

Modification of SiO₂ from Rice Husk with Polyvinyl Alcohol (PVA) as Adsorbent of Congo red dye

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Abstract

Rice husk is an agricultural waste abundant in silica (SiO₂). SiO₂ extracted from rice husk has a porous structure, so it has the potential to be used as an adsorbent material. To increase the stability and adsorption capacity, SiO₂ was modified by adding Polyvinyl Alcohol (PVA) and used to removal Congo red dye. SiO₂ extraction was carried out through a calcination process at three different temperatures, namely 600, 700, and 800 °C. The XRD spectra show that the three SiO₂ extracted samples show an amorphous structure, where the smallest crystallite size is SiO₂ calcined at a temperature of 600 °C. Morphological analysis and elemental composition using SEM-EDX showed the presence of a polymer layer on the surface of the material, which functions as a distribution medium for SiO₂ particles. In addition, an increase in carbon (C) content was detected after the addition of PVA, confirming the presence of PVA in the composite. The optimum conditions for the adsorption process of Congo red dye by SiO₂-PVA were obtained at pH of 1, contact time of 75 minutes, and Congo red dye concentration of 75 mg/L. Experimental data showed that the Langmuir isotherm model was most suitable for describing adsorption behavior compared to the Freundlich model, with an adsorption capacity of 14.31 mg/g. These results indicate that SiO₂-PVA has the potential as an alternative adsorbent for the treatment of wastewater containing dyes.

Keywords: rice husk, SiO₂, adsorption, Congo red dye, isotherm

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Abstrak (Indonesian)

Sekam padi merupakan limbah pertanian yang kaya akan kandungan silika (SiO₂). SiO₂ yang diekstraksi dari sekam padi memiliki struktur berpori, sehingga berpotensi digunakan sebagai material adsorben. Untuk meningkatkan kestabilan dan kapasitas adsorpsi, silika dimodifikasi dengan penambahan *Polyvinyl Alcohol* (PVA) dan digunakan untuk menyerap zat warna Congo Red. Ekstraksi silika dilakukan melalui proses kalsinasi pada tiga suhu berbeda, yaitu 600, 700, dan 800 °C. Spektra XRD menunjukkan bahwa ketiga SiO₂ hasil ekstraksi menunjukkan struktur amorf, dimana ukuran kristal paling kecil adalah SiO₂ hasil kalsinasi pada temperatur 600 °C. Analisis morfologi dan komposisi unsur menggunakan SEM-EDX menunjukkan keberadaan lapisan polimer di permukaan material, yang berfungsi sebagai media distribusi partikel SiO₂. Selain itu, peningkatan kandungan unsur karbon (C) terdeteksi setelah penambahan PVA, yang mengonfirmasi keberadaan PVA dalam komposit. Kondisi optimum untuk proses adsorpsi zat warna Congo red oleh SiO₂-PVA diperoleh pada pH 1, waktu kontak 75 menit, dan konsentrasi larutan 75 mg/L. Data eksperimen menunjukkan bahwa model isotherm Langmuir paling sesuai untuk menggambarkan perilaku adsorpsi dibandingkan model

Freundlich dengan kapasitas adsorpsi 14.3 mg/g. Hasil ini mengindikasikan bahwa SiO₂-PVA memiliki potensi sebagai alternatif adsorben untuk pengolahan limbah yang mengandung zat warna.

Kata Kunci: sekam padi, SiO₂, adsorpsi, zat warna Congo red, isotherm

INTRODUCTION

Water contamination caused by organic contaminants is a significant environmental issue today [1,2]. Various industrial sectors such as textiles, paper, soap, pharmaceuticals, tanning, and paints contribute to this pollution, especially due to the use of dyes in their production processes [3,4]. Among the various dyes, azo dyes are the most commonly used, accounting for around 50% to 70% of the total dyes applied in industry [5,6]. These dyes have an aromatic structure interconnected through azo bonds (-N=N-), making them difficult to decompose naturally. It is estimated that 1–20% of the dyes used in the dyeing process are wasted as waste, and if this waste enters the environment without proper treatment, it can have a negative impact on aquatic ecosystems and human health [6].

