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Textile Dyes Removal by ZSM-5 from Bangka Kaolin

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Abstract. The ZSM-5- adsorption capacity, which is synthesized directly from Bangka kaolin, has been thoroughly to remove the basic dye in a textile waste solution. The Langmuir and Freundlich isotherm models were used to analyze and measure the results of experimental adsorption data from the red adsorption of Congo Red (CR), Auramin (Au), Methylen Blue (MB), Azin (Az), Rodamin B (RB) on ZSM-5 at a temperature of 40 ^{0}C. The ZSM-5 adsorption capacity of Bangka kaolin showed dye absorption : Congo Red, Auramin, Methylen Blue, Azin and Rodamin B respectively were 129,157, 134, 205 and 210 mg/g. Therefore, ZM-5 of Bangka kaolin is respectively good adsorbent to remove the textile base dye waste.

Keyword : Bangka-kaolin-ZSM-5, Textile-basic-dyes, Adsorption, Removal-dyes-wastewater

1. Introduction

Organic dyes and pigments are widely used in industry to dye their production materials, and produce waste [1]. The industry not only produces dye waste but is mixed with other materials such as dispersants, salts, emulsifiers and heavy metals. This mixture makes the dyestuff waste stable in aqous waste [2] and is generally considered to be highly toxic to aquatic systems or ponds. Even some of these dyestuff wastes are reported to cause a fatal effect on living and human bodies that intentionally or unintentionally consume [3].

Many dyes used in the textile industry, some of which are often used in are Congo Red for red dye, Auramin for yellow dye, methylen blue for blue, Azin for orange, Rodamin B for blue color. This dyes can be grouped dyes of diazo and xanthat compounds, this group dye is a water soluble compound with very high solubility (above 3.5 g / 100 mL). These dye compounds as crystals are stable in the form of acid or anionic and cationic salts [4]. The dye also causes significant toxicity to humans and animals that can cause burning eyes, irritation of the skin, gastrointestinal tract and respiratory tract. Based on the above analysis, the dye waste treatment is very important before being discharged to water bodies such as rivers or ponds [5], this is due to high solubility and its stability so that it is difficult to be processed in a simple way.

The usual treatment of the above textile dyestuffs is difficult to treat in order to obtain acceptable concentrations according to the existing rules for dye waste allowed to be fed into rivers or lakes. Special treatment should be used, usually by physics and chemistry methods, it is still very expensive and complicated to be widely used primarily for developing countries. Several commonly used processes are sonochemically degradation [6], photochemistry [7], electrochemistry [8], coagulation and flocculation [9], membrane separation [10], adsorption [11] and oxidation or ozonation [12, 13]. Some of these processes are an option which needs to find an appropriate economic process for removing the textile dyestuff. The adsorption process is a common and appropriate process to be developed into textile dyestuff processing and this process of recent years has attracted great attention. The application of several applications of cheap adsorbents for the processing of dyes and the removal of metal ions has been reviewed and offered [14]. Interestingly, the adsorption method has been commercially proven to be more economically efficient and feasible to remove textile dyes from wastewater.

In this study, ZSM-5 synthesis was prepared using Kaolin Bangka raw material without activation method with calcination which means the direct process of kaolin without using the template, the result is used as adsorbent for dyestuff waste, previously the result of ZSM-5 is characterized by scanning electron

microscopy (SEM), FTIR spectroscopy, BET surface area and XRD and further study of adsorption, kinetics and thermodynamic.

2. Material and Methods

2.1 Adsorbent preparation
ZSM-5 was synthesized directly from Bangka kaolin without pretreatment and without using organic template. The synthesis procedure of ZSM-5 was adopted from Hartanto, D., et al [15, 16], taking the recipe for Si/Al molar ratio was 60.

2.2 Adsorbate preparation
The dyes materials were used in this experiments are The dye Congo red [C.I name = Direct Red 28, chemical formula weight = 696.65, λmax-498nm]; Auramin; Methylen Blue; Azin and Rhodamine-B [C.I. name = Basic violet 10, chemical formula weight = 479.02, λmax-543nm]. The stock dyes solution have concentration are 500 mg/L, that was prepared by dissolving 0.5 g of dye powder in 1000 ml of distilled water. Different dilutions ware made any ranging from 60 to 250 mg/L, were then prepared using distilled water.

2.3 Adsorption Isotherm
The relationship between the amount of adsorbate absorbed by the concentration of adsorbate in solution at equilibrium and temperature conditions can be expressed by adsorption isotherm. In isothermal adsorption, in that circumstance the amount of adsorbate (molecule adsorbed) is a function of pressure (if its molecule in the form of a gas) or as a function of concentration at a constant temperature state.

The isotherm adsorption process can be understood through isotherms produced between the contact time (20-180 minutes) and the amount of adsorbate absorbed by the adsorbent mass unit at constant temperature (298 K). In the results of this study used experimental data analysis using Langmuir isotherm adsorption model, Freundlich,. The applicability and suitability of the isotherm equation to the equili-brium data were compared using the values of the correlation coefficients, R^2.

