

Adsorption of Remazol Brilliant Blue R Using Amino-Functionalized Organosilane in Aqueous Solution

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ABSTRACT

Synthesis of amino functionalized organosilane (AFOS) via UV-irradiation method has been conducted. In this research, the precursors for synthesizing AFOS are 3-glycidiloxypropyl trimethoxysilane (GPTMS) and ethylene diamine. Synthesis of amino functionalized organosilane was performed for 16 h monitored by Infra-red spectroscopy technique. The morphological structure of AFOS was observed by scanning electron microscopy. The adsorption process was conducted in the batch method using Remazol Brilliant Blue R (RBBR) as anionic dye model. In this study, the pH, contact time and dyes concentration were varied to determine the optimum pH, kinetic and isotherm adsorption. Based on the calculation, the kinetic rate of the RBBR adsorbed onto AFOS was determined by k_2 (pseudo second-order). Moreover, the isotherm study showed that the Langmuir model fitted for the adsorption of RBBR onto AFOS with Q_m by 21.3 mg g^{-1} .

Keywords: adsorption; dye; removal; organosilane; Remazol Brilliant Blue R

ABSTRAK

Telah dilakukan sintesis organo-silan terfungsionalisasi amin (AFOS) melalui metode irradiasi-UV. Pada penelitian ini, prekursor untuk mensintesis AFOS adalah 3-glisidiloksipropil trimetoksisilan (GPTMS) dan etilen diamin. Sintesis organo-silan terfungsionalisasi amin dilakukan selama 16 jam yang dipantau melalui teknik spektroskopi infra-merah. Struktur morfologi dari AFOS diamati menggunakan Scanning Electron Microscopy (SEM). Proses adsorpsi dilakukan menggunakan metode batch menggunakan zat warna remazol brilliant blue R sebagai model. Pada penelitian ini, pH, waktu kontak dan konsentrasi zat warna divariasikan untuk menentukan kondisi pH optimum, kinetika, isothermal adsorpsi. Berdasarkan perhitungan, laju kinetik dari adsorpsi RBBR pada AFOS ditentukan oleh k_2 (Pseudo second-order). Selain itu, studi isotherm menunjukkan bahwa model Langmuir cocok untuk adsorpsi RBBR pada AFOS dengan nilai Q_m sebesar 21.3 mg g^{-1} .

Kata Kunci: adsorpsi; penghilangan; zat warna; organo-silan; remazol brilliant blue R

INTRODUCTION

The human population growth rate increase every year [1]. These increases have an impact on the growing need for clean water. However, fresh water supply dwindles due to environmental pollution. It is the result of the industrial growth, particularly food and textile industry in developing countries [2]. Moreover, wastewater treatment technology is inadequate. Therefore, an effective, efficient and inexpensive material or waste water treatment technique for sustainability is urgently needed.

Dyes are used in several industrial sectors such as paint, coating, automotive, textile, food and many others. In the industrial sector, especially in textile usually employed synthetic dye. The synthetic dyes are difficult to remove than natural dye since it has good chemical structure stability [3-4]. Moreover, wastewater containing

synthetic dye has high toxicity for human and aquatic biota. Thus, it should be removed before disposal process [5]. Some conventional process for dye removal, such as coagulation [6], reverse osmosis [7], chemical oxidation [8], ion exchange [9], ozonation [10], electrochemical [11] are reported as high-cost method and technique and less efficient. Therefore, adsorption process is very favorable, since it does not need high technology, easy to operate, and relatively low cost. The synthetic dye is intensively used in textile industry, paper, ink, and leather [12]. One of the synthetic dye mostly consumed in the textile industry is Remazol Brilliant Blue R (RBBR). The RBBR synthetic dye, with chemical formula $\text{C}_{22}\text{H}_{16}\text{N}_2\text{NaO}_{11}\text{S}_3$, has excellent chemical stability make it difficult to degrade by the chemical or conventional process. This compound is categorized as an anionic dye, due to it has $-\text{SO}_3^-$ group. Some adsorbent employed by

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previous work such as Pineapple leaf and lime peel powder [13], mangosteen peel based activated carbon [14] and other waste natural resources to maintain RBBR in aqueous solution gave a positive impact on the environmental. However, they are less stable and required long contact time.

