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Removal of Methylene Blue from Aqueous Solution with Silica Gel Extracted from Coal Fly Ash

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The textile industry has the potential to cause water pollution due to dye waste such as methylene blue (MB). Many methods have been developed to overcome the challenges of dye removal, one of which is the adsorption method. This study introduces silica gel (SiG) as an effective material for MB dye wastewater treatment. The silica source was extrated from coal fly ash (FAC) which has a relatively high silica content, making it a potential silika source for silica gel synthesis. The silica gel formed was characterized using FTIR and SEM. The FTIR characterization results showed that the synthesized SIG had adsorption in the wavelength range of 3500-3400 cm^{-1} and 1700-1600 cm^{-1}, indicating the presence of silanol and siloxane groups. The SEM results showed that the SiG morphology was spherical, non-agglomerated and had a fairly uniform particle size. This study also investigated the effect of pH, contact time, and adsorbent dosage on the MB dye adsorption process. The optimum conditions were obtained at pH 11, contact time 75 minutes and adsorbant dosage 0,1 gram with an adsorption efficiency of 89,49%. Therefore, this synthesized silica is promising for the efficient removal of MB dye from aqueous solutions and offers practical applications in wastewater treatment.

Keywords: adsorption, coal fly ash, silica gel, methylene blue

1. Introduction

The rapid development of various industrial technologies to meet human needs can also lead to environmental problems such as water pollution. The textile industry is a growing industry with the potential to cause water pollution due to the excessive use of textile dyes, this industry releases the highest level of dye wastewater and contributes more than 50% of the dye waste found in the environment worldwide [1]. Approximately 700.000 tons of synthetic dyes are produced every year and more than 100.000 types of dyes are used in the textile industry. Most of these dyes are considered harmful to aquatic life, stable against light and difficult to degrade; moreover the pollution caused by these dyes will reduce light penetration into the water. This will affect the photosynthetic activity of aquatic plants and reduce the availability of oxygen and nutrients for these organisms [2].

Methylene blue (MB) is a cationic dye from the azo group, used for dyeing cotton, silk, fabric and wool. Methylene blue contained in wastewater can have adverse effect on human health, such as an increased heart rate, vomiting, shock, eye and skin irritation, jaundice, and others. The discharge of wastewater without prior treatment results in high concentration of Chemical Oxygen Demand (COD) and increased toxicity in the water system. Therefore, these negative impacts need to be minimized by special treatment of wastewater before discharge into the environment [3].

Several methods have been developed to remove methylene blue from wastewater, such as biological methods (using enzymes and microorganisms), chemical methods (using oxidation processes) and physicochemical methods (adsorption). Among these treatment methods, adsorption is known as a potential alternative method for removing methylene blue pollutants in wastewater, due to its relatively high efficiency, simplicity, cost-effectiveness, and low energy consumption. In recent years, new adsorbents such as activated carbon [4], grapheme [5], membranes [6], and mesoporous silica [7] have been developed to remove organic and inorganic pollutants from wastewater through the adsorption process. However, adsorbents derived from natural materials have their own appeal, because this type of adsorbents are environmentally friendly and can be obtained from industrial waste.

Silica gel is a porous inorganic material that has advantages such as an active surface sites in the form of silanol (Si-OH) and siloxane (Si-O-Si) groups, large pores, small particle size and a unique surface area. The presence of silanol (Si-OH) groups on the surface of silica gel is related to its ability to adsorb methylene blue dye. In additional, silica gel stable, non-toxic, and low cost, so these advantages have the potential to be applied as an adsorbent in the treatment of methylene blue dye waste. Silica gel can be synthesized from materials with a high silica content, including fly ash, which is waste from the coal combustion process in power plants. The silica content in fly ash is approximately 60-70% [8].This is why fly ash waste has the potential to be an alternative source of silica for adsorbent utilization.

