RESEARCH ARTICLE

Synthesis of Sodium Alginate-g-poly(Acrylic acid-fumaric acid) hydrogel and Application as Adsorbent of Amoxicillin from Aqueous Solution

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ABSTRACT

Removal of drug from aqueous solution is of very significance because of their possible reverse effects on living organisms and elaborated resistance of the bacteria. The study deals with amoxicillin (AMX) removal utilizing hydrogel. The Sodium alginate-g-poly(Acrylic acid-fumaric acid) hydrogel was described through fourier transform infrared (FT-IR)), field emission scanning electron microscopy (FESEM), and UV-visible spectroscopy. The adsorption study was done to estimation the impact of (10-100 mg L^{-1}) conc. of AMX Optimization seem the best % removal at 97.40% at con-centration 100 mg L^{-1}, and contact time 2 hours. study two isotherm model Freundlich, Langmuir model the best fitted Freundlich isotherm (R2 = 0.9772) and study three type of kinetic model first order, second order and Elchovich model and the best result at second order (R2 = 0.9041).

Keywords: Adsorption, Amoxicillin AMX drug, Isotherm, Kinetic model, Removal.

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INTRODUCTION

Nowadays, pharmaceuticals are considered one of the most important water pollutants because of their widespread use. Pharmaceuticals are classified as a class of health care products and are used all over the world to enhance human health.[1-6] They are also applied in animal care and in agriculture, where antibiotics are released into wastewater and consider very dangerous materials.[7-9] AMX is an antibiotic with widespread use in veterinary and human medicine due to poor metabolism in the organism, where very large amounts of amoxicillin are discharged into effluents.[10-12] Therefore, there are several effective ways to remove drugs from wastewater, including ozone, photo-oxidation, and adsorption. The adsorption process is one of the simplest, easiest, and cheapest methods used to remove pollutants, especially medicines, from water and sludge for use on very high efficiency, cheap and easy to prepare surfaces.[13,14]

In this research, a very highly effective hydrogel surface was used to remove amoxicillin, where several techniques were used, including FTIR, FESEM, where the effect of AMX drug concentration, adsorption isotherms Kinetic model were studied.

EXPERIMENTAL PART

The calibration curve, solutions of different AMX drug concentrations was prepared via sequential dilutions. The absorbance values of these solutions was measured at the carefully chosen λ_{max} value as shown in Figure 1. The calibration in the range concentration that falls in the region of Lambert Beer law was employed. The chemical structure of AMX ($C_{16}H_{19}N_3O_5S$) and molar mass (365.40 g/mol) drug. The maximum absorbance

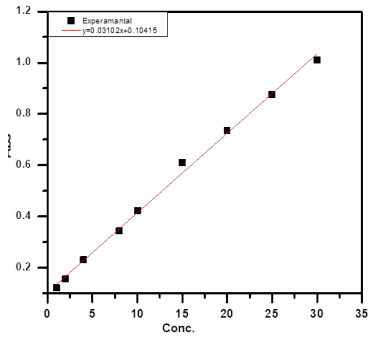

Figure 1: Calibration curve for the AMX drug

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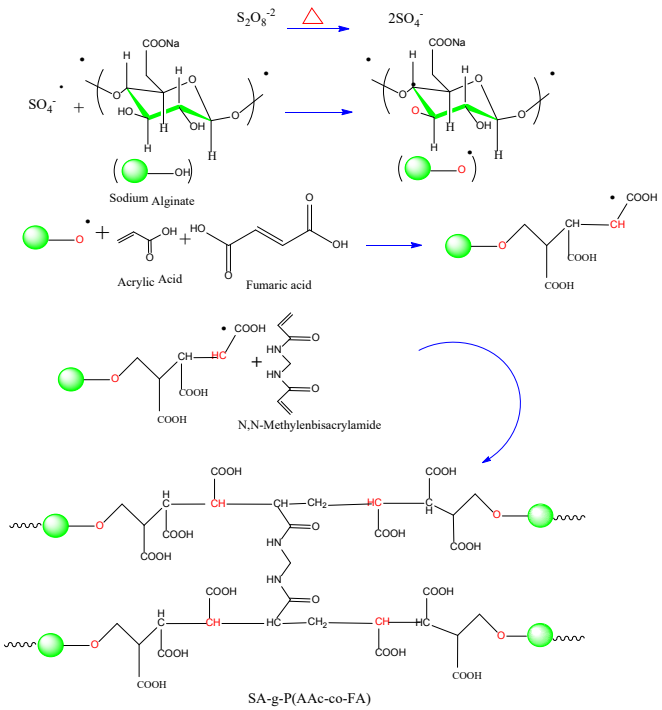
Preparation of Hydrogel



