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Synthesis of Graphene Oxide-Coated Mesoporous Silica with Cetyltrimethylammonium Bromide (CTAB) Template for Methylene Blue Adsorption

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Abstract

The synthesis of graphene oxide-coated mesoporous silica (MS_GO) with the template surfactant (Cetyltrimethylammonium Bromide) CTAB has been carried out. The effect of combining mesoporous silica and graphene oxide was studied by knowing the bonds, functional groups and crystalline structures. Functional groups C=O and C=C were formed at wave numbers 1,722, 1,617 and 1,647 cm^{-1}, respectively, as a characteristic of GO compounds. XRD data showed that MS_GO has a more amorphous structure than graphite and GO due to the incorporation of silica onto the graphene oxide surface. The MS_GO synthesis was also applied as an adsorbent for methylene blue dye in water. The adsorption results showed that MS_GO was more effective than pure GO. The percent adorption efficiency (R %) of MS_GO against 10 ppm methylene blue was 93.1 % while that of pure GO was 91.5 %. The addition of mesoporous silica to graphene oxide makes MS_GO adsorbent more effective in adsorption dyes than pure GO, this is supported by the larger total surface area of BET MS_GO was 161.066 $m^2 \cdot g^{-1}$, while that of pure GO was 103.818 $m^2 \cdot g^{-1}$.

Keywords: Mesoporous silica, Graphene oxide, CTAB, Adsorption, Methylene blue, Fungtional group, Crystallinity

Introduction

Porous material is a solid material that has pores so it has a large surface area. The group includes porous materials, for example, zeolites, porous carbon and mesoporous silica. Mesoporous silica is a mesoporous form of silica and is a recent development in nanotechnology. These mesoporous materials consist of inorganic metal oxides, such as silica or alumina, and the pore sizes range between 2 and 50 nm [1]. The synthesis of mesoporous silica is very interesting because it shows a very regular and stable mesoporous structure, well-developed, a broad and uniform pore system with a narrow distribution and a large volume, high chemical and hydrothermal properties and with easily modified surface area [2].

Mesoporous silica is usually synthesized using surfactants to form aligned and orderly assemblies, which are used as metal oxide templates, followed by removal of the template. Surfactant is very important in the synthesis because it will determine the size of the pores formed [1]. Some surfactants can be intercalated into the interlayer space of layered materials such as clay, graphite, graphite oxide and graphene oxide. Graphene oxide (GO) can be used as an adsorbent which is very useful in the adsorption of heavy metal ions and dyes with its large surface area. GO functional groups can be used as linking or nucleation points for other materials combined with the synergistic effect of the decorated material, and can increase the adsorption capacity and selectivity of the adsorbent [3].

The use of silica and graphene as adsorbents has been widely studied. Han *et al.* [4], investigated the synthesis of mesoporous silica from iron ore tailings for efficient adsorption of methylene blue. The mesoporous silica obtained showed a specific surface area of 544.68 $m^2 \cdot g^{-1}$ with well-developed porosity. As an adsorbent, it showed good monolayer adsorption with an adsorption capacity of 192 $mg \cdot g^{-1}$ for methylene blue dye in aqueous solution [4]. Saman *et al.* [5], also investigated the synthesis and characterization of CTAB-silica nanocapsules and their adsorption to Pd(II) ions in aqueous solution. The results obtained showed that the maximum adsorption capacity of Pd(II) ions to SiNC-CTAB obtained from the adsorption isotherm was 124.50 $mg \cdot g^{-1}$ [5]. Li *et al.* [6], examined the adsorption of methylene blue from aqueous solutions by magnetic graphene oxide/humic acid composites. The results obtained showed that an adsorption reaction occurred at 45 °C with 30 mg of adsorbent to adsorb 5.0 $mg\cdot L^{-1}$ MB in 100 mL of solution, with a maximum MB adsorption capacity of about 59.00 $mg\cdot g^{-1}$ [6].

In this research, the synthesis of mesoporous silica in the GO interlayer space will be carried out using the CTAB (Cetyltrimethylammonium Bromide) surfactant as a template and pore printer. The mesoporous silica will be sandwiched between adjacent graphene oxide sheets, and 1 mesoporous row is parallel to the space between the graphene layers in the GO interlayer. Graphene has interesting properties, synthesis of templates from mesostructures in the space between GO layers will form new graphene-based composites with unique morphology and unusual properties [7]. Mesopore silica that has been synthesized with GO will be applied as an adsorbent for the organic pollutant methylene blue in water.