Material based silica has excellent potential to use as adsorbent since it has a porous structure, relatively stable in high temperature and can be designed. Kareem et al. [15] were utilized mesoporous silica for dye removal in aqueous solution. They reported that mesoporous silica has high adsorption capacity since the presence of pores and high surface area. Moreover, according to adsorption kinetic study, the adsorption process was categorized as chemisorption which it is more beneficial than physisorption. In our study, we used organosilane compound as the silica source. To improve chemical interaction between dye and adsorbent in aqueous solution, the amino functionalized compound is introduced. In our hypothesis, by introducing amines group will make the ionic interaction between RBBR and adsorbent more suitable.

3-glycidiloxypropyl trimethoxysilane, one of an organosilane compound, has oxirane ring as their active site for further modification. Opening oxirane ring is easy to achieve where Putzien et al. [16] applied room temperature for curing reaction. Several researchers have used reflux method to react the oxirane and amine compounds [17-18]. However, it has a disadvantage because of it required high energy. Another alternative is by using UV light irradiation as an initiator for opening oxirane ring. Based on our literature review, this method was not yet employed for the synthesis of amino functionalized organosilane. Moreover, the investigation of using these compounds for anionic dye removal in aqueous solution is not yet studied. Thus, our study focused on the synthesis of amino functionalized organosilane as synthetic-sorbent and determined their performance in dye removal.

EXPERIMENTAL SECTION

Materials

3-glycidiloxypropyl trimethoxysilane (GPTMS) was used as organosilane compound and obtained from Sigma-Aldrich, Singapore. Amine functional group compound was ethylene diamine (Merck). Toluene was purchased from Merck. Sodium hydroxide (NaOH, Merck) and hydrogen chloride (HCl, Merck) was used as a pH conditioner. Remazol Brilliant Blue R was purchased from dyes shop in Surakarta and used as anionic dyes without further purification.

Instrumentation

Fourier Transform Infra-Red (FTIR) IR-Prestige 21 was used as chemical analysis. Morphology of product was analyzed using Scanning Electron Microscopy (SEM) Vega3 LM Tescan. Uv-Vis PerkinElmer Lambda 25 Spectroscopy was used to determine adsorbent performance in dye removal.

Procedure

Synthesis of Amino-Functionalized Organosilane (AFOS)

Synthesis of AFOS compound was conducted via UV irradiation method with 365 nm of wavelength. About 1 mmol of GPTMS (0.24 g) was reacted with 2 mmol ethylene diamine (0.12 g) in the flask. Opening oxirane ring was initiated by UV 365 nm. In this study, the applied reaction time was 12 h with medium constant stirred. The final product was washed using toluene to remove unreacted GPTMS and ethylene diamine several times and dried in 60 °C for 12 h to complete the curing process. The product was crushed and sieved to pass 150 mesh sieve. The fine powder AFOS was restored in a desiccator before used.

Characterization

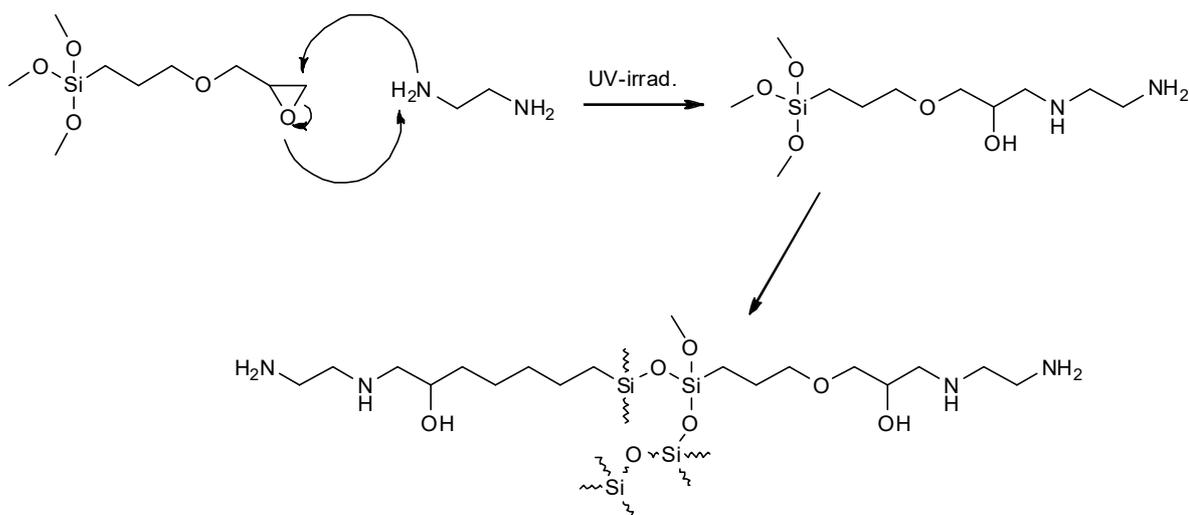
The AFOS compound was characterized using Fourier transform infrared (FTIR) spectrometer. The spectrum was recorded using IR Prestige-21 SHIMADZHU with the number of scans 45 times. The sample was prepared in pellet form by mixing the sample with KBr powder. Morphology of the sample was analyzed using scanning electron microscopy (SEM) Vega3 LM Tescan.

