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Adsorption of anionic dye onto ZSM-5 zeolite-based bio membrane: Characterizations, kinetics and adsorption isotherm

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Abstract

In this study, we report polyvinyl alcohol/carboxymethyl cellulose/gelatin/ZSM-5 zeolite (PVA/CMC/GEL/ZSM-5) membrane for anionic dye (rhodamine B, Rh B) removal from aqueous solution. The prepared membrane was characterized using different techniques such as Fourier transform infrared spectroscopy (FT-IR), X-ray diffraction (XRD), scanning electron microscopy (SEM), optical microscopy (OM), universal testing machine (UTM) and water contact angle respectively. XRD, FT-IR and SEM analysis indicates successful

incorporation of zeolite into PVA/CMC/GEL membrane. The improved hydrophobicity of the zeolite loaded membrane was confirmed by contact angle analysis. The Rh B removal efficiency of zeolite loaded PVA/CMC/GEL membrane was investigated through batch adsorption technique. The effect of different parameters such as initial dye concentration, zeolite dosage, contact time, temperature and pH on the adsorption was examined. Rh B dye adsorption onto the membrane followed Freundlich isotherm model. The kinetic studies revealed that Rh B dye adsorption on the membrane could be explained using pseudo-second-order model. Finally, the recyclability test revealed that the membrane exhibits good recycle efficiency and is stable after 6 recycle.

Keywords: *ZSM-5 zeolite; Gelatin; Carboxymethyl cellulose; Adsorption; Rhodamine B*

1. Introduction

Water is a basic resource which is essential for life [1]. Although our planet earth covers 71% of water, only 2% of its water is available as pure water [2, 3]. Due to anthropogenic activity, most of our water bodies get contaminated with dyes, heavy metals, microorganisms etc.[4, 5]. The industries responsible for escalating the chemical contaminant in water are textiles, cosmetics, paper, leather, pharmaceuticals, plastic, paints and varnish [6]. Dyes are one of the major sources of water pollution [7]. Dyes are organic compound which are mainly used to colour textiles, foods etc.[8, 9]. Most of the synthetic dyes are toxic, carcinogenic and mutagenic [10]. High chemical stability, large molecular weight and low biodegradability makes dyes a potent water pollutant. Even at low concentration, dyes can cause serious health issues to human as well as animals [11, 12]. Therefore, the removal of organic dyes from aqueous solution was extremely important for the environment and its species [13]. Ion-exchange, solvent extraction, precipitation, filtration, ozonation, adsorption, evaporation, reverse osmosis, flotation, electrochemical etc. are different techniques which are commonly used for the removal of dye from aqueous solution [14-18]. Adsorption process received special attention from researchers as its simple, cheap, efficient and possess good regeneration ability [19]. Large numbers of adsorbents are available for the removal of contaminants from waste water [20, 21]. Synthetic adsorbents such as carbon, clay, silica etc. have been successfully employed for the removal of both cationic and anionic dye from water [22, 23]. However, high cost and low regeneration capacity of synthetic adsorbent limits its industrial applications [24, 25].

Zeolites are naturally occurring aluminosilicate. Here the tetrahedral TO_4 ($\text{T} = \text{Si}, \text{Al}$) unit combine together to form a 3D structure with interconnected channels and voids [26, 27]. Compared with other adsorbents, zeolite possesses some superior properties such as high surface area, porosity, thermal stability, ion-exchange capacity and so on [28, 29]. These special features make them a unique material for water treatment [30, 31]. Zeolite is a good adsorbent for the removal of heavy metal from aqueous solution [32]. Zeolites based polymer composites have been successfully employed for the removal of toxic contaminants from water [33]. There are few studies on the removal of toxic dyes from wastewater using zeolites.

