RESEARCH ARTICLE

Removal of Indigo Carmine Dye from Aqueous Solutions by Low-cost Surface: Investigation of Adsorption Properties

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ABSTRACT

This work was conducted to study the blue dye removal from textile company wastewater by the adsorption process using a continuous system (hydrogel composite). The process of preparing Graphene Oxide/Poly (acrylic amide/Formic acid) GO/P(AM-FA) hydrogel surface depends on the principle of swelling hydrogel behavior and pH-dependent, which in turn becomes a high possibility for the surface swelling of the hydrogel to remove the Indigo Carmine dye very quickly from the aqueous solution. Several techniques were used to determine the properties of the prepared surface before and after the adsorption process, including X-ray diffraction (XRD), thermogravimetric analysis (TGA)). Several factors affecting the adsorption process were studied, including (equilibrium time, pH effect, surface weight effect, ionic strength effect, and temperature effect). It was found that the best equilibrium time was after one hr. When studying the Effect of weight, it was found that with increasing weight, the percentage of removal increased and the efficiency of adsorption decreased, where the best weight was 0.05 g. The thermodynamic factor was studied, and it was found that the enthalpy value is negative, and through the negative value of Gibbs free energy, it was found that the reaction is spontaneous

Keywords: Adsorption process, Dye, Hydrogel, Indigo Carmine, Sewlling, Thermodynamic.

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INTRODUCTION

Wastewater from the textile finishing industry can strongly impact the aquatic environment because it is discharged in high quantities and may contain many biorecalcitrant contaminants; even if these pollutants are found in very low concentrations, their impact is dangerous on humans, plants, and animals. Therefore, drinking water is contaminated with many pollutants, such as the waste of pharmaceutical industries, pesticides, and dyes.[1-3] Dyes, especially organic dyes, are widely used in tanning, textiles, papers, paint, and other liquid industries. Large quantities of sewage containing dyes are produced.[4,5] As most dyes contain an aromatic structure, this aromatic composition has very high toxicity and is difficult to decompose-hazardous effects on the environment. One of the most important treatment methods is coagulation, sedimentation, filtration, and absorption.[6,7] The maximum significant contamination treatment in the aquatic environment is hydrogels, which have been used extensively over the past years due to their very high ability to remove pollutants. They can be easily prepared from inexpensive, non-toxic, and chemically stable materials. On swelling, it was used in many applications, and the main importance is used to remove dyes.

EXPERIMENTAL PART

Adsorption Experiments

Some factors affecting the adsorption process were studied, such as equilibrium time (2–300 minutes), pH effect (3-12), GO/P(AM-FA) hydrogel weight effect 0.01–0.07 gm, tempera-

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ture effects (5–35°C), and ionic strength effects (0.01–0.1 g/L) on removing dye using hydrogel surface, and all these experimental factors were performed. In a volumetric flask of 20 mL in a volume of 10 mL DW a solution of dye, in a volume of 10 mL, with an initial concentration of 25 mg/L was mixed with a weight (0.05 g) of hydrogel and placed in a water bath quickly. A total of 300 rpm for one hour until equilibrium is reached. After that, the remaining concentration was separated by centrifuge at 4000 rpm, and a UV-Visible spectrophotometer measured the absorbance at a wavelength of 610 nm. The adsorption efficiency was calculated through equation (1), and the removal ratio was also calculated through equation (2)

$$q_e = (V.(Co - Ce))/W \quad ... (1)$$

$$\% R = (Co - Ce)/Co \times 100 \quad ... (2)$$

RESULTS AND DISCUSSION

Thermogravimetric Analysis (TGA)

In this research, the TGA was used to determine the prepared surface's thermal decomposition and thermal stability. The sample was heated from 39–906°C under an atmosphere at a rate of 10°C min^{-1}. The surface heating method GO/P(AM-co-FA) depends on the loss of weight by up to 2.75% in the range of 90-230°C due to the presence of moisture.[8] The process of heating the compound in the range of 220–351°C leads to a weight loss of 44.59% which can be due to pyrolysis, caused by the low degree of polymerization due to the breaking of chemical bonds with the removal of hydroxyl and carboxyl groups in the form of carbon dioxide and carbon dioxide, so that[9] the loss of the weight of the compound within the thermal