Anionic dye adsorption

Anionic dye adsorption effect of adsorbent amounts. About 20, 40, 60, 80 and 100 mg of AFOS was added into 10 mL of 50 ppm dye solution at pH 3. The dye removal process was carried out for 10 min and the amount of dye adsorbed was evaluated using PerkinElmer Lambda 25 UV-Vis spectrometer.

Effect of pH condition. Optimum absorption pH condition was performed by varying dye solutions pH for 3, 5, 7, 9 and 11. The pH is controlled by adding NaOH or HCl solutions. 20 mg of adsorbent is incorporated into 10 mL of pH conditioned dye solution and then shaken for 10 min. The absorption dyes were filtered immediately, then analyzed the concentration of dye adsorbed.

Isotherm and kinetic adsorptions. Concentrations of RBBR dyes were varied 10, 30, 50, 70 and 90 ppm which made from 1000 ppm standard solution. 20 mg of AFOS was added in 10 mL dye solutions then shaken



Scheme 1. Proposed chemical reaction in the amino-functionalized organosilane forming

using shaker machine for 10 min. After absorption process, dye solution was filtered immediately then analyzed using PerkinElmer Lambda 25 UV-Vis spectrometer. Isotherm adsorption (Langmuir and Freundlich models) was calculated using equation 1 and 2.

$$\frac{C_e}{q_e} = \frac{1}{K_L \times Q_m} + \frac{1}{Q_m} C_e \quad (1)$$

$$\ln q_e = \ln K_F + \frac{1}{n} \ln C_e \quad (2)$$

Analogue work was carried out to determine kinetic adsorption. About 20 mg of AFOS was added into 10 mL of 50 ppm dye solution and shaken for 2, 4, 6, 8, 10, 12, 14 and 16 min. The adsorbed Dye solution was analyzed using PerkinElmer Lambda 25 UV-Vis spectrometer. Absorption kinetic was evaluated following pseudo first-order and pseudo second-order using equation 3 and 4, respectively.

$$\log(q_e - q_t) = \log q_e - \frac{k_1 t}{2.303} \quad (3)$$

$$\frac{t}{q_t} = \frac{1}{q_e^2 k} + \frac{1}{q_e} t \quad (4)$$

RESULT AND DISCUSSION

Synthesis of Amino-Functionalized Organosilan

Synthesis of amino functionalized organosilane was performed via UV-Irradiation method. This method is a green approach where they did not use solvent and high energy. Putzien et al. [16] also used this method for curing reaction in oxirane opening ring. Proposed reaction mechanism of amino functionalized organosilane was showed by Scheme 1. Oxirane ring is a reactive group and makes it easy to react with other

