RESEARCH ARTICLE

Preparation, Characterization, and Adsorption Potential of the Eco-friendly Surface to Remove the Basic Dye from an Aqueous Solution

Aseel M. Aljeboree[1]*, Noor A. Al-lamy[2], Mohanad A. Mohammed[3], Fatima S. Fahim[4], Zuhair I. Al Mashhadani[5], Ahmed K. O. Aldulaim[6], Emad S. Abood[7], Shahad M. Qasim[8]

[1]*Department of Chemistry, College of Sciences for Girls, University of Babylon, Hilla, Iraq*
[2]*Al-Manara College for Medical Sciences, Amarah, Iraq*
[3]*College of Pharmacy, Al Farahidi University, Baghdad, Iraq*
[4] *Department of Anesthesia Techniques, Al-Mustaqbal University College, Babylon, Iraq*
[5]*Al-Nisour University College, Baghdad, Iraq*
[6]*Department of Pharmacy, Al-Zahrawi University College, Karbala, Iraq*
[7] *Department of Medical Physics, Hilla university college, Babylon, Iraq*
[8]*Department of Chemistry, College of Education, University of Al-Qadisiyah, Diwaniya, Iraq*

Received: 05th February, 2022; Revised: 24th April, 2022; Accepted: 17th May, 2022; Available Online: 25th June, 2022

ABSTRACT

The work deals with one of the applications of adsorption from solution. It deals with the study of the removal of Indigo Carmine dye on the selected eco-friendly surface (Graphene Oxide/poly (acrylic amide / Formic acid)). This research relied on preparing a GO/P(AM-FA) hydrogel surface that can be prepared from available and inexpensive materials with high efficiency in removing the Indigo Carmine dye from its aqueous solution. The physical and chemical properties of the surface were studied through several important techniques, including (fourier transform infrared (FTIR), field emission scanning electron microscopy (FESEM), and transmission electron microscopy (TEM)). Two types of Langmuir and Freundlich adsorption isotherms were studied and through the results. It was found that it obeys the Freundlich model depending on the value of ($R^2 = 0.9276$) for heterogeneous surfaces. Also, the adsorption kinetics of first order and second order were studied, and the results found that it obeys the second-order depending on the value of ($R^2 = 0.9999$).

Keywords: Adsorption, Equilibrium, Indigo Carmine dye, Isotherm, Kinetic, Removal.

International Journal of Pharmaceutical Quality Assurance (2022); DOI: 10.25258/ijpqa.13.2.18

How to cite this article: Aljeboree AM, Al-lamy NA, Mohammed MA, Fahim FS, Al Mashhadani ZI, Aldulaim AKO, Abood ES, Qasim SM. Preparation, Characterization, and Adsorption Potential of the Eco-friendly Surface to Remove the Basic Dye from an Aqueous Solution. International Journal of Pharmaceutical Quality Assurance. 2022;13(2):182-186.

Source of support: Nil.

Conflict of interest: None

INTRODUCTION

Hydrogels have been used over the past years and in many different applications. Hydrogels consist of three-dimensional networks of hydrophilic polymers that can absorb water in large quantities and thus can swell.[1-4] The hydrogel has a very high ability to absorb pollutants inside and remove the dye. It is also characterized by chemical stability, non-toxic, and high stability against light, so it was used to remove many pollutants, the most important of which are dyes. As is well known, industrial dyes and their products are considered among the most dangerous toxic pollutants that cause many problems for the living organisms in the water.[5,6] These dyes spread in groundwater and drinking water and accumulate in large quantities and be a constant pollutant for water because they do not easily degrade biologically due to their aromatic structures.[7-10] Therefore, the complete removal of these toxic dyes from industrial waste has become more important because of their harmful effects on humans, animals, and plants.[11-13] Most of the dyes are used in the dyeing of papers, fabrics, leather, and other textiles. Several methods have been adopted to remove dyes from their aqueous solution: photo-oxidation, separation, Extraction, coagulation, photocatalytic, ozonation, and adsorption.[14-16] It was found that adsorption is one of the most important techniques in removing dyes easily, in an economical, inexpensive, and effective way. Using cheap, available, and inexpensive surfaces that can be easily prepared from them (activated carbon, polymers, carbon nano, clays, graphene oxide, chitosan, and hydrogel.[17,18] This study was based on the preparation of one of the surfaces that are very effective in removing dye, which can be prepared from cheap

**Author for Correspondence:* annenayad@gmail.com

and inexpensive materials. The adsorption isotherms and adsorption kinetics were studied.