Habiba et al. [34] developed poly(vinyl alcohol)/chitosan/zeolite composite for the removal of methyl orange, congo red and chromium (VI) from wastewater. The authors observed that zeolite incorporated composite has high removal efficiency (94%) at an initial dye concentration of 125 mg/L. Therefore, it was suggested as a potential candidate for wastewater treatment. The same group [35] fabricated poly(vinyl alcohol)/chitosan/zeolite composite through electrospinning method. They observed that the addition of zeolite improved the physical and adsorption properties of the membrane. Baberi et al. [36] fabricated PVA/4A zeolite membrane for methylene dye removal from water. They found that the prepared membrane exhibits high adsorption capacity (9.18 mg/g) towards methylene blue. Another group [37] developed an economical membrane from cellulose/bentonite zeolite. Their studies revealed that 99% of the brilliant green dye was removed using the composite. The negative charge on the zeolite is responsible for attracting the cationic dye. Zeolite containing both micro and mesopores are termed as hierarchical zeolites and are superior to conventional zeolite. Therefore, in our previous studies we have synthesized hierarchical zeolite and applied it for the development of polymer membranes. Our group have successfully employed PVA/CMC/zeolite for methylene blue dye adsorption from aqueous solution. Maximum removal efficiency (99%) was achieved at 0.3 g of zeolite dosage [13]. Due to the electrostatic attraction between the negatively charged dye molecule and positively charged membrane. Similarly, PVA/agar/ZSM-5 zeolite membrane exhibits good Congo red dye removal efficiency (89%). The high surface area and porosity of zeolite will enhanced the adsorption capacity PVA/agar membrane [20].

The aim of the present study is to synthesis hierarchical zeolite and employed it for the fabrication of PVA/CMC/GEL membrane for Rh B removal from aqueous solution. The polymers used in this study was eco-friendly and non-toxic polymers such as (polyvinyl alcohol) PVA, (carboxymethyl cellulose) CMC and gelatin. The prepared membrane was

characterized using different techniques such as XRD, FT-IR, SEM, TGA and OM. The Rh B adsorption property of membrane was studied by varying parameters such as pH, temperature, time, adsorbent amount and initial dye concentration. The kinetic and adsorption isotherm were used to determine the mechanism of dye adsorption. Finally, the recyclability tests were also studied.

2. Experimental section

2.1 Materials

Polyvinyl alcohol (PVA, (C_2H_4O , hydrolysis degree: 99%) was obtained from Ajax Finechem Pvt. Ltd. (Thailand). Rhodamine B ($(C_{28}H_{31}ClN_2O_3)$, Rh B) and gelatine was purchased from Merck (Thailand). The cross-linking agent glutaraldehyde ($C_5H_8O_2$; GA) was purchased from Loba Chemie Products Limited. (Thailand). Carboxy methyl cellulose (CMC, $(C_6H_7O_2(OH)_x(OCH_2COONa)_y)_n$) and chitosan ($C_{18}H_{35}N_3O_{13}$) was provided by Chemipan (Thailand). Tetra propyl ammonium hydroxide ($((CH_3CH_2CH_2)_4NOH$, TPAOH), tetraethyl orthosilicate ($C_8H_{20}O_4Si$, TEOS) and aluminum isopropoxide ($C_9H_{21}AlO_3$, AIP) were procured from Sigma Aldrich Co. Ltd (India). All the chemicals were analytical grade and were used without further modification. The polymer solution and dye solution were prepared in distilled water.

2.2 Preparation of ZSM-5 zeolite

The synthesis procedure for hierarchical zeolite is as follows; initially 0.13 g of aluminium source (AIP) and structure directing agents (tetra propyl ammonium hydroxide) were mixed in distilled water under constant stirring at room temperature for 3 h. To the properly mixed solution, silica source i.e., tetraethyl orthosilicate was added and the mixture was further stirred to form homogenized solution. This was followed by the addition of 2 g of chitosan (meso template). The resultant mixture was sonicated for 1 h in water bath to get a uniform dispersion. The mixture was then transferred into an autoclave and aged at $100^\circ C$ for 6 h for further crystallisation. The product was collected by filtration, washed with distilled water, dried in oven, and then calcined at $550^\circ C$ for 5 h in a muffle furnace to remove the meso/micro template.