Scheme 1: Preparation of (Sodium alginate-g-poly (Acrylic acid-fumaric acid) hydrogel

of AMX happens at wavelengths of 230 nm. The 1000 mg L^{-1} drug solutions and respective diluted working solution were prepared freshly via weight and dissolving via 1.0 g of AMX in 1000 mL elementary flasks (Scheme 1).

Effect of Initial Drug Concentration:

A series of several concentrations of AMX drug of 100 L was utilized in this study (10–100) ppm, was adding to elementary flask in the presence of 0.05 g of hydrogel these sequence were putting in a shaker water bath for 2 hours, at pH 7.2; temp. 25°C; weight of hydrogel 0.05 gm for 100 mL after that the supernatant were separated by centrifuge and measured the remaining concentration via utilizing spectrophotometer UV-v at the λ_{max} 230 nm for drug.

The adsorption efficiency was calculated from equation 1:[15]

$$qe = \frac{(C_0 - C_e) * V_L}{m_{gm}} \quad (1)$$

q_e = Quantity of AMX adsorbed per unit weight of hydrogel (mg/g). C_o= Primary drug conc. (mg.L^{-1}), C_e= Equilibrium conc. drug (mg.L^{-1}).m = weight of hydrogel (g). (E%) of the drug was estimation on the basis of reduction in absorbance at λ_{max}.[16]

$$E\ \% = \frac{C_0 - C_e}{C_0} * 100 \quad (2)$$

C_0 and C_e are primary and equilibrium drug concentration, at the same order.

RESULTS AND DISCUSSION

Fourier Transform Infrared Spectroscopy (FTIR)

The hydrogel was characterized through spectroscopy FTIR. from 4000–400 cm^{-1} with a resolution of 1 cm^{-1}. The FTIR spectra of hydrogel before and after AMX adsorption are look in Figure 2, It is observed from the figure after the adsorption process that no new pick appears, only there is a slight change in the intensity of adsorption and this is evidence of the occurrence of the adsorption process and the adsorption process is of a physic sorption.[17,18]

Field Emission Scanning Electron Microscopy (FESEM)

FESEM of the hydrogel surface before the adsorption process noted that the surface contains many cavities and irregular assemblies. However, after the adsorption process occurred, the surface became smooth and smooth, and this is evidence of the loading of the drug on the surface and the occurrence of the adsorption process,[19-21] as appear in Figure 3.

Effect of Initial Concentration of AMX Drug

Figure 8 shows the plots of amounts of AMX drug adsorbed (qe) and removal (R%) of AMX several initial concentrations of AMX drug C^o at various experimental conditions. From the figure, it can be look that the removal E% of AMX drug decreased through in-creasing in the concentrations of AMX and found the removal percentage E% decrease from 97.87% to 82.11% but also the adsorption capacity of AMX rise with increased initial drug concentration and found the adsorption efficiency increase from 18.21 to 168.22 mg/g. Because when the initial concentration drug increases, the number of collisions between drug and the hydrogel increases, which improves the adsorption method. The influence of AMX drug concentration on hydrogel efficiency was found to be of considerable significance for the AMX used.[22-25]

Adsorption Model

Freundlich Isotherm: The Freundlich equation is one of the most significant utilized models in the case of solution adsorption.[26,27] This Isotherm accepts that the surface of the hydrogel is heterogeneous because of the variance energy levels for adsorption sites Freundlich isotherm model has been defined in equation.[28,29]

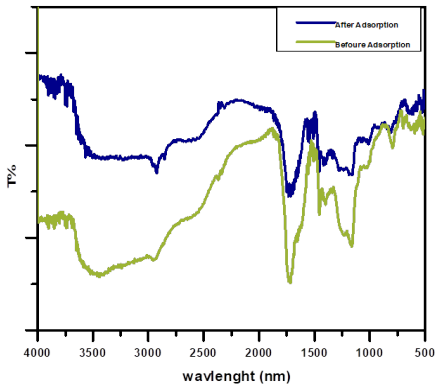


Figure 2: FTIR spectrum of hydrogel before and after adsorption of AMX drug.