EXPERIMENTAL PART

Preparation of Standard Solution

All materials used in this research are of high purity. Merck (Germany). The standard solution of Indigo Carmine dye (1000 mg/L) resulted from dissolving (1.0 g) in 1000 ml of distilled water. A series of dilute solutions were made to obtain the dilute concentrations used in the experiment.

Preparation of GO/P(AM-FA) Hydrogel

The first step in making (AM-co-FA) hydrogel is to prepare the solutions of the substances that will be used during the hydrogel's installation. 2% of MA was used when prepared in deionized water, and 4% of acrylic amide was used. A total of 40% of MA and 80% of the acrylic acid were combined with GO in a ratio of 1:10. The mixture is thoroughly stirred before adding the cross-linker agent MBA (0.20 mol/L), which is added to the initiator KSP (0.0361 mol/L) while the mixture is still stirring. After that, the mixture is poured into polyethylene test tubes and exposed to nitrogen gas for 10 to 15 minutes, depending on the temperature. The temperature is raised in a hot water bath from 45 to 65°C while the tubes are still in the bath. The following is the temperature rise: A temperature range of 45°C for an hour, 55°C for 2 hours, and 65°C for two hours were tested. After cooling and removing the hydrogel from the tubes, it is cut into 6mm-long pieces. After that, perform one-week ethanol and distilled water wash to remove all non-reactive monomers. Then dry at room temperature, then in an electric oven at 50 to 60°C until a constant weight is obtained; repeat as necessary.

Adsorption Experiments

Indigo Carmine dye solution adsorption experiments were performed at 25 °C at 200 rpm. The adsorption experiments were carried out in a volumetric flask of 20 mL in a volume of 10ml of distilled water, and the weight of the GO/P(AM-FA) hydrogel surface was used (0.05 gm). The solution was placed in a water bath shaker for 60 minutes until the equilibrium state was reached. resolution was by UV-Visible spectrophotometer. The adsorption isotherms and adsorption kinetics were studied under the same optimum conditions. The adsorption efficiency was calculated through the equation (1):

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

RESULTS AND DISCUSSION

FTIR

The FTIR was used to characterize the hydrogels before and after the surface adsorption process on the dye, as shown in Figure 1. Therefore, we notice a clear and significant difference in the adsorption intensity between GO/P(AM-co-AF)). Hydrogels before the adsorption process with the dye show a clear decrease in the FTIR spectra in the intensity of the bands adjacent to the adsorption. This confirms that the adsorption process on this dye is weak due to the chemical composition and the acidic nature of dye.[19] Also, the surface contains acidic aggregates that lead to a difference in absorption intensity.

FESEM

Based on the physical properties, porosity, and nature of the prepared surface, as well as the shape and size of the particles and the way they are distributed on the surface, the FESEM technique is studied before the adsorption process for each P(AM-co-FA) and after the graphene loading process GO/P(AM-co-FA). It was found that the hydrogel surface contains many heterogeneous holes. After graphene oxide loading, the surface became less rough. Many cavities were reduced due to graphite oxide loading,[20] which confirms the adsorption process's success as shown in Figure 2.