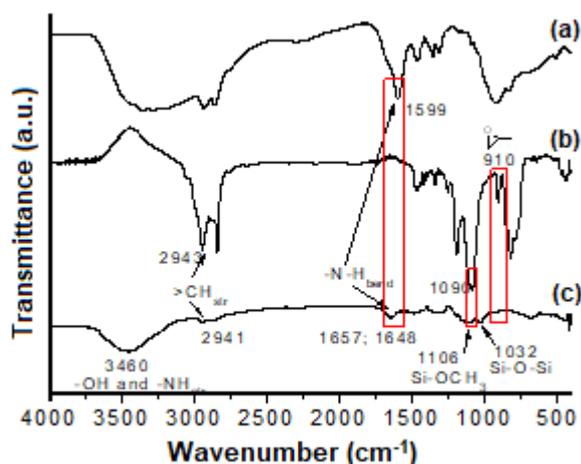


Fig 1. Infra-Red spectrum of (a) GPTMS, (b) ethylene diamine and (c) Amino functionalized organosilane

compounds. In this study, opening ring of oxirane group was initiated by radically through UV-Irradiation. When the ring has open, amine groups from ethylene diamine was easy to donor their electron and forming a good bond with a carbon atom in oxirane group of GPTMS showed in Scheme 1. During the oxirane ring opening reaction takes place, the formation of siloxane network of Si-O-Si also occurs. This is evidenced by the loss of infra-red characteristic absorption of oxirane ring at around 910 cm^{-1} , (Fig. 1). Moreover, the presence of a new absorbance band at around 1032 cm^{-1} corresponding to the siloxane (Si-O-Si) characteristic band has been observed. The appearance of this absorbance band due to polymerization has occurred between silane group and formed siloxane network. However, Si-OCH₃ typical absorbance band was also found but shift to 1106 cm^{-1} from their raw material at around 1090 cm^{-1} .

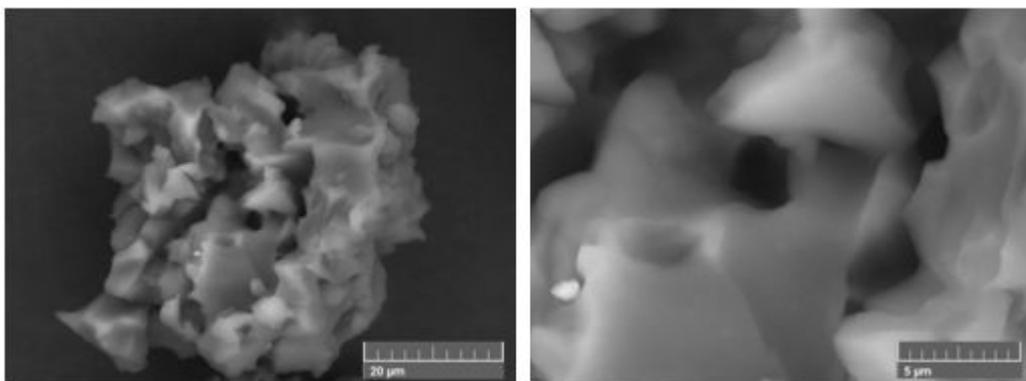


Fig 2. Surface morphology of amino functionalized organosilane

Ethylene diamine as the functionalized compound has an absorbance band at around 1599 cm^{-1} corresponding to amine bending vibration. After synthesis of amine functionalized organosilane compound, this absorbance band was split into two peaks at 1657 and 1648 cm^{-1} , due to the formation of secondary amine resulted by reacting of the primary amine group with oxirane group. The appearance of these both peaks evidenced that this compound has two amine groups, one is at the end of amine functionalized organosilane chain and the other one as a linker of ethylene diamine and an organosilane, see Scheme 1. A broad absorbance band at 3460 cm^{-1} corresponded to hydroxyl vibration overlapped with amine stretching vibration. The hydroxyl group is generated from opening oxirane ring reaction. In summary, it can be concluded that the proposed structure was strongly supported by FTIR data.

Morphological Study

Scanning electron microscopy (SEM) is a strong technique to morphologically analysis of materials surfaces structure and porosity [19]. Morphology of amino functionalized organosilane can be found in Fig. 2. Based on visual observation, it was clearly seen that this compound has pores in their structure. They have poor pores distribution, however, due to the irregular siloxane network. Moreover, their surface looks rough. It is very beneficial for dye adsorption where the surfaces roughness in adsorbent will make it easy to contact with another compound.

