RESEARCH ARTICLE

Role of Hydrogel and Study of its High-Efficiency to Removal Streptomycin drug from Aqueous Solutions

Nadher D. Radia[1*], Suhair M. H. Kamona[2], Hadeer Jasem[3], Russul R. Abass[4], Samar E. Izzat[5], Mohammed S. Ali[6], Sukaina T. Ghafel[7], Aseel M. Aljeboree[8]

[1*] *Department of Chemistry, College of Education, University of Al-Qadisiyah, Diwaniya, Iraq*
[2] *Al-Manara College for Medical Sciences, Amarah, Iraq*
[3] *Medical Instrumentation Techniques Engineering Department, Al-Mustaqbal University College, Babylon, Iraq*
[4] *Medical Lab. Techniques Department, College of Medical Technology, Al-Farahidi University, Baghdad, Iraq*
[5] *Al-Nisour University College, Baghdad, Iraq*
[6] *Department of Dentistry, Al-Zahrawi University College, Karbala, Iraq*
[7] *Scientific Research Center, Al-Ayen University, Thi-Qar, Iraq*
[8] *Department of Chemistry, College of Sciences for Girls, University of Babylon, Hilla, Iraq*

Received: 25th February, 2022; Revised: 09th April, 2022; Accepted: 26th May, 2022; Available Online: 25th June, 2022

ABSTRACT

Adsorption plays an effective role in pollution control, water treatment, and reuse. In this study, the removal of Streptomycin drug from aqueous solution was adopted by preparing a hydrogel surface for drug absorbent. The poly (PVA-AAM) hydrogel surface was prepared by relying on the polymerization of free radicals, using KPS as a starting material for free radicals, and using the cross-linking agent (AMB). The surface was studied and characterized by using several techniques such as (FTIR, TEM, AFM, and BET). Several important factors were studied, including (the effect of equilibrium time, the ionic strength, and the effect of temperature). The adsorption efficiency decrease when the concentration of salt increase, and the best weight (0.05 g) give higher adsorption capacity and best pH in the acidic medium. The study showed that adsorption efficiency increases with decreasing temperature through the adsorption process at different temperatures. The change in the values of thermodynamic functions (Entropy, Enthalpy, and Gibbs free energy) was calculated and found the reaction spontaneous and exothermic process. The adsorption isotherms were studied through the results, and it was found that it obeys the Freindlich model

Keywords: Drug, Exothermic process, Hydrogel, Pharmaceutical, Removal, Spontaneous.

International Journal of Pharmaceutical Quality Assurance (2022); DOI: 10.25258/ijpqa.13.2.13

How to cite this article: Radia ND, Kamona SMH, Jasem H, Abass RR, Izzat SE, Ali MS, Ghafel ST, Aljeboree AM. Role of Hydrogel and Study of its High-Efficiency to Removal Streptomycin drug from Aqueous Solutions. International Journal of Pharmaceutical Quality Assurance. 2022;13(2):160-163.

Source of support: Nil.

Conflict of interest: None

INTRODUCTION

Pollution is the harmful changes that occur in environmental systems due to the introduction of polluting materials such as industrial waste, hospitals, and pharmaceuticals to get rid of these materials because of their physical and chemical side effects on the environment lead to water pollution.[1,2] The toxic substances that change the properties of water that contain heavy elements such as lead, cadmium, copper, and others. Also, sewage water contains dangerous chemicals and bacteria that cause dangerous diseases. Drugs are considered one of the most dangerous pollutants affecting water's chemical and physical properties.[3-6] Therefore, the country's pollution has gained attention due to the damage it causes to the biological systems of the natural environment. These drugs may be highly toxic, cause allergies, genetic mutations, and cancer diseases and thus affect human health.[7-10] Some physical and chemical techniques have contributed to reducing pollution and getting rid of pollutants, whether liquid or solid. These techniques include extraction, ion exchange, precipitation, filtration, and evaporation, but these techniques are limited in use because of their high cost. Adsorption occupies an important position among the techniques used to get rid of pollutants because it is an effective and easy way to use and depends on the use of surfaces available from natural or industrial sources of low cost. Most of these surfaces are very effective for the removal of pollutants because they depend on the surface area and high porosity.[11,12] Many examples of surfaces used in the adsorption process include coal, clay, activated

**Author for Correspondence: nadhir.dhaman@qu.edu.iq*

carbon, carbon nano-tube, silica gel, graphite oxide, and hydrogels.