Congo red dye is a dye frequently used in the textile industry. This dye belongs to the benzidine-based anionic diazo group and is known to have fast coloring ability, so it is often chosen in the textile production process [2]. However, Congo red dye has the potential to cause negative impacts on the environment and health because it can trigger allergic reactions, is converted in the body into carcinogenic benzidine, and is toxic to various organisms [7,8]. The disposal of wastewater containing Congo red dye must be managed meticulously to prevent adverse effects on human health and environmental sustainability.

Various techniques have been used for dye removal, such as coagulation-flocculation [9], filtration [10], photodegradation [11], ozone treatment [12], electrochemical advanced oxidation [13], and adsorption [14]. Among these treatment techniques, adsorption is the most widely used method for pollutant treatment due to its easy operation, low cost, and high efficiency [15]. However, the need for effective adsorbents from various natural and synthetic options increases the attractiveness of this technique in terms of cost-effectiveness and efficiency [16]. Various adsorbents have been used for dye adsorption, such as bentonite and hydroxyapatite [17], activated carbon [18], Chitosan [19], and SiO₂ [20].

Rice husk is one of the most promising natural materials for use as an adsorbent. Rice husk is available in large quantities, especially in agricultural countries. Rice husk consists of organic matter, such as cellulose and lignin, and mineral components, including SiO₂,

alkali, and other elements. The ash content ranges from 10 to 20%, mostly 87–97% SiO₂ [21]. The high content of SiO₂ in rice husk makes it a suitable raw material as an adsorbent [22]. The surface of SiO₂ has silanol groups (-OH), which play an important role in interacting with pollutant molecules through hydrogen bonds or electrostatic interactions.

Amorphous SiO₂ is more reactive than crystalline SiO₂, but its use as an adsorbent is still constrained by its brittle nature, agglomeration, instability under extreme conditions, and difficulty in separation and regeneration. Polyvinyl alcohol (PVA) as a doping is a promising alternative because PVA acts as a binding matrix that can increase the stability, mechanical strength, and sustainability of SiO₂ in solution. On the other hand, the presence of hydroxyl groups (-OH) in PVA allows interactions with pollutants and is environmentally friendly [23–25].

This study aims to synthesize SiO₂ modified with PVA for the removal of Congo red dye. Rice husk as agricultural waste is an abundant, cheap, and environmentally friendly source of SiO₂. PVA is a polymer that is easily degraded in the environment and is non-toxic. Modification with PVA improves adsorption efficiency. In addition, the silica-PVA composite is safer for the environment [26,27]. Adsorption was carried out using the batch method. The effects of solution pH, contact time, and Congo red dye concentration were studied to obtain optimum conditions for the adsorption process.

MATERIALS AND METHODS

Materials

The materials used in this study were rice husks taken from Tanah Bumbu district, South Kalimantan. Chemicals include Polyvinyl alcohol (PVA), Congo red dye, NaCl, NaOH, and HCl from Merck, Germany.

Methods

Preparation of SiO₂

Rice husks are cleaned of impurities. Then calcination is carried out at temperatures of 600, 700, and 800 °C for 5 hours. Calcined rice husks are ground and sieved with a size of 200 mesh.

Synthesis of SiO₂-PVA

A total of 10 g of SiO₂ was mixed with 10 g of PVA into 50 mL of distilled water, and the mixture was stirred with a magnetic stirrer at a temperature of 70 °C

until homogeneous (± 1 hour). Then dried in an oven at a temperature of 110 °C for ± 3 hours. SiO₂-PVA was characterized and used for the adsorption process.

Characterization of SiO₂-PVA

X-ray Diffraction (XRD Rigaku Miniflex 600 Japan) was used to analyze the crystallite structure of SiO₂-PVA. The morphology and elemental composition were determined using Scanning Electron Microscopy - Energy Dispersive X-ray Spectroscopy (SEM-EDX Tescan Vega 3). Congo red dye concentration analysis was performed using a UV-Vis spectrophotometer (Orion Aquamate 8000).