2.3 Membrane fabrication

To the freshly prepared PVA solution, 1 wt% CMC and 1.5 wt% gelatine was added. The solution was continuously stirred for 5 h to get uniform dispersion. Into the above solution, different weight percentage of zeolite which was previously sonicated in water bath was added

and the solution was stirred for whole day. The cross-linking of the polymers was achieved by addition of GA and 3% HCl. Finally, the solution was poured into clean petri dish to form uniform thickness film. The membrane was dried at room temperature for 5 days and peeled off from the petri dish. For comparison studies, the plain membrane was prepared in similar manner without the addition of zeolite. The membrane loaded with 0, 1, 3 and 5 %. Zeolite was denoted as PCGZ-0, PCGZ -1, PCGZ -2 and PCGZ -3 respectively. The membrane thickness was measured by screw gauge at different positions and the thickness was found to be 28 ± 2 μm .

Table 1: The composition of the casting solution

Sample code	PVA	CMC	GEL	ZSM-5 zeolite
PCGZ-0	9	1	1.5	0
PCGZ-1	9	1	1.5	1
PCGZ-2	9	1	1.5	3
PCGZ-3	9	1	1.5	5

2.4 Characterisation

The surface morphology of the membrane was performed using scanning electron microscope (FEI Quanta 450) with an accelerating voltage of 5 kV and optical microscope (Olympus BX43 series) instrument. FT-IR spectroscopy was used to analysis the chemical composition of the membrane (Jasco 4700). The spectral was scanned in the range of $450\text{-}4000\text{ cm}^{-1}$ with a resolution of 4 cm^{-1} . The tensile strength and elongation at break were calculated by UTM (Universal testing machine, QC-506M1-204) with 10 kN load capacity and a crosshead speed of 10 mm/min. The hydrophilic/hydrophobic nature of the membranes was accessed using contact angle analysis (Drop Meter SCA data physics). Here, a small water droplet was released using inverted needle and droplet was directly placed on the membrane. The droplet was captured using microscope which was coupled with camera. The measurement was done in triplicate and average value was presented. Swelling experiment was carried out to understand the durability of membrane in water. Initially, membrane was weighed and subsequently immersed in 50 mL of distilled water for 48 h at room temperature. At different intervals, the swollen sample is removed from water, wiped and weight is noted. The process was continued until a constant weight is observed.

The percentage of swelling calculated by

$$\text{Swelling ratio} = \frac{W_s - W_d}{W_d} \times 100 \quad \text{————— (1)}$$

where W_d and W_s are the weight of dry and swollen membrane.

2.5 Adsorption experiment

The adsorption capacity of the PVA/CMC/GEL/ZSM-5 membrane was examined using a model dye; Rh B. For adsorption studies the membrane was agitated with 60 mL of Rh B solution and after certain time period it was separated, filtered and the absorbance was measured using UV-visible spectrophotometer at a wavelength of 554 nm. The percentage dye removal efficiency (R%) and equilibrium adsorption capacity (q_e) were evaluated using the equations.

$$q_e = \frac{(C_0 - C_e)V}{W} \quad \text{————— (2)}$$

$$R(\%) = \frac{C_0 - C_e}{C_0} \times 100 \quad \text{————— (3)}$$

Here C_0 and C_e represent the initial (mg/L) and equilibrium (mg/L) concentrations of Rh B respectively; V (L) is the volume of dye solution and W (g) is the weight of the adsorbent.

2.6 Reusability

The reusability test is an important factor for practical applications. The recyclability study was carried out by mixing 0.1 g of the membrane with 50 mL of dye solution to ensure the maximum contact between the membrane and dye molecules. Later the dye laden membrane was taken out, wash several times with distilled water and stirred with 1 % HCl. The selection of solvent was based on the desorption efficiency of Rh B dye form the membrane. After the desorption process, the membrane was separated, washed, dried and reused for next cycle. The process is repeated for another six cycle to evaluate the reusability property of the membrane [30].