EXPERIMENTAL PART

Chemicals

All chemicals used in this study have (acrylic acid (AA)) and are purchased from Sigma-Aldrich. N, N⁻-methylene-bisacrylamide (MBA) and potassium persulfate (KPS) from Kemiou Chemical Reagent China. Spontaneous and NaOH were purchased from Merck. PVA was obtained from Fluka. All materials utilized were analytical grade pure and utilized without further purification.

Preparation of Poly (PVA-AMA) Hydrogel

Preparation of Polyvinyl Alcohol Solution

Dissolving 2 gm of polyvinyl alcohol in 30 mL of distilled water with continuous stirring until all the substance is dissolved in the distilled water.

Preparation of Acryl Amide Solution

A quantity of 4 g from AAm is dissolved in 5 mL of distilled water and added to the reaction mixture with constant stirring.

Preparation of Sodium per Sulfate Solution

Dissolve an amount of about 0.05 gm from KPS in 1-mL of distilled water and add to the reaction mixture a drop–drop.

Preparation of MBA Solution

Dissolve an amount of about 0.05 gm from MBA in 1 mL of distilled water with continuous stirring until the substance is dissolved.

Batch Adsorption Experiments

All adsorption experiments are carried out in a 10 mL conical flask with 0.05 g hydrogel and 10 mL of solutions drug. To get isotherms of adsorption, 0.05 gm of the hydrogel is got in touch with 10 mL of several conc. of solutions drug (10–100 mg/L). The put-in shaker water bath at 160 r/min for 1-hour at 20°C. When the adsorption process attainment equilibrium time, the appropriate solution is gathered to filter utilizing a filter paper and then estimated via UV–Visible spectrophotometer at 275 nm.

Furthermore, additional experiments are too carry out to detect the effect of solution pH on the adsorption method. The impact of solution pH on drug onto hydrogel is estimated via adjusting values of solution pH. Utilized in the experiment is 0. 1 N HCl and NaOH by the primary conc. of 100 mg/L of drug. All adsorption experiments are performed to ensure a single variable. The adsorption capacity of drug is calculated in the equation:

$$q_e = \frac{V(C_e - C_0)}{m} \quad (1)$$

RESULTS AND DISCUSSION

Fourier Transform Infrared (FTIR)

The infrared spectrum shows characteristic peaks of the prepared hydrogel surface within (1050–3500 cm^{-1}) wide band (3433 cm^{-1}) range, showing the interaction between the stretchy vibrations of the (O-H) group in the carboxyl and hydroxyl groups and the (N-H) group in the acryl amide and cross-linking agent (MBA). Also, an infrared spectrum of the adsorbed drug appears on the hydrogel surface after the adsorption process, as shown in Figure 1, where we notice that no new peak appears, only a slight difference in the intensity of adsorption, a clear evidence of the occurrence of physical adsorption due to weak bonds and the success of the adsorption process.[13,14]

Atomic Force Microscopy (AFM)

The technique AFM uses the nature of the surface, the average thickness of the surfaces, the average roughness (Ra), the mean root square roughness (Rq), and the extent of surface distribution and homogeneity, which are the most factors to describe the hydrogel surface topography,[15] as shown in Table 1

TEM

The surface properties were studied through the TEM technique to know the surface's nature and porosity. Through the results, the surface porosity was smooth. This is due to the strong correlation between the polymeric chains by means of the cross-linking agent,[16,17] as shown in Figure 2.