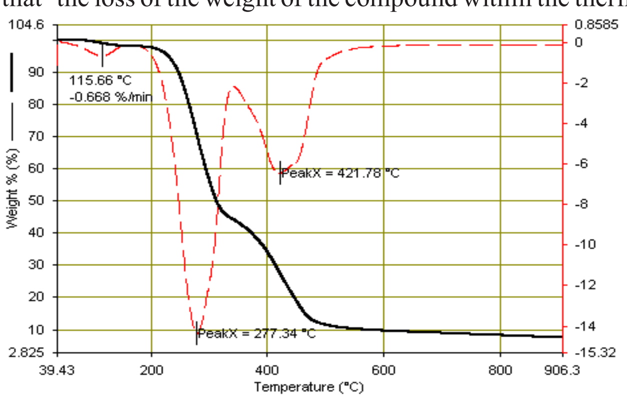

Figure 1: TGA analysis of GO/P(AM-co-FA) hydrogel

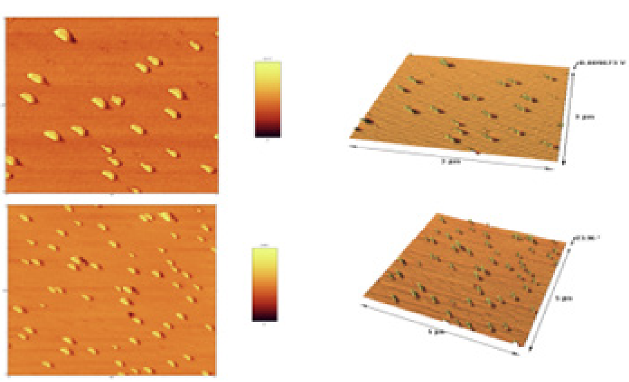

Figure 2: A 2D and 3D AFM images for GO/P(AM-co-FA)

range 421–600°C by 17.14% is caused by the breaking of the cross-linked polymeric chains of the adjuvant compound as shown in Figure 1.

Atomic Force Microscopy (AFM)

The AFM was used to determine and measure the roughness and topography of the hydrogel surface, as shown in Figure 2. The D3-dimensional image of the GO/P(AM-co-FA) hydrogel and the result shows through the three-dimensional image that the prepared surface has a roughness and a clear roughness.

Transmission Electron Microscopy (TEM)

The TEM technology was used for the prepared surface after loading graphite oxide on the polymer (GO/Poly (AM-co-FA). Its adsorption on an Indigo Carmine dye, where it was observed dark black spots aggregated evidence of dye loading on the surface and the occurrence of the adsorption process[10] as appear in Figure 3.

Effect of pH

One of the maximum important parameters on which the adsorption method depends is the Effect of solution pH that determines the interaction between the adsorbent and the adsorbant. As the pH does not depend only on the surface charge of the hydrogel, it depends mainly on the degree of ionization and the nature of the surface.[11-14] The surface of the prepared GO/P(AM-co-FA) hydrogel contains negative groups such as -COOH and -OH. These groups change with the change in the pH of the solution. We notice that increasing or decreasing the pH of the solution induces a change in the pH of the removal percentage of the dye and a change in the adsorption efficiency. Through the results, we determine the optimal pH 3.[15-18] The effective removal of the dye was in the acidic medium in this study, as shown in Figure 4.

Effect Weight of the Hydrogel

This study was based on taking different weights from the GO/P(AM-FA) hydrogel surface (0.01, 0.02, 0.03, 0.04, 0.05, 0.06, and 0.07 gm) that were used to determine the effect of the adsorbent dose as shown in Figure 5. It was observed that by