Determination of pH_{pzc}

A total of 0.2 g of the SiO₂-PVA was added to several 100 mL conical flasks, each containing 50 mL of a 0.1 M NaCl solution. The pH of the solution was adjusted within a range of 2 to 12 using 0.1 M HCl and NaOH. The mixtures were then agitated on a shaker at 120 rpm for 48 hours. The pH_{pzc} was identified from a plot of the initial pH versus the change in pH ($\Delta\text{pH} = \text{pH}_{\text{final}} - \text{pH}_{\text{initial}}$) [28].

Determination of optimum adsorption conditions

Three variables are used to optimize the adsorption of Congo red dye, namely solution pH (1–8), dye concentration (15–90 mg/l), and contact time (15–90 minutes). The Congo red dye solution volume used was 25 mL with a SiO₂-PVA weight of 0.02 g. The calculation of the absorption capacity uses the following formula:

$$q_e = \frac{V(C_0 - C_t)}{m} \quad (1)$$

C_0 represents the initial concentration of metal ions (mg/L), while C_t denotes the final concentration (mg/L), respectively. V refers to the volume of the solution (L), and m indicates the mass of the adsorbent used (g).

RESULTS AND DISCUSSION

Proximate Analysis of Coal Waste and Palm Kernel Shell

The calcination of rice husk is intended to eliminate organic matter and obtain silica (SiO₂). As shown in **Figure 1**, the SiO₂ produced at 800 °C appears whiter than that obtained at 600 and 700 °C. This is due to the initial transformation of the SiO₂ structure from amorphous to crystalline.

At low calcination temperatures, rice husks are brown or yellowish in color because they still contain organic matter such as lignin and cellulose. As the calcination temperature increases, these organic materials burn and decompose, so that the color of the

rice husks changes to white or light gray, indicating the formation of silica. If the calcination temperature is too high, there is a risk of sintering or changes in the silica crystal structure. Therefore, choosing the right calcination temperature is very important.

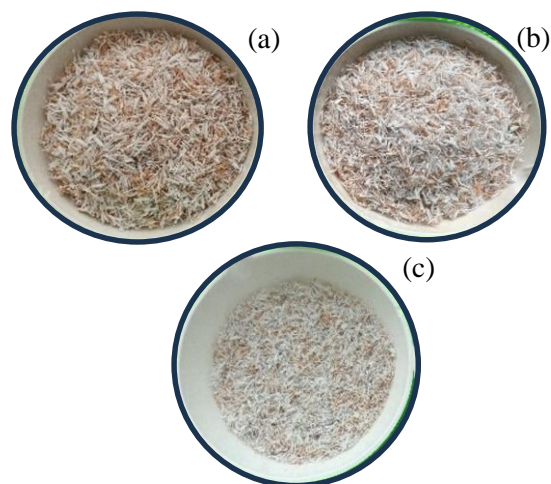


Figure 1. SiO₂ from calcination at temperature (a) 600, (b) 700 dan (c) 800 °C

Characterization of SiO₂-PVA

The XRD analysis of SiO₂ is presented in **Figure 2**. The calcination temperatures include 600, 700, and 800 °C. Extraction of amorphous silica from rice husk involves calcination or pyrolysis processes, usually in the 550–750 °C [29]. At temperatures <600 °C, SiO₂ generally remains in an amorphous (irregular) form. This structure tends to have a high surface area and is mesoporous. At high temperatures (>800 °C), SiO₂ can begin to crystallize, forming phases such as cristobalite or tridymite [21].