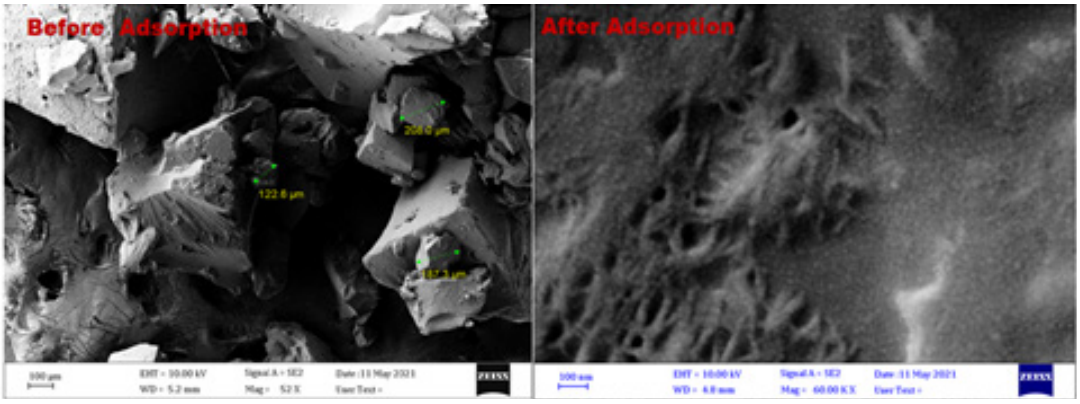


Figure 3: FESEM of hydrogel before and After adsorption

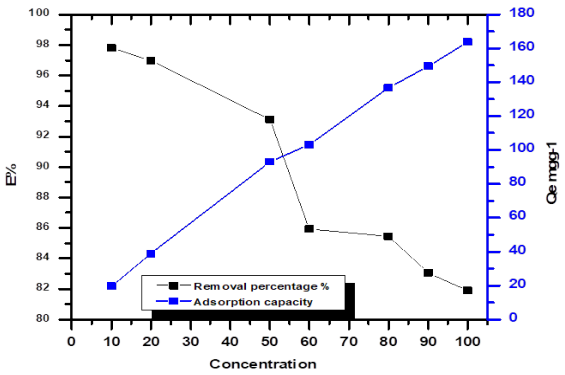


Figure 4: Effect of concentration of AMX drug adsorption utilizing hydrogel (weight of hydrogel 0.05 g, 25°C, and pH 7.2).

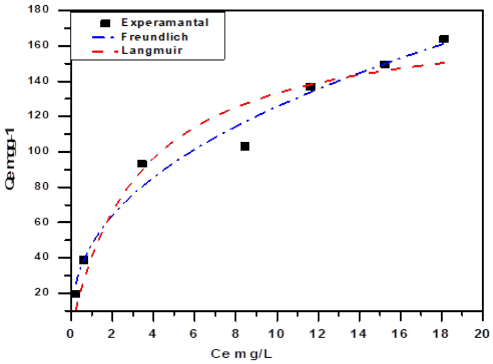


Figure 5: Different isotherm absorption model patterns non-linear fit for absorbing the AMX drug on hydrogel, primary conc. = 100 mgL^{-1}, Temp. = 25°C, the mass of hydrogel = 0.05 g

$$q_e = K_f C_e^{1/n} \quad (3)$$

Langmuir Isotherm: isotherm Langmuir has a widespread use to absorb contaminants from the solution liquid.[30,31] Langmuir single-layer models adsorption isotherm are applicable to solid-liquid adsorption methods.[32] Here, adsorption Langmuir model has been defined in equation 6.

