Synthesis and Adsorption of Alginate and Starch-Based Hydrogels for Cationic Dye from Aqueous Solution: Thermodynamic and Isotherm Modeling Non-linear

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Received: July 13, 2023
Accepted: January 22, 2024

DOI: 10.22146/ijc.86908

Abstract: *This work prepared new eco-friendly biocomposite hydrogels by graft co-polymerization from starch (ST), sodium alginate (SA), acrylamide (AM), and acrylic acid (AC). The performance of starch graft-poly(acrylamide-co-acrylic acid) (ST-g-p(AM-co-AC)) and sodium alginate graft-poly(acrylamide-co-acrylic acid) (SA-g-p(AM-co-AC)) absorbent hydrogels was evaluated for efficient dye removal from aqueous solution due to their unequal network structure and a restricted number of the hydrophilic groups. Adsorption characteristics of the as-prepared hydrogels were tested for methylene blue (MB) as an adsorbate. The removal percentage increases when increased the adsorbent doses of both hydrogels due to the increase of active sites of the hydrogel. The isotherm models and thermodynamic studies of MB dye on hydrogels have been assessed at several conditions like adsorbent dosage, solution temperature, and equilibrium time. The equilibrium results followed the Freundlich model. The thermodynamic parameter indicated that MB dye adsorption on hydrogels was endothermic and spontaneous. On the basis of the obtained result, the hydrogels are environmental and expansive adsorbent that might be a reliable alternative to elimination dyes from aqueous solution.*

Keywords: *hydrogel; dye; starch; alginate; isotherm; thermodynamic*

■ INTRODUCTION

Adsorption by using eco-friendly, low-cost, and efficient adsorbent is a successful method for dyes removal from aqueous solution. Thus, various works have been done for the preparation of efficacious and low-expense adsorbents from biological, inorganic, and natural wastes. A composite of starch, sodium alginate, cellulose, acrylamide, acrylic acid, and hydroxyapatite is one of the safe substances, which can show the required properties as eco-friendly and efficacious adsorbent for the elimination of dyes from aqueous solution [1-4].

Dyes are utilized as significant industrial raw materials in several implementations like textile, paper, and plastic industries. The improper discharge of dye including wastewater streams into the environment that can lead to many ecological difficulties. In overall, dyes can be classified into anionic, cationic, and non-ionic categories. Different studies have confirmed that cationic dyes are more damaging because they simply bind surfaces [5-6]. Methylene blue (MB) is one of the synthetic cationic dyes that is extensively utilized in industries like pesticides, paper, cosmetics, textile, and printing due to its high stability and solubility in water [7-10].

Hydrogels are appealing biomaterials for three-dimensional networks and engineering applications. The making ready of hydrogels utilizing alginate and gelatin supply cross-linked hydrophilic polymers that can swell but non-dissolve in water. Several hydrogels have a set of functional groups (ionic), like hydroxyl, carboxylic acid, sulfonic acid, and amine groups, which make the hydrogel attractive to extremely absorbable organic and inorganic pollutants [11-14]. Acrylamide-derived hydrogels have received major attention for being utilized as assist carriers in biomedical engineering [15-17]. The researchers used a group of adsorbents made from alginate, starch, chitosan, and others, and the removal percentage was not satisfying [18-19].

In this research, environmentally friendly hydrogels based on natural polymers (starch and sodium alginate) were prepared to remove the toxic MB dye. The effect of several factors affecting the adsorption process for the removal MB dye, such as equilibrium time, temperature solution, adsorbent's dose, and regeneration of hydrogels are studied. Adsorption isotherms and several thermodynamic parameters are achieved.

■ EXPERIMENTAL SECTION

Materials

Acrylamide (AM, 98%, Sigma Aldrich) and acrylic acid (AC, 98%, Aldrich) were utilized as a monomer, while sodium alginate (SA, 99%, Aldrich) was utilized as the backbone for grafting of AM and AC fragments. *N*,*N*-methylene bis acrylamide (MBA, 99%, Aldrich) and potassium per-sulphate (KPS, 97%, Aldrich) were used as cross-linked and redox initiators in that order. MB (Sigma Aldrich) was a cationic dye. All substances were pure chemically of analytical grade and utilized directly as received without previous purgation.