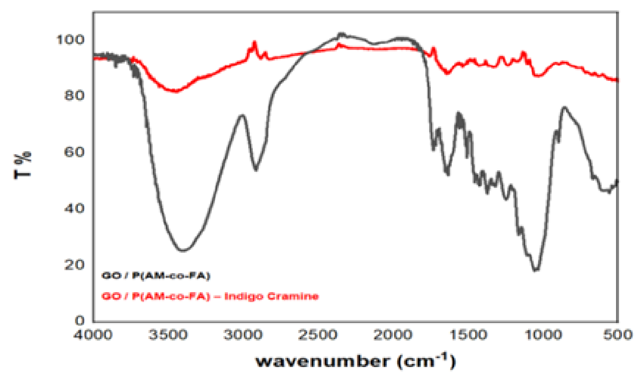

Figure 1: FTIR spectra of GO/P(AM-co-FA) hydrogels (a) before and (b) after adsorption of Indigo Carmine.

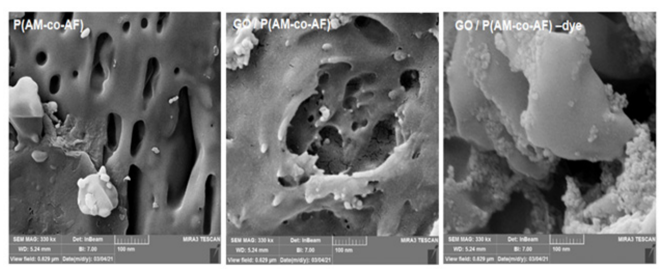

Figure 2: FESEM image of GO/P(AM-co-FA) before and after adsorption of Indigo Carmine.

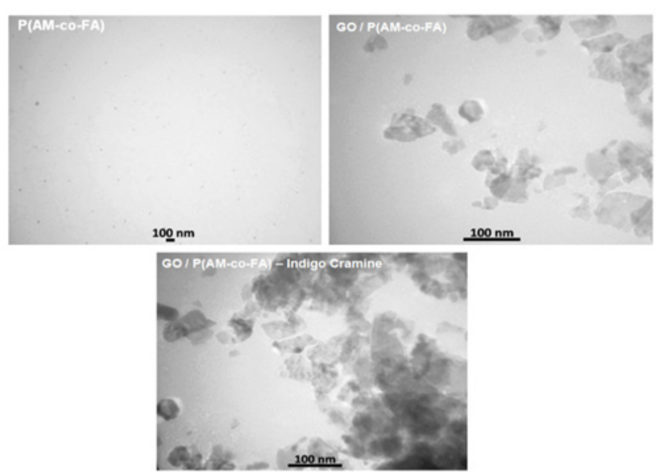

Figure 3: TEM image a- p (AM-co-FA), b- GO/p(AM-co-FA), c- GO/p(AM-co-FA)- Indigo Carmine

Table 1: Kinetic adsorption factor of Indigo Carmine dye adsorption onto the hydrogel

Pseudo- first-order					
R^2	q_e *(mg/g)*	$k_1(min^{-1})$	*Intercept*	*Slope*	
0.446	0.810	0.002	1.418	-0.017	
Pseudo- second-order					
h $(mg.\ g^{-1}.\ min^{-1})$	R^2	q_e *(mg/g)*	$k_2\ (g.\ mg^{-1}.min^{-1})$	*Intercept*	*Slope*
21.008	0.9999	1.419	0.299	0.048	0.029

Table 2: constant Isotherms and the correlation coefficients of Isotherm Langmuir and Freundlich of Indigo Carmine dye adsorption onto the hydrogel

Langmuir equation			*Freundlich equation*		
K_L	q_m	R^2	K_F	*n*	R^2
-0.003	-84.034	0.0134	0.308	1.036	0.9280

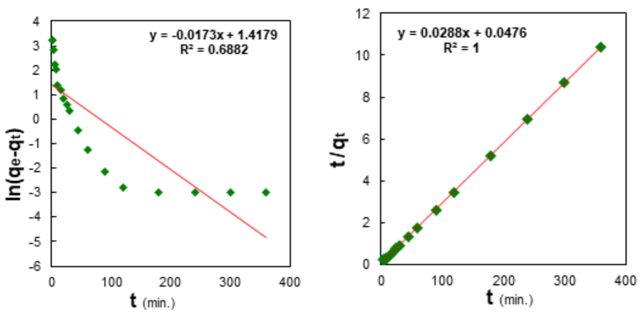

Figure 4: Adsorption kinetics of first-order (a). second-order (b) of Indigo Carmine dye onto the hydrogel