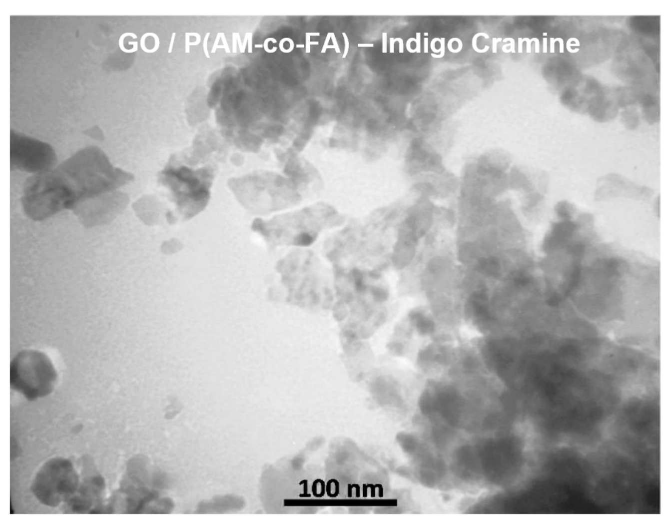

Figure 3: TEM image of GO/P(AA-co-AM) hydrogel

increasing the weight of the hydrogel surface, the percentage removal of Indigo Carmine dye increases significantly more pronounced, but with increasing weight, the adsorption efficiency decreases.[8,19,20] Increasing the weight of hydrogel surface from 0.01 to 0.07 gm, the removal percentage increased from 21.45% to 27.22%, and the adsorption efficiency decreased from 5.5 to 1.21 mg/g.

Effect Ionic Strength

There is strong competition between salt and molecules in the solution to capture the adsorption sites on the GO/P(AM-FA) hydrogel surface. The ionic strength factor significantly affected removing the dye at all optimal conditions.[8] When using different concentrations of 0.01–0.1 g/L of salts, the adsorption efficiency of the Indigo Carmine dye was studied, and the best adsorption efficiency when using NaCl salt Is shown in Figure 6.

Effect of Solution Temperature

Figure 7 shows the solution temperature's Effect on the adsorption dye in the chain from 5 to 35°C. The GO/P(AM-FA) hydrogel adsorption capacity increases when the solution temperature decreases from 5 to 35°C. It is found that the lower temperature is to the advantage of adsorption and that the adsorption is an endothermic reaction.[19] The parameter of thermodynamic (ΔH), (ΔG), and (ΔS) were evaluated using equations 3 and 4.

$$\ln Xm = \frac{-\Delta H}{RT} + constant \quad \text{... (3)}$$

$$\Delta G = -RT \ln K \quad \text{... (4)}$$

Table 1 shows the estimation values of the parameter of thermodynamic of adsorption dye onto hydrogel. Tables 2 and Figure 8 appear the determination of the maximum adsorbed quantity of dye adsorption onto hydrogel. The enthalpy value indicates adsorption endothermic. One possible explanation of the adsorption endothermic of dye onto hydrogel that is solvated in water. For the dye to be adsorbed, they have to

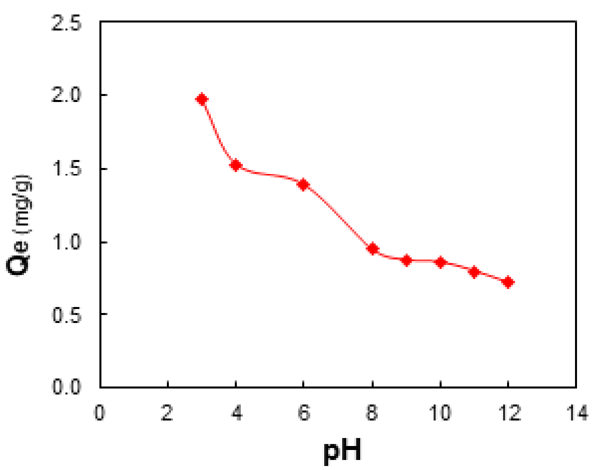

Figure 4: Effect of pH solution of the removal Indigo Carmine dye onto the hydrogel