Instrumentation

To determine the morphology and size of the prepared samples, the powder was analyzed by different techniques. The crystal structure of hydrogel nanocomposite of hydrogel was characterized by utilizing a XRD Bruker AXS Gumby (Germany) in a range scan of 5°–140°. The morphology of hydrogel nanocomposite before and after adsorption was observed by field-emission scanning electron micrographs (FESEM) utilizing TESCAN (Czechia). Transmission electron microscope (TEM) measurements utilized Leo (Germany) while the UV–vis spectra of the MB solution were recorded utilizing a spectrophotometer Shimadzu 1700 from Japan.

Procedure

Adsorption experiments

All adsorption experiments were conducted at a speed constant of 200 rpm by means of conical flasks 100 mL including 0.1 g of two hydrogels in 100 mL of MB dye at 25 °C. The MB solution (10–100 mg/L) with several quantities of hydrogels (0.03–0.12 g) and at various temperatures (283, 293, and 303 K). The conical flasks were shaken for 30 min to allow enough gestate time for equilibrium to be established among the hydrogels and MB solution. UV-vis spectrophotometer (λ_{max} = 604 nm) was used to determine the residual concentration of MB. The amount of MB dye adsorbed at equilibrium Q_e (mg/g) and removal percentage are calculated in Eq. (1) and (2):

$$Q_e = \frac{(C_0 - C_e) \times V_L}{m_g} \quad (1)$$

$$\%E = \frac{C_0 - C_e}{C_0} \quad (2)$$

where C_0 is initial concentration, C_e is equilibrium concentrations, Q_e is adsorption capacity, %E is percentage of removal, m_g is the mass of adsorbent and V_L is the volume of solution.

Preparation of starch graft-poly(acrylamide-co-acrylic acid) hydrogel

Ecofriendly biopolymer hydrogel was prepared by co-polymerization method. The grafting reaction among ST, AM, and AC. The ST gelatinizes under the effect of shear forces and moisture, which devastates the hydrogen bonds among starch. The structure of ST grafting reaction with two monomers (AM and AC) produces the network structure. The ST and two monomers were mixed by stirring ultrasonic. In the presence of a crosslinking (MBA) and initiator (KPs), free radicals were generated on the ST molecular chain, which reacted with the double bond of AM and AC. To develop swelling of the ST-p(AM-co-AC), a NaOH solution was added throughout the last step of the reaction. NaOH can change the amide groups (O=C–NH_2) on the side chains of polyacryl amide into a more hydrophilic carboxylate (–COONa) group at 65 °C [20].

Preparation of sodium alginate graft-poly(acrylamide-co-acrylic acid) hydrogel

The SA-p(AM-co-AC) sorbent by free radical co-polymerization method. The AM, SA, and AC precursors were dissolved in deionized water. Briefly, AC (0.1 g in 5.0 mL), AM (0.6 g in 5.0 mL), and SA (2.0 g in 20.0 mL) were homogenized via ultrasonic. The SA solution was added dropwise to AM and AC solution under constant stirring at room temperature. After that,

2.00 mL crosslinking MBA 0.05 g and 3.00 mL Initiator KPS 0.03 g were added to the mature solution for 3 h for stirring to ensure complete mixing of solutions followed via purging with N_2 gas for 3 min. The homogeneous mixture was heated at 70 °C for 3 h, which led to the synthesis of SA-p(AM-co-AC). The hydrogel product was cooled and washed in water and dried at 65 °C in an oven to obtain powder use in the experiment.

Swelling measurement of hydrogel

The behavior of sensitive pH swelling (%) of graft and co-polymeric hydrogels was thoroughly tested at 25 °C and solution pH 4. Exactly the weight dries of hydrogels were put into 200 mL DW, followed by recording its weight. After 1–10 h, the weight remains constant. The hydrogel from water was wiped out with paper before to determine weight. The calculated swelling percentage by Eq. (3):

$$\text{Swelling}(\%) = \frac{W_s - W_i}{W_i} \quad (3)$$

where W_s is weights of swollen hydrogel and W_i is weights of dry hydrogel.