3. Results and discussion

3.1 Characterization of PVA/CMC/GEL/ZSM-5 membrane

Figure 1 illustrate the FT-IR spectra of plain PVA/CMC/GEL and zeolite loaded PVA/CMC/GEL membrane. The broad band observed at 3280 cm^{-1} is characteristics of -OH stretching of PVA, CMC and gelatine respectively. The peak located at 2920 cm^{-1} can be attributed to C-H stretching of polymers. The two bands observed at 1660 and

1420 cm^{-1} corresponds to asymmetric and symmetric stretching of -COOH group of PVA and CMC respectively. The absorption peak at 1065 cm^{-1} is assigned to Si-O stretching of zeolite. The main peak observed at 520 cm^{-1} corresponds to Si-O-Si and Al-O-Si respectively. The emergence of new peaks at 1106, 802 and 441 cm^{-1} for zeolite loaded membrane corresponds to Si-O and Al-O bond stretching [38-41]. The surface morphology of membrane was observed through SEM technique. As can be seen in Figure 2, zeolite incorporation has a significant effect on the surface roughness of PVA/CMC/GEL membrane. It is also evident that membrane loaded with high percent of zeolite (5wt%) has greater particle distribution. However, at high zeolite dosage (>5wt%), the particles tend to agglomerate and thereby reduce its adsorption ability. Therefore, we choose 5 wt% zeolite as the optimum zeolite dosage. These observations were complementary to optical microscopic analysis and confirmed the successful incorporation of zeolite in the membrane (Figure 3) [38]. X-ray diffraction pattern of plain PVA/CMC/GEL and zeolite loaded PVA/CMC/GEL is displayed in Figure 4. The semi crystalline nature of polymers was observed at $2\theta = 19.9^\circ$. The diffraction peak at $2\theta = 24.3^\circ$ and 25.6° for zeolite loaded membrane is attributed to the MFI structure of zeolite. It can also observe that with increase in zeolite dosage, the intensity of characteristic peak of zeolite at $2\theta = 24.3^\circ$ increases. Meanwhile, a diminishing intensity was noted for peaks at $2\theta = 19.9^\circ$ [34, 42, 43]. The wettability of the membrane was monitored by contact angle analysis and the results are presented in Figure 5. Owing to its hydrophobic nature, zeolite will decrease the hydrophilic nature of PVA/CMC membrane. Therefore, on increasing the zeolite percentage, the value of water contact angle also increases and a maximum contact angle was observed (85°) for 5 wt% zeolite loaded membrane. Consequently, the water droplet on the membrane become spherical with zeolite dosage. Swelling is an important parameter for adsorption application. The swelling capacity of membrane depends on cross-linking density, pH and temperature. PVA, gelatine and CMC are hydrophilic polymers and therefore it has greater affinity for water. Hence, we observed high degree of swelling (97%) for PVA/CMC/GEL/ZSM-5 membrane. However, the addition of zeolite decreases the swelling. This is ascribed to the hydrophobic nature of zeolite. The membrane with desirable mechanical properties is preferred for water treatment application. This prompted us to examine the mechanical property of PVA/CMC/GEL/ZSM-5 membrane. The effect of ZSM-5 zeolite on the mechanical properties of PVA/CMC/GEL membrane is shown in Table 1. It can be seen that tensile strength of the membrane increases with increase in zeolite loading. This implies

that zeolite particle acts as filler and thereby strengthen the polymer matrix. However, a reduction in elongation at break value was noted with zeolite content [44, 45].

