacity when the surface is fully covered with dye molecules which assists in the comparison of adsorption performance (Hui et al., 2005).

Table 3. The parameter used in Langmuir and Freundlich model.

Dyes	Langmuir model			Freundlich model		
	b (mg^{-1})	kb ($mg\ g^{-1}$)	R^2	n	kf	R^2
Methylene blue	-0.049	0.016	0.375	0.31	0.012	0.690
Remazol brilliant blue	19.493	1.867	0.972	1.60	2.304	0.990
Disperse orange	6.042	0.621	0.835	1.82	0.843	0.735
Vinyl sulfone	-2.014	0.067	0.898	0.31	0.001	0.950

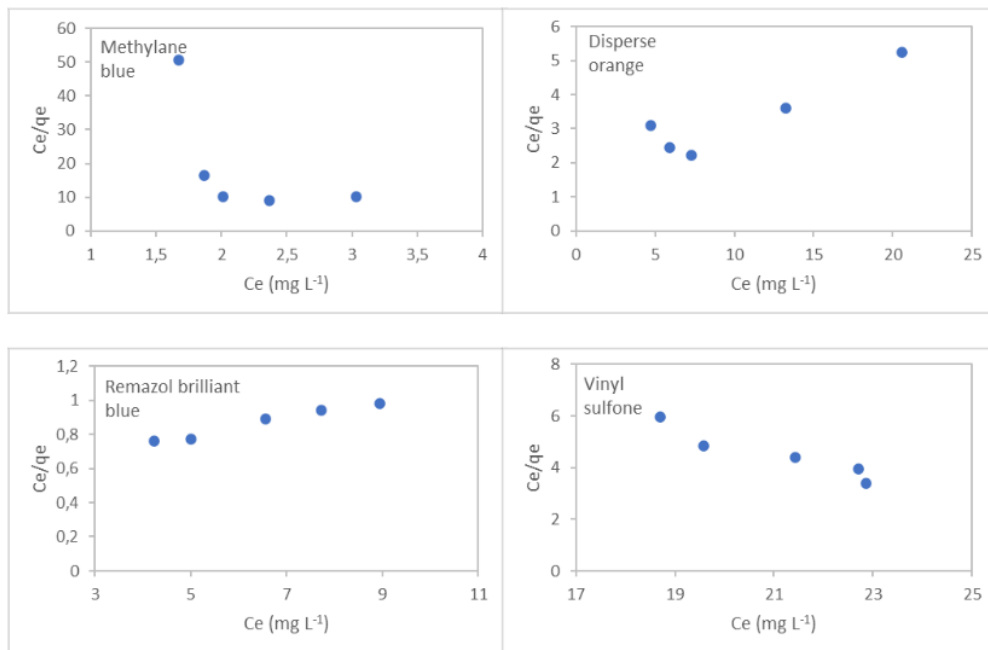


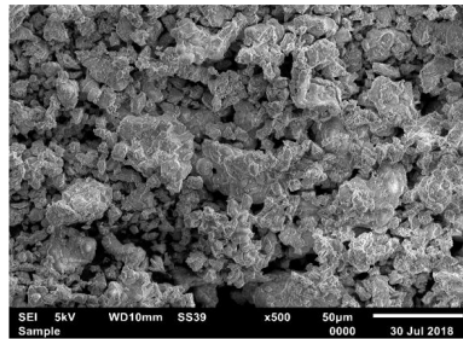
Figure 2. Langmuir sorption isotherms of dyes onto coral reef.

In explaining the heterogeneous systems, the empirical equation of the Freundlich isotherm is used. From this equation, see equations (5 & 6), K_f and n values roughly indicate the adsorption capacity of the adsorbent and the adsorption intensity, respectively. K_f is a Freundlich constant that shows the strength of the relationship between adsorbate and adsorbent, while the n value indicates adsorption favorability. The value of n is 1 to 10 and generally represents good adsorption (Vadivelan & Vasanth Kumar, 2005).

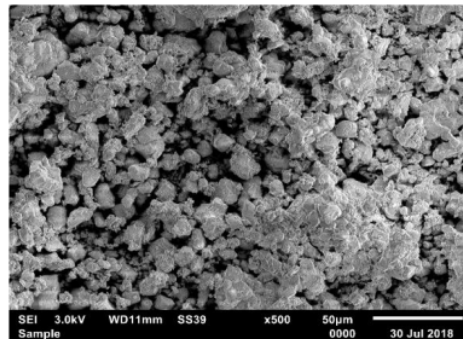
Table 3 shows the value of Langmuir and Freundlich adsorption isotherm. The b value of RBB is the highest, followed by DO. It was indicated that the affinity of RBB is stronger than DO, and the lowest affinity was reached to VS. It is worth noting that the RBB has a higher b and K_b value that indicates RBB follows the Langmuir model adsorption. Hence, the thread of dye sorption from this model was $RBB > DO > VS > MB$. The equilibrium isotherm curves in Figure 2. were plotted between the amount of dye adsorb from the single dye solution against the equilibrium concentration. The results showed that different types of dye give different types of adsorption. In addition, the n and K_f value of RBB from the Freundlich model is higher than others. It was indicated that coral reef has good sorption to RBB in both model adsorption isotherm.

Mechanism of Adsorption Process

In this study, SEM analysis was used to monitor the changes in the surface structure of coral reefs after the experiments. Figure 3 shows the surface structure before and after the adsorption experiment with RBB. It was shown that there were any changes in SEM analysis, indicating that something happened with the adsorbent. After the adsorption experiment, the particles looked tidier and uniform, flattened, and there were holes between one particle to particles. Based on the Langmuir equation model, the RBB has higher b and K_b values. Therefore, the dye might be bound to the particle's surface and thus create holes between particles.



(a)



(b)

Figure 3. SEM analysis of *G. columna* before (a) and after adsorbing Remazol brilliant blue (b).

4. CONCLUSION

Coral reef waste was used to remove some dyes solutions as a low-cost adsorbent. Based on percentage removal and coefficient distribution value, the selectivity sequence of dye adsorption onto coral reef occurred in the following order: $RBB > DO > VS > MB$. The high content of minerals is a major component of this adsorbent. Future studies are needed to evaluate the effectiveness of this adsorbent and some factors affecting it, including pH, temperature, reaction time, and adsorbent dose. These results indicated the potential usefulness of coral reef waste or coral-like adsorbents as a low-cost adsorbent, especially for removing RBB from the environment.

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