The result conducted by kurniasih (2025), showed a comparison of methylene blue adsorption using fly ash and silica gel extracted from fly ash as adsorbent. The adsorption capacity of the silica gel was 151.33 mg/g, the value is greater than the fly ash adsorption capacity with a value of 95.81 mg/g [9]. Based on these results, in this study, coal fly ash from PT Imip Morowali was modified with hydrochloric acid (FAC) and silica gel (SiG) was extracted using the sol-gel method. Then, the silica gel adsorption capacity of methylene blue dye was tested by determining the optimum conditions for pH, contact time and silica gel dosage.

2. Materials and Methods

2.1. Materials

Coal fly ash was obtained from PT. Imip Morowali Industrial Park, Morowali Regency, Central Sulawesi, Indonesia. Hydrochloric acid 37%, NaOH, and methylene blue supplied by Merck were used in this study, and distilled water.

2.2. Methods

2.2.1 Preparation and Activation of Coal Fly Ash

The cleaned fly ash was activated with 6 M hydrochloric acid for 4 hours at 90°C and filtered. The residue was rinsed with distilled water until the pH as neutral. The precipitate obtained was dried at 120°C for 3 hours to produce activated fly ash (FAC). Sodium silicate extraction was performed by dissolving FAC in 4 M NaOH and heating at 90°C for 1 hour. Then filtered to obtain the sodium silicate solution. The solution was neutralized with 4 M HCl and aged for 24 hours at room temperature to form hydrogel. The silica gel (SiG) obtined was separated from the filtrate and dried at 80°C for 12 hours. FAC and SiG materials were characterized using Fourier Transform-Infra Red Spectrophotometer (FTIR, Prestige-21, Shimadzu) for infrared scanning from 4000-400 cm^{-1}. Surface morphology and elemental composition of SiG were characterized using Scanning Electron Microscopy–Energy Dispersive *X*-Ray (SEM-EDX, Joel Jsm 6360LA Phenom-BSD Detector).

2.2.3 Batch Adsorption Studies

The batch adsorption system was performed using orbital shaker with 120 rpm. Batch adsorption test were conducted to analyze the effect of pH (3-13), contact time (15-90 min) and silica gel dosage (0,025-0,125 grams) on the adsorption process. Furthermore, 0.1 gram of SiG was added to a 100 ppm MB solution and the pH of the solution was adjusted by adding 1 M HCl or 1 M NaOH according to the pH above. Then, it was shaken based on the contact time. The mixture was filtered and the adsorbance of the filtrate was determined using a UV-Vis Spectrophotometer at the maximum wavelength of MB. The amount of MB adsorbed on SiG can be determined by calculating the adsorption capacity (q) and adsorption efficiency (%*R*) with the formula:

$$q = \frac{V \text{ x } (Co - Ce)}{\text{m}} \quad (1)$$

$$\%R = \frac{Co - Ce}{Co} \text{ x } 100\% \quad (2)$$

With *q* (mg/g) represents the adsorption capacity, C_0 and C_e (mg/L) represents the initial and final concentrations, m (g) represents the mass of SiG and V (L) represents the volume of MB solution.

3. Result and Discussion

Silica gel extraction from coal fly ash is carried out in several reaction stages. First, the fly ash is activated using 1 M HCl. Acid activation aims to remove metallic oxide impurities. The extraction process with 4 M NaOH using the reflux method produces a sodium silicate solution (Na_2SiO_3). Then, the solution was neutralized with 4 M HCl. The addition of acid to the sodium silicate solution is an important step to substitute Na^+ ions with H^+ which produces a gel-like solid. This treatment aims to separate the silica particles bound to water moleculs called silica hydrosol or metasilicic acid, which is a white solid, as shown in **Figure 1**. The reaction of adding acid to the sodium silicate solution is described by the reaction below