$$q_e = \frac{q_0 K_L C_e}{1 + K_L C_e} \quad (4)$$

q_e refers to the quantity absorbed in each unit mass of the hydrogel at equilibrium ($mg.g^{-1}$), Ce concentration Equilibrium of the adsorbent in the solution following absorption (mg/L), qo involves the Empirical constant Langmuir representing the greatest absorption efficiency ($mg.g^{-1}$) (Figure 4). Furthermore, K_L empirical Langmuir constant (Lmg^{-1}) and Freundlech model reflected the good fitting to absorbe the AMX onto hydrogel as appear R^2 (0.9773) values and K_F rise by increased temperature of the adsorption (Table 1). The values of the model parameter are provided of the diagram of (Qemg/g) vis (Cemg/L) (Figure 5) and (Table 1).

Table 1: The Freundlech, Langmuair, the model factors for AMX drug absorbed onto hydrogel at 25°C.

Isotherm models	*Parameters*	*AMX*
Langmuir	qm ($mg.g^{-1}$)	178.578 ± 20.092
	KL($L.mg^{-1}$)	0.2931 ± 0.1241
	R2	0.9333
Freundlech	KF	47.772 ± 5.377
	1/n	0.4197 ± 0.0445
	R2	0.9773

Kinetic Models

The kinetics adsorption give details and gives information about mechanics of the adsorption (Figure 6). In this study, three models of kinetics adsorption were utilized: first, second, and Elcovich models.

$$q_t = q_e [1\text{-}\exp(kf\, t)] \quad (5)$$

The model of the kinetics adsorption process might also be called in second order equation[33].The non linear form of t equation is expressed as:

$$qt = \frac{K2qe2t}{1 + K2qet} \quad (6)$$

The structure nonlinear of the Elcovich model (Chemi-sorption model kinetic) [49] model as appear in equation 7:

$$qt = \frac{1}{\beta}\ln(\alpha\beta) + \frac{1}{\beta}\ln t \quad (7)$$

Table 2: Kinetic model first model, second model, and Elcovich model correlation coefficients for AMX drug adsorption on to hydrogel

Model	*Equation*	*Parameters*	*Value*
First model	qt = qe [1-exp (kf t)]	Kt(min^{-1})	0.1797 ± 0.0285
		qe(calc)(mgg^{-1})	155.114 ± 3.4333
		R2	0.5351
Second model	$qt=\frac{K2qe2t\ \ K2qe2t}{1+K2qet1+K2qet}$	K2(g/mg/min)	0.3103 ± 0.0410
		qe(calc)(mgg^{-1})	165.903 ± 2.563
		R2	0.9045
Elcovich model	$qt=\frac{11}{\beta\beta}1\beta\ln(\alpha\beta)+\frac{1}{\beta}\ln t\ \frac{1}{\beta}\ln t$	α (mg g-1 min^{-1})	5.340 ± 1.321
		β (g min^{-1})	50.576 ± 4.374
		R2	0.8751

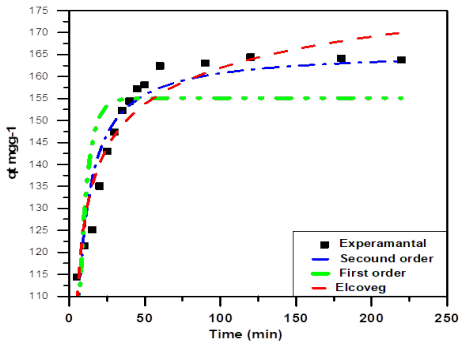


Figure 6: First-order reaction kinetics, second order, and Elcovich model of AMX drug adsorption on hydrogel.

The model of kinetic result from the three model appear in Table 2. The nonlinear plots of qt vs. t for several initial concentrations drug demonstrated best consistency among the experimental and estimation values qe. Also, the kinetic model second-order has higher R2 than the First model and Elcovich. As a result, the adsorption is good fit to the model second than the model of first and Elcovich.[27,34] Kinetics adsorption are related with the intra-particle diffusion concept.

CONCLUSION

- In this study, a hydrogel surface with very high efficiency was used to remove amoxicillin
- Two types of adsorption isotherms have been studied, and the best obey is the Frendlig isotherm.
- Three Kinetic models First model, second model and Elcovich model were studied, where the best obedience was second model
- The adsorption efficiency rises with increasing conc. of AMX drug, while the percentage E% removal decreases with increasing concentration.

