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Recycling of Broken Glasswares Into an Adsorbate to Remove Both Cationic and Anionic Dyes for Waste Water Treatment

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Abstract
Chemically modified broken laboratory glass wares were proved to act as an efficient adsorbent for both anionic and cationic dyes from waste water. The glassware wastes were chemically modified as silica gel for cationic dyes adsorption. The silica gel is again modified with amphiphile for anionic dye adsorption. Both silica gel and modified silica gel were characterized with Scanning Electron Microscopy, Energy Dispersive X-ray Analysis and Fourier Transform Infrared Spectroscopy. The adsorptions of the dyes were confirmed visually as well as using UV- visible spectroscopy. The adsorption capacities of the modified silica gel were quantified using model cationic and anionic dyes. The adsorption was found to be 97.84 mg/g for cationic and 196.14 mg/g for anionic dyes by the modified silica gel. The efficiency of the adsorption was quantified at various dosages, pH, initial concentrations and contact time. The mechanism of the adsorption is predicted by applying mathematical models. This study demonstrated an efficient way of recycling the non-biodegradable broken glassware waste for the waste water treatment by modifying as an efficient adsorbate for both anionic and cationic dyes. This study doubly contributes towards the environmental protection by recycling hazardous broken glass wares an adsorbate towards clean water.

Keywords: Broken glassware, Silica gel, CTAB, Methylene Blue, Methyl Orange and Adsorption

Introduction
Glasses are a translucent and versatile materials that is most essential and commonly used because it can be easily made into a variety of shapes and used for many applications. After the end use, glasses are also one of the waste materials produced and it is difficult to eliminate because the glasses are non-biodegradable in nature [1]. The glass waste may be in the form of powder or solid which is depending on the source of generation of glass waste. Mostly glass powder composed 81% of silica, 12% of boron oxide, 4% of sodium oxide and 2% of aluminium oxide, these compositions varies depending on the various sources which includes manufacturing, production and availability and far more [2]. Generally, most of the glass wastes are used in a construction industry as a fine aggregate replacement in concrete and clicker replacement in cement production and also some of the glass wastes are chemically modified for various applications [3-6]. Even though the material is often sterilized throughout the recycling process, recovered waste glasses cannot typically be utilized for products that contact edible items due to health concerns. Hence upcycling can be the better option to decrease the pollution caused by waste glass.

The waste water is either liquid or water carrying waste, which is disposed from so many sources including residences, institutions, commercial and industrial establishments. Its discharge is often oxygen containing wastes, inorganic chemicals, toxic compounds, pathogenics that are not only affecting the environment, but also has huge health risks [7]. Among all sources of wastewater, textile industries have a significant effect because the waste water produced from this industry is highly carcinogenic in nature. Textile disposals have huge impact on environment due to the waste dyes, when the textile fibres are cleaned or washed after adding the dye, which contributes crucially to the water pollution. This textile waste water not only contains dye, but also contains other components including suspended solids, metallic solids, etc [8]. There are so many methods available commercially but they have limitations because of high cost and difficult to operate. Lot of researches are going on towards the development of low-cost adsorbent materials.

Many studies were reported on the adsorbents based on silica gel which were prepared from various sources to treat waste water [9-11]. This study aims at preparing the silica gel from laboratory broken glassware for cationic dyes, since it has a negatively charged surface. For making the silica gel which can also be able to adsorb anionic dyes, the prepared silica gel should have capacity towards anionic dyes adsorption. Hence, an attempt was made to chemically modify the borosil glass and investigate its adsorption kinetics and equilibrium studies.

Materials and Methods

Chemicals

Waste borosil glass (laboratory broken glass) was collected from Department of Chemistry, Anna university. Sodium hydroxide (NaOH, 99% purity), cetyltrimethylammonium Bromide (CTAB, 99% purity), methylene blue (MB) dye and methyl Orange (MO) dye were purchased from Sisco Research Laboratories Pvt. Ltd, and used without purification and Distilled water.

Preparation of Silica Gel

Finely crushed glass powder and NaOH at 1:3 (w/w) ratio is calcined at 400°C for 4 hours. The sodium silicate formed through alkali decomposition was dissolved in 50 ml of boiling water. The resulting solution was then filtered. The pH of the solution was adjusted about 0.5 – 2.0 by adding 3 M hydrochloric acid drop by drop to the solution while stirring at 450 rpm. The gel was formed and allowed to age for 18 hours, after which it was filtered and dried at 80°C (figure1) [12].