3. Results and Discussion

3.1 ZSM-5 from Bangka Kaolin
ZSM-5 synthesized from Bangka Kaolin and It were characterized using X-ray diffraction (XRD) with a Philips X'Pert Powder Cu Kα radiation source (λ = 1.54 Å) in the 2θ between 5-40 °[17]. Characteristic of XRD is show at fig 1, that representative samples of kaolin (a) and sample ZSM-5 result of synthesis with Bangka kaolin (b). Figure 1(a) shows the peak at 2θ about 12, 20-27 and 35-40 °. Base on Fig 1a. Kaolinite is the main content of the kaolin with it have height peak XRD pattern [18]. Fig 1.b did not indicate kaolin content again and that only have ZSM-5 crystal phase, it show difractogram peaks only XRD patter of ZSM-5 synthesized And the height 2θ pattern showed sharp peaks with high intensity. Therefore, crystallinity of ZSM-5 have height crystallinity.
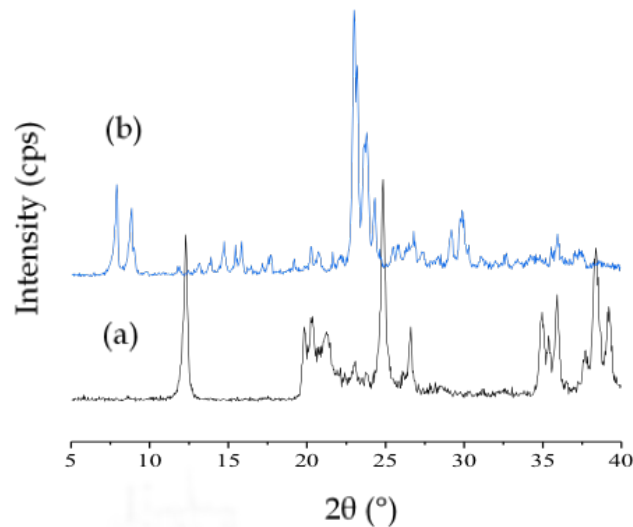
Figure 1. Difraktogram (a) kaolin and (b). ZSM-5 synthesis from Bangka

The surface area of ZSM-5 synthetic from Bangka kaolin calculation uses the BET (Brunauer, Emmet and Teller) calculation model with the assumption that the gas forms an infinite number of layers above a surface [19, 20]. Based on calculations using the BET model, the surface area of the catalyst sample is obtained. The result of isothermal adsorption of nitrogen gas at temperature 77K for determination of surface area of one zeolite with ZSM-5 Si/Al 50 shown that surface area a 333 m2/g and a pore diameter of 5.72 A and 20-28 A Figure 2.
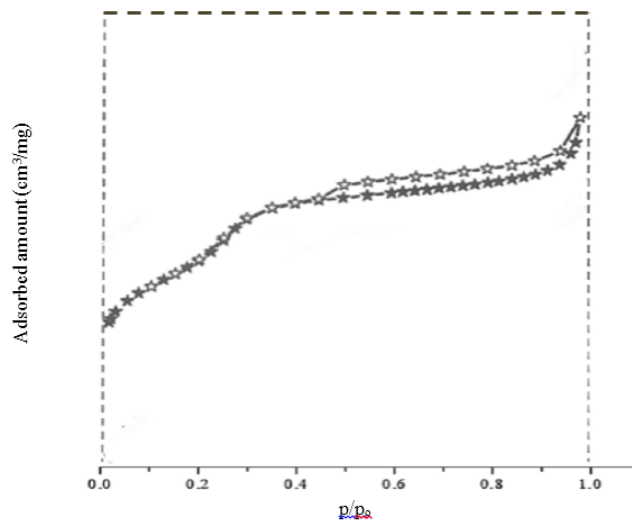
Figure 2. Pore and surface area ZSM-5 synthesis from Bangka kaolin

The surface area of ZSM-5 calculation uses the BET (Brunauer, Emmet and Teller) calculation model with the assumption that the gas forms an infinite number of layers above a surface [21, 22]. Based on calculations using the BET model, the surface area of the catalyst sample is obtained. The result of isothermal adsorption of nitrogen gas at temperature 77K for determination of surface area of one zeolite

with ZSM-5 Si/Al 50 shown that surface area a 333 m2/g and a pore diameter of 5.72 A and 20-23 A figure 2.

3.2 Batch Adsorption Studies

Effect of Initial dye concentration, loading catalyst and effect time contact. In order to determine the optimum initial concentration of the target dye, its value was varied (for both dye under study) in the range 50-150 mg/L, while keeping the temperature, adsorbent mass and the contact time constant at 303 K, 0.010 g and 180 min respectively. The effect of initial concentration on the removal of CR, Au, MB, Az and RB by the adsorbent is indicated in Table 1. It is evident from the table that percentage of CR, Au, MB, Az and RB removal decreases with increase in dye concentrations; however actual amount of the dye adsorbed is increased. This is due to increase in dyes concentration, surface area and active sites of the adsorbent were saturated and hence percentage removal decreases [23, 24].