REFERENCES

1. Safa A. Yassin, A.H.A.k., and Firas H. Abdulrazzak Enhance the Growth of Multi- walled Carbon Nanotubes from Coal by Catalytic Chemical Flame Deposition. IOP Conf. Series: Materials Science and Engineering, 2019.571:1-3.
2. Maleki A, Mohammad M, Emdadi Z, Asim N, Azizi M, Safaei J. Adsorbent materials based on a geopolymer paste for dye removal from aqueous solutions. Arabian Journal of Chemistry. 2020;13(1):3017-3025.
3. Azuma T, Otomo K, Kunitou M, Shimizu M, Hosomaru K, Mikata S, Mino Y, Hayashi T. Removal of pharmaceuticals in water by introduction of ozonated microbubbles. Separation and Purification Technology. 2019;212:483-489.
4. Layth S J, Aljeboree, M.A., Removal of Heavy Metals by Using Chitosan/ Poly (Acryl Amide-Acrylic Acid)Hydrogels: Characterization and Kinetic Study. Neuro Quantology, 2021. 19(2):31-37.
5. Waleed K. Abdulsahib, S.H.G., Nadher D. Radia, Layth S. Jasim, New Approach for Sulfadiazine Toxicity Management using Carboxymethyl Cellulose Grafted Acrylamide Hydrogel. Int. J. Drug Deliv Technol. 2020;10(2):259-264.
6. Falah H. Hussein, A.M.A., Zeyad O. Musa, Firas H. Abdulrazzak, Mohammed Bassim Alqaragoly and Ayad F. Alkaim, Is it Photocatalytic Degradation of Textile Dyes a Friendly Method? Methyl Violet Dye as a Model for Application in Aqueous Solutions in the Presence of Commercial TiO2. Int J Recent Technol Eng. (IJRTE), 2019. 8(2S3).
7. Radhi, I.M., F.H. Abdulrazzak, and T.A. Himdan, Influence of water in size of Synthesized Carbon Black Nanoparticles from Kerosene by Flame Method. IOP Conf. Series: Mater. Sci. Eng.. 2019. 571.
8. Aseel M Aljeboree, A.N.A., Colorimetric determination of Amoxicillin using 4-Aminoantipyrine and the effects of different parameters. J. Phys. Conf. Ser. 2019;12(5): 052067.
9. Aljeboree, A.M. and A.F. Alkaim, Comparative Removal Of Three Textile Dyes From Aqueous Solutions Byadsorption: As A Model (Corn-Cob Source Waste) of Plants Role in Environmental Enhancement. Plant Arch. 2019;19(1):1613-1620.
10. Aljeboree, A.M. and A.N. Alshirifi, Oxidative coupling of Amoxicillin using 4-Aminoantipyrine: Stability and higher sensitivity. J. Phys. Conf. Ser. 2019;1294(5):052001.
11. Aseel M Aljeboree, A.N.A., Determination of Phenylephrine Hydrochloride and Amoxicillin in a Binary Mixture using Derivative Spectrophotometry Methods. Int. J. Pharm. Qual., 2019;10(3):168-177.
12. Aljeboree, A.M., Abbas, A. S., Removal of Pharmaceutical (Paracetamol) by using CNT/ TiO2 Nanoparticles. J. Glob. Pharma Technol., 2019;11(1):199-205.