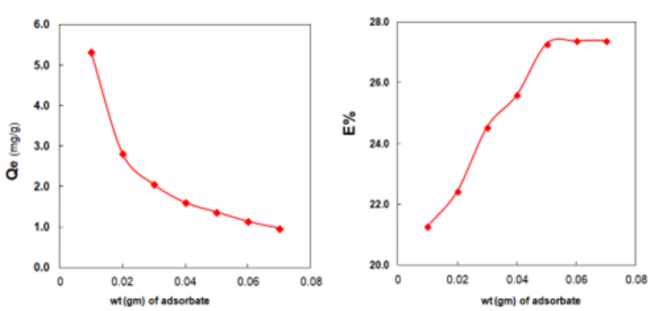

Figure 5: Effect of weight hydrogel of adsorption of Indigo Carmine dye

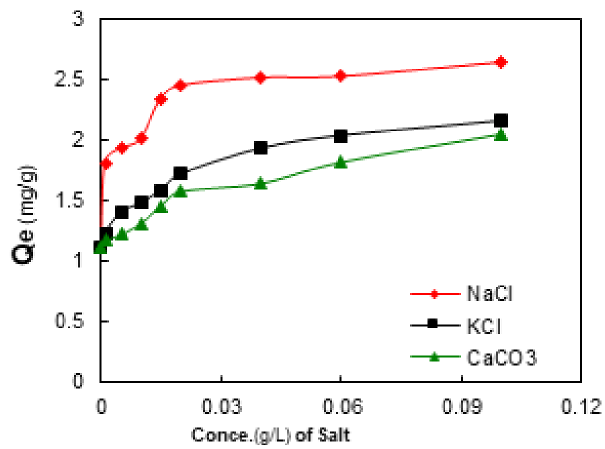

Figure 6: Effect of Ionic strength of adsorption of dye onto the hydrogel

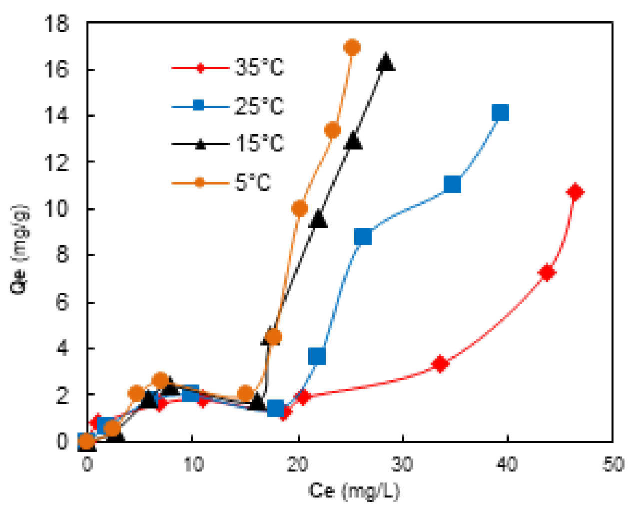

Figure 7: Adsorption isotherms of dye onto hydrogel at several temperatures

Table 1: Thermodynamic parameters for Indigo Carmine dye adsorption onto the hydrogel

ΔH° (KJ/mol)	*ΔG° (kJ/mol)*	*ΔS° ($J.mol^{-1}. K^{-1}$)*	*Equilibrium constant*
21.25	-1.23	4.127	1.643

Table 2: Effect of temperature solution of the maximum adsorbed quantity of Indigo Carmine dye adsorption on hydrogel

		$C_e = 7.142$	
T(K)	*1000/T(K⁻¹)*	X_m	$\ln X_m$
288	3.472	1.700	0.530
293	3.413	1.900	0.641
298	3.356	2.300	0.832

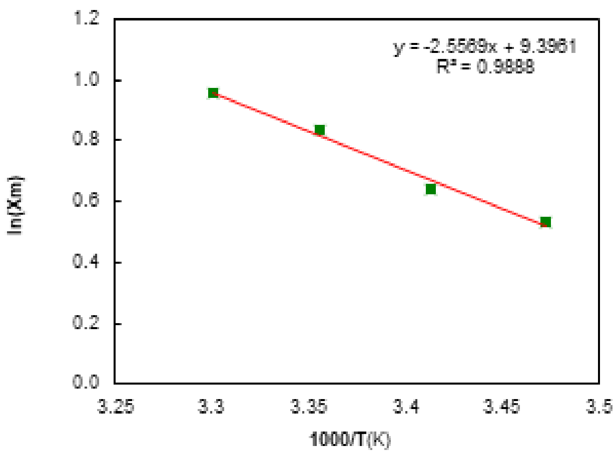

Figure 8: Plot of ln X_m against the reciprocal absolute temperature of dye adsorption on to hydrogel

lose part of their shell hydration. The dehydration way dye and the adsorbent hydrogel requires energy.[7] So, the dehydration methods supersede the endothermic adsorption ways. The values of ΔS positive and the very small negative values of ΔG have also been considered as the consequence of the RG dye's diffusion into the chemical hydrogel of the adsorbent.[20-22]

CONCLUSION

- The best adsorption efficiency of the Indigo Carmine dye was in the acidic medium at pH 3.
- The maximum removal percentage of Indigo Carmine dye at the weight (0.05 gm) where it was found that the removal percentage increased and the adsorption efficiency decreased with increasing weight.
- The adsorption efficiency increases when the temperature decreases.
- The efficiency of adsorption increases, where does the concentration of salts increase, and the best salt to be used is salt NaCl.
- The enthalpy value is positive the reaction is endothermic and spontaneous depending on the importance of Gibbs free energy.

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