■ RESULTS AND DISCUSSION

FESEM images of SA-g-p(AM-co-AC) and ST-g-p(AM-co-AC) hydrogels and the morphologies surface of hydrogels are shown in Fig. 1. It was observed that the surface of SA-g-p(AM-co-AC) hydrogel composite exhibit a regular surface embedded in matrix with the ingathering of SA. This can be referred to the fact that the alkyl chains of AC could reduce the interaction among hydrophilic groups and improve the agreement of SA with the matrix polymeric [21]. The hydrogel set micrometric stacked so soft hunks with shapes several that their edges were sunk together in a path that a particular isolated particle could barely be seen in Fig. 1(a) and 1(b).

Compared with the ST-g-p(AM-co-AC) hydrogel, numerous crinkles and ball shape which normally aggregated together and made larger particles with sundry nanometer sizes were plainly observed in the micrographs of ST-g-p(AM-co-AC) hydrogel and several pores appear on the surface. FESEM of ST-g-p(AM-co-AC) in Fig. 1(c) and 1(d) and portrayed close-packed

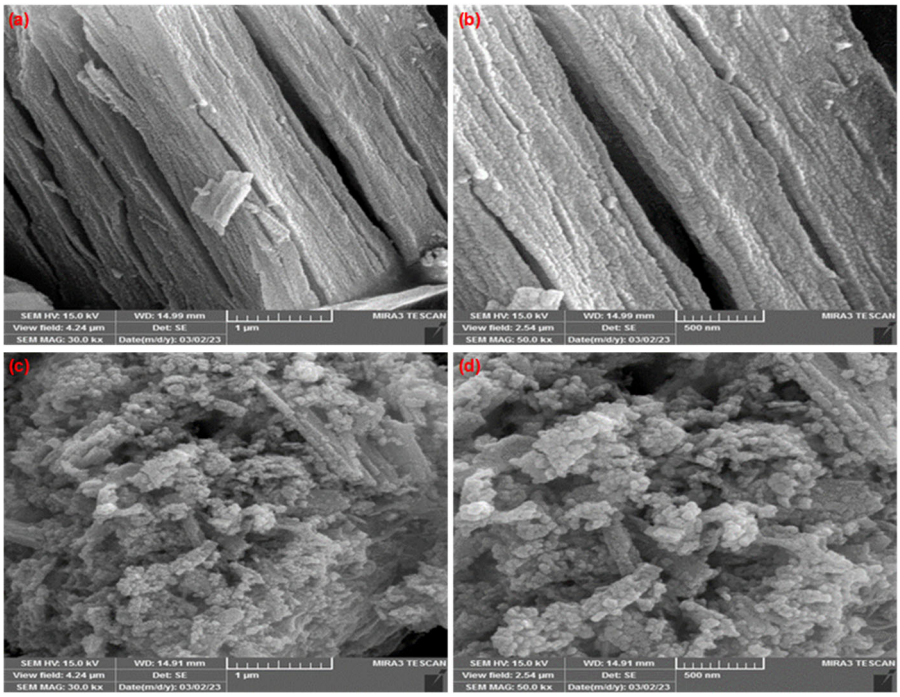

Fig 1. FESEM image of SA-g-p(AM-co-AC) at (a) 1 μm, (b) 500 nm and ST-g-p(AM-co-AC) hydrogels at (c) 1 μm, (d) 500 nm

small spherical particles which evidenced as white globes, particularly at 500 nm indicating the SA that increase the degree of improved morphologies of the surface [22-23].

Moreover, TEM images have been used to add extra set proof in support of the formation of SA-g-p(AM-co-AC). Also, the results proved the monomers could effectively produce uniform and tiny particles, which were most agglomerated (Fig. 2(a)). Therefore, TEM image of ST-g-p(AM-co-AC) in Fig. 2(b) reveal the ST to be black ball shape and non-equally sparse at the size range of 50 nm [24].

