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ORIGINAL ARTICLE

Removal of Amoxicillin from Aqueous Solutions by using Synthesized Highly Hydrogel Surface as a Good Adsorbent

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KEYWORDS

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Removal;
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ABSTRACT: Because of the potential for reversible effects on living organisms and bacterial elaboration resistance, removing drugs from aqueous solutions is critical. Deals with the amoxicillin AMX removal trial using hydrogel. (F-TIR), (F.E-SEM), and UV-visible spectroscopy were used to describe the hydrogel of sodium alginate-g-poly (Acrylic acid-fumaric acid). The purpose of the adsorption investigation was to determine the impact of (10-100 mg L^{-1}) conc. of AMX Optimization appear to have the best percentage percent removal at 97.40 percent at concentration 100 mg L^{-1}, and contact duration 2hr. Take a look at two isotherm models. The second order model (R2= 0.9041) outperforms the Freundlich, Langmuir (R2= 0.9772), and three types of kinetic models (first order, second order, and Elchovich).

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 [1-5] They are also applied in animal care and in agriculture, where antibiotics are released into wastewater and consider very dangerous materials [6-8]. Amoxicillin 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 [9-11]. Therefore, there are several effective ways to remove drugs from wastewater, including ozone, photo oxidation and adsorption. These methods are characterized as simplest, easiest and cheapest used to remove pollutants, especially medicines, from water and sludge for use on very high efficiency, cheap and easy to prepare surfaces [12, 13]. 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 concentration of AMX drug, adsorption isotherms and Kinetic model were studied.

MATERIALS AND METHODS

The calibration curve, solutions of different AMX drug concentrations was prepared via sequential dilutions. The values of absorbance of these solutions were measured at the carefully chosen λmax value as appear in Figure 1.

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The calibration in the concentration range is linear according to Beer-Lambert law. The chemical structure of AMX ($C_{16}H_{19}N_3O_5S$). The maximum absorbance of AMX happens at wavelengths of 230 nm. By weighing and dissolving 1.0 g of AMX, the 1000 mg L^{-1} drug solutions and their diluted working solutions were prepared fresh in 1000 mL elementary flasks. (Figure 2).

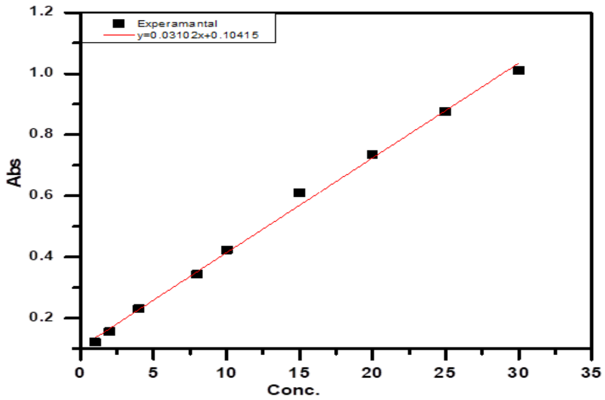

Figure1. Calibration curve for the AMX drug.