Effect of Adsorbent Ratio

The ratio of adsorbent influence on the dye removal. Based on literature review, the higher of adsorbent ratio increased effectivity to dye removal, since the increasing of sorption site in the adsorbent surfaces [13,20-21]. It was manifestly seen in Fig. 3, the addition of high ratio amino functionalized organosilane

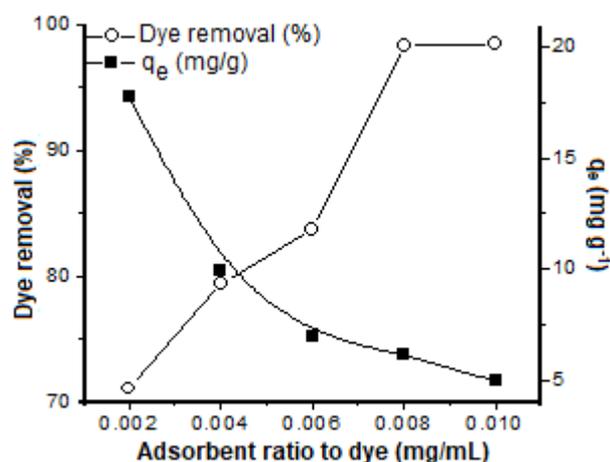


Fig. 3 Effect of adsorbent ratio to dye removal

compound has high dye removal percentages by 98.5%. The addition of 0.02, 0.04, and 0.06 g adsorbent slightly increased dye removal percentages by 71, 79, and 83%, respectively. However, the dye removal dramatically increased after addition of 0.08 g adsorbent (98%). However, in terms of adsorption capacity, the high value was observed by using a small amount of adsorbent (0.02 g). Moreover, It was observed that by using a small amount of adsorbent (0.02 g) able to remove more than 50% of dyes in aqueous solution with short contact time (10 min). Since economic reason, we decided to use 0.02 g of the adsorbent amount for another investigation, such as to determine optimum pH condition, contact time and isotherm adsorption.

Effect of pH

Solution state (in this case are pH) is significant influencing on adsorption process. In this study, the pH condition is varied by 3, 5, 7, 9, and 11. It can be seen in Fig. 5, best adsorption process was achieved at acid condition (pH = 3) and decrease by increasing of pH. However, the dye removal percentages increased again