The XRD patterns of ST-g-p(AM-co-AC) and SA-g-p(AM-co-AC) hydrogels are shown in Fig. 3. The explicit of ST-g-p(AM-co-AC) in Fig. 3(a) displayed diffraction peaks at $2\theta = 20.2°$ conformable to (020) amorphous pattern reflections characteristic of ST. The co-polymer of SA-g-p(AM-co-AC) in Fig. 3(b) displayed a no crystalline plane. SA grafted co-polymer appears a diffraction band of intensity lower at $2\theta = 21°$ (020) for non-crystalline SA. Our results confirm the grafting of two monomers onto ST and SA. The diffraction band in ST and SA grafted co-polymer at $2\theta = 26.1°$ suggested the existence of some crystalline ST and SA allomorph that was not fully destroyed throughout the preparation, limiting the accessibility of COO– groups throughout the copolymerization reaction [25].

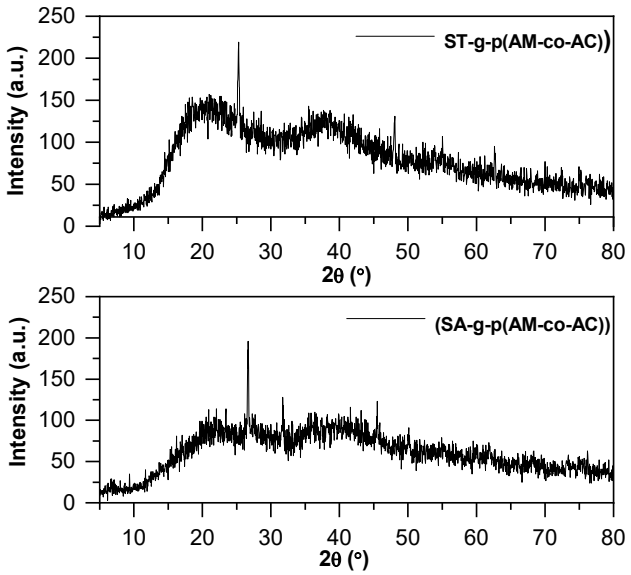

Fig 3. XRD of two hydrogels

Effect of Equilibrium Time

The effect of equilibrium time on the removal percentage (%) of MB dye is studied by changing its value from 2 to 60 min, as appears in Fig. 4. It was observed that the value of removal percentage (%) of MB dye was raised

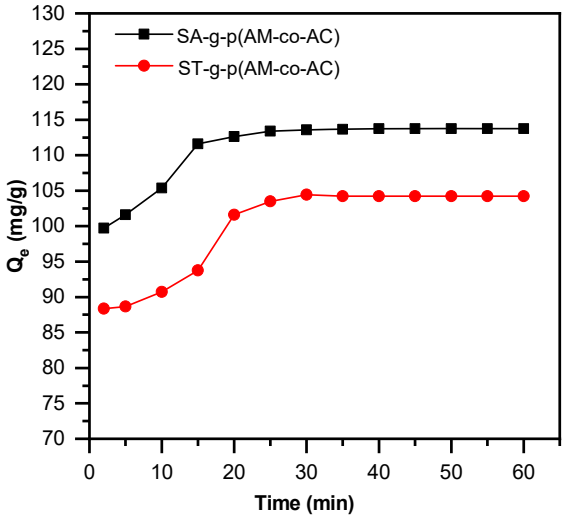

Fig 4. Effect of equilibrium time of SA-g-p(AM-co-AC) and ST-g-p(AM-co-AC) hydrogels onto MB dye

