RESEARCH ARTICLE

Antibiotics Removal by Adsorption onto Eco-friendly Surface: Characterization and Kinetic Study

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ABSTRACT

This study includs the preparation of the hydrogel using the free radical polymerization method with the possibility of using it as an adsorption surface for a drug streptomycin from its aqueous solution. The structural and thermal properties of the hydrogel surface by using several techniques [Transmission electron microscopy (TEM), field emission scanning electron microscope (FESEM), fourier transform infrared spectroscopy (FTIR)] were study. Where the best weight of the hydrogel surface was (0.05 g) when the weight of surface increased, the removal percentage increased and the adsorption efficiency decreased, and the equilibrium time was one hour. The adsorption kinetics were studied, and it was found that the adsorption behavior follows the kinetics. The second order is based on the value of (R^2= 1) compared with the first order.

Keywords: Adsorption, Antibiotics, Pharmaceutical, Streptomycin drug, Removal, kinetic.

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INTRODUCTION

Antibiotics have received widespread attention, but over time there has been increased concern about the accumulation of antibiotic residues in the aquatic environment. Which can lead to the stimulation of genes and resistance to antibiotics upon prolonged exposure and at relatively low concentrations, which affect the organisms present in the aquatic environment.[1] Pollution mainly comes from liquid waste from sewage treatment plants and water from pharmaceutical factories and hospitals. Therefore, there are several ways to remove these pollutants from water, the most important of adsorption.[2-4] The adsorption process is the collection of molecules or ions in their gaseous or liquid state on the surface of a solid. The material that undergoes adsorption on the surface is called (Absorbent) and the surface on which adsorption occurs is called (Absorbate) and the adsorption may lead to the formation of only one molecular layer on the surface and it is called single-molecular adsorption or there are several molecular layers on the surface and it is called multi-layer adsorption.[5-7] Adsorption is applied as a separation process on a large scale in the industrial economy and in daily life, and it is similar in its work to the ion exchange technology, where it depends on the presence of a group called (Sorption processes) to separate the mixture of liquids.[8-12] Adsorption can be classified according to the type of forces that bind ions, atoms or molecules adsorbed on the surface of the adsorbent material, and it is classified into physical adsorption and chemical adsorption.[13,14] The adsorption process depends on the use of low-cost and available surfaces that can be prepared easily, such as activated carbon, carbon nano-tube, clay, graphite oxide, hydrogel, sawdust and others. In this study, it relied on the preparation of the hydrogel surface, which is highly efficient and can be prepared from available materials and can remove drugs from water.

EXPERIMENTAL PART

Preparation of Hydrogel

Polyvinyl alcohol is dissolved (2 g) in 30 mL distilled water with continuous stirring for two hours. Acrylic acid solution is prepared by dissolving polyvinyl alcohol (3 mg) in 10 mL of distilled water, then adding sodium per sulfate resulting from dissolving (0.04) in 1-mL of distilled water slowly added in the form of drops. Then a solution thermo scientific pierce tetramethylethylenediamine (TEMED) is added as a reaction accelerator, and finally the cross-linking agent Methylenebisacrylamide (MBA) is added to the reaction mixture. The reaction mixture is placed at a temperature (75°C) for a period of time, the polymer is washed several times, then dried and ground to obtain the powder used in this experiment.

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Determination of λ_{max} and Calibration Curve for Streptomycin Drug

A standard solution of a drug is prepared by dissolving 1.0 g of drug in distilled water in volume 1000 mL to obtain a concentration 1000 mg/L. The maximum wavelength of a drug is determined where the spectrum of a drug is recorded using UV-vis be spectroscopy within the range of (200–800 nm) using a cell of quartz thickness (1 cm) and found that the value of the wavelength ((λ_{max} 275 nm) as appear in Figure 1 (a). To draw a calibration curve, a series of solutions is prepared, absorbance is measured, and a calibration curve is determined by drawing between absorbance and concentration as appear in Figure 1(b).