Preparation of Modified Silica Gel

500 ml of 1.0 mmol CTAB solution and 10 mmol prepared silica gel was stirred overnight. The resulting solution was filtered and the retained solid materials were dried for 3 hours at 120°C (figure1) [13].

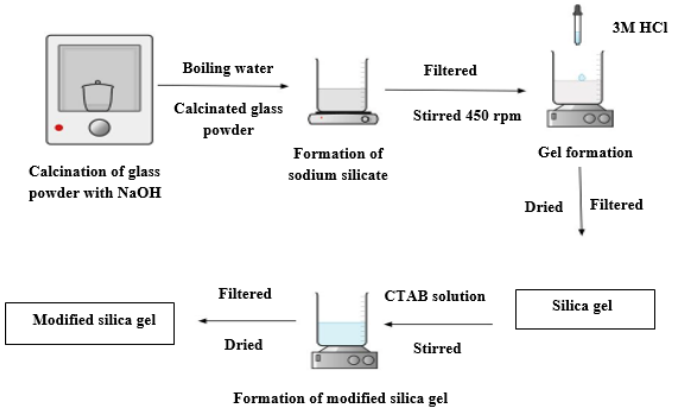

Figure 1: Schematic Representation for the Process of Formation of Silica Gel and Modified Silica Gel

Characterization

The TESCAN VEGA3 XMU Scanning Electron Microscopy (SEM) was used to observe the surface morphology of both silica gel and modified silica gel. The SEM operates at a resolution of 10 KV and is capable of capturing external morphology, orientation, and crystal structure. The FT-IR spectra of glass powder, silica gel, and modified silica gel were obtained using the Thermo Nicolet iS50 with an inbuilt ATR in KBr mode, covering a range of 400 - 4000 cm^{-1}. The thermal stability of the adsorbent was analyzed by the Diamond TG/DTA and was utilized in a nitrogen atmosphere and it was carried out over a temperature range of 30-700 °C with a heating rate of 10 °C/min.

Adsorption Experiments

The prepared silica gel and modified silica gel adsorbents were used to analyse the adsorption of MB and MO dyes from aqueous solution and the experiment was carried out using batch adsorption with varying process parameters such as initial concentration of dye solution (10-35 mg L^{-1}), pH (3 - 11), contact time (0 - 200 min.) and adsorbent dosage (2 - 14 mg) for both dyes. Initially 1000 mg L^{-1} of stock solution was prepared for both dyes. For upcoming experiments, the dye solutions of MB and MO were diluted to required concentration. The dye solution was kept in orbitek shaker for about 90 min, then 50 ml of the dye solution was used to analyse with varying parameters. Once the adsorption process was completed then the adsorbent from the dye solution was separated (desorption) using centrifugation, and the final concentrations of MB and MO in aqueous solutions were analyzed by JASCO V-730 UV-VISIBLE spectroscopy at 661.5 nm and 464 nm for MB and MO, respectively. The removal rate of dye (R) and equilibrium adsorption capacity (q_e) were calculated using Eqs (1) and (2) respectively [14].

$$R = \frac{C_o - C_e}{C_o} \times 100 \qquad (1)$$

$$q_e = (C_o - C_e) \times \frac{V}{m} \qquad (2)$$

Where C_0 (mg L^{-1}) is the initial concentration of dye, C_e (mg L^{-1}) is the equilibrium concentration of the dye, V (L) is the volume of the dye solution and m (mg) is the mass of the adsorbent.

Adsorption Isotherms

Generally, adsorption isotherms were used to analyse the interaction between adsorbent and adsorbate; which was also used to determine the adsorption capacity of adsorbent. In this study, Langmuir and Freundlich models were used to describe the mechanism of adsorption. The Langmuir model assumes the surface is homogeneous so monolayer adsorption is possible and the process is reversible. Linear form of Langmuir isotherm model was represented by the following equation (3) [12].

$$\frac{C_e}{q_e} = \frac{1}{K_L q_m} + \frac{C_e}{q_m} \qquad (3)$$

Where, Ce (mg/L) is equilibrium concentration of adsorbate, q_e (mg/g) is the amount of dye adsorbed per gram of adsorbent at equilibrium state, K_L (L/mg) is Langmuir constant related to adsorption capacity, and q_m (mg/g) is the maximum adsorption capacity.