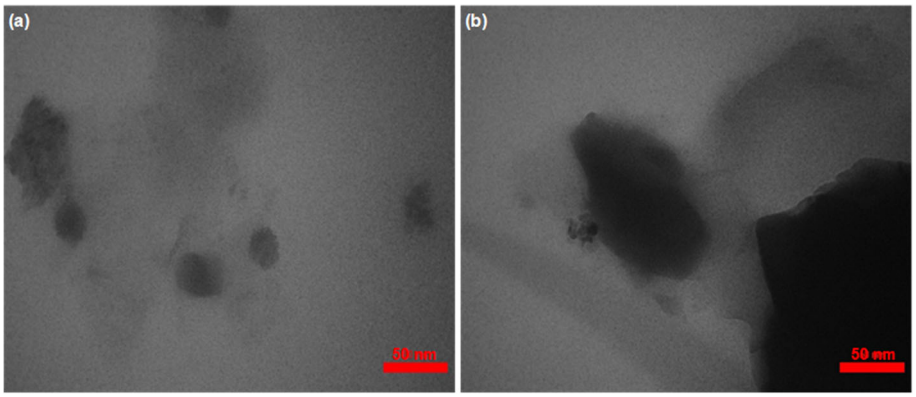

Fig 2. TEM images of (a) SA-g-p(AM-co-AC) and (b) ST-g-p(AM-co-AC)

with an increase of contact time and was turned onto constant after 30 min. Therefore, the best contact time for dye removal was found to be 30 min. At the most favorable time, equilibrium was established among the MB dye present in the solution and its quantity adsorbed on the surface. Therefore, an increase of contact time does not change the removal value percentage of MB dye in aqueous solution [26].

Effect of Adsorbent Dose

Hydrogels are considered a highly important factor with respect to adsorption performance. The influence of adsorbent doses on the percentage adsorption of dye was studied for initial concentration of 100 mg/L for MB dye and an equilibrium time of 30 min, as shown in Fig. 5. It can be seen that %E increased when increased the adsorbent doses of both hydrogels surfaces as improved with further increases in the weight of hydrogels. This can be attributed to increased active sites of the hydrogel surface to interact with functional groups in MB dye, resulting in higher %E. The adsorption capacity of MB was studied and appears in Fig. 5. As can be clearly, the adsorption efficiency decreased from 286.76–80.78 and 249.67–75.67 mg/g with increasing the adsorbent dose of hydrogels [27]. The optimum condition for the removal of MB dye at mass adsorbent of hydrogels 0.08 g, pH of solution 7.0, initial concentration of MB dye 100.00 mg/L, solution temperature 20 °C and equilibrium time 35 min. These conditions show the highest adsorption capacity of SA-g-p(AM-co-AC) (113.76 mg/g) and ST-g-p(AM-co-AC) (104.234 mg/g).

Effect of Temperature and Thermodynamic

Temperature is one of the most effective factors that could safely affect the behavior of the adsorption of MB dye. In this context, the influence of temperature solution on the adsorption capacity of MB on hydrogels was studied in 283, 293 and 303 K at the same optimum conditions shown in Fig. 6. With an increase in the temperature, MB dye removal capacity increased at a temperature of 303 K. This phenomenon indicates that the adsorption of dye is endothermic. The thermodynamic factors like as change of entropy (ΔS), change of enthalpy (ΔH), change (ΔG) by utilizing Eq. (4) and (5) [28-29]:

$$\Delta G = -RT \ln K_{eq} \quad (4)$$

$$\ln X_m = -\frac{\Delta H}{RT} + \text{Cons.} \quad (5)$$

where R is ideal gas constant (8.314 J/mol K), T is

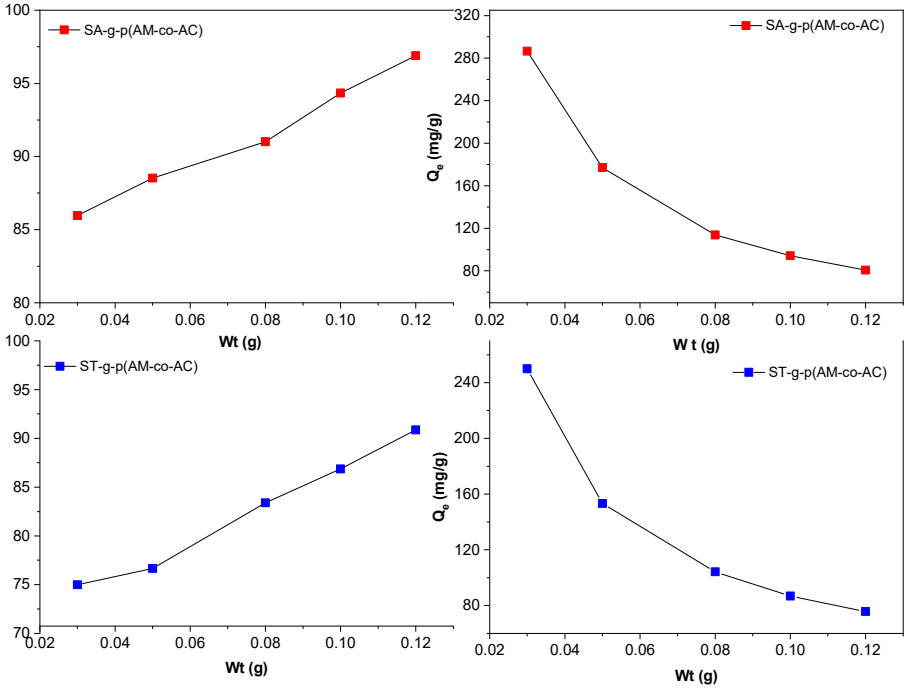

Fig 5. Effect of adsorption dosage of ST-g-p(AM-co-AC), and SA-g-p(AM-co-AC) hydrogels onto MB dye

temperature (K), and K_{eq} is thermodynamic equilibrium. The ΔH and ΔS were obtained from the slope, the intercept from the van't Hoff equation, and all the thermodynamic factors appear in Table 1. The ΔG value for MB dye adsorption on hydrogels was negative, indicating the adsorption process was spontaneous. With the rise in solution temperature, the ΔG value gradually increases, corroborating that the high-temperature prelature would increase the adsorption efficiency of dye. In addition, the ΔH value positive corroborative that the adsorption of dye was endothermic [30-31].

Adsorption Isotherm

Freundlich nonlinear model

The non-linear form of the Freundlich isotherm model is given via Eq. (6):

$$q_e = K_f C_e^{1/n} \quad (6)$$

where q_e is amount adsorbed per unit weight of adsorbent at equilibrium (mol/g or mg/g), K_f is empirical Freundlich constant or efficiency factor (L/mg), C_e is equilibrium concentration (mg/L or mol/L), and 1/n is Freundlich exponent. If the value of n = 1 is unity, adsorption is linear; if below unity, chemical adsorption and if above unity, then physical adsorption occurs [32].

Langmuir nonlinear model

The isotherm Langmuir is generally used for dye adsorption from liquid solutions. The nature of the adsorption process was derived via isotherm Langmuir calculated by Eq. (7) [33].