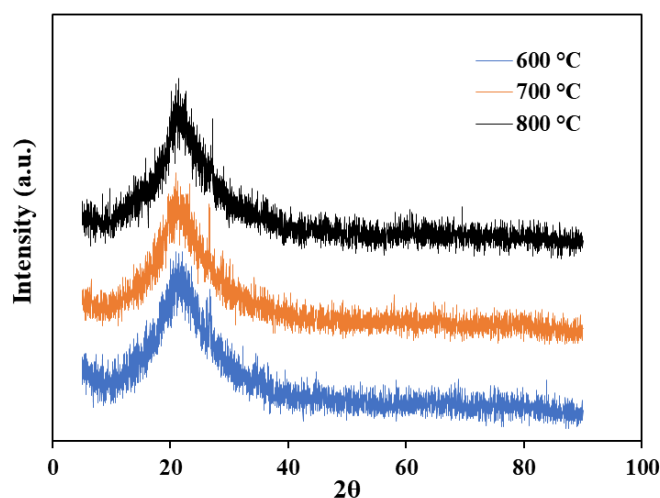


Figure 2. XRD spectra of SiO₂

The other study showed that at a temperature of 700 °C, there was a change from the amorphous phase to the crystalline phase [30]. This may be because the presence of metal oxides such as K_2O acts as a catalyst for the phase change reaction. The wide and high peaks are observed at the angle of $2\theta = 22.3^\circ$, which is the 002 crystal plane. Increasing the temperature can cause the SiO_2 particles to aggregate, which affects the particle size distribution. In addition, it can remove the

$-OH$ (silanol) groups on the SiO_2 surface, which reduces the reactivity on the surface.

The study demonstrates that SiO_2 derived from rice husk at calcination temperatures of 600, 700, and 800 °C retains an amorphous structure, exhibiting a broad peak in the region of $2\theta = 21-23^\circ$. Typically denotes quartz, specifically JCPDS No. 29-0085. This is further corroborated by the appearance of a peak at $2\theta = 26.6^\circ$.