However, the Freundlich isotherm model assumes that the surface is heterogeneous and multilayer adsorption is possible. The linear form of this isotherm model was represented by the following equation (4) [12].

$$\ln q_e = \ln K_F + \frac{1}{n} \ln C_e \qquad (4)$$

Where K_F is Freundlich constant and 1/n is the intensity of adsorption. The favorable adsorption condition is that value of 'n' is greater than 1.

Results and discussion

Characterization

Fourier Transform Infrared (FTIR) Spectroscopy

The FT-IR spectrum was used to analyse the functional group of glass powder, silica gel and modified silica gel (figure 2). For glass powder, the peaks at 953.47 cm^{-1}, 770.62 cm^{-1} and 427.18 cm^{-1} are due to siloxane rings, Si-O-Si symmetric stretching and Si-OH stretching respectively. For silica gel, the peak at 3375.57 cm^{-1} in the spectrum is due to OH stretching vibration of H_2O, peak at 1632.85 cm^{-1} shows the presence of H-O-H bending, peaks at 1053.57 cm^{-1}, 945.10 cm^{-1}, 792.82 cm^{-1}, 543.31 cm^{-1}, 433.08 cm^{-1} indicates that the presence of Si-O-Si asymmetric stretching, Si-OH stretching, vibration of Si-O symmetric stretching from Si-O-Si, siloxane rings and Si-O-Si bending vibration respectively [15]. For modified silica gel, there are three new peaks. The peaks at 2921.65 cm^{-1} and 2852.12 cm^{-1} are due to C-H bending in carbon chain of CTAB. Peak at 1467.81 cm^{-1} is due to C-H bending in alkane and peak at 3379 cm^{-1} is due to O-H asymmetric bending at siloxane group and C-N of tertiary amine [13].

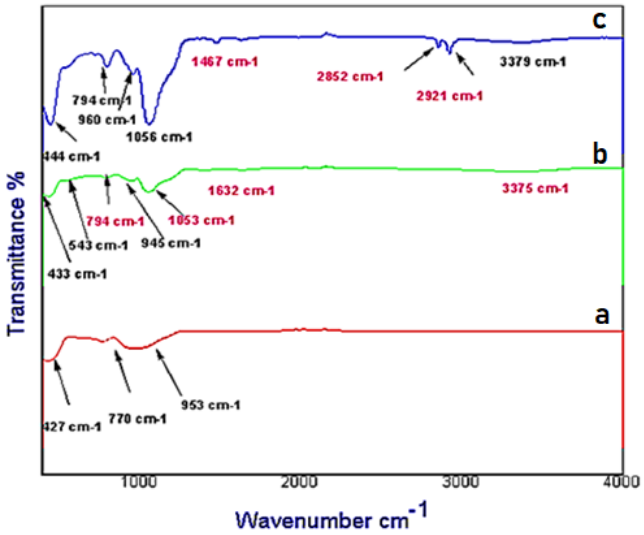

Figure 2: FTIR Spectrum for (A) Glass Powder, (B) Silica Gel, (C) Modified Silica Gel 400 – 4000 Cm^{-1}, Kbr Pellet

SEM and EDAX

The surface morphology of glass powder, silica gel and modified silica gel were analyzed using SEM images. The SEM result shows that the waste glass powder and silica gel have irregular shape and size. The surface was found to be smooth without pores (figure 3a & b), but the morphology has changed during the glass powder to silica gel conversion, the sharp edges of glass powder changed into smooth edges. Whereas, the modified silica gel shows fine aggregates

on the surface with the size of around 500 nm (figure 3c). The EDAX result shows that glass powder containing 52% of silica and 38% oxide and small amounts of other elements, and silica gel containing 65% of silica and 35% of oxide and small amounts of other elements. The modified silica gel containing 52% of silica, 31% of oxide, 9% of carbon, 2% of nitrogen and 7% of bromine and small amounts of other elements, (figure 4c) which is standing as a concrete evidence for the chemical modification of silica gel with CTAB.