Figure 1. FT-IR spectra of PVA/CMC/GEL/ZSM-5 membranes: (a) PCGZ-0; (b) PCGZ -1; (c) PCGZ -2; (d) PCGZ -3

Figure 2. SEM images of different zeolite loaded PVA/CMC/GEL membranes

Figure 3. Optical microscopic images of different zeolite loaded PVA/CMC/GEL membranes

Figure 4. XRD pattern of different zeolite loaded PVA/CMC/GEL membranes: (a) PCGZ-0; (b) PCGZ -1; (c) PCGZ -2; (d) PCGZ -3

Figure 5. Water contact angle of different zeolite loaded PVA/CMC/GEL membranes

Figure 6. Degree of swelling of different zeolite loaded PVA/CMC/GEL membranes

Table 1. Mechanical properties of PVA/CMC/GEL/ZSM-5 membrane

Samples	Tensile Strength (N/mm ²)	Elongation at break (%)
PCGZ-0	10.69	62.17
PCGZ-1	33.34	35.96
PCGZ-2	52.73	34.50
PCGZ-3	63.15	28.65

3.2 Adsorption experiments

The effect of zeolite dosage on the Rh B dye removal was investigated and the result is shown in Figure 7(a). It can be noted that on increasing the amount of zeolite from 1 to 5%, the adsorption capacity of the membrane increases and a maximum adsorption efficiency (97%) was noted for 5 wt% loaded membrane. The rapid enhancement observed for zeolite loaded membrane is due to the large surface area and availability of more active sites present on the adsorbent [46, 47]. Scheme 1 show the schematic representation of different percentage of zeolite on PVA/CMC/GEL membrane will interact with Rh B dye molecule. However, no significant improvement in the removal efficiency was noted for membrane loaded with zeolite percentage greater than 5 wt%. This could be due to the agglomeration of zeolite particle on the surface of membrane. The initial dye concentration plays a major role in controlling the removal percentage and adsorption capacity of membrane. The effect of initial dye concentration on the dye removal ability of PVA/CMC/GEL/ZSM-5 zeolite membrane is show in Figure 7(b). On increasing the initial dye concentration, the effective collision between the membrane and Rh B molecule increases. This leads to enhancement of adsorption capacity. Meanwhile, a reduction in removal efficiency was noted with increase in initial concentration. Similar trend was reported by Liu, Han and Vimonses [48-50] and it was attributed to the reduction of adsorption sites on the membrane. pH will affect the charge and surface binding site of an adsorbent. Therefore, we have varied the pH of solution from 2 to 10 and studied its effect on Rh B removal (Figure 7(c)). At low pH, the solution contains large number of H⁺ ions and thereby protonate the membrane. Consequently, the membrane will repel positive charge Rh B dye molecule and the adsorption capacity decreases. Furthermore, the excess H⁺ on the aqueous solution will compete with the cationic dye (Rh B) for the same adsorption site. This is also responsible for the low adsorption capacity at acidic pH. However, at basic pH, the membrane has greater tendency to attract positively charged cationic dye through electrostatic

attraction. The maximum adsorption capacity of Rh B on the membrane at basic pH was 10 [51, 52]. Figure 7(d) show the effect of contact time on Rh B dye removal efficiency of PVA/CMC/GEL/ZSM-5 zeolite membrane. In the initial contact time, due to the availability of the large number of free adsorption sites, the Rh B dye adsorption occurs very rapidly. This is reflected in the adsorption capacity value ($q_e=4.85$). After 1.30 h, the adsorption capacity increases only slowly with contact time and finally after 3 h it reaches equilibrium. This is due to the saturation of binding site [53-55]. Temperature plays an important role on the dye adsorption process. We have monitored the dye adsorption capacity of the membrane at different temperature range i.e., 30 to 60 °C (Figure 7(e)). The result revealed that on increasing the temperature from 30 to 60°C, the adsorption capacity decreases. This indicates that adsorption of Rh B onto the membrane is exothermic in nature [51].

Scheme 1: Rhodamine B dye molecules adsorbed on the membrane

Figure 7. The effect of different parameters such as a) zeolite dosage; b) initial dye concentration; c) pH d) contact time; and e) temperature on the removal of Rh B dye using PVA/CMC/GEL/ZSM-5 membrane (Experimental condition: adsorbent dosage = 5 wt%, initial dye concentration = 10 ppm, contact time = 180 min, pH=10 and temperature= 30°C)

3.3 Adsorption isotherm

The adsorption isotherm is employed to explain the distribution of adsorbate between the solid and liquid. In this study, the relationship between the adsorbate and the adsorbent was understood using two well-known isotherm models namely Langmuir and Freundlich isotherms