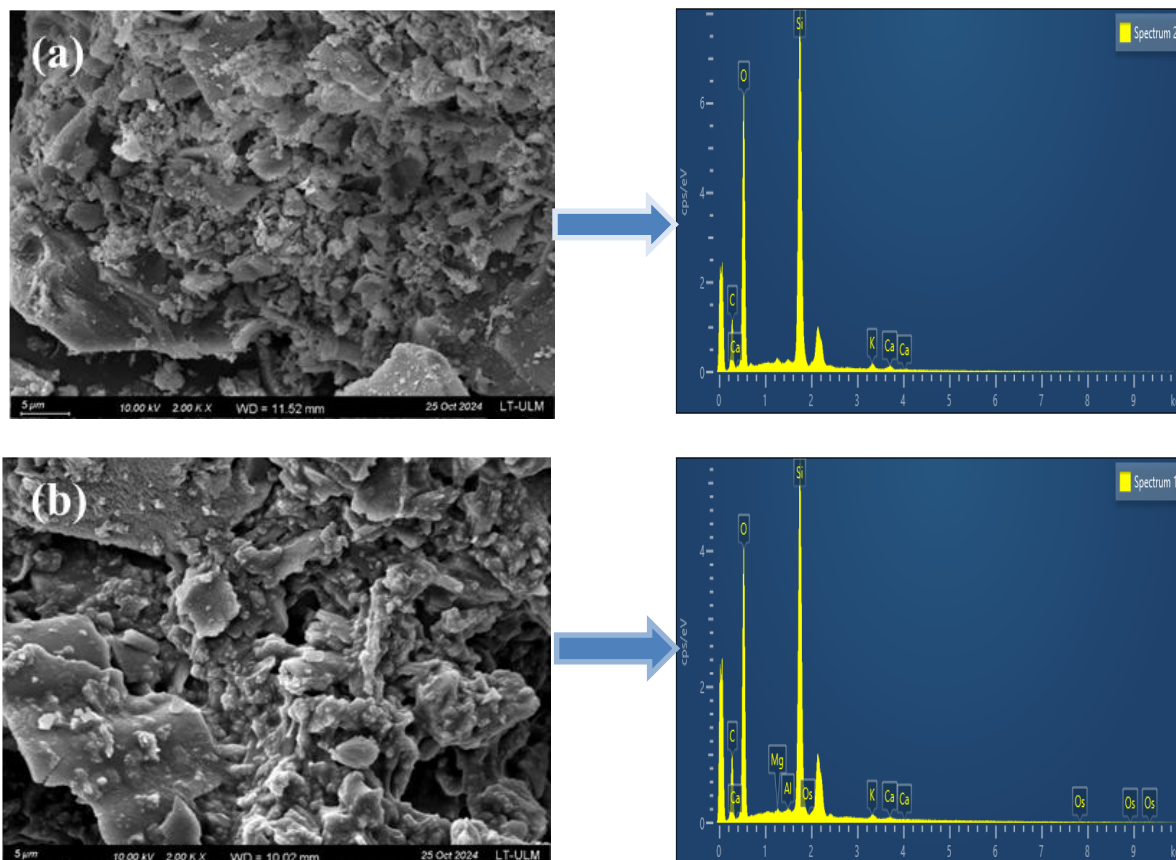


Figure 3. SEM-EDX of (a) SiO_2 and (b) PVA- SiO_2

The calculation of the crystallite size of SiO_2 using the Scherrer equation is presented in **Table 1**. SiO_2 at a calcination temperature of 600 °C has the smallest size. Increasing the calcination temperature encourages crystal growth through the reorganization of the atomic structure, which causes small particles to combine into larger particles. [21,30]. Furthermore, SiO_2 from calcination at a temperature of 600 °C is used for the adsorption process.

Figure 3 is the result of the analysis of the morphology and composition of the elements of SiO_2 and SiO_2 -PVA using SEM-EDX with a magnification of 2000X. The morphology of SiO_2 appears rough, slightly porous, and tends to form agglomeration, while SiO_2 -PVA shows a layer of polymer from PVA

and SiO_2 grains distributed on the surface. The composition of SiO_2 and SiO_2 -PVA is presented in **Table 2**. The presence of carbon signifies the existence of organic molecules within SiO_2 , suggesting an amorphous structure. The percentage of C in SiO_2 -PVA increases due to the addition of C from PVA. The addition of PVA also has an impact on reducing the percentage of SiO_2 . [30].

Table 1. Crystallite size of SiO_2

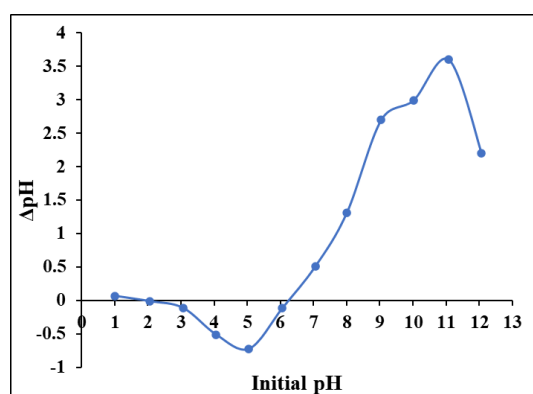
| Calcination temperature (°C) | Crystallite size (nm) |
|---------------------------------|--------------------------|
| 600 | 7.57 |
| 700 | 8.38 |
| 800 | 7.95 |

Table 2. Elemental composition of SiO₂ and SiO₂-PVA from EDX

| Elements | Weight (%) | |
|----------|------------------|-----------------------|
| | SiO ₂ | SiO ₂ -PVA |
| C | 27.99 | 32.61 |
| O | 43.33 | 41.17 |
| Si | 26.60 | 24.54 |
| K | 1.12 | 0.82 |
| Ca | 0.96 | 0.48 |
| Other | - | 0.38 |