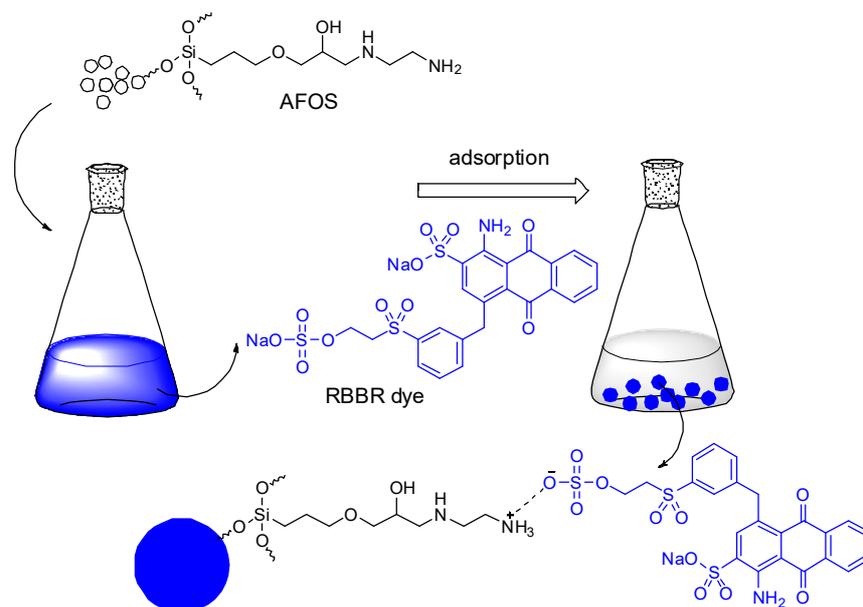


Fig 4. Illustration of RBBR dye interaction with AFOS compound

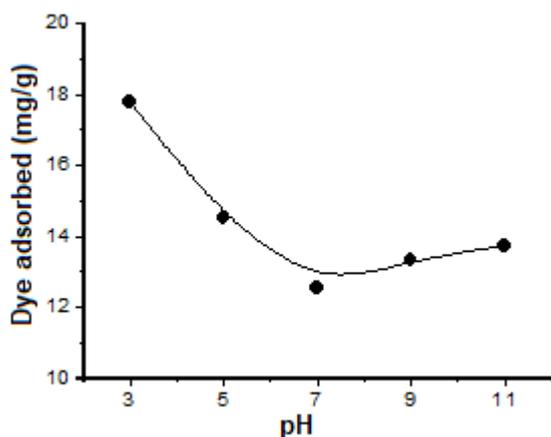


Fig 5. Effect of pH condition to dye removal

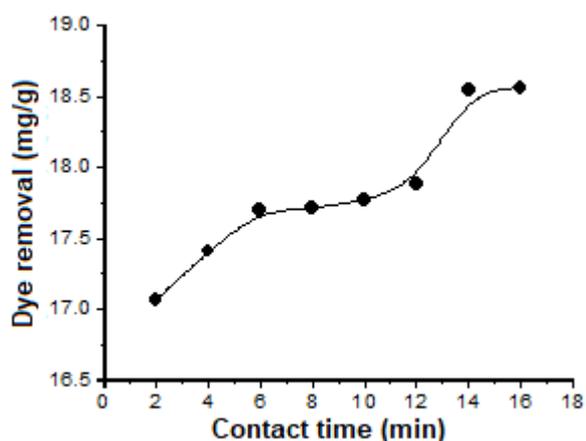


Fig 6. Effect of contact time on dye removal

when it reaches basic condition (pH = 9 and pH = 11), but it is not higher than an acid condition. At acidic condition (pH = 3), the amine group of AFOS was protonated by the presence of H^+ to form $-NH_3^+$. In this case, RBBR, an anionic dyes compound, is easy to ionic interact with the AFOS as illustrated in the Fig. 4. Consequently, the dye removal value increased. This phenomenon leads adsorbent more suitable to adsorb the dye. At basic condition, however, the presence of OH^- will make RBBR competing to ionic interact with the adsorbent. Thus, the dye removal value will not higher than in acid condition.

Effect of Contact Time

Contact time is an important factor in dye removal process. The dye adsorbed into amino functionalized

organosilane increased with the length of contact time, until it reaches an equilibrium state. Effect of contact time on dye removal was showed in Fig. 6.

The study of contact time to dye removal was conducted at short period over 16 min with interval 2 min. The dye adsorbed from 2 until 12 min was slightly increased and indicating it reach equilibrium phase [22-24]. Based on this study, more than 50% dye removal was achieved at the short time. It means that between adsorbent and dye has fast interaction. It is very beneficial to industrial field, especially textile industry due to the short time contact and small amounts of dye adsorbent. Thus, it has high efficiency to adsorb the dye, where at 2 min contact time was reach 68.29%, or there is 17.07 mg of dye adsorbed into 1 g of adsorbent. The short contact time has an impact on

economic value due it will reduce the operational cost to dye removal.

Adsorption Kinetic

The kinetic study is an important and useful technique to analyze dye adsorbed into adsorbent mechanism [25-26]. In this study, kinetic used pseudo first-order and pseudo second-order approach. It can be seen in Fig. S1 that adsorption of this study are followed to pseudo-second-order while the linier regression correlation coefficients (R^2) of pseudo-second-order models gave a value 0.998 and compared to pseudo first-order reach 0.881. It means that the adsorption mechanism was according to chemisorption. Chemisorption is very favorable than physisorption, due to dye-adsorbent interaction more suitable.