$Na_2SiO_{3(aq)} + 2HCl_{(aq)} \rightarrow H_2SiO_{3(s)} + 2NaCl_{(aq)}$

$H_2SiO_{3(s)} \rightarrow SiO_{2(s)} + H_2O_{(aq)}$

Figure 1. Physical form FAC and SiG

3.1. Characteristics of Silica Gel from Coal Fly Ash

The characterization of the synthesized silica gel was demostrated using FTIR in the range of 4000-400 cm^{-1}. This aims to determine the presence of silanol and siloxane groups on the sample surface. The FTIR spectra of the FAC and SiG samples are shown in **Figure 2**. It can be seen that the FTIR spectrum profiles of both samples are almost similar. The stretching vibration of the hydroxyl group corresponds to the absorption band appearing in the range of 3500-3400 cm^{-1}, indicating the presence of O-H groups originating from the silica gel constituents, specifically the silanol groups (Si-OH). The presence of hydroxyl groups (O-H) in the sample is also strengthened by the presence of absorption in the bending vibration of the O-H group bound to silica with an absorption band of 1700-1600 cm^{-1} [10]. The next absorption is illustrated by the 1100-1040 cm^{-1} band, caused by bending vibrations that indicate the stretching of the Si-O-Si symmetry, and the stretching oscillation of the siloxane group (Si-O-Si) symmetry also appears at the absorption band of 444.46 cm^{-1}. Furthermore, the symmetry stretching vibration of the siloxy group (Si-O) in the 467 cm^{-1}. absorption band proves the presence of the Si-O-Si group in the TO_4 structure [11].

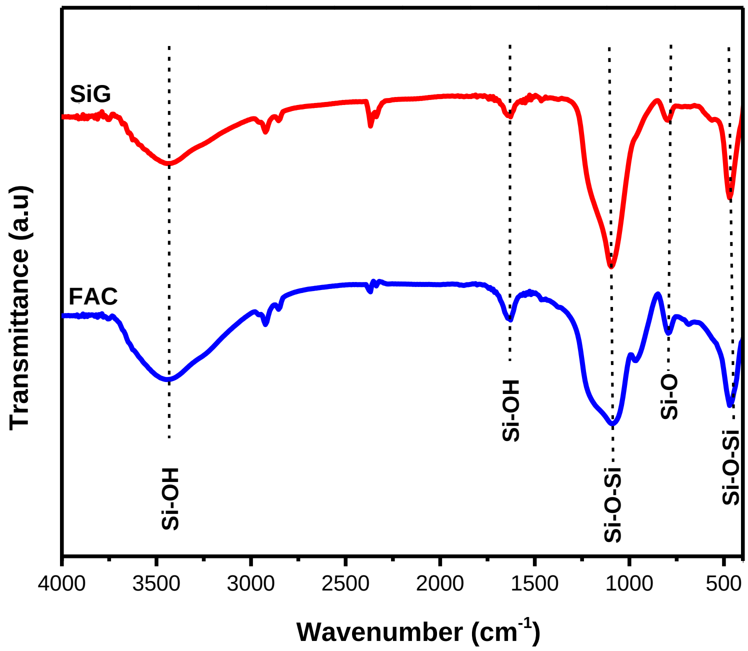

Figure 2. The FTIR spectrum of FAC and SiG

In SiG, the absorption of the siloxane group exhibits a strong and sharp intensity compared to FAC. This indicate that the Si-O-Si group is not affected by other groups contained in FAC. Furthermore, the FAC absorption at the wavenumber of 787 cm^{-1} was not found in SiG, where the absorption indicates the presence of a Si-O-Al bond [9]. The results of this FTIR spectrum significantly verify that silica extracted from coal fly ash has silanol dan siloxane groups.