Adsorption of Congo red dye using SiO₂-PVA

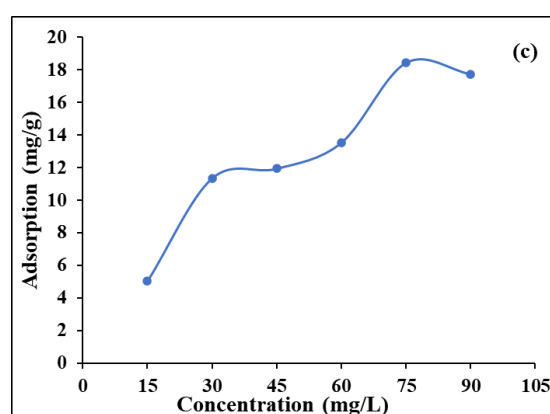
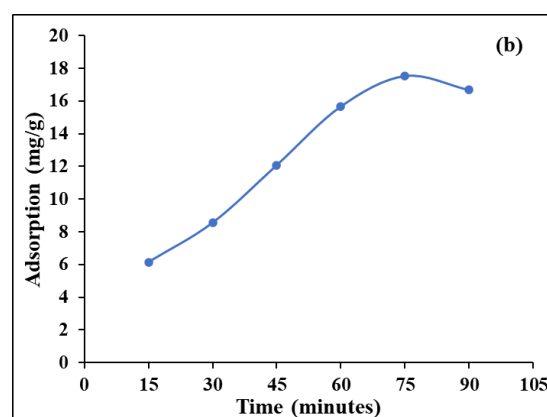
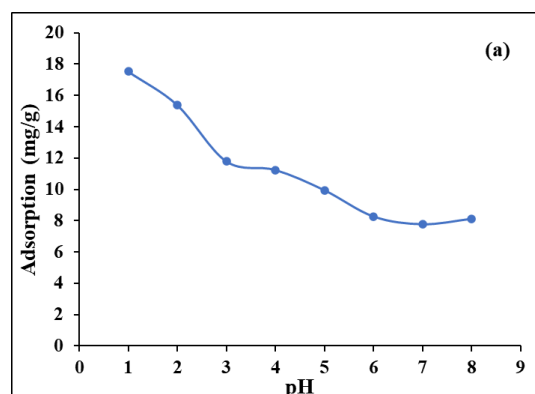
The pH_{pzc} is the pH at which the surface of a material is neutral (i.e., the number of positive charges equals the number of negative charges). In this study, the pH_{pzc} of SiO₂-PVA was 6.2 (**Figure 4**). When the solution pH is greater than the pH_{pzc}, the material surface becomes positively charged, thus tending to attract negative ions, and if the solution pH > pH_{pzc}, then there is a repulsion between SiO₂-PVA and the Congo red dye [1,31].

**Figure 4.** pH_{pzc} of SiO₂-PVA

As shown in **Figure 5(a)**, the optimal pH for Congo red adsorption by SiO₂-PVA is pH 1. Under this highly acidic condition, below the point of zero charge (pH_{pzc}), SiO₂-PVA carries a positive surface charge, while Congo red bears a negative charge due to its sulfonate groups (-SO₃⁻), leading to strong electrostatic attraction. In contrast, under basic conditions, electrostatic repulsion occurs, resulting in reduced adsorption efficiency. Similar findings have been reported in other studies, where acidic pH was also identified as optimal for Congo red adsorption, such as with activated carbon (pH 2) [32], chitosan-MnFe₂O₄ (pH 2) [31], and chitosan-graphene (pH 3) [14].

Figure 5(b) shows that contact time significantly influences the adsorption capacity of SiO₂-PVA for Congo red dye. In the initial stages, numerous active sites on the adsorbent surface are still available,

allowing dye molecules to bind readily. As time progresses, these active sites become increasingly occupied, leading to a slower adsorption rate. The optimal contact time was determined to be 75 minutes. Beyond this point, a decline in adsorption capacity was observed, possibly due to the desorption of some Congo red molecules back into the solution [14,32].

**Figure 5.** Effect of (a) pH, (b) contact time, and (c) concentration

The weakening of the adsorbate–adsorbent interaction is likely influenced by physical factors such as stirring. During the adsorption process, an increase in dye concentration initially leads to a higher

adsorption capacity, as the number of Congo red molecules is relatively low, and many active sites on the adsorbent surface remain unoccupied. However, as these sites become limited, the adsorption rate decreases due to increased competition among dye molecules for available active sites [34,35]. As shown in Figure 4(c), the optimum dye concentration was 75 mg/L, with an adsorption capacity of 18.44 mg/g.

Adsorption isotherm

The Langmuir and Freundlich isotherms are the most commonly utilized models in the adsorption process. The Langmuir isotherm characterizes a uniform adsorbent surface where adsorption transpires in a monolayer. This differs from the Freundlich isotherm, where adsorption may take place in many layers (multilayer). The formulations for the Langmuir and Freundlich adsorption isotherms are as follows: [31,33].

$$\frac{C_e}{q_e} = \frac{C_e}{q_m} + \frac{1}{K_L q_m} \quad (2)$$

$$\log q_e = \log K_F + \left(\frac{1}{n}\right) \log C_e \quad (3)$$

The maximum adsorption capacity is denoted by q_m (mg/g). K_L (L/mg) is the Langmuir isotherm constant, while K_F represents the Freundlich constant with units of (mg/g)(mg/L). The value of $1/n$ indicates the adsorption intensity and the degree of surface heterogeneity at varying concentrations [36].