The effect of contact time on the removal of CR Au, MB, Az and RB by the adsorbent is indicated in Figure 3. It is indicated that uptake of the dye is rapid in the beginning and then it becomes constant. The adsorption curves are single, smooth, and continuous leading to saturation and indicate the possible monolayer coverage on the surface of adsorbents by the molecules. In fig3. It give figure of height capacity of absorption adsorpt varying dyes diluted in water [25, 26, 27].

Table 1. The effect of initial concentration and loading of adsorbent

Parameter	Absorption Efficiency by intial concentration (%)			Amount of Absorbent (mg/g)			Absorption Capacity mg/g
Initial Concentration	50 mg/l	100 mg/l	150 mg/l	20 mg	30 mg	40 mg	Average
CR	98.2	84.0	65.3	194.5	120.0	194.5	129
Au	78.6	82.5	84.0	125	215	193	157
MB	57.0	59.1	59.5	85.0	235	285	134
Az	56.0	55.2	58.1	183	235	221	205
RB	50.0	59.5	58.0	150.0	197.5	435	210
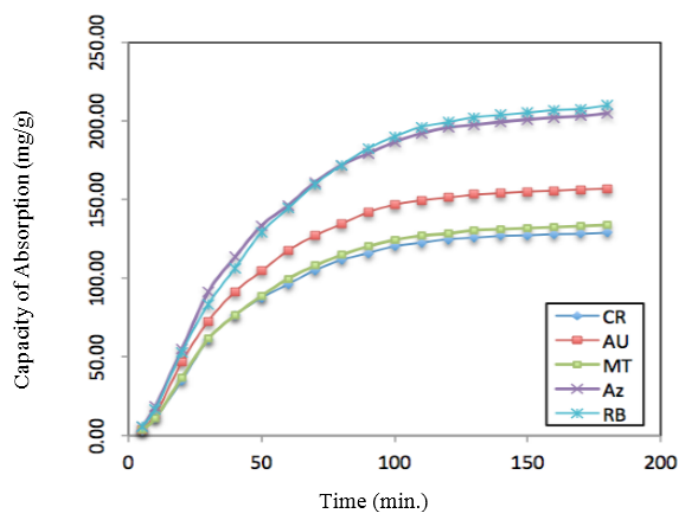
Figure 3. The Influence Time Absorption and Absorption Capacity

Table 2. Different Adsorption Isotherm model parameters for the   adsorption of CR, Au, MB, Az and RB on ZSM-5

Methods	Parameters	CR	Au	MB	Az	RB
Langmuir Isotherm	R^2	0.993	0.9971	0.9944	0.9984	0.9982
	K_L	0.009	0.007	0.008	0.005	0.004
	Q_m(mg/g)	129.201	157.21	134.21	205.33	210.21
Freundlich Isotherm	R^2	0.949	0.973	0,954	0.9873	0.9883
	K_f	13.6×10^4	4.9×10^6	8.1×10^4	8.1×10^4	4.6×10^6
	n	0.48	0.56	0.54	0.54	0.54

Thermodynamic parameters are important in adsorption studies; they provide a better understanding of the effect of temperature on the adsorption process. The thermodynamic parameters are calculation and the negative values of $\Delta H°$ obtained in the adsorption of both CR, Au, MB, Az and RB dye onto ZSM-5 signify that the adsorption process was exothermic in nature. It can be proven from the results of research conducted showed CR, Au, MB, Az and RB adsorption process using ZSM-5 decreased with increasing temperature adsorption [28, 29].

The $\Delta H°$ value was calculated to be -188,201 kJ/mol (CR), -132 kJ/mol (Au), -144kJ/mol (MB), -161kJ/mol (Az) and -161.414 kJ/mol (RB). In the other hand, the positive values of $\Delta S°$ indicate that increase in randomness occurred at solid-solution interface during the adsorption process. This indirectly shows the affinity of adsorbent toward dye molecules [30,31]. $\Delta G°$ values were negative at all temperatures studied, inferring that the adsorption was spontaneous in nature. While a higher negative value indicates that the process is energetically preferred over other adsorption mechanisms. Since the values of $\Delta G°$ decreased with increasing temperature, it suggests that at higher temperature, the driving force was less, resulting in lower adsorption uptake [32, 33].

4. Conclusion

The ZSM-5 adsorption capacity of Bangka kaolin showed dye absorption: Congo Red, Auramin, Methylen Blue, Azin and Rodamin B respectively were 129, 157, 134, 205 and 210 mg/g, the adsorption phenomena are allow Langmuir model. Therefore, ZSM-5 of Bangka kaolin is a relatively good adsorbent to remove the textile base dye waste.

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