The surface morphology of the SiG adsorbent was evaluated using SEM which covers the roughness, porosity, and elemental abundance found on the SiG surface. **Figure 3 (a)-(b)** shows the surface morphology of SiG at 5000x and 10.000x magnification, respectively. Based on these images, the morphology of the SiG adsorbent produced is spherical, not-aggregated, and has a relatively uniform particle size. Additionally, a larger size, a smooth surface and a dark texture are also seen [12]. Furthermore, the average particle size was determined using Image J software. The results of this analysis are displayed in **Figure 3 (c)** which graphically shows the particle size distribution with an average particle size of 1762 nm or 1.7 µm. the use of NaOH in the silica extraction from fly ash plays a role in the impact of silica micro particle formation on mono disparity and narrow size distribution. Polar NaOH can also increase the solubility of sodium silicate, thereby helping the growth of strong silica nuclei in polydisparity [13].

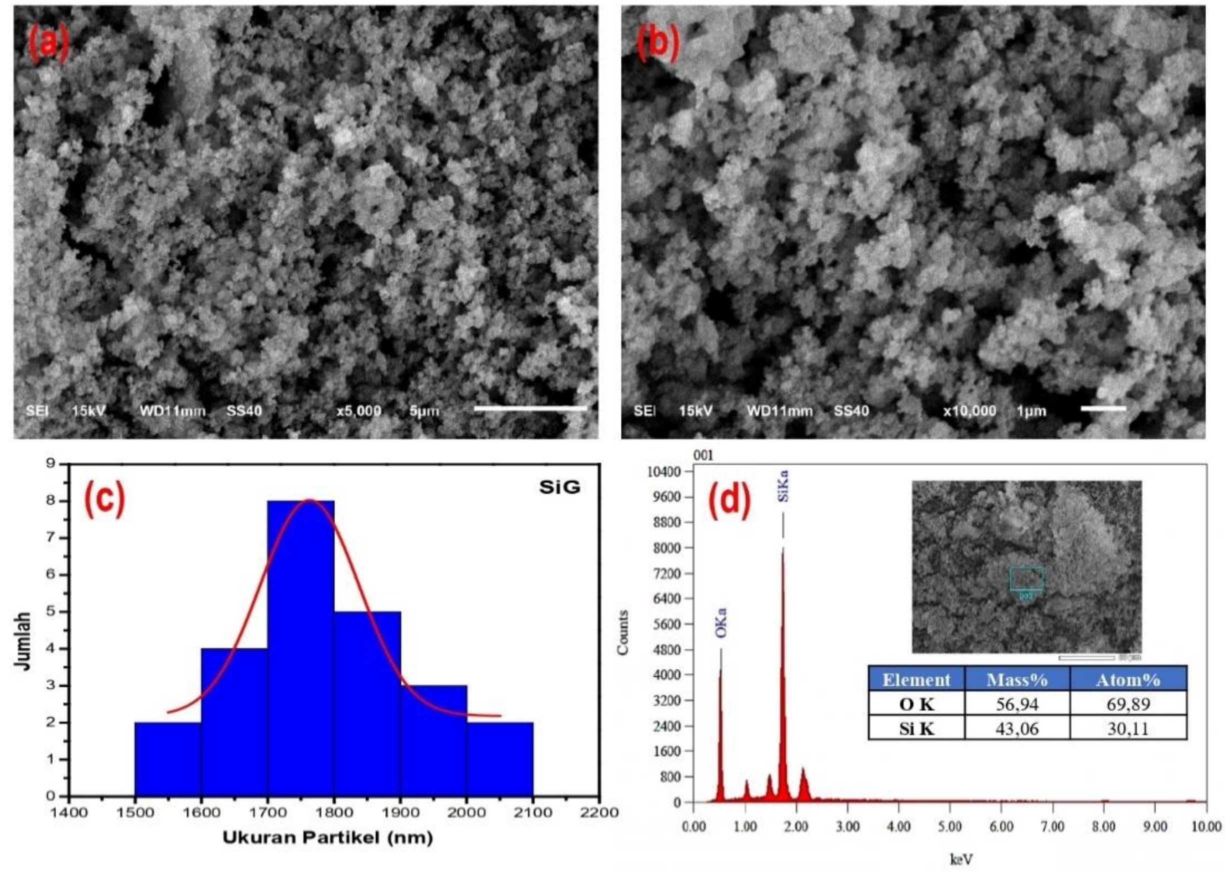

Figure 3. SEM image of SiG (a) 5000x and (b) 10.000x magnification.