The adsorption of Congo Red dye by SiO₂-PVA adheres to the Langmuir isotherm model. This can be seen from the higher determination coefficient (R^2) value in data analysis using the Langmuir model compared to the Freundlich model (Table 3). This finding suggests that the surface of SiO₂-PVA is homogeneous, where all adsorption sites have similar and equivalent characteristics, and there is no interaction between adsorbed molecules.

Table 3. Isotherm parameters of Congo red dye adsorption using SiO₂-PVA

| Parameters | Isotherm | |
|------------------|----------|------------|
| | Langmuir | Freundlich |
| R^2 | 0.916 | 0.591 |
| K_L (l/mg) | 0.1225 | - |
| q_m (mg/g) | 14.31 | - |
| K_F (g/mg.min) | - | 2.202 |
| n | - | 2.193 |

Various assumptions can be made to explain the interactions between SiO₂-PVA and the Congo Red dye. Hydrogen bonding is likely to occur between the

-OH groups of PVA and the silanol groups (Si-OH) of SiO₂ with the -NH₂ and -SO₃⁻ groups present in the Congo red molecule. Congo Red dye is an anionic dye (containing -SO₃⁻ groups), electrostatic interactions may also form between the negative charges of Congo red molecules and the positive charges on the adsorbent surface. In addition, weak Van der Waals interactions are also likely to occur between the Congo red molecules and the surface of SiO₂-PVA [37-39].

Langmuir parameters such as maximum adsorption capacity and Langmuir constant provide a quantitative description of the efficiency and affinity of the adsorbent to Congo red dye, obtained 14.31 mg/g and 0.1225. Several studies show the same phenomenon, that the adsorption process of Congo red dye follows the Langmuir isotherm, namely using bentonite adsorbent [35] and ZnO-SiO₂ [40]. Figure 6 shows the Langmuir and the Freundlich isotherm curves.

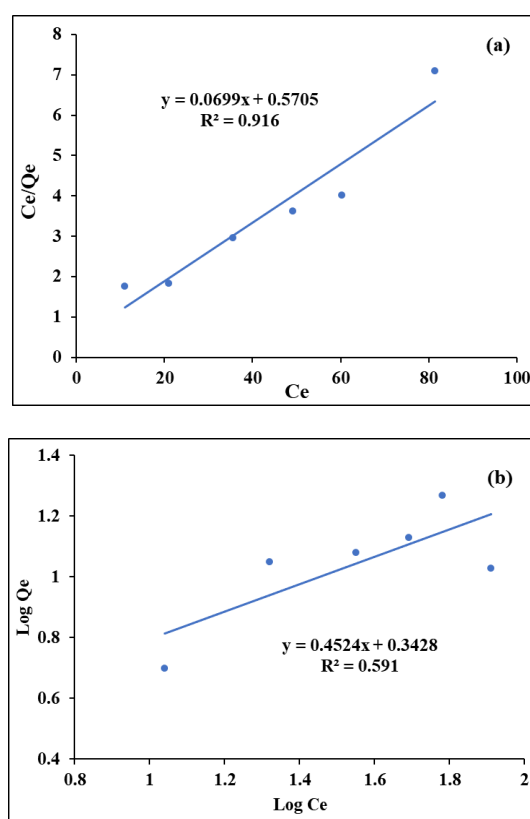


Figure 6. (a) Langmuir and (b) Freundlich isotherm model curves for Congo red dye

CONCLUSION

In this study, SiO₂ was successfully extracted from rice husk through a calcination process at 600, 700, and 800 °C. The XRD characterization showed that SiO₂ calcined at 600 °C had the most amorphous structure. To improve the stability and adsorption capacity, SiO₂

was modified using polyvinyl alcohol and used as an adsorbent for Congo red dye. The optimum adsorption conditions were obtained at pH 1, contact time of 75 minutes, and 75 mg/g dye concentration. The adsorption data best fit the Langmuir isotherm model, with a maximum adsorption capacity reaching 14.31 mg/g.

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