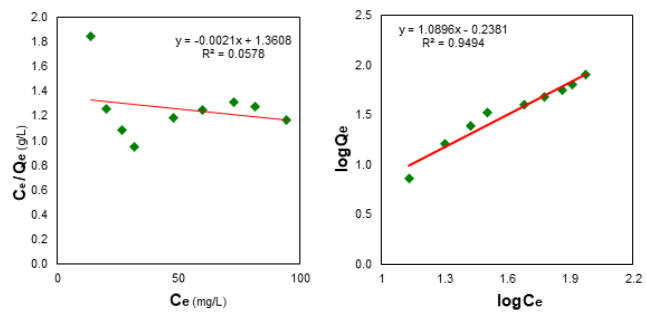

Figure 5: Effect of adsorption model of Isotherm Langmuir (A), Isotherm Freundlich (B) of Indigo Carmine dye adsorption on hydrogel

TEM

TEM technique was studied for the Surface (P(AM-co-FA) before and after the graphite oxide loading process (GO/P(AM-co-FA)). It was observed that after loading the graphite oxide, some dark spots appeared on the (GO) loading on the surface; after loading the dye on the surface by the adsorption process, we noticed the appearance of large dark spots gathered, confirming the success of the adsorption process,[21] shown in Figure 3.

Adsorption Kinetics

The design of the adsorption system is considered one of the utmost important parameters that control the kinetic properties that depend on the adsorption speed. Therefore, studying the percentage of dye removal depends mainly on the kinetic properties, and the effective effects of dye removal also depend on the reaction time.[22] To determine the mechanism of control of the adsorption processes, we use adsorption kinetics like a chemical reaction and surface adsorption or penetration mechanisms that determine the effective factors on reaction speed.[10,23,24] Liters of different times (5-300 minutes), pH solution 6, and weight of GO/P(AM-FA) hydrogel 0.05 gm using a water bath shaker at a speed of 300 rpm. The efficiency of adsorption depends mainly on the equation first-order kinetic. Its use depends on its use when adsorption is in a boundary layer; the equation second-order kinetic applied to the adsorption chemical process is the adsorption method's control.[25,26] The experimental result study used the first order and second order. Table 1 displays that adsorption of Indigo Carmine dye by using hydrogel is achievable because of the R^2 value increase via the second-order compared to the first order as appearing in Figure 4.[18,27]

Isotherm Adsorption

The adsorption capacity explains equilibrium results describing the interaction between the adsorbents and the adsorbents. Two types of Freundlich and Langmuir adsorption isotherms were used in this study, where the Langmuir isotherm is for adsorption of homogeneous surfaces and monolayers. At the same time, the Freundlich isotherm depends on the adsorption of multi-layered heterogeneous surfaces with a non-uniform distribution of heat adsorption on surfaces. Langmuir and Freundlich adsorption isotherms equations.[28] Isotherm Freundlich of multilayer and heterogeneous adsorption. Isotherm linear as appears in equation (2):

$$\ln q_e = \ln K_f + 1/2\ C_e \qquad ...\ (2)$$

The isotherm Langmuir linear as appears in equation (3):

$$C_e/q_e = 1/(q_m.K_L)+(1/q_m)\ C_e \qquad ...\ (3)$$

Figure 5 shows the result from the adsorption method acquired at 25°C. The result shows that the adsorption method of dye onto GO/P(AM-FA) hydrogel of the adsorbent is analogical to the isotherm Freundlich Table 2 depicts the R^2 value and constants isotherm Langmuir and Freundlich.[29]

CONCLUSION

This study relied on removing the textile dye Indigo Carmine using a surface GO/P(AM-FA) from an aqueous solution, and the adsorption isotherms were studied, and it was found that the isotherm Freundlich model best value (R^2=0.9280) comparative with isotherm Langmuir model(R^2=0.0134). The adsorption kinetics were also studied, and it was found that it obeys the second-order kinetics.