$$q_e = \frac{q_m K_L C_e}{1 + K_L C_e} \quad (7)$$

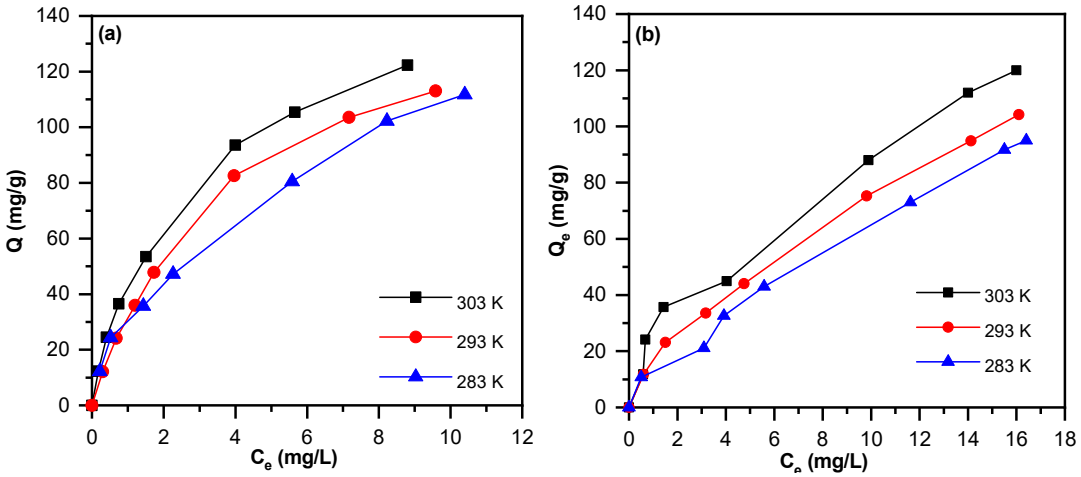

Fig 6. Influence of solution temperature on the adsorption of MB dye on to (a) SA-g-p(AM-co-AC), (b) ST-g-p(AM-co-AC)

Table 1. Thermodynamic functions ΔG, ΔS, and ΔH of MB dye adsorbed on the hydrogels

(ST-g-P(AM-co-AC) adsorbent/MB adsorbate				
Thermodynamics parameters T (K)	K_e	ΔG (kJ/mol)	ΔH (kJ/mol)	ΔS (J/K mol)
283	17.2656	−6.7026		
293	8.5938	−5.2399	29.9304	−61.626
303	7.5000	−5.0758		
(SA-g-p(AM-co-AC) adsorbent/MB adsorbate				
Thermodynamics parameters T (K)	Ke	ΔG (kJ/mol)	ΔH (kJ/mol)	ΔS (J/K mol)
283	17.6470	−6.7540		
293	17.3529	−6.9517	12.0802	−36.5941
303	17.0588	−7.1459		

where q_e is amount adsorbed per unit weight of hydrogel at equilibrium (mg/g), C_e is concentration equilibrium (mg/L), q_m is maximum adsorption capacity (mg/g), and K_L is Langmuir constant (L/mg). The coefficients of determine (R^2) and model isotherm parameter from the nonlinear regressive method appear in Tables 2 and 3. A comparison of nonlinear fitted curves from experimental data and two isotherms models at 25 °C. A plot of q_e vs C_e is shown in Fig. 7 where the value 1/n and K_F are found in the slope and intercept of the non-linear regressions [34-36].

Study Regeneration of Hydrogels

Reactivation and regeneration studies of the hydrogels are very important when assessing commercial applications. In acidic medium, protons (H^+) can displace ions MB dye that is attached to hydroxyl or carboxyl of hydrogels. To avoid introducing other impurity molecules, the hydrochloric acid solution as an eluent to reactivate hydrogel. In the regeneration of the adsorbent experiment, the dye-loaded hydrogels were immersed in solution HCl (0.01 M) for 2 h, filtered and washed several times in water. The adsorbent was then dried at 65 °C for 12 h to obtain the hydrogel regenerated. As shown in Fig. 8, hydrogels adsorbent regenerated has