Adsorption Study

The adsorption experiments were conducted at room temperature, and the equilibrium time of (1 hour) with pH of drug solutions (6.2). Utilizing a concentration (100–2000 mg/L) of the drug and an amount of hydrogel weight of (0.05 g) was added, the samples were quickly placed in a water bath shaker at 390 rpm during (10 minutes). The solution was separated using Centrifuge and the concentration of drug was measured before and after the adsorption process using UV-vis spectrometer wavelength 275 nm. The adsorption kinetics were also studied at the same optimum conditions for the experiment. Take the drug concentration 1000 mg/L by (10 mL) of drug solution at room temperature and the weight 0.05 g of the hydrogel using different times from (5–120 minutes). The adsorption efficiency was calculated using the equation.

$$Qe = \frac{Co - Ce}{Co} \frac{V}{W} \quad (1)$$

RESULTS AND DISCUSSION

FTIR

To determine the changes in the ratio of the effective groups between atoms, the technique of FTIR is studied. Potassium bromide (KBr) is used after adding the surface to it and a spectrometry before and after the adsorption process. As shown in the Figure 2, the absorption reaches its peak the broad band shown at 3350 cm^{-1} belongs to the stretching (-COOH) vibrations, and the other vibrations described in 3020–2850 are due to the aliphatic and aromatic (C-H) asymmetric stretching vibrations. It was also found that no new peak appeared, only a difference in the intensity of absorption, evidence of the occurrence of the physical type due to weak bonds.[14-16]

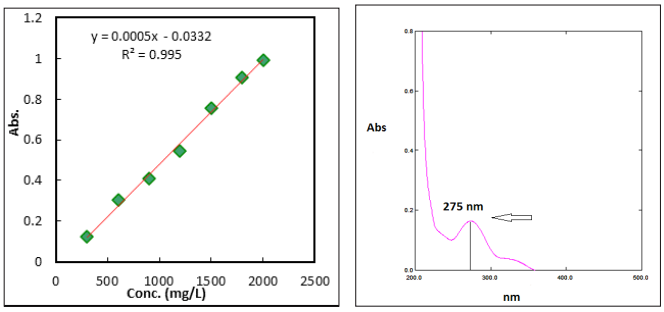

Figure 1: a) UV-visible absorption spectrum to the drug, b) Calibration curves of drug

FESEM

Using FESEM technique to know the nature and size of the particles on the surface and their porosity before and after the adsorption process. The prepared surface is rough containing many cavities and a sponge-like structure due to its high functional groups, which include carbonyl, carboxylate and hydroxyl.[17,18] After the adsorption process, we notice the fullness of the active sites of the surface, as well as the accumulation of materials inside the pores of the pores, as appear in Figure 3.

Thermo Gravimetric Analysis (TGA)

TGA illustrated the thermal behavior of the hydrogel it has Relative stability, and the curve appeared to lose 3% at range (39.7–100)°C at Which is attributed to the evaporation of water molecules and missing about 14% from Weight in range (230–290)°C decomposition of (-$CONH_2$ and -COOH) groups this regarded second-step decomposition as appear in Figure 4.[19-21]

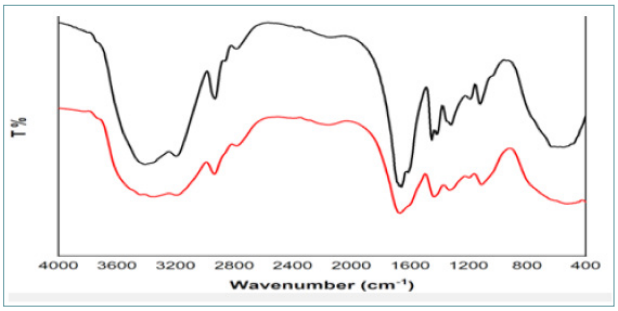

Figure 2: FTIR spectra to determination drug before and after adsorption