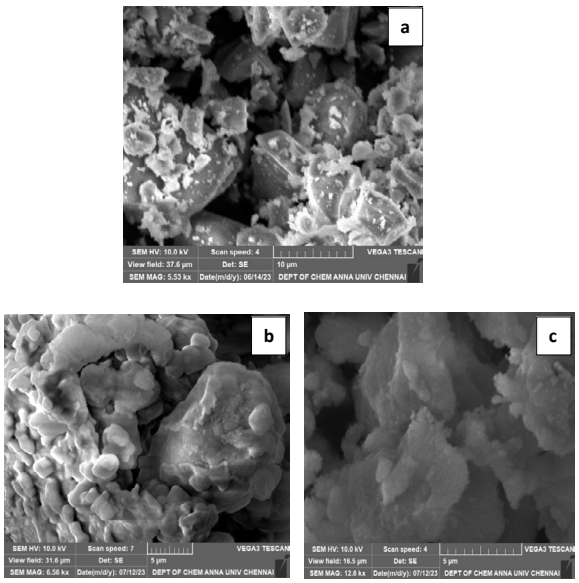

Figure 3: SEM Images of a) Glass Powder, Scale:10 µm b) Silica Gel, Scale:5 µm c) Modified Silica Gel, Scale:5 µm

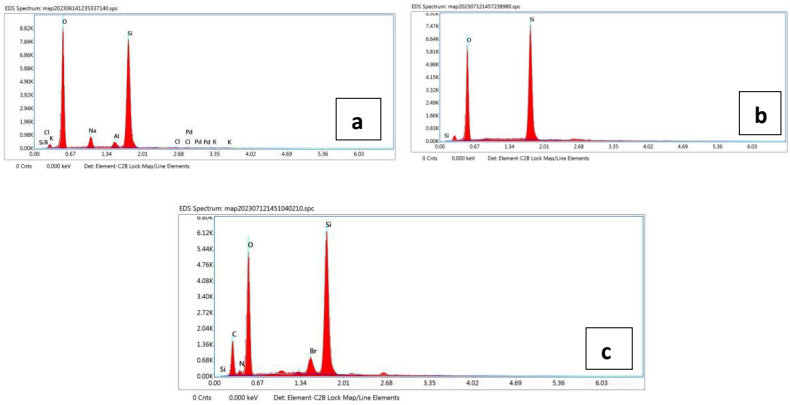

Figure 4: EDAX Images of A) Glass Powder, B) Silica Gel and c) Modified Silica Gel

Effects of Various Parameters on Dye Adsorption

Effect of Adsorbent Dosage

The effect of dosage of both silica gel and modified silica gel as adsorbent on the adsorption of MB and MO dyes was analyzed over a range of 2 - 14 mg. Fixed concentration (10 ppm) solutions of MB and MO were prepared as used for this study. The result shows that the maximum removal efficiency occurred for 10 mg of adsorbent for both dyes (figure 5). However more than 10 mg dosage, the removal efficiency becomes constant because of partial agglomeration of adsorbents that will affect the availability of active sites of adsorbents [9].

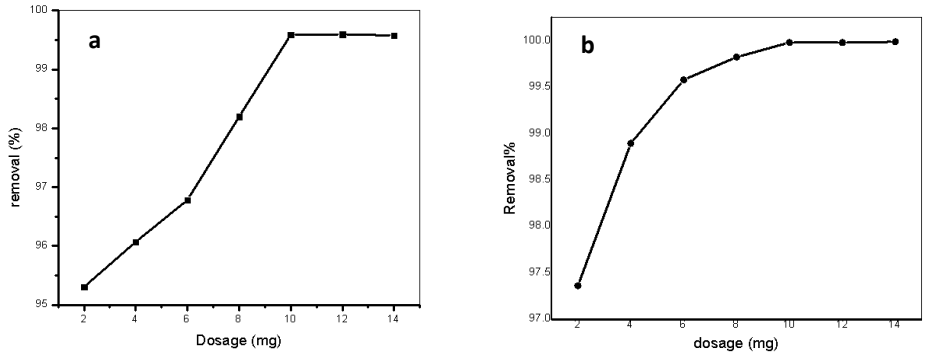

Figure 5: Plot of Dosage Vs. % Removal of a) Methylene Blue (MB) dye Vs. Silica gel, and b) Methyl Orange (MO) Dye Vs. Modified Silica Gel