REFERENCE

1. R. Saravanan, VKG, T. Prakash, V. Narayanan, A. Stephen, *Synthesis, characterization and photocatalytic activity of novel Hg doped ZnO nanorods prepared by thermal decomposition method.* J. Mol. Liq., 2013;88-93.
2. X. Liang, Y.X., X. Tan, L. Wang, Y. Sun, D. Lin, Y. Sun, X. Qin, Q. Wang, *Heavy metal adsorbents mercapto and amino-functionalized palygorskite: preparation and characterization.* Colloids Surf. A, 2013:98-105.
3. Atyaa, A.I., Radhy, N.D., Jasim, LS, *Synthesis and Characterization of Graphene Oxide/Hydrogel Composites and Their Applications to Adsorptive Removal Congo Red from Aqueous Solution.* Journal of Physics: Conference Series, 2019. (1):012095.
4. Layth S J and Aljeboree, M.A., *Removal of Heavy Metals by Using Chitosan/ Poly (Acryl Amide-Acrylic Acid)Hydrogels: Characterization and Kinetic Study.* NeuroQuantology, 2021. (2):31-37.
5. Yu, F., et al., *Adsorptive removal of antibiotics from aqueous solution using carbon materials.* Chemosphere, 2016. :365-385.
6. Aljeboree, A.M., Alkaim ,A. F., Abdulrazzak, F. H., Abbas, A. S., Alshirifi, A. N., *Spectrophotometric Determination of Pharmaceutical by Oxidative Coupling of 4-Aminoantipyrine: A Short Review.* Journal of Engineering and Applied Sciences, 2019. (2):5561-5569.
7. Kamil, A.M., Mohammed, H.T., Alkaim, A.F., Hussein, F.H., *Adsorption of Congo red on multiwall carbon nanotubes: Effect of operational parameters.* Journal of Chemical and Pharmaceutical Sciences, 2016. (3):1128-1133.
8. I. Ali, M.A., T.A. Khan, *Low cost adsorbents for the removal of organic pollutants from wastewater.* J. Environ. Manag., 2012. :170-183.
9. Aljeboree, A.M., Abbas, A.S., *Removal of Pharmaceutical (Paracetamol) by using CNT/ TiO2 Nanoparticles.* Journal of Global Pharma Technology, 2019. (1):199-205.
10. Safaa H. Ganduh, R.Q.K., Makarim A. Mahdi, Aseel M Aljeboree, Layth S. Jasim, *Selective Spectrophotometric Determination of 4-amino Antipyrine Antibiotics in Pure Forms and their Pharmaceutical Formulations.* International Journal of Drug Delivery Technology, 2021. (2):371-375.
11. Vahid Saadattalab, A.S.a.H.G., *Effect of CNTs and nano ZnO on physical and mechanical properties of polyaniline composites applicable in energy devices. Progress in Natural Science: Materials International* Progress in Natural Science Materials International, 2016. (1):22-23.
12. Aljeboree, A.M., *Adsorption and Removal of pharmaceutical Riboflavin (RF) by Rice husks Activated Carbon. International* Journal of Pharmaceutical Research 2019. (2):255-261.
13. Behera SK, K.H., Oh JE, Park HS., *Occurrence and removal of antibiotics, hormones and several other pharmaceuticals in wastewater treatment plants of the largest industrial city of Korea.* . Sci Total Environ. 2011. :4351-60.
14. F. Long, J.L.G., G.M. Zeng, L. Chen, X.Y. Wang, J.H. Deng, Q.Y. Niu, H.Y. Zhang, X.R. Zhang, *Removal of phosphate from aqueous solution by magnetic Fe-Zr binary oxide.* Chem. Eng. J., 2011. :448-455.
15. Aseel M. Aljeboree, A.F.A., Ali loay, hanadi m algburi, *Photocatalytic Degradation of Textile Dye Crystal Violet Wastewater using Zinc Oxide as a Model of Pharmaceutical Threat Reductions.* Journal of Global Pharma Technology, 2019. (02):138-143.