The elemental composition analysis was carried out using EDX which provides clear information about the elements contained in the sample. By scanning different positions of the sample, it will produce a spectrum that shows the presence and quantity in the form of a percentage of each element present in the sample. **Figure 3 (d)** shows the EDX spectrum of SiG. Based on the spectrum, the sample contains silicon (Si) and oxygen (O) elements. This confirms the presence of SiO_2 compounds in the SiG adsorbent [14]. This result also does not show the presence of other element spectra such as the results of the EDX spectrum of fly ash, such as Al_2O_3 and Fe_2O_3 [9], so that the pure SiG adsorbent contains SiO_2 without any other metal oxide impurities.

3.2. Adsorption of Methylene Blue Dye with SiG

Before the adsorption of methylene blue with SiG adsorbent began, a preliminary study was conducted to determine the appropriate adsorbate concentration and better adsorption without being disturbed by other parameters. Adsorbate concentrations of 20 ppm and 100 ppm were used for this preliminary study. The results of the adsorption capacity of both concentrations can be seen in **Figure 4 (a)**, which shows that with a high adsorbate concentrations, the SiG adsorbent is able to adsorb with a greater adsorption efficiency. From these results, the methylene blue concentration used to determine the optimum adsorption is a concentration of 100 ppm.

The influence of solution pH is a crucial factor in the adsorption process because pH affects the interaction between the adsorbate and functional groups on the adsorbent surface. The addition of H^+ ions or OH^- ions during solution pH adjustment will affect the ionization process of functional groups on the adsorbent surface. Functional groups will be protonated or deprotonated by changes in pH and can cause changes in the surface charge of molecules in adsorption. Furthermore, pH will also affect the ionization process of dye and regulate their interactions with the active sites of the adsorbent. Therefore, MB adsorption was studied to investigate the effect at pH 3-13 as can be seen in **Figure 4 (b)**. As shown in **Figure 4 (b)**, the adsorption efficiency increased from 55.87 to 85.39% with increasing pH from 3 to 11, then decreased at pH 13 to 81.06%, the decrease in adsorption efficiency at higher pH is caused by MB molecules being unable to interact with the SiG pore structure because of steric hindrance. This result is in accordance with the research of Khushnood (2024) which shows that the optimum pH condition for MB adsorption is in a solution with pH > 7 (pH 11) [15].