Effect of pH

The pH of the dye solution in the adsorption process has a significant effect on the adsorption mechanism. In this observation, the removal efficiency was analysed for both MB and MO dyes at various pH ranges from 3 to 11 (figure 6) with the constant initial dye concentration (25 ppm), time (90 minutes) and adsorbent dosage (10 mg). For MB dye the maximum removal efficiency was observed at pH 11, may be because at higher pH, the presence of silicate ions is more, enhancing the adsorption efficiency through electrostatic attraction. Whereas at lower pH, the presence of (H+) ions are more, which may compete with MB dye, which significantly affects the removal efficiency [16]. For MO dye the high removal efficiency was observed at pH 5, because of the positive surface of modified silica gel that is due to presence of the amine head group of CTAB amphiphiles [13]. Whereas at higher pH, the efficiency is low that is due to electrostatic repulsion. At pH 3, the MO dye forms complex so it changes its colour, so at pH 3, removal % couldn't be determined.

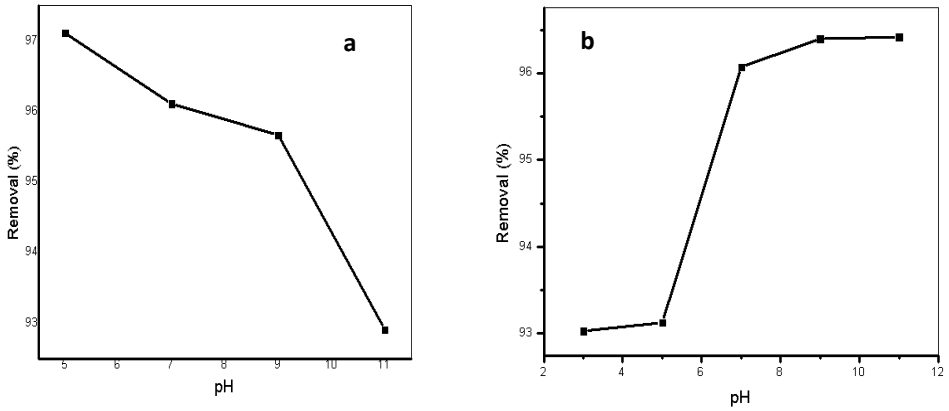

Figure 6: Plot of pH Vs. % Removal of a) Methylene Blue (MB) Dye and b) Methyl Orange (MO) Dye

Effect of Initial Concentration of Dye

In this study, the % removal at fixed adsorbent dosage of silica gel and modified silica gel (10 mg) with various initial concentrations (5 - 35 ppm) of the MB and MO at dyes were analyzed (figure 7). For MB and MO dyes, the % removal was found to decrease from 98.9% to 66.5% & 99.9% to 99.5% respectively. The decrease in % removal for MB is higher than MO, may be because of the difference in mechanism of adsorption of modified silica gel. At low concentration, as expected the removal efficiency high, may be because the number of adsorbate molecules is less than or equal to the available sites. With increase in concentration of dye, the number of available adsorbates decreases so the removal efficiency is decreasing.

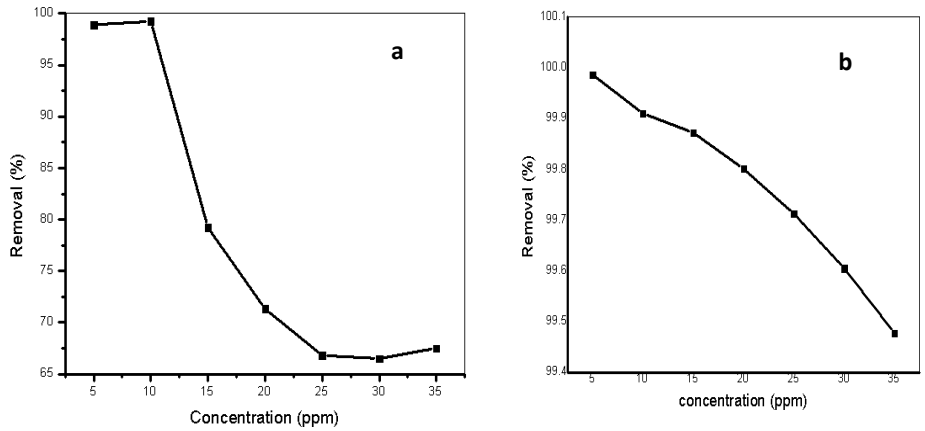

Figure 7: Plot of Initial Concentration of Dye Vs. % Removal of a) Methylene Blue (MB) Dye and b) Methyl Orange (MO) Dye