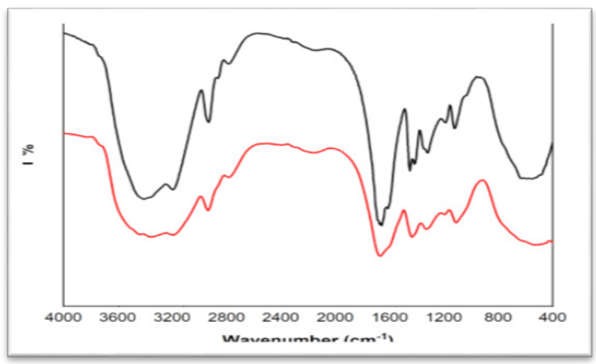

Figure 1: FT-IR spectra of hydrogel before and after adsorption

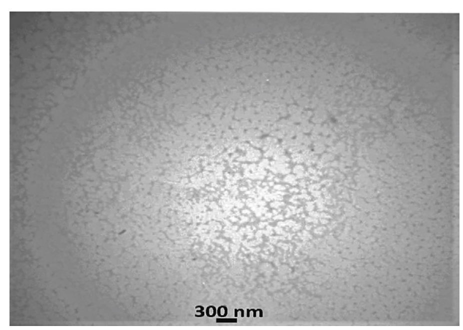

Figure 2: TEM image of the hydrogel surface

Table 1: Statistical value of roughness of hydrogel

Statistical value of roughness	*Hydrogel*
(Ra) nm	6.692
(Rq) nm	7.970
(Rsk) nm	0.5903
(Rku)	2.384
nm Rp	23.96
Rv nm	14.41

Surface Area Analysis

The hydrogel surface properties such as pore size, surface area, and pore diameter were studied. So, use the pore volume (BJH) and the adsorption isotherms (BET) adsorption-desorption. According to the classification of (BJH), the hydrogel follows the classification of the TV type, which indicates that the adsorption is multi-layered. The isotherm is of the type (H3), and this indicates that the surface pores are in the form of non-solid aggregates. They contain pores in their composition.[2,18] as shown in Figure 3.

Freundlich Isotherm

The isotherm Freundlich was based on heterogeneous and multi-layer adsorption. The linear equation appears in equation 2.

$$\ln q_e = \ln K_f + \frac{1}{2} C_e \quad (2)$$

Langmuir Isotherm

Isotherm Langmuir is based on two assumptions: adsorption energy was constant during the method, and adsorption of adsorbates occurs on a homogeneous hydrogel via monolayer adsorption.[19] The linear equation appears in equation 3.

$$\frac{C_e}{q_e} = \frac{1}{q_m \cdot K_L} + \left(\frac{1}{q_m}\right) * C_e \quad (3)$$

Figure 4 shows the adsorption isotherm of a drug onto a hydrogel surface at room temperature. The results applied two models of adsorption isotherms (Friendlich, Langmuir). It was found that it obeys the Friendlych isotherm depending on the value of (R2), and this indicates that the surface is multi-layered.[20-22]

Effect of Temperature and Calculation of Thermodynamic Functions of Adsorption Process

The adsorption of a drug on the hydrogel surface was studied at different temperatures (5, 20, 25, and 30°C). Through the results shown in Figure 5 and Table 2, the process of adsorption of a drug on the hydrogel increases with a decrease in temperature, meaning that the adsorption process is exothermic, and this is attributed to the fact that the increase in temperature leads to an increase in the solubility of minutes The drug is in water. This is a strong drop between the drug molecules and the active sites on the surface. Hence, the adsorption process favors low temperatures to increase the adsorption efficiency.[21,22]

The thermodynamic functions were calculated, and through the results shown in Table 2, It was found that the enthalpy value is negative, indicating that the reaction is exothermic, and through the value of Gibbs free energy, the value is negative that the reaction is spontaneous.[23]

Effect of Ionic Strength on the Adsorption

The effect of the ionic strength on the adsorption of a drug on the hydrogel surface was studied. In this study, the use of different weights of salts (NaCl, KCl, $CaCO_3$) was adopted under the experimental conditions of temperature, concentration, pH, and surface weight. The results showed in Figure 6 that the amount of drug adsorption on the surface of the hydrogel decreases with increasing salt concentration, due to the effect of competition between drug molecules and salt-positive ions on the active sites of the surface, as these salts increase the solubility of the drug in the solution.[24,25]

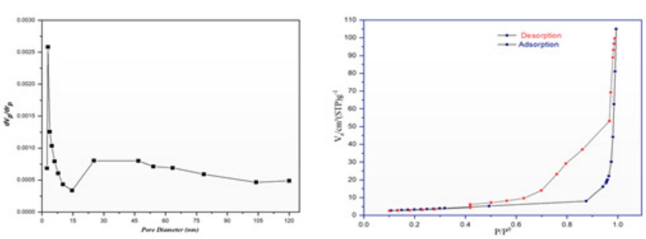

Figure 3: surface area analysis of the hydrogel

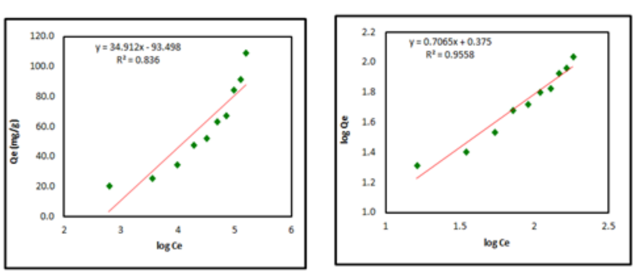

Figure 4: Adsorption isotherms of the drug adsorption at 25 °C.