Preparation of hydrogel

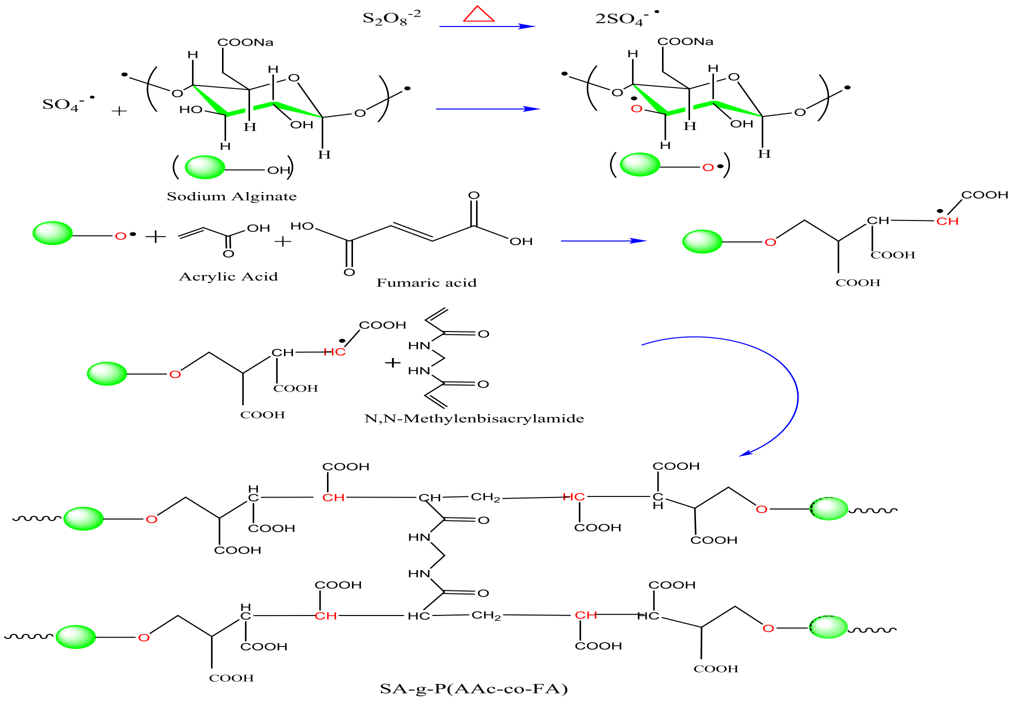

Figure 2. (Hydrogel of sodium alginate-g-poly(Acrylic acid-fumaric acid) preparation.

Effect of initial drug concentration

A series of several concentrations of AMX drug of 100 mL was utilizing 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 water bath shaker for 2hr, pH= 7.2 ; temp. 25 C; weight of hydrogel 0.05 gm for 100 ml After that, The remaining concentration was determined using a spectrophotometer after the supernatant was centrifuged at the λ_{max} 230 nm for drug. The adsorption efficiency was calculated from equation (1): [14]

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

q_e = The amount of AMX adsorbed per gram 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 determined using the decrease in absorbance at λ_{max} [15]

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

RESULTS AND DISCUSSION

FTIR

The hydrogel was studied using FTIR spectroscopy from 4000 to 400 cm^{-1}, with a resolution of 1 cm^{-1}. Figure 3 shows the FTIR spectra of hydrogel before and after AMX adsorption. The fact that no new pick appears after the adsorption process, simply a slight change in the degree of adsorption, proves the adsorption process' presence, this is proof of the adsorption process' occurrence, which is of a physic sorption. [16, 17].

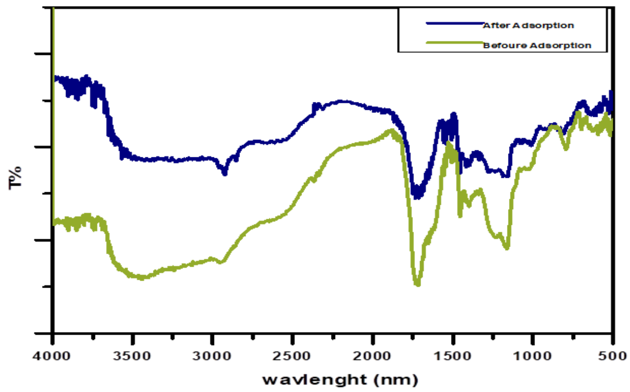

Figure 3. FT-IR hydrogel spectrum before and after AMX drug adsorption.

FESEM

Before the adsorption process, the hydrogel's surface has numerous voids and uneven assemblies, while after the adsorption process, the surface has few voids and uneven assemblies, the surface became smooth and smooth, indicating that the drug was loaded on the surface and the adsorption process occurred. [18, 19] as appear in Figure 4.

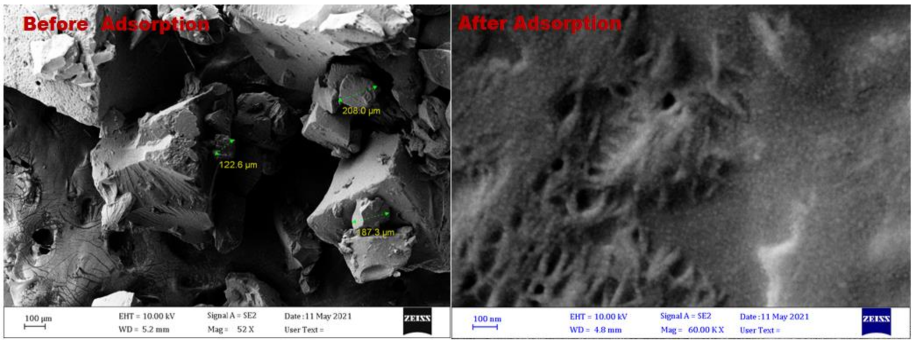

Figure 4. Before and after adsorption FESEM of hydrogel.