Effect of Contact Time

The effect of adsorption capacity of both silica gel and modified silica gel based on contact time was analyzed (figure 8). For this experiment 10 ppm of MB and 50 ppm of MO dye solutions was prepared separately at fixed adsorbent dosage (5 mg). The difference in the dosage of the dyes was taken to get a clear visible change of the dyes from the initial concentration. The adsorbant quantity was kept minimal to ensure the saturation of the adsorption. The adsorption saturation is attained in first 3 minutes for MB and in 15 minutes for MO. The maximum adsorption occurs for MB 3 minutes and for MO 15 minutes after that it reaches the equilibrium, this shows that initially there are more active sites available, once the adsorption sites are occupied it reaches its equilibrium state. The difference in time to attain equilibrium also suggests a different mechanism of adsorption happens in the adsorbents.

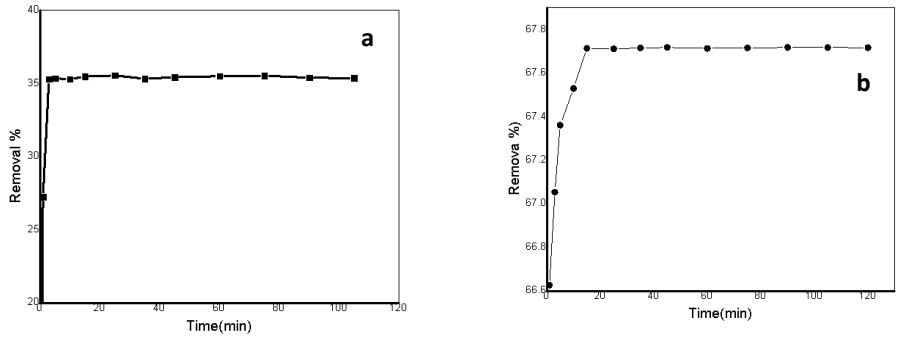

Figure 8: Plot of Contact Time (Min.) Vs. % Removal of a) Methylene Blue (MB) Dye and b) Methyl Orange (MO) Dye

Adsorption Isotherm

The Langmuir and Freundlich models were developed to describe the mechanism of adsorption. The adsorption isotherms of Langmuir and Freundlich models are shown in (figure 9) and table 1. For MB dye the correlation coefficient R^2 = 0.8809 of Langmuir model is almost nearer to 1 when compared to Freundlich model (correlation coefficient 0.76), so the adsorption isotherm of MB and silica gel adsorbent assumes that the adsorption is monolayer meaning that once the dye molecule is adsorbed by adsorbent there is no further adsorption occurs at the same site. Whereas for MO, the correlation coefficient is R^2 = 0.97 of Freundlich model is nearer to 1 so that the adsorption MO and modified silica gel adsorbent assumes that the multilayer adsorption is possible with varying levels of binding energies.

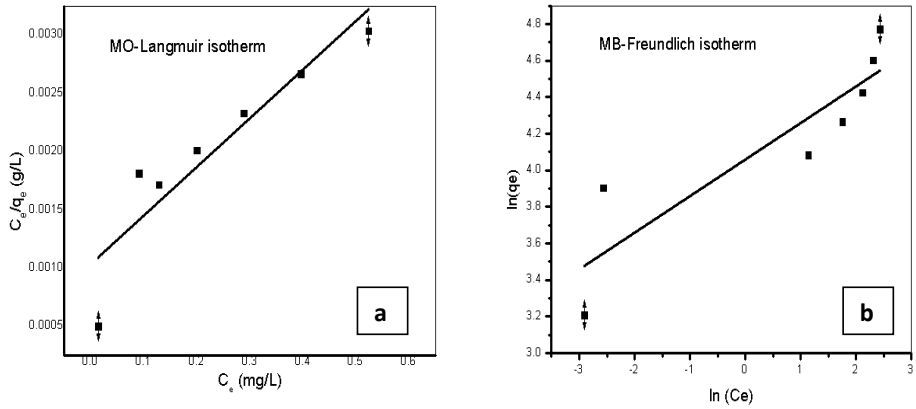

Figure 9: Fitting of Adsorption Isotherm Models of Methyl Orange by (a) Langmuir Adsorption and (b) Freundlich Adsorption Isotherm, Methylene Blue by (c) Langmuir Adsorption and (d) Freundlich Adsorption Isotherm