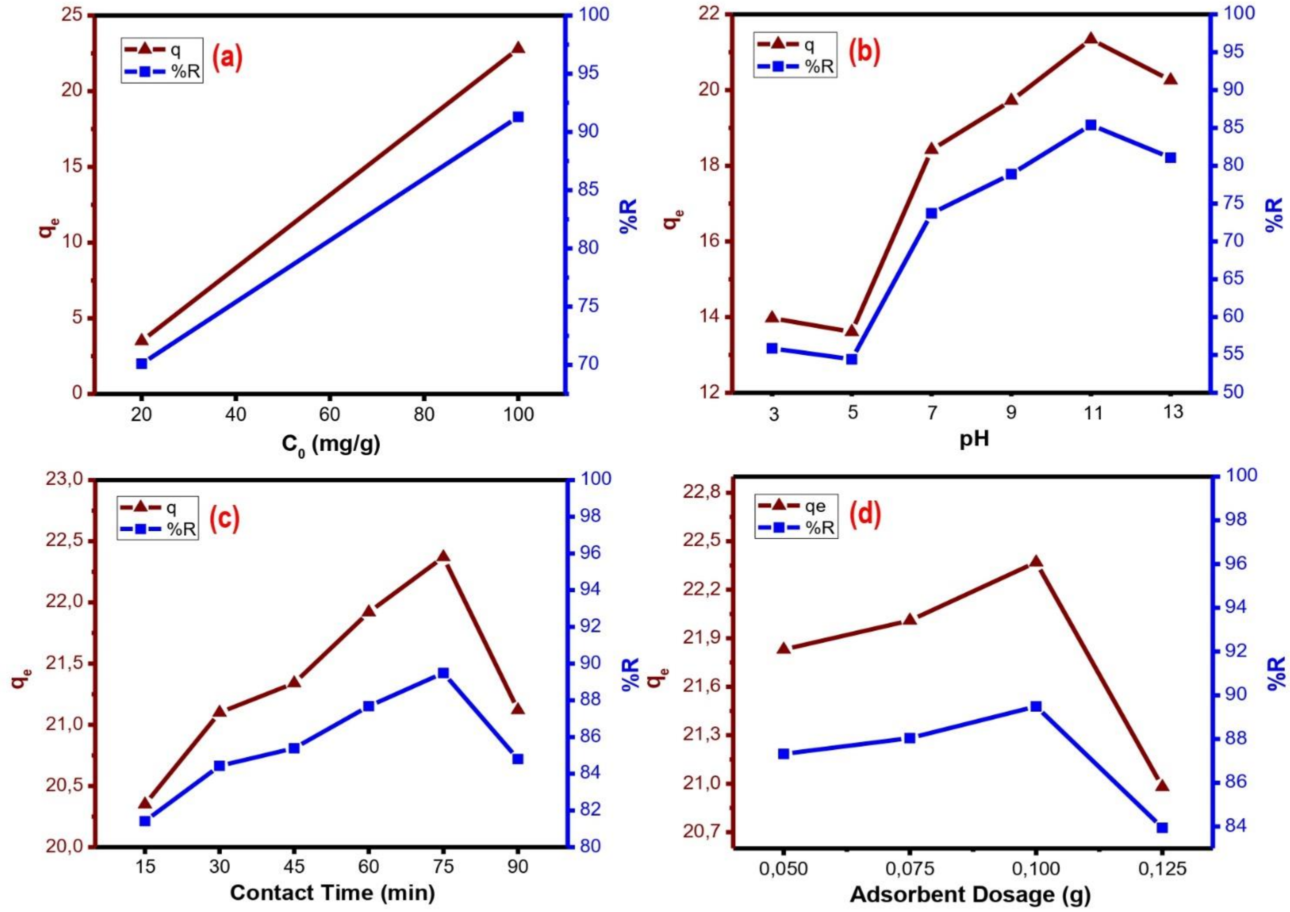

Figure 4. Effect of (a) initial MB concentration, (b) pH, (c) contact time, (d) adsorbent dosage

The adsorption mechanism of MB with SiG likely originates from hydrogen bonding and electrostatic interactions between the silanol and siloxane functional groups in SiG and cationic MB. It is probable that at higher pH, the protonation of the silanol and siloxane groups gradually decrease. Therefore, the increase in negative charge on SiG is proportional to the increase in pH. Consequently, the adsorption process is influenced by electrostatic interactions between the negatively charged functional groups in SiG and cationic MB, according to the predicted mechanism in **Figure 5**. Meanwhile, at low pH, the presence of H^+ ions in water can compete with cationic MB for the free active zone in SiG. Furthermore, the possibility of strong hydrogen bonding interaction that the difficult to break between the hydroxyl groups and the cationic MB at low pH can also affect the decrease in MB adsorption capacity [16], [17]. Based on these result, the optimum pH for MB adsorption was found to be pH 11, which will be used for further adsorption.