13. Firas H. Abdulrazzak, A.M.A., Tariq H. Al Mgheer, , Israa Mohammed, Aseel M. Ajobree, Ayad F. Alkaim, Takialdin A. Himdan, Falah H. Hussein, Novel Coronavirus 2019-nCoV Selectivity and Activity between Asians and Europeans Populations: A Review. Int. J. Psychosoc. Rehabilitation., 2020;24(05).
14. Liqaa H. Abd, R.A., Aseel M. Aljeboree, Firas H. Abdulrazzak, Falah H. Hussein and Ayad F. Alkaim, Role of Semiconductors (Zinc Oxide as a Model) for Removal of Pharmaceutical Tetracycline (TCs) from Aqueous Solutions in the Presence of Selective Light. Int J Recent Technol Eng. (IJRTE), 2019. 8(2S3): p. DOI : 10.35940/ijrte.B1270.0782S319.
15. Doğan M, Alkan M, Demirbaş Ö, Özdemir Y, Özmetin C. Adsorption kinetics of maxilon blue GRL onto sepiolite from aqueous solutions. Chem. Eng. J. 2006;124(1-3):z89-101.
16. Ahmad R. Studies on adsorption of crystal violet dye from aqueous solution onto coniferous pinus bark powder (CPBP). Journal of hazardous materials. 2009;171(1-3): 767-773.
17. Abdulrazzak FH. Enhance photocatalytic activity of TiO2 by carbon nanotubes. Int. J. Chemtech Res. 2016;9(3): 431-443.
18. Alkaim AF, Alqaraguly MB. Adsorption of basic yellow dye from aqueous solutions by activated carbon derived from waste apricot stones (ASAC): equilibrium, and thermodynamic aspects. Int. J. Chem. Sci. 2013;11(2):797-814.
19. Jassm AM, Hussein FH, Abdalrazak FH, Alkaim AF, Joda BA. Synthesis and characterization of carbon nanotubes by modified flame fragments deposition method. Asian J Chem. 2017;29(12):2804-2808.
20. Atyaa AI, Radhy ND, Jasim LS. Synthesis and characterization of graphene oxide/hydrogel composites and their applications to adsorptive removal congo red from aqueous solution. J. Phys. Conf. 2019;1234(1)012095. IOP Publishing.
21. Alkaim AF, AM A. White marble as an alternative surface for removal of toxic dyes (Methylene Blue) from aqueous solutions. Int. J. Adv. Sci. and Technol. 2020;29(5):5470-5479.
22. Aljeboree AM, A.. Adsorption and Removal of pharmaceutical Riboflavin (RF) by Rice husks Activated Carbon. Int. J. Pharm. Res.. 2019;11(2):255-261.
23. 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.
24. Nandi BK, Goswami A, Purkait MK. Adsorption characteristics of brilliant green dye on kaolin. Journal of hazardous materials. 2009 Jan 15;161(1):387-395.
25. Ahmed MJ. Adsorption of quinolone, tetracycline, and penicillin antibiotics from aqueous solution using activated carbons. Environ. Toxicol. Pharmacol. 2017;50:1-10.
26. Crini G, Peindy HN, Gimbert F, Robert C. Removal of CI Basic Green 4 (Malachite Green) from aqueous solutions by adsorption using cyclodextrin-based adsorbent: kinetic and equilibrium studies. Sep. Purif. Technol. 2007;53(1):97-110.
27. Mahde BW, Radia ND, Jasim LS, Jamel HO. Synthesis and characterization of polyacrylamide hydrogel for the controlled release of aspirin. J. Pharm. Sci. 2018;10(11):2850-2854.
28. Ho YJ, Eichendorff J, Schwarting RK. Individual response profiles of male Wistar rats in animal models for anxiety and depression. Behav. Brain Res. 2002;136(1):1-2.
29. Özacar M, Şengil İA. A kinetic study of metal complex dye sorption onto pine sawdust. Process Biochem. 2005;40(2):565-572.
30. Langmuir I. The constitution and fundamental properties of solids and liquids. Part I. Solids. J. Am Chem. Soc. 1916;38(11):2221-2295.
31. Langmuir I. The adsorption of gases on plane surfaces of glass, mica and platinum. J Am Chem. Soc. 1918 Sep;40(9):1361-1403.
32. Freundlich H, Heller W. The adsorption of cis-and trans-azobenzene. J. Am Chem. Soc. 1939;61(8):2228-2230.
33. Ho YS, McKay G, Sorption of dye from aqueous solution by peat. S Chem. Eng. J, 1998;70:115.
34. Layth S. Jasim, N.D.R., Hayder O. Jamel Synthesis and Characterization of Poly (Acryl Amide - Maleic Acid) Hydrogel: Adsorption Kinetics of a Malachite Green from Aqueous Solutions. Eurasian Journal of Analytical Chemistry 2018;13(1b):74.