Methylene blue (MB) is a very important basic dye and is relatively inexpensive compared to other dyes. High doses of MB can cause nausea, vomiting, abdominal and chest pain, headache, excessive sweating and hypertension. In addition, methylene blue can also cause irritation to the digestive tract if swallowed, cause cyanosis if inhaled, and irritate the skin if touched by the skin [8].

Materials and methods

Materials

Graphite, sulfuric acid (H_2SO_4), sodium nitrate ($NaNO_3$), potassium permanganate ($KMnO_4$), aquadamine, hydrogen peroxide (H_2O_2), cetyl trimethylammonium bromide (CTAB), sodium hydroxide (NaOH), tetraethoxysilane (TEOS) and Methylene Blue (MB).

Methods

Synthesis of graphene oxide

GO synthesis was carried out using the hummers method. The 4.5 g of graphite powder was dissolved in 4.5 g of sodium nitrate and 225 mL of sulfuric acid 98 % with vigorous stirring at 70 °C for 1 h. The mixture was cooled in an ice bath to a temperature below 5 °C with continuous stirring. Then, 9 g of potassium permanganate were added slowly to the mixture ensuring that the temperature did not exceed 20 °C. The mixture was stirred in an ice bath for 1 h. The mixture was then stirred at 35 °C for 5 h with continuous stirring [9]. To this mixture, 414 mL of aquadamine was added slowly to avoid temperatures above 100 °C. The resulting mixture was stirred for 1 h at 98 °C. Thereafter, 845 mL of warm aquadamine was added to dilute the mixture, followed by the addition of 5 % hydrogen peroxide to the mixture to reduce the unwanted reactants and dissolve the salt. The precipitate was left overnight and separated from the filtrate. The precipitate was washed several times with a solution of 10 % hydrochloric acid and warm aquadamine. Then, the product was dried in oven at 60 °C to get a brown GO [9].

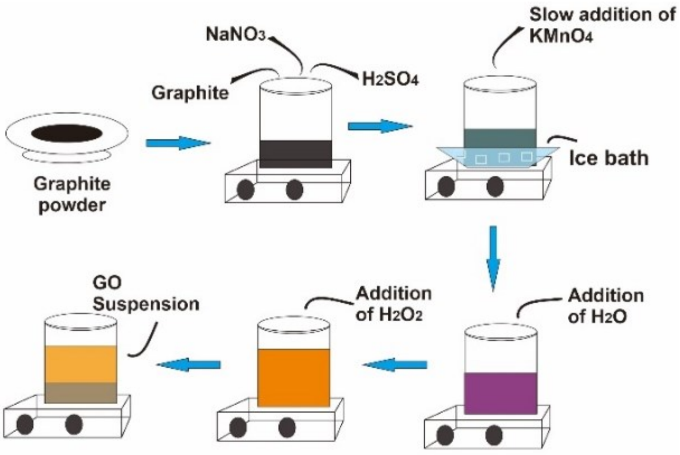

Figure 1 GO Synthesis with Hummers method.

Synthesis of mesoporous silica/graphene oxide

Prepared GO intercalated surfactant cetyl trimethylammonium bromide (CTAB). The 1.456 g of CTAB was dissolved in 40 mL 0.1 M of sodium hydroxide solution (NaOH). The 0.2 g of GO powder was weighed and then immersed in the CTAB hydroxide solution at 25 °C for 4 days. After immersion, the suspension was filtered and washed with water to remove unintercalated CTAB and NaOH. The resulting GO-CTAB was dried at a relative humidity of about 60 % for 7 days.

GO-CTAB (0.1 g) dry powder was stirred in 0.36 g dodecylamine (if using hexylamine or decylamine, the molar weight is equal to the molar weight of dodecylamine) for 2 h at 45 ℃ followed by addition of 4.28 mL tetraethoxysilane (TEOS). The resulting suspension was stirred for further 4 h at 45 ℃. The samples were air dried under 60 % relative humidity for 20 days to form the synthesized MS_GO [7].