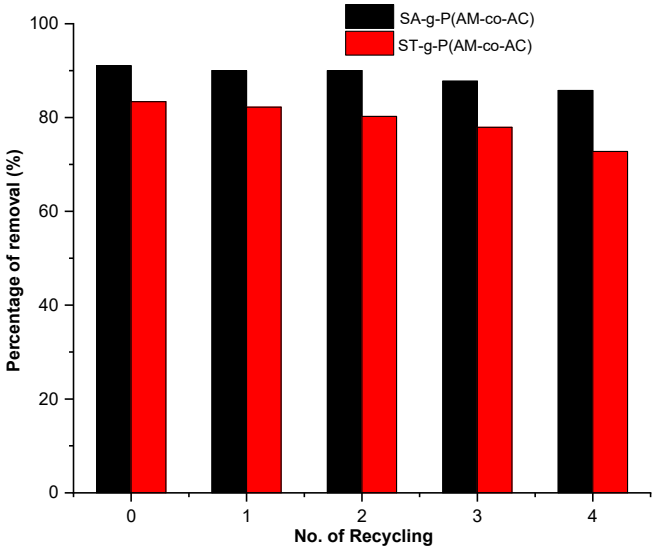

Fig 8. Effect of reactivation and regeneration of hydrogels

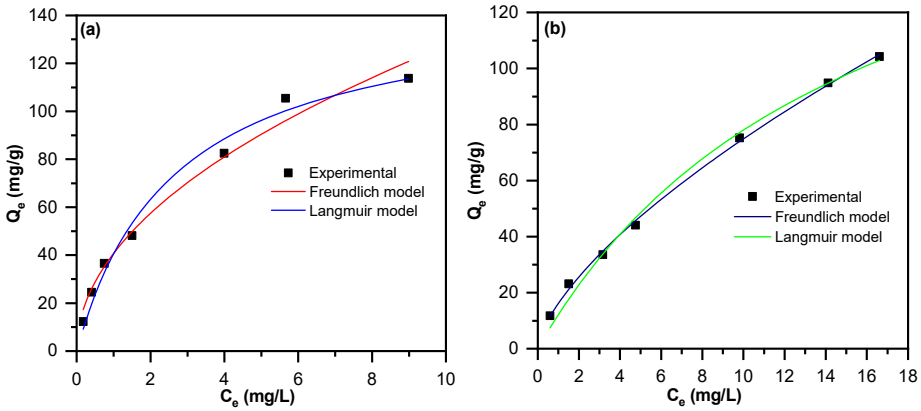

Fig 7. Effect nonlinear model isotherm on the adsorption of MB dye onto (a) SA-g-p(AM-co-AC) and (b) ST-g-p(AM-co-AC)

Table 2. Effected factors from nonlinear models of MB onto ST-g-p(AM-co-AC)

ST-g-p(AM-co-AC)			
Freundlich	K_f	40.8077	±2.9131
	1/n	0.4940	±0.0400
	R^2	0.9987	
Langmuir	q_m (mg/g)	146.8970	±10.6780
	K_L (L/mg)	0.3789	±0.0724
	R^2	0.9345	

Table 3. Effect nonlinear factors isotherm of MB on SA-g-p(AM-co-AC)

SA-g-p(AM-co-AC)			
Freundlich	K_f	16.1769	±0.6301
	1/n	0.6655	±0.0155
	R^2	0.9988	
Langmuir	q_m (mg/g)	198.1720	±22.3230
	K_L (L/mg)	0.0649	±0.0128
	R^2	0.8987	

chemically stable properties, and the percentage removal MB dye is in the range of 90.0 ± 2% to 83.4 ± 2%. In addition, the physical morphology of the hydrogels remains stable after five cycles from adsorption–desorption [35,37].

■ CONCLUSION

The percentage removal and adsorption efficiency of MB dye increase with increased solution temperature and contact time, but adsorption capacity (Q_e) decreased with the increased adsorbent dose. The better contact time for equilibrium is to be reached at a constant interval of 30 min. The adsorption capacity of MB dye was decreased from 286.76–80.78 and 249.67–75.67 mg/g when the absorbent dose of SA-g-p(AM-co-AC), and ST-g-p(AM-co-AC) hydrogels increase. All adsorption method is considered spontaneous and the ΔG value is negative, ΔS value is positive, and the ΔH is endothermic. The best Q_e is determined from the isotherm Freundlich model for two hydrogels. Regenerated hydrogels via HCl solution washing and their performance of adsorption remains stable after several reused. Thus, hydrogel is ecofriendly, green materials, low-cost, simple preparation, and has high removal efficiency of 90.0 ± 2% to 83.4 ± 2% for two hydrogels.

■ ACKNOWLEDGMENTS

The authors would like to thank the Department of Chemistry, College of Sciences for Girls, University of Babylon, Hilla, Iraq and the laboratories used inside the university affiliated with the Iraqi Ministry of Education.

■ CONFLICT OF INTEREST

All authors declare that they have no conflicts of interest.

■ AUTHOR CONTRIBUTIONS

Aseel Mushtaq Aljeboree: supervision, conceptualization, methodology, software, validation; writing-reviewing and editing. Ayad Fadhil Alkaim: data curation, writing-original draft preparation, visualization, investigation. All authors read and approved the final manuscript.

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