Effect of initial concentration of AMX drug

Figure 5 look the plot the amounts of AMX drug adsorbed (qe) and removal (R%) of AMX several initial concentration of AMX drug C^o at various experimantal conditions. From the Figure, it can be look that the removal E% of AMX drug de-creased 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 increase initial drug concentration and found the adsorption efficiency increase from (18.21 to 168.22

mg/g). because when the number of collisions increases with the increase in the initial concentration among drug and the hydrogel increasing, that get better the adsorption method. For the AMX utilized, there was a substantial effect of AMX drug concentration on hydrogel efficiency. [20-23].

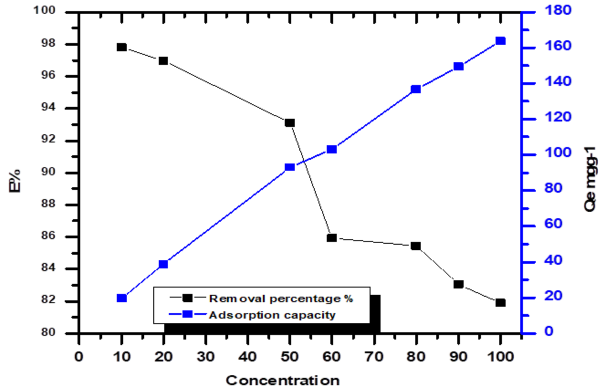

Figure 5. Effect of AMX drug adsorption concentration in a hydrogel (25°C, pH 7.2, weight of hydrogel 0.05 g).

Adsorption model

Freundlich isotherm

The Freundlich equation is one of the utmost significant utilized models in the case of adsorption of solution. [24]:This Isotherm accepts that the surface of the hydrogel is hetero-geneous because of the variance energy levels for adsorption sites Freundlich isotherm model has been defined in equation [25, 26]

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

Langmauir Isotherm: isotherm Langmauir has a widespread use to absorbe contaminants from the solution liquid [27, 28]. The adsorption isotherm of Langmuir single-layer models can be applied to solid-liquid adsorption methods [29]. Here, adsorption Langmauir model has been defined in equation (6).

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

qe is for the amount absorbed (mg g^{-1}), Ce stands for the adsorbent content in the solution after absorption (mg/L), and qo stands for the Empirical constant Langmauir, which represents the highest absorption efficiency (mg g^{-1}). Furthermore, the Freundlech model and the KL empirical Langmauir constant (L mg^{-1}) indicated a strong fit to absorb AMX onto hydrogel, R2 (0.9773) values and KF increase as adsorption temperature rises (Table 1), the model parameter values are shown in the figure of (Qemg g^{-1}) vs. (Cemg L^{-1}) (Figure 6) and (Table 1).

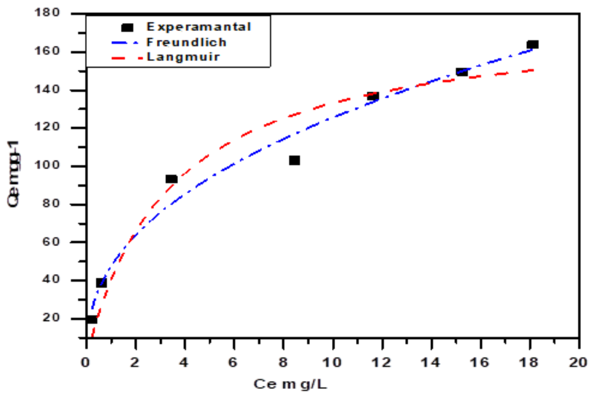

Figure 6. Different non-linear isotherm absorption model patterns for absorbing the AMX medication on hydrogel, main concentration = 100 mg L^{-1}, temperature = 25°C, hydrogel mass = 0.05 g

Table 1. At 25°C, the model variables for AMX medication absorbed on to hydrogel (Freundlech and Langmuair).