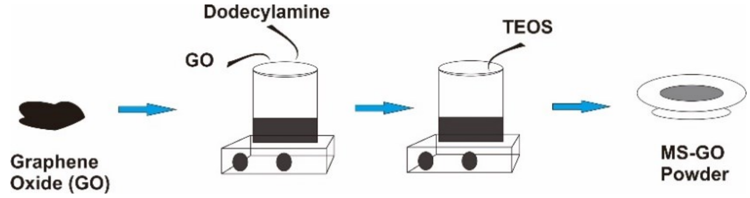

Figure 2 Synthesis mesoporous silica with GO.

Characterization

The synthesized graphene oxide-coated mesopore silica was characterized to determine its crystalline phase by X-Ray Diffraction, functional groups and formed by Fourier Transform Infrared (FTIR) spectra.

Adsorption with methylene blue

Graphene oxide powder, and mesoporous silica/graphene oxide (MS_GO) as much as 10 mg were each put into 20 mL of methylene blue solution with various concentrations of 10, 20, 30, 40 and 50 ppm. The optimum concentration obtained is then used as a reference for the adsorption of time variation 5, 10, 15, 20, 25, 30 and 60 min. The Erlenmeyer flask covered with plastic, was shaken for 120 min at 155 rpm. The adsorbent is separated from the solution by filtering using filter paper. The absorbance of obtained filtrate was then measured it absorbance with a UV-Vis spectrophotometer with a wavelength of 664 nm [10].

The percentage of MB adsorption efficiency by MS_GO adsorbent was calculated using the equation:

$$R\% = \left(\frac{C_o - C_e}{C_o}\right) x100\%$$

where, R % is the adsorption efficiency, C_o (mg·L^{-1}) is the initial pollutant concentration, C_e (mg·L^{-1}) is the pollutant concentration after the adsorption process [10].

Results and discussion

Synthesis of graphene oxide

The synthesis of graphene oxide (GO) from graphite was prepared by modified hummers method using a mixture of sodium nitrate, potassium permanganate ($KMnO_4$) and sulfuric acid (H_2SO_4). The sodium nitrate in the Hummers method has an oxidizing effect, helping H_2SO_4 and $KMnO_4$. In addition, (there is the) addition of H_2O_2 to stop the oxidation reaction by reducing the remaining permanganate and manganese dioxide to manganese sulfate which is colorless during the reaction [11].

The main oxidation step of this method is understood to be the oxidative peeling of graphite by green dimanganese heptoxide (Mn_2O_7) and permanganyl cation (MnO^{3+}) formed from the reaction between permanganate and concentrated sulfuric acid. Although permanganate is a strong oxidizing agent, manganese heptoxide (Mn_2O_7) is a real active reagent that is built up during the reaction of permanganate and sulfuric acid. The temperature needs to be controlled during the synthesis, because manganese heptoxide tends to react explosively when exposed to temperature at 55 ℃ [12].

The results of the synthesis using the Hummer graphene oxide method from graphite will form epoxy, hydroxyl and carboxyl functional groups as seen in the results of functional group analysis using FTIR. GO contains various functional groups in its structure, such as carboxyl, hydroxyl and epoxy groups, which can chelate with heavy metal ions and dyes so that they are very useful in the adsorption process [3].

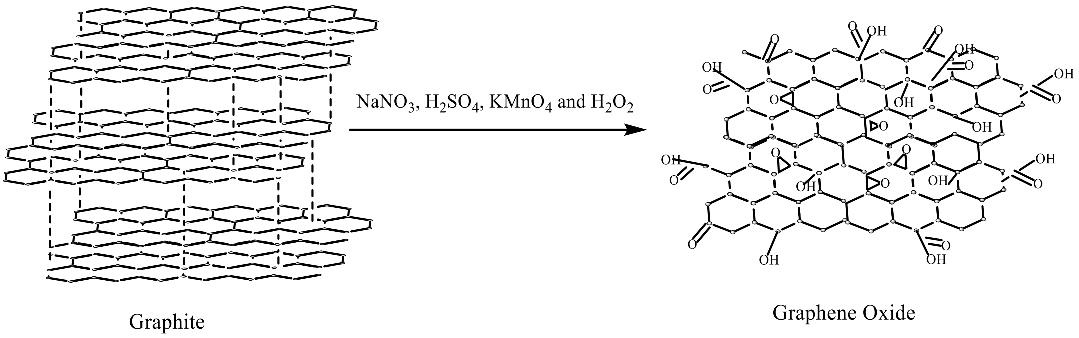

Figure 3 Graphene oxide formation reaction from graphite.