Table 1 shows the kinetic parameters for the adsorption of RBBR onto AFOS calculated from kinetic fit linear models. As summary, the k_1 and k_2 of the RBBR adsorbed onto AFOS were $7.37 \times 10^{-2} \text{ min}^{-1}$ and $14.7 \times 10^{-2} \text{ g mg}^{-1} \text{ min}^{-1}$, respectively. Since the adsorption of RBBR follows the pseudo second-order, the adsorption rate is determined by k_2 . The rate of adsorption was influenced by several factors such as the presence of an active site on the adsorbent surface, pore availability, adsorbent properties (acid or base), pH condition, and many other [27]. However, since the adsorption rate is determined by k_2 , the most influential factor on the adsorption of RBBR onto AFOS is the presence of active site such as amines group on the AFOS surfaces. Herald et al. [23] reported that there are four step processing of dye adsorbed into adsorbent, which is: (1) dye transport into adsorbent surface in aqueous solution, (2) diffusion of dye into adsorbent surface region, (3) intraparticle diffusion via adsorbent pores and (4) dye binding at adsorbent active site. In this study the amount of dye binding at adsorbent active site expressed as adsorption capacity (q_e) calculated from pseudo second-order fitting linier is 18.9 mg g^{-1} . This value agree with the experimental as shown in Table S2 in the supplementary data.

Adsorption Isotherm

Adsorption isotherm study was evaluated using Langmuir and Freundlich models approaches. The Langmuir model is used to describe homogeneous adsorption on mono-molecular sorption process [13,28-29]. Langmuir model will generate monolayer maximum adsorption capacity (Q_m) and Langmuir constant (K_L), in another side n and Freundlich constant (K_F) value are resulted by plotting Freundlich model. All of the data derived from Langmuir and Freundlich models were listed in Table 2. In this study, we employed

Table 1. Kinetics parameters for the adsorption of RBBR onto AFOS

Kinetic models	Parameters	Values
Pseudo first-order	k_1 ($\times 10^{-2} \text{ min}^{-1}$)	7.37
	R^2	0.881
Pseudo second-order	k_2 ($\times 10^{-2} \text{ g mg}^{-1} \text{ min}^{-1}$)	14.7
	q_e (mg g^{-1})	18.9
	R^2	0.998

Table 2. Langmuir and Freundlich isotherm parameters for the adsorption of RBBR onto AFOS

Adsorption isotherm	Parameters	Values
Langmuir	Q_m (mg g^{-1})	21.3
	K_L	1.78
	R^2	0.992
Freundlich	n	2.29
	K_F	4.13
	R^2	0.876

two adsorption isotherm such as Langmuir and Freundlich models, since they are most frequently used to study the adsorption isotherm. The plot curves of the models was shown in Fig. S2 (see supplementary data). According to the linier regression correlation (R^2), the best mechanism of RBBR adsorbed onto AFOS was fitted to Langmuir model indicated by R^2 of Langmuir higher than Freundlich. It means that the equilibrium adsorption of RBBR onto AFOS was appropriate to Langmuir model assumed the monolayer coverage of RBBR is accepted. This is correlated with kinetic study fitting to pseudo second-order where the chemisorption is favorable on the adsorption of RBBR onto AFOS. However, the RBBR covering AFOS to form multilayer probably occurs by considering the R^2 value of Freundlich plot models reaches 0.876. It is also supported by n value which equals to 2.29 ($n > 1$, favorable). Both Langmuir and Freundlich models could explain the isotherm adsorption of RBBR onto AFOS. However, the monolayer coverage strongly occurs in this study.

As mentioned before that the isotherm adsorption mechanism is strongly appropriate to Langmuir models. From the plotting curves of C_e versus C_e/q_e is also obtained the monolayer maximum adsorption capacity (Q_m). The Q_m value of RBBM adsorbed onto AFOS is 21.3 mg g^{-1} . The calculated Q_m value is not quite different with the experiment data. It means that the Langmuir model fit to describe the mechanism on the RBBR adsorption. Unfortunately, the Q_m value is much lower compared to previous work conducted by other researchers. However, it should be noted that this adsorbent require a short contact time among the listed references in this report [13,28].

CONCLUSION

The synthetic amino functionalized organosilane has been successfully synthesized which proved by chemical analysis. This compound has good potential as synthetic-sorbent due to the kinetic study showed that it followed the pseudo-second-order kinetic model which means the adsorption mechanism is chemisorption. Based on Langmuir and Freundlich adsorption isotherm model, it was summarized that the AFOS has Q_m by 21.3 mg g^{-1} and the adsorption process was categorized as favorable sorption due to the $n > 1$.

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