Langmuir isotherm

Langmuir isotherm model indicates monolayer adsorption on the homogeneous surface. The linear form of Langmuir isotherm model is as follows [52]

$$\frac{C_e}{q_e} = \left(\frac{C_e}{q_m}\right) + \left(\frac{1}{K_L * q_m}\right) \quad \text{————— (4)}$$

where q_e and C_e corresponds to the equilibrium adsorption capacity (mg/g) and equilibrium concentration of adsorbate (mg/L) respectively. K_L is Langmuir constant and is related to the affinity of binding site. q_m is monolayer adsorption capacity (mg/g). The value of q_e and K_L were obtained from the slope and intercept of the linear plot C_e/q_e against C_e (Figure 8). The nature of adsorption is related to dimensionless constant R_L (separation factor) which is mathematically represented as follows;

$$R_L = \frac{1}{1 + K_L C_0}$$

The value of R_L between 0-1 indicates a favorable adsorption. If the value of R_L is not in the aforementioned range it is considered as unfavorable ($R_L > 1$), linear ($R_L = 1$) and irreversible ($R_L = 0$).

Freundlich isotherm

Freundlich isotherm model assumes a multilayer adsorption and thus suggests a heterogeneous adsorbent surface [56]. The linearized form of the equation is

$$\ln(q_e) = \ln K_F + \frac{1}{n} * \ln(C_e) \quad \text{————— (5)}$$

Here C_e is the equilibrium concentration of Rh B solution (mmol/L), q_m is the maximum absorption capacity (mg/g), q_e is the amount of Rh B dye adsorbed at equilibrium and K_L is a Langmuir isotherm constant. The value of $q_{\max}$ and K_L values are calculated from the intercept and slope of the graph plotted against $\ln q_e$ versus $\ln C_e$ (Figure 8).

The value of $1/n$ is used to predict the favourability of the adsorption. The value between 0 and 1 implies a favourable adsorption. If the value is greater than zero then it is considered as irreversible whereas $1/n = 0$ represent an unfavourable adsorption. The values of isotherm parameters are shown in Table 2. It can be seen clearly from table 2 that adsorption data fitted well in the Freundlich model. Also, the correlation coefficient (R^2) for Freundlich model ($R^2:0.999$) fitted better than that of Langmuir model ($R^2:0.97$). The result thus implies that the adsorption of Rh B dye onto PVA/CMC/GEL/ZSM-5 zeolite membrane is heterogeneous in nature [50, 57].

Figure 8. Isotherm model plots for the adsorption of Rh B dye removal using PVA/CMC/GEL/ZSM-5 membrane (Experimental condition: adsorbent dosage = 5 wt%, initial dye concentration = 10 ppm, contact time = 180 min, pH=10 and temperature= 30°C)

Table 2. Parameter values of adsorption isotherm

Experimental	Langmuir isotherm			Freundlich isotherm		
$q_e, \text{exp (mg g}^{-1}\text{)}$	R^2	$q_{\text{max}}(\text{mg g}^{-1})$	K_L	R^2	$K_F(\text{mg g}^{-1})$	n
4.85	0.977	25.57	0.476	0.999	8.2	2.2

3.4 Adsorption kinetics

In order to predict the adsorption mechanism of Rh B onto PVA/CMC/GEL/ZSM-5 zeolite membrane, both pseudo-first-order (PFO) and pseudo-second-order (PSO) models were used. The linearized form of PFO and PSO models are expressed by equation 6 and 7.