Mesoporous graphene oxide-coated silica synthesis (MS_GO)

The synthesis of graphene oxide-coated mesopore silica (MS_GO) with CTAB template has been carried out. The template was synthesized in the GO interlayer space involving the use of an intercalated ammonium cation surfactant and a neutral amine as co-surfactant to direct the hydrolysis and condensation of inorganic precursors (tetraethoxysilane, TEOS) in the GO interlayer space. The confinement effect provided by the GO interlayer space makes the mesoporous silica skeleton sandwiched between adjacent graphene sheets, and 1 mesoporous line is parallel to the GO interlayer space. Template synthesis of mesostructures in the interlayer space of GO will form new graphene-based composites with unique morphology and unusual properties [7].

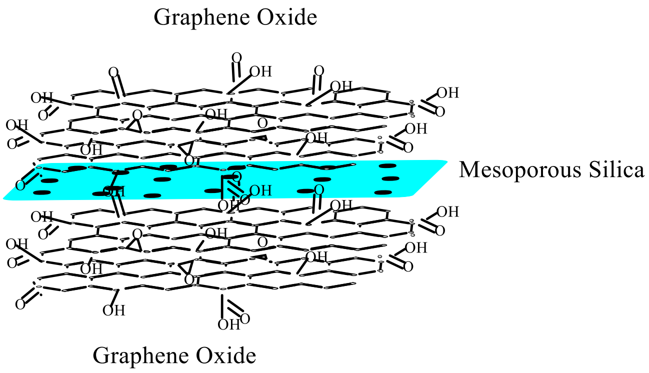

Figure 4 Silica in the GO interlayer space.

The porous silica material can be grown on graphene oxide (GO) sheets to form a composite with a sandwich structure. The SiO_2/graphene has the characteristics of high specific surface area, good electrical conductivity and excellent mechanical properties. Selection of the right method for the manufacture of graphene/silica composites can produce a synergistic effect with improved physical and chemical properties for high performance. On the one hand, composites can be made through noncovalent bonds between GO and silica [13].

Characterization

Graphene oxide (GO) and graphene oxide-coated mesoporous silica (MS_GO) samples have been synthesized and analyzed for their characteristics using FTIR (Fourier Transform Infrared) to determine their functional groups, and XRD (X-Ray Diffraction) to determine the crystalline phase, as well to test the adsorption ability toward methylene blue dye.

Functional group analysis with FTIR (Fourier Transform Infrared)

FTIR analysis was carried out to determine the functional groups and bonds formed during the synthesis process. The samples analyzed by FTIR included graphite before becoming GO, graphene oxide (GO) and GO which had been synthesized with silica (MS_GO). The Graphite, GO and MS_GO FTIR results are shown in **Figure 5**.

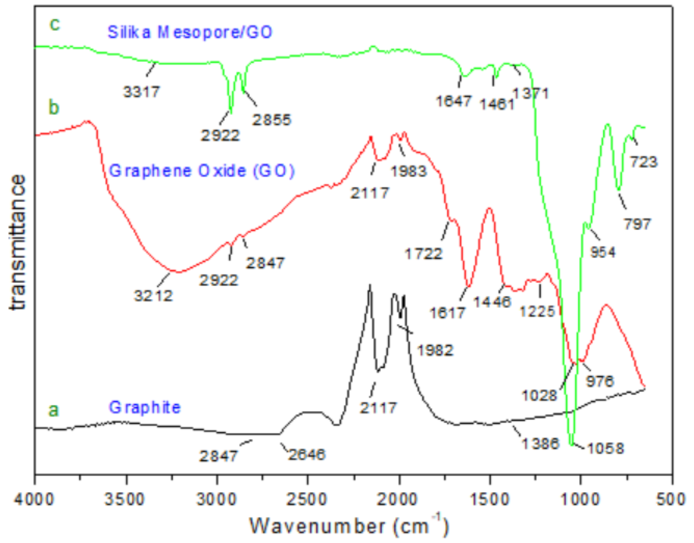

Figure 5 FTIR spectrum of a) graphite, b) GO and c) MS_GO.