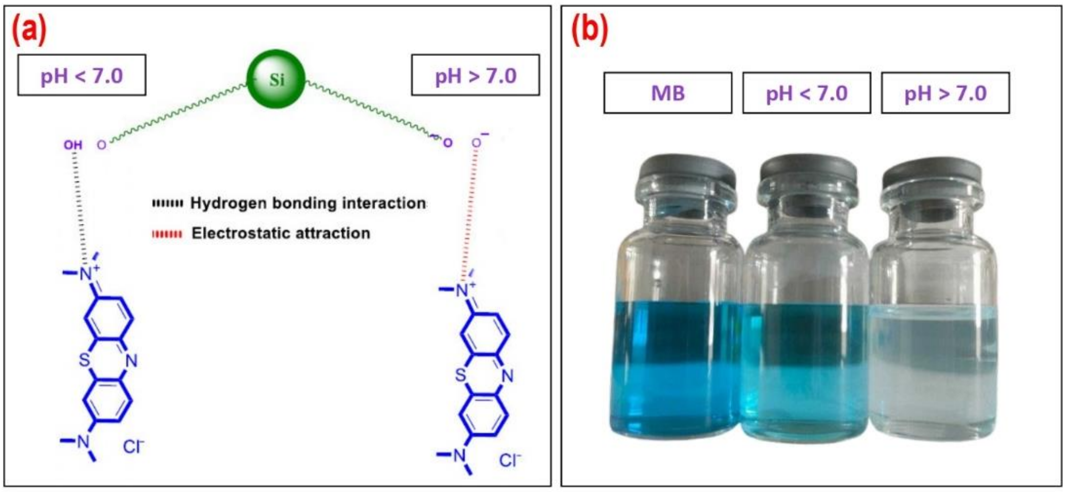

Figure 5. (a) Proposed governing mechanism of MB adsorption on SiG [16], (b) the difference in color of MB solution, after adsorption of MB in acidic and alkaline conditions

The length of MB adsorption contact time will affect the number of opportunities for MB adsorbate to approach the active site of the adsorbent which will increase the adsorption efficiency. To determine the optimum contact time, the experiment was carried out at pH 11 and adsorption was observed for intervals of 15 to 90 minutes as shown in **Figure 4 (c)**. The adsorption curve illustrates that the MB adsorption efficiency initially increase with increasing contact time because there is sufficient time for MB molecules to be adsorbed to the SiG surface. However, it eventually decreases and there is no further increase when the mixture reaches equilibrium [18]. So the optimum contact time for MB adsorption in this study is at 75 minutes with an adsorption efficiency of 89.49%.

The effect of adsorbent dosage on MB adsorption was also explored by varying SiG from 0.05 to 0,125 grams. The adsorption curve can be seen in **Figure 4 (d)** which shown that there is an increase in adsorption capacity along with the amount of SiG adsorbent dosage with the highest adsorption capacity at a dose of 0,1 grams, but the value decreases at a larger SiG dose. This is because at a large SiG dose, more active sites of the adsorbent are in the system, even though the amount of MB remains constant, so that MB molecules are not enough to saturate the much more active sites available at higher doses. In additional, the possibility of clumping at higher doses can reduce the effective surface area and cause competition between adjacent binding sites and decrease its adsorption capacity [19].

4. Conclusion

This study investigated the adsorption of MB from aqueous solution by preparing silica gel extracted from coal fly ash as an adsorbent through a batch adsorption process. FTIR and SEM-EDX characterization methods were used to elute the proposed adsorbent. FTIR results confirmed the presence of silanol and siloxane vibrations in the obtained SiG. The surface morphology of SiG was spherical, non-agglomerated with fairly uniform particle size, and EDX results showed that SiG contained silicon and oxygen elements without any other impurities. In the adsorption process, the optimum adsorption conditions were determined by changing various parameters such as pH, contact time, and adsorbent dosage. The optimum pH was obtained at pH 11, contact time 75 minutes and adsorbent dosage 0.1 gram with an adsorption efficiency of 89.49%. Therefore, SiG has the potential as an applicable adsorbent in the treatment of MB dye waste.

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