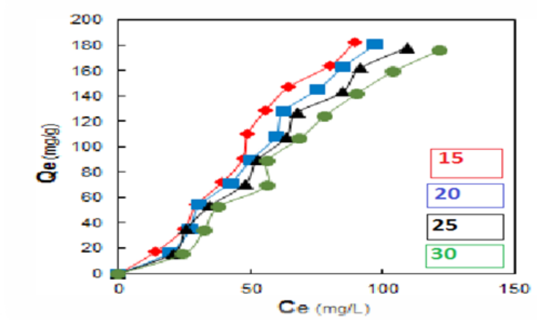

Figure 5: Effect of different temperatures of the adsorption drug on hydrogel.

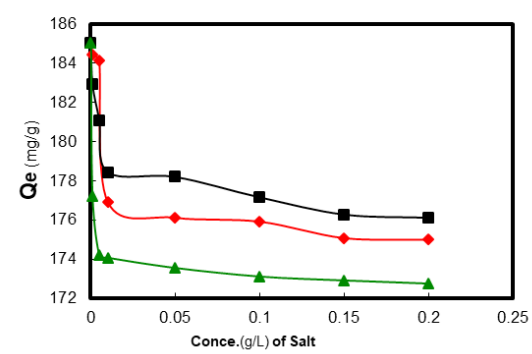

Figure 6: Effect of Ionic Strength on the adsorption onto hydrogel

Table 2: Thermodynamic parameters of the adsorption process of drug onto the hydrogel

Drug	*ΔH (kJ.mol-1)*	*ΔG (kJ.mol-1)*	*ΔS (J.mol-1. k-1)*	*(Keq)*
Streptomycin	-18.082	-2.984	-50.664	1.362

CONCLUSION

- The adsorption process of a drug on the hydrogel increases with a decrease in temperature
- The enthalpy value is negative, indicating that the reaction is exothermic, and through the value of Gibbs free energy, the value is negative that the reaction is spontaneous.
- the adsorption capacity of drug adsorption on the hydrogel surface decreases with increasing salt concentration.
- It was found to obey the Friendlych isotherm depending on the value of R2.