Based on **Figure 5**, it is known that graphite, GO and MS_GO have CH_3 absorption at wave numbers 2,922, 2,855 and 2,847 cm^{-1} [14]. **Figure 5(a)** graphite spectrum shows a weak -OH absorption at a wave number of 2,646 cm^{-1}, the bond streching of C≡C alkynes at wavenumber of 2,117 cm^{-1} and for C-H, CH_2 bond at wavenumber of 1,386 cm^{-1} [15]. The GO spectrum **Figure 5(b)** shows the presence of a wide -OH hydroxyl group at the wavenumber 3,500 - 3,200 cm^{-1} [9,16,17]. There is also the C=O carboxylic group at a wavenumber of 1,722 cm^{-1}, the carboxyl group, the C=C alkene at wavenumber 1,617 cm^{-1} [9,16,17], the C-O epoxy group at wavenumber 1,225 cm^{-1} [9,16], and the C-H trans group at 976 cm^{-1} [15]. In the MS_ GO spectrum **Figure 5(c)** shows the absorption of the N-H amine group at the wavenumber 3,317 cm^{-1} [18], and the stretch C-N group at the wavenumber 1,461 cm^{-1} which is formed due to the addition of CTAB [17]. There is also the group alkene C=C at wavenumber 1,647 cm^{-1} [9,16,17], C-H and CH_2 groups at wavenumber 1,371 cm^{-1} [15] Si-O-Si group bonds at wavenumber 1,058 cm^{-1} [14,16], the trans C-H bond is said to be 954 cm^{-1} [15] and the Si-C bond at wavenumber 797 cm^{-1} [20].

XRD analysis (x-ray diffraction)

Graphite powder, GO and MS_GO were analyzed for their crystalline and amorphous phases by X-ray diffraction. The difactogram of the sasmple analysis results is shown in **Figure 6**.

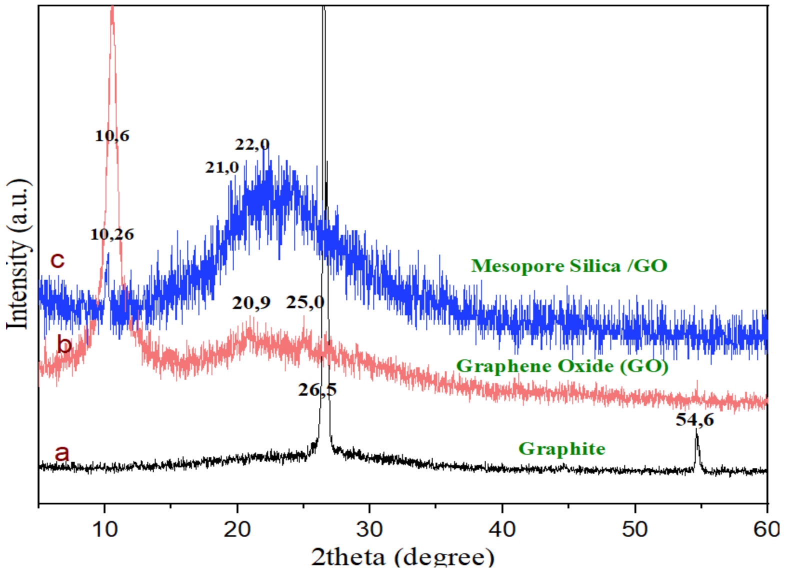

Figure 6 XRD difactogram (a) graphite, (b) GO and, (c) MS_GO.

Based on the XRD difactogram **Figure 6** graphite has the sharpest 2θ peak than GO and MS_GO. The graphite pattern of **Figure 6(a)** has a sharp peak at 2θ = 26.5 ° which corresponds to the reflection of the (002) plane and a slight peak at 2θ = 54.6 ° for the (004) plane with distances d of 3.36 and 1.68 Å [21]. **Figure 6(b)** the GO pattern shows a sharp peak at 2θ = 10.6 ° and corresponds to the plane reflection (001) with a distance of d = 8.34 Å. Small peaks are also seen at 2θ = 20.9 and 25.0 ° with a distance d of 4.25 and 3.55 Å, respectively. The increase in the distance between the fields in graphene oxide is caused by the presence of oxygen functional groups and water molecules into the structure of the carbon layer [22]. This increase in interplanar distance has been widely reported. The characteristic range of GO generally ranges between 0.7 and 0.8 nm but may vary slightly at higher or lower values, depending on the level of functionality. **Figure 6c** MS_GO pattern has an amorphous crystal structure and has 2θ peaks which are lower than graphite and GO. The decrease in the intensity of the diffraction peak (002) at 2θ = 10.25 ° indicates a long-range order decrease in MS_GO due to the incorporation of silica onto the graphene oxide surface [23]. The MS_GO pattern also shows peaks at 2θ = 21.0 and 22.0 with a distance d of 4.02 and 4.22 Å, respectively.