Table 1: Adsorption Isotherm parameters of the Dyes

Dyes	LANGMUIR			FREUNDLICH		
	q_m (mg/g)	K_L (L/mg)	R^2	n	K_F (L/g)	R^2
MB	97.84	0.93	0.87	5.00	57.85	0.76
MO	196.14	4.95	0.82	1.83	234.55	0.97

Adsorption Kinetics

The kinetic study is used to analyse the possible mechanism of dye adsorption onto the adsorbent. For these experiments the initial concentrations of MB (10 ppm) and MO (50 ppm) with 100 mL volume and the fixed adsorbent dosage (5 mg) were used. The pseudo-first and second-order equations, were used to analyze the adsorption process and the linear form is represented by the following equations (5) & (6) [16].

$$\ln(q_e - q_t) = \ln q_e - k_1 t \qquad (5)$$

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{t}{q_e} \qquad (6)$$

Where, q_e and q_t (mg/g) are the adsorption capacities at equilibrium and time t (min) respectively, k_1 and k_2 (g/mg/min) are the rate constant for pseudo first order and pseudo second order respectively.

For both dye adsorptions the pseudo-second order correlation coefficient R22=0.99 is nearer to 1 shown in figure10 and table 2. So, it assumes that the adsorption is based on chemisorption or chemical adsorption meaning that the adsorption rate is determined by the adsorption capacity rather than concentration of adsorbate.

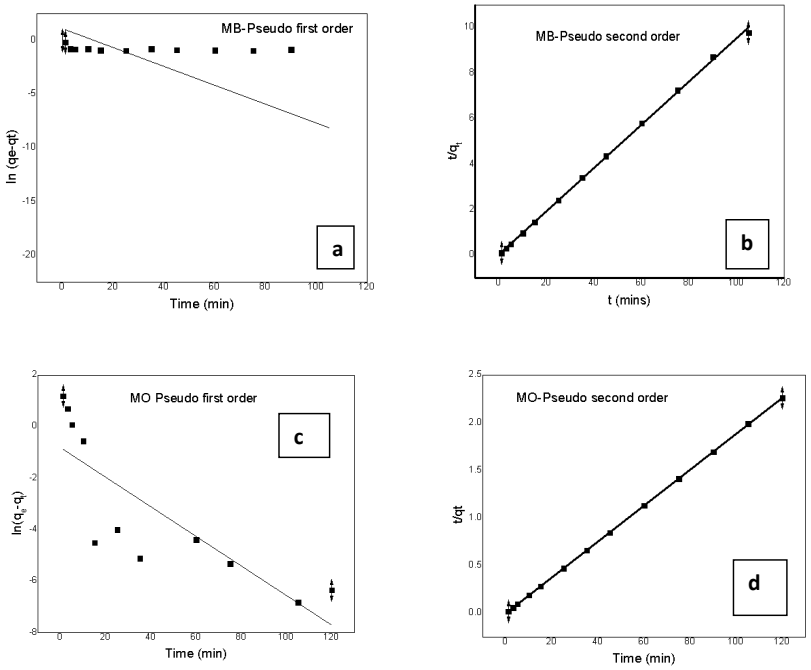

Figure 10: Adsorption Kinetics of Methylene Blue (a) Pseudo First Order and (b) Pseudo Second Order and Methyl Orange by (c) Pseudo First Order and (d) Pseudo Second Order

Table 2: Parameters of pseudo first and second order

Pseudo first order			Pseudo second order		
$q_{e,cal}$ (mg/g)	k_1 (min^{-1})	R_1^2	$q_{e,cal}$ (mg/g)	k_1 (min^{-1})	R_2^2
11.38	11.38	0.28	10.53	2.43	0.99