16. Aseel M. Aljeboree, A.F.A., *Role of plant wastes as an ecofriendly for pollutants (Crystal Violet dye) removal from Aqueous Solutions* Plant Archives 2019 (2):902-905.
17. Thitame, P.V. and S.R. Shukla, *Adsorptive removal of reactive dyes from aqueous solution using activated carbon synthesized from waste biomass materials.* Int. J. Environ. Sci. Technol. , 2016. :561-570.
18. Aseel M Aljeboree, R.A.M., Makarim A. Mahdi, Layth S. Jasim, Ayad F. Alkaim, *Synthesis, Characterization of P(CH/ AA-co-AM) and Adsorptive Removal of Pb (II) ions from Aqueous Solution: Thermodynamic Study.* NeuroQuantology 2021. (7):137-143.
19. Banerjee, S. and M.C. Chattopadhyaya, *Adsorption characteristics for the removal of a toxic dye, tartrazine from aqueous solutions by a low cost agricultural by-product.* Arabian Journal of Chemistry, 2017. :S1629-S1638.
20. Soleyman Hosseinzadeh, H.H., Shahryar Pashaei, Zahra Khodaparast, *Synthesis of magnetic functionalized MWCNT nanocomposite through surface RAFT co-polymerization of acrylic acid and N-isopropyl acrylamide for removal of cationic dyes from aqueous solutions.* Ecotoxicology and Environmental Safety 2018. :34-44.
21. Adeyemo, AA, I.O. Adeoye, and O.S. Bello, *Adsorption of dyes using different types of clay: a review.* Appl Water Sci,, 2017. :543-568.
22. Waleed K. Abdulsahib, Shg, Makarim A. Mahdi, Layth S. Jasim, *Adsorptive Removal of Doxycycline from Aqueous Solution using Graphene Oxide/Hydrogel Composite.* International Journal of Applied Pharmaceutics, 2020. (6):100-106.
23. Konicki, W., Sibera, Daniel, Mijowska, Ewa, Lendzion-Bielu, Zofia, Narkiewicz, Urszula, *Equilibrium and kinetic studies on acid dye Acid Red 88 adsorption by magnetic ZnFe2O4 spinel ferrite nanoparticles.* Journal of Colloid and Interface Science, 2013. :152-160.
24. Aljeboree A.M., A.A.N., Alkaim A.F, *Removal of pharmaceutical amoxicillin drug by using (Cnt) decorated clay/ fe2 o3 micro/ nanocomposite as effective adsorbent: Process optimization for ultrasound-assisted adsorption.* International Journal of Pharmaceutical Research, 2019. (4):80-86.
25. Mhammed Alzayd, A.A., Atyaa, A.I., Radhy, N.D., Jasim Al-Hayder, L.S., *A new adsorption material based GO/PVP/ AAc composite hydrogel characterization, study kinetic and thermodynamic to removal Atenolol drug from wast water.* IOP Conference Series: Materials Science and Engineering, 2020. (6):062023.
26. Abbas N. Karim, L.S.J., *Synthesis and Characterization of Poly (CH/AA-co-AM) Composite: Adsorption and Thermodynamic Studies of Benzocaine on from Aqueous Solutions* International Journal of Drug Delivery Technology 2019. (4):558-562.
27. Aljeboree, A.M., Alkaim A F, *Removal of Antibiotic Tetracycline (TCs) from aqueous solutions by using Titanium dioxide (TiO2) nanoparticles as an alternative material.* Journal of Physics: Conf. Series 2019.052059.
28. Gamal Owes El-Sayed, T.Y.M., and Ashraf Abd-Allah Salama,

Batch Adsorption of Maxilon Red GRL from Aqueous Solution by Natural Sugarcane Stalks Powder. Environmental Chemistry. 2013.514154.

29. M. Ajbary, AS, V. Morales-Flórez, R. Esquivias, *Removal of basic yellow cationic dye by an aqueous dispersion of Moroccan stevensite.* Appl. Clay. Sci., 2013;46-51.