Gas Sorption Analyzer (GSA)

Gas Sorption Analyzer (GSA) is a gas adsorption mechanism on the surface of a solid material at various constant pressures and temperatures (isotherms). GSA analysis with the BET surface model was carried out to determine the surface area and porosity of the synthesized compounds.

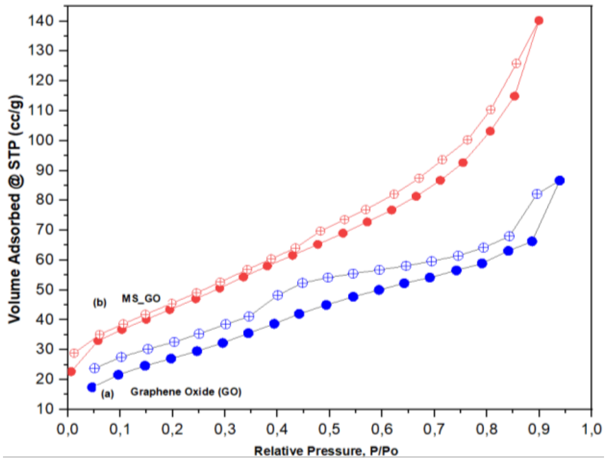

Figure 7 Nitrogen adsorption-desorption isotherms (a) GO and (b) MS_GO.

Based on **Figure 7** the calculation of the specific surface area is carried out through the adsorption isotherm using Brunauer-Emmett-Teller (BET) analysis. The results of the analysis showed that the MS_GO and GO isotherms are type IV (according to the IUPAC classification) the presence of mesopores [7,24]. A characteristic hysteresis loop of relative pressure (P/P0) higher than 0.8 was observed, and it can be noted that the formation of the pore structure was caused by the accumulation of nanoparticles. With increasing GO, more pores are formed by the accumulation of nanoparticles, which means that the adsorption performance of GO-SiO_2 increases [25].

The total surface area of BET of pure GO compound in **Figure 7(a)** is 103.818 $m^2 \cdot g^{-1}$, while the total surface area of BET MS_GO in **Figure 7(b)** is larger than with pure GO was 161.066 $m^2 \cdot g^{-1}$. The results of the GSA analysis are presented in **Table 1**.

Table 1 Table of analysis results with Brunauer-Emmett-Teller (BET).

Sample	**BET surface area ($m^2 \cdot g^{-1}$)**	**BJH Adsorption ($cc \cdot g^{-1}$)**	**Total Pore Volume ($cc \cdot g^{-1}$)**	**Average Pore Size ($cc \cdot g^{-1}$)**
Graphene Oxide (GO)	103.818	50.490	0.134324	2.58786
MS_GO	161.066	86.496	0.217444	2.70007

Based on **Table 1** MS_GO has a larger surface area than pure GO, the results show that the formation of MS_GO on graphene sheets can prevent graphene particulate aggregation, through weakening the π-π stacking interactions between graphene sheets [24]. MS_GO average pore size looks bigger (2.7 $cc\cdot g^{-1}$) than pure GO (2.58 $cc\cdot g^{-1}$). MB is a medium sized molecule therefore, relatively large pore size allows faster mass transfer of MB to the interior of MS_GO particles.

MS_GO adsorption on methylene blue

The adsorption test on methylene blue used 2 samples, namely graphene oxide and graphene oxide-coated mesoporous silica to determine the adsorption ability of pure GO before adding silica and GO after being synthesized with silica. The results of GO and MS_GO adsorption on methylene blue were determined based on the percentage of adsorption efficiency (% R).