Proposed Adsorption Mechanism

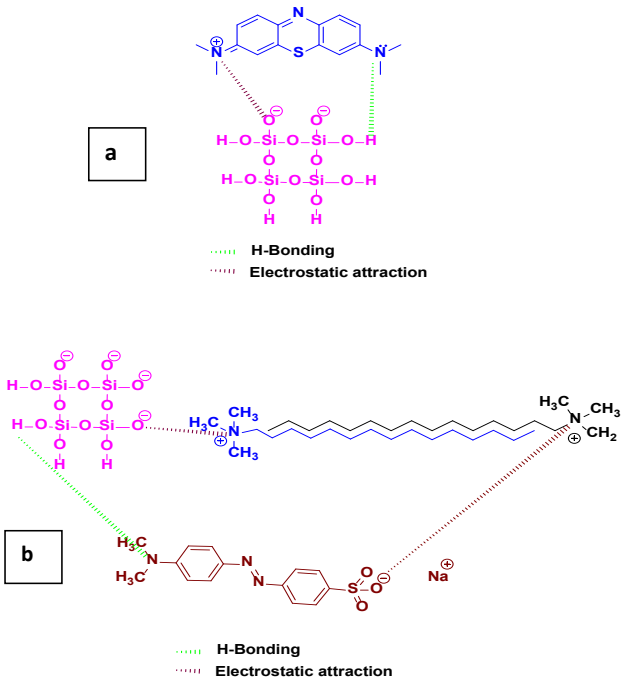

Figure 11: Schematic Representation of Adsorption Mechanism of (a) Methylene Blue on Silica Gel and (b) Methyl Orange on Modified Silica Gel

The proposed adsorption mechanism for MB and silica gel adsorbent is mainly due to electrostatic attraction between more silanol groups which are deprotonated in the form of Si-O- and the cationic dye of MB. There is a hydrogen bonding between amine group of MB dye and the silanol group (figure11) [12]. However, the adsorption mechanism of MO and modified silica gel is also mainly due to electrostatic attraction between anionic MO dye and positively charged modified silica gel. The CTAB amphiphiles were added on the silica gel for formulating modified silica gel, the bi-layer formed by CTAB on silica gel in modified silica gel, makes the modified silica gel surface to positively charged and one more possibility is the hydrogen bonding between amine group of MOs and the silanol group of modified silica gel (figure11) [13].

Dyes	Adsorbent	Adsorption capacity (mg/g)	References
MB	Fly Ash - based Geopolymer	37.04	[17]
	Wood ashes	50.00	[18]
	Natural Zeolite	23.60	[19]
	CO_3O_4/SiO_2 nanocomposite	53.87	[20]
	Silica gel (this work)	**97.84**	
MO	Calcinated organic matter-rich clays from Egypt	34.48	[21]
	Organic matter-rich clays from Egypt	41.67	[21]
	Biochar from chicken manure	39.47	[22]
	Coffee waste/cetylpyridinium chloride	62.50	[23]
	Modified silica gel (this work)	**196.14**	

Table 3: Comparison of Adsorption Capacity with Different Adsorbent for MB and MO

This study is for comparing the adsorption capacity of adsorbent, determined using Langmuir adsorption isotherm with previously available adsorbents which is reported in the previous literature (Table 3). This clearly indicated that the silica gel modified gel has the good adsorbent capacity.

Conclusion

In this study, the broken waste borosil glass is chemically modified to form silica gel and the modified silica gel is used for dye adsorption from wastewater. The silica gel is modified with amphiphiles CTAB to adsorb anionic and cationic dyes from waste water. The adsorption capacities of the modified silica gel were quantified using model dyes like Methylene blue (MB) (Cationic dye) and Methyl Orange (MO) (Anionic dye). The adsorption efficiency was found to increase with increasing the adsorbent dosage for both dyes. The percentage removal of dye was increased with an increase in dosage of both dyes. For MB dye at higher pH value and MO dye at lower pH value the adsorption increases with decreasing the contact time. The adsorption capacity of both silica gel and modified silica gel were determined using an isotherm model and that is 97.84 and 196.14 mg/g for MB and MO respectively. The best isotherm model for MB was Langmuir isotherm meaning that the adsorption is monolayer coverage with completely homogeneous surface and for MO the best isotherm was Freundlich isotherm meaning that the adsorption occurs multilayer with heterogeneous surface [17-23]. For kinetic models both MB and MO on silica gel and modified silica gel obeys the pseudo second order meaning that the adsorption is based on chemisorption or chemical adsorption.

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Declarations

There was no funding from the funding agencies for the above-mentioned research work. There are no Competing Interests.

Ethical Approval

It is not applicable for the reported research work.

Availability of data and materials

Data Availability Statement: No Data associated in the manuscript.

Conflict of Interest

There are no conflicts to declare.

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