Isotherm models	Parameters	AMX
Langmauir	qm (mg g^{-1})	178.578± 20.092
	K_L(L mg^{-1})	0.2931±0.1241
	R^2	0.9333
Freundlech	K_F	47.772±5.377
	1/n	0.4197±0.0445
	R^2	0.9773

Kinetic models

The kinetics of adsorption provides details and information on the adsorption mechanics. Three kinetics adsorption models were used in this study: first models, second models, and the Elcovich model.

$$qt = qe\,[1-\exp(kf\,t)] \qquad (5)$$

The kinetics adsorption process model is also known as a second order equation[30] . The nonlinear form of the t equation is as follows:

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

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

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

Table 2 shows the kinetic model results from the three models. For numerous starting drug concentrations, nonlinear plots of qt vs. t revealed the best concordance between experimental and estimation values qe. In addition, Elcovich's and the first model's R2 are lower than the second-order kinetic model's. As a result, the second model is better adapted to the adsorption than the first, and Elcovich prefers it. Kinetics adsorption is related with the intra-particle diffusion concept. (Figure 7)

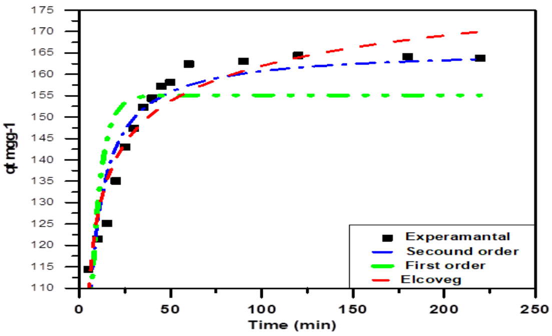

Figure 7. AMX drug adsorption on hydrogel (first and second order reaction kinetics, and Elcovich model).

Table 2. AMX drug adsorption on hydrogel, Elcovich model and correlation coefficients utilizing first and second order reaction kinetics

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

CONCLUSIONS

1- In this study, to eliminate amoxicillin, a hydrogel surface with a high effectiveness was utilized.

Two types of adsorption isotherms have been studied, and the best obey is the Frendlig isotherm.2-

3- Three kinetic first and second order reaction kinetics, as well as the Elcovich model, were investigated, with the second model proving to be the most effective.

4-The adsorption efficiency rises with increasing conc. Of AMX drug, with increasing concentration, the percentage E percent removal drops.

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REFERENCES

1. Layth S., J Aljeboree M.A., 2021. Removal of Heavy Metals by Using Chitosan/ Poly (Acryl Amide-Acrylic Acid)Hydrogels: Characterization and Kinetic Study . Neuro Quantology. 19(2), 31-37.

2. Yahya A., Faleh N.D.R., 2021. 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. 790, 012062.

3.Wenyan Jiang L.Z., Xiaoming G., Mei Y., Yiwen L., Yijun W., Yousen Zh., Guangtao W., 2019. Adsorption of cationic dye from water using an iron oxide/activated carbon magnetic composites prepared from sugarcane bagasse by microwave method. Environmental Technology. 2. DOI: 10.1080/09593330.

4. Santos S.C.R.O., Boaventura A.F.M., 2019. Bentonitic clay as adsorbent for the decolourisation of dyehouse effluents. J Clean Prod. 126, 667-67.

5. Sakin O.A., Belal H.M., Arbi H.M., 2019. Adsorption thermodynamics of cationic dyes (methylene blue and crystal violet) to a natural clay mineral from aqueous solution between 293.15 and 323.15 K. Arabian Journal of Chemistry. 11(5), 615-623.

6. Israa M., Radhi F.H.A., Takialdin A., 2019. Himdan Influence of water in size of Synthesized Carbon Black Nanoparticles from Kerosene by Flame Method. IOP Conf. Series: Materials Science and Engineering. 571.

7. Aljeboree A.M., 2019. Colorimetric determination of Amoxicillin using 4-Aminoantipyrine and the effects of different parameters. Journal of Physics: Conference Series. 12(5), 052067.

8. Aljeboree A.M., 2019. Comparative removal of three textile dyes from aqueous solutions by adsorption: as a model (corn-cob source waste) of plants role in environmental enhancement. Plant Archives. 19(1), 1613-1620.

9. Aljeboree A.M., Alshirifi A.N., 2019. Oxidative coupling of Amoxicillin using 4-Aminoantipyrine: Stability and higher sensitivity. Journal of Physics: Conference Series. 1294(5), 052001.

10. Aljeboree A.M, Alshirifi A.N.A., 2019. Determination of Phenylephrine Hydrochloride and Amoxicillin in a Binary Mixture using Derivative Spectrophotometry Methods. International Journal of Pharmaceutical Quality Assurance. 10(3), 168-177.

11. Aljeboree A.M., Abbas A.S., 2019. Removal of Pharmaceutical (Paracetamol) by using CNT/ TiO_2 Nanoparticles. Journal of Global Pharma Technology. 11(1), 199-205.