$$\log(q_e - q_t) = \log q_e - \frac{K_1 t}{2.303} \quad \text{————— (6)}$$

$$\frac{t}{q_e} = \frac{1}{K_2 q_e^2} + \frac{t}{q_e} \quad \text{————— (7)}$$

where q_t represents the amount of Rh B adsorbed on PVA/CMC/GEL/ZSM-5 zeolite membrane at time 't' and q_e is the amount of Rh B adsorbed on PVA/CMC/GEL/ZSM-5 zeolite membrane at equilibrium (mg/g) respectively. K_1 and K_2 are pseudo-first-order and pseudo-second-order rate constant with unit min^{-1} and g/mg min respectively. The equilibrium adsorption capacity (q_e) and kinetic parameter such as rate constant (K_1 and K_2) were determined from the slope and intercept of the linear plot of PFO ($\log (q_e - q_t)$ vs t) and PSO (t/q_t vs t). The value of kinetic parameters (K_1 , K_2 , q_e and R^2) for dye adsorption is listed in Table 3. From the table 3, it is clear that the adsorption data fitted well with PSO with high correlation value ($R^2=0.97$). It can be also noted that the experimental and calculated q_e values was in good agreement with pseudo-second-order. Therefore, PSO model is most suitable to explain the kinetic of Rh B adsorption on PVA/CMC/GEL/ZSM-5 zeolite membrane than PFO model which has low correlation coefficient (R^2) [48, 58, 59].

Figure 9. PFO and PSO kinetic models related to the adsorption of Rh B on PVA/CMC/GEL/ZSM-5 zeolite membrane (Experimental condition: adsorbent dosage = 5 wt%, initial dye concentration = 10 ppm, contact time = 180 min, pH=10 and temperature= 30°C)

Table 3. Summary of parameter values of adsorption kinetics for Rh B onto PVA/CMC/GEL/ZSM-5 zeolite membrane

Experimental		PFO		PSO		
$q_e, \text{exp (mg g}^{-1}\text{)}$	$K_1(\text{min}^{-1})$	$q_e \text{ (mg g}^{-1}\text{)}$	R^2	$K_2 \text{ (g mg}^{-1} \text{ min}^{-1}\text{)}$	$q_e \text{ (mg g}^{-1}\text{)}$	R^2
4.85	-0.020	4.64	0.63	0.0031	6.17	0.97

3.5 Reusability

Reusability tests is very essential for practical applications. The recyclability of PVA/CMC/GEL/ZSM-5 zeolite membrane is shown in Figure 10. It can be seen that even after six successive cycles, the membrane possesses high removal efficiency. A slow decrease in the removal efficiency was noted on moving from first to sixth recycle. This is due incomplete removal of adsorbed dye from the membrane.

Figure 10. Reusability of Rh B dye after adsorption from PVA/CMC/GEL/ZSM-5 membrane

Conclusion

In this study, we developed ZSM-5 zeolite incorporated PVA/CMC/GEL membrane and applied it as an adsorbent for Rh B removal from the aqueous solution. The membrane was characterized using different techniques such as XRD, FT-IR, SEM, optical microscopy, contact angle and UTM. We observed that the with the addition of zeolite, the physical properties (mechanical, crystallinity etc.) as well as the adsorption properties of the membrane gets improved. The batch adsorption studies shows that initial dye concentration, pH, zeolite dosage and temperature play a major role in the Rh B dye removal using PVA/CMC/GEL/ZSM-5 zeolite membrane. The maximum adsorption was achieved at pH-10 with initial dye concentration-10 ppm. The adsorption data was well fitted with the Freundlich isotherm and pseudo-second-order model. Recyclability studies shows that membrane retains good adsorption capacity (91%) even after six cycles. Our result thus suggests that PVA/CMC/GEL/ZSM-5 zeolite membrane is an efficient adsorbent for the dye removal from aqueous solution.

Declarations

Ethics approval and consent to participate

The article we have submitted to the journal for review is original, has been written by the stated authors and has not been published elsewhere. The images that we have submitted to the journal for review are original, and has not been published elsewhere. This manuscript has not been submitted to, nor is under review at, another journal or other publishing venue. The

authors have no affiliation with any organization with a direct or indirect financial interest in the subject matter discussed in the manuscript.

Competing interests

The authors don't have any conflicts of interests.

Consent for publication

The authors confirm the consent for publication

Availability of data and material

All data generated or analysed during this study are included in this published article

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Compliance with ethical standards

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