REFERENCES

1. Zaheer, Z., Al-Asfar, Aisha, Aazam, Elham Shafik, Adsorption of methyl red on biogenic Ag@Fe nanocomposite adsorbent: Isotherms, kinetics and mechanisms. Journal of Molecular Liquids, 2019. 283: p. 287-298.
2. Abdulsahib WK, Ganduh SH, Mahdi MA, Jasim LS. Adsorptive removal of doxycycline from aqueous solution using graphene oxide/hydrogel composite. Int J Appl Pharm. 2020;12(6):100-106.
3. Yahya A. Faleh, N.D.R., Removal of Metformin hydrochloride from Aqueous Solutions by using Carboxymethyl cellulose-g-poly(acrylic acid-co-acrylamide) Hydrogel: Adsorption and Thermodynamic Studies. IOP Conf. Series: Earth and Environmental Science 2021. 790: p. 012062.
4. Kamil, A.M., Mohammed, H.T., Alkaim, A.F., Hussein, F.H., Adsorption of Congo red on multiwall carbon nano-tubes: Effect of operational parameters. Journal of Chemical and Pharmaceutical Sciences, 2016. 9(3): p. 1128-1133.
5. Saeed U J, A.A.K., Saad M, Iftikhar A, Guohua S, Cheng-Meng C, Rashid A, Removal of azo dye from aqueous solution by a low-cost activated carbon prepared from coal: adsorption kinetics, isotherms study, and DFT simulation. Environmental Science and Pollution Research, 2020. 1: p. https://doi.org/10.1007/s11356-020-11344-4.
6. Aseel M. Aljeboree, N.A.A., Mohammed B. Alqaraguly,Makarim A. Mahdi, Layth S. Jasim, Ayad F. Alkaim, Adsorption Of Pollutants By Using Low-Cost (Environment-Friendly): Equilibrium, Kinetics And Thermodynamic Studies: A Review. Sys Rev Pharm 2020. 11(12): p. 1988-1997.
7. Qusay K. Mojar Alshamusi, AAMA, Makarim A. Mahdi, Layth S. Jasim and Aseel M. Aljeboree, Adsorption Of Crystal Violate (CV) Dye In Aqueous Solutions By Using P(Pvp-Co-Aam)/Go Composite As (Eco-Healthy Adsorbate Surface) : Characterization And Thermodynamics Studies. Biochem. Cell. Arch., 2021. 12: p. 2423-2431.
8. M. Ghaedi, AA, M. H. Habibi and A. R. Asghari, Removal of Malachite Green from Aqueous Solution by Zinc Oxide Nanoparticle Loaded on Activated Carbon: Kinetics and Isotherm Study. J. Ind. Eng. Chem.,, 2014. 20(17).
9. 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. 1234(1): p. 012095.
10. Hu XJ, Wang JS, Liu YG, Li X, Zeng GM, Bao ZL, Zeng XX, Chen AW, Long F. Adsorption of chromium (VI) by ethylenediamine-modified cross-linked magnetic chitosan resin: isotherms, kinetics and thermodynamics. Journal of hazardous materials. 2011 Jan 15;185(1):306-314.
11. Al-Gubury, H.Y., Fairooz, N.Y., Aljeboree, A.M., Alqaraguly, M.B., Alkaim, A.F., Photcatalytic degradation n-undecane using coupled ZnO-Co2O3. International Journal of Chemical Sciences, 2015. 13(2): p. 863-874.
12. Putra, EK, et al., Performance of activated carbon and bentonite for adsorption of amoxicillin from wastewater: Mechanisms, isotherms and kinetics. Water Research. 2009;43(9):2419-2430.
13. Hadi, Z.A., Aljeboree, A.M., Alkaim, A.F., Adsorption of a cationic dye from aqueous solutions by using waste glass materials: Isotherm and thermodynamic studies. International Journal of Chemical Sciences, 2014. 12(4): p. 1273-1288.
14. 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. 19(7): p. 137-143.
15. 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. 19(2): p. 31-37.
16. Chayid, M.A., Ahmed, Muthanna J., Amoxicillin adsorption on microwave prepared activated carbon from Arundo donax Linn: Isotherms, kinetics, and thermodynamics studies. Journal of Environmental Chemical Engineering. 3(3): p. 1592-1601.
17. Mohammed A. Jawad, A.J.K., Nadher D. Radia, Role of Sodium Alginate-g-poly (Acrylic acid-fumaric acid) Hydrogel for Removal of Pharmaceutical Paracetamol from Aqueous Solutions by Adsorption International Journal of Pharmaceutical Quality Assurance, 2021. 12(3): p. 202-205.
18. 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. 9(4): p. 558-562.
19. Choy, K.K.H., J.F. Porter, and G. McKay, Langmuir Isotherm Models Applied to the Multicomponent Sorption of Acid Dyes from Effluent onto Activated Carbon. J.Chem. Eng. Data. , 2000. 45 p. 575-584.
20. Tian, W., et al., Negative adsorption in the isotherm adsorption experiments of low-adsorption coal and shale. Natural Gas Industry B, 2019. 6(1): p. 44-50.
21. Gouamid M., OMR, and Bensaci M. B., Adsorption Equilibrium, Kinetics and Thermodynamics of Methylene Blue from Aqueous Solutions using Date Palm Leaves. Energy Procedia, 2013. 36: p. 898-907.
22. Alidadi, H., et al., Enhanced removal of tetracycline using modified sawdust: Optimization, isotherm, kinetics, and regeneration studies. Process Safety and Environmental Protection. 117: p. 51-60.
23. Ghaedi, M., Sadeghian, B., Pebdani, A.A., Sahraei, R., Daneshfar, A., Duran, C., Kinetics, thermodynamics, and equilibrium evaluation of direct yellow 12 removals by adsorption onto silver nanoparticles loaded activated carbon. Chem. Eng. J., 2012. 187: p. 133-141.
24. 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. 928(6): p. 062023.
25. Mansouri, H., Carmona, R.J., Gomis-Berenguer, A., Souissi-Najar, S., Ouederni, A., Ania, C.O., Competitive adsorption of ibuprofen and amoxicillin mixturesfrom aqueous solution on activated carbons. . J. Colloid Interface Sci. , 2015. 449: p. 252-260.