12. Firas H., Abdulrazzak A.M.A., Tariq H. Al M., Israa M., Ajobree A.M., Ayad F.A.,Takialdin A. H., Falah H.H., 2020. Novel Coronavirus 2019-nCoV Selectivity and Activity between Asians and Europeans Populations: A Review. International Journal of Psychosocial Rehabilitation. 24(5), 2829-2837.

13. Liqaa H., Abd R.A., Aljeboree Aseel M., Firas H., Abdulrazzak Falah H.H., Ayad F. A., 2019. Role of Semiconductors (Zinc Oxide as a Model) for Removal of Pharmaceutical Tetracycline (TCs) from Aqueous Solutions in the Presence of Selective Light. International Journal of Recent Technology and Engineering (IJRTE). 8(2S3). DOI : 10.35940/ijrte.B1270.0782S319.

14. Doğan M., Alkan M., Demirbas O., Ozdemir Y., Ozmetin C., 2006. Adsorption kinetics of maxilon blue

GRL onto sepiolite from aqueous solutions. Chem Eng J. 124, 89-101.

15. Ahmad R., 2009. Studies on adsorption of crystal violet dye from aqueous solution onto coniferous pinus bark powder (CPBP). J Hazard Mater. 171, 767-773.

16. Abdulrazzak F.H., 2016. Enhance photocatalytic Activity of TiO_2 by Carbon Nanotubes. International Journal of Chem Tech Research. 9(3), 431-443

17.Ayad F., Alkalim M.B.A., 2013. Adsorption of basic yellow dye from aqueous solutions by activated carbon derived from waste apricot stones (ASAC): equibrium , and thermodynamic aspects. Int J Chem Sci. 11(2), 797-814.

18. Ali M., Jassm B.A.J., Firas H., Abdulrazzak A., Alkaim F., Falah H.H., 2017. Synthesis and Characterization of Carbon Nanotubes by Modified Flame Fragments Deposition Method. Asian Journal of Chemistry. 29(12), 2804-2808.

19. Ayad F. Alkaim, A.M.A., 2020. White Marble as an Alternative Surface for Removal of Toxic Dyes (Methylene Blue) from Aqueous Solutions. International Journal of Advanced Science and Technology. 29(5), 5470 - 5479.

20. Aljeboree A.M., 2019. Adsorption and Removal of pharmaceutical Riboflavin (RF) by Rice husks Activated Carbon. International Journal of Pharmaceutical Research. 11(2), 255-261.

21. Abdulsahib W.K., Ganduh S.H., Mahdi M.A., Jasim L.S., 2020. Adsorptive removal of doxycycline from aqueous solution using graphene oxide/hydrogel composite. International Journal of Applied Pharmaceutics. 12(6), 100-106.

22. Nandi B.K., Purkait M.K., 2009. Adsorption characteristics of brilliant green dye on kaolin. J Hazard Mater. 161, 387-395.

23. Ahmed M.J., 2017. Adsorption of quinolone, tetracycline, and penicillin antibiotics from aqueous solution using activated carbons: Review. Environmental Toxicology and Pharmacology. 50, 1-10.

24. Crini G., Peindy H.N., Gimbert F., Robert C., 2007. Removal of C.I. basic green 4, (malachite green) from aqueous solutions by adsorption using cyclodextrin-based adsorbent: kinetic and equilibrium studies. Purif Technol. 53, 97-110.

25. Ho Y.S., Porter J.F., McKay G., 2002. Individual response profiles of male Wistar rats in animal models for anxiety and depression. Water Air Soil Pollut. 141, p. 1-12.

26. Özacar M., Şengil İ.A., 2003. Adsorption of reactive dyes on calcined alunite from aqueous solutions. J Hazard Mater. B98, 211–224.

27. Langmuir I., 1916. The constitution and fundamental properties of solides and liquids. Part I. Solids J Am Chem Soc. 38(11), 2221-2295.

28. Langmuir I., 1918. The adsoption of gases on plane surfaces of glass, Mica and platinum. J Am Chem Soc. 40(9), 1361–1403.

29. Freundlich H.W., 1939. The Adsorption of cis- and trans-Azobenzene. J Am Chem Soc. 61, 2228-2230.

30.Ho Y.S., McKay G., 1998. Orption of dye from aqueous solution by peat. S Chem Eng J. 70, 115.