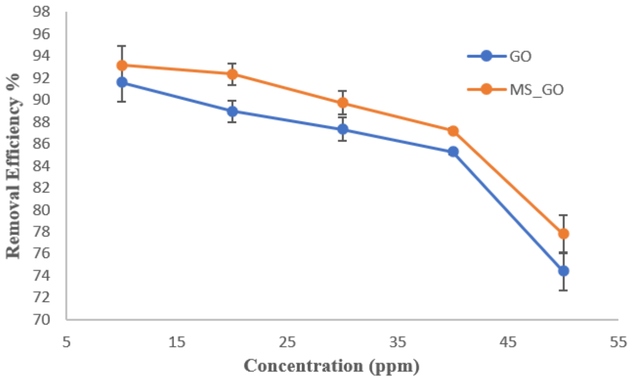

Figure 8 Graph of methylene blue adsorption results by GO and MS_GO concentration variations.

Based on **Figure 8**, the graph of the adsorption results of methylene blue by GO and MS_GO shows that adsorption with MS_GO has higher adsorption efficiency and adsorption capacity than GO. The addition of mesopore silica to graphene oxide makes this MS_GO adsorbent has a larger and more regular surface area than GO. Mesoporous materials have large specific surface area, rich surface functional groups, which have great application potential in the fields of catalysis, adsorption and separation [4]. On the other hand, the functional groups contained on the MS_GO surface allow MB-adsorbent interactions including π-π bonding, electrostatic and van der Waals interactions [26]. The results also increase the adsorption rate of MB. The concentration of methylene blue also affects the adsorption efficiency of GO and MS_GO. In **Figure 8**, the graph of the percent adsorption efficiency (R %) shows that at higher concentration of MB, the (R %) decreases. The decrease in the percentage of absorption is caused by the increasing number of pollutants that exceed the number of available adsorption sites [3]. GO and MS_GO adsorbents showed the best absorption to remove MB at an initial concentration of 10 ppm.

In the adsorption of methylene blue, in addition to variations in concentration, the contact time of the adsorbent were also varied with variations in time of 5, 10, 15, 20, 25, 30 and 60 min. The results of adsorption with variations in contact time are presented in **Figure 8**.

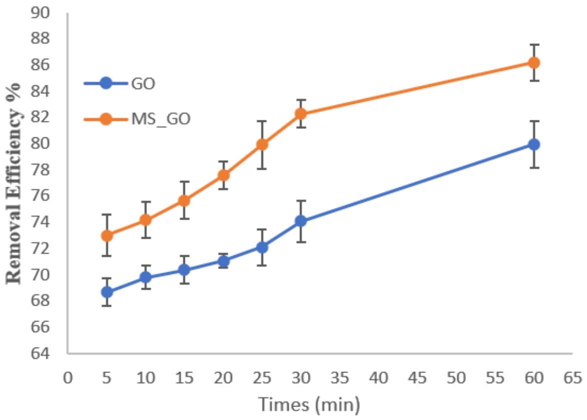

Figure 9 Graph of methylene blue adsorption results with variations in contact time.

Based on **Figure 9**, the adsorption graph of the contact time variation shows that (R %) are directly proportional to the contact time. The graph shows that the longer the contact time of the adsorbent with methylene blue, the higher the (R %) will be. The best time variation occurs at minute 60 which has an absorption efficiency for GO of 79.9 % and MS_GO of 86.1 %.

Conclusions

In this research, the synthesis of graphene oxide-coated mesopore silica with a CTAB template has been carried out. The results of sample analysis with FTIR showed the formation of the C=O and C=C groups at wave numbers 1,722, 1,617 and 1,647 cm^{-1}, and Si-O-Si groups at wave number 1,058 cm^{-1}, respectively. The sample analysis with XRD showed differences in crystallinity of graphite, GO and MS_GO, where MS_GO has a more amorphous structure than graphite and GO due to the incorporation of silica onto the graphene oxide surface. The results of the synthesis were also tested for adsorption on methylene blue dye and obtained better adsorption with MS_GO than with pure GO. The percent adsorption efficiency (R %) of MS_GO against 10 ppm methylene blue was 93.1 % while GO was 91.5 %. The addition of mesopore silica to graphene oxide makes this MS_GO adsorbent has a larger and more regular surface area than GO, and more active sites than GO. The addition of mesopore silica to graphene oxide makes MS_GO adsorbent have a larger surface area than pure GO with a total surface area of BET MS_GO was 161.066 $m^2 \cdot g^{-1}$, while that of pure GO was 103.818 $m^2 \cdot g^{-1}$. MS_GO also has more active sites than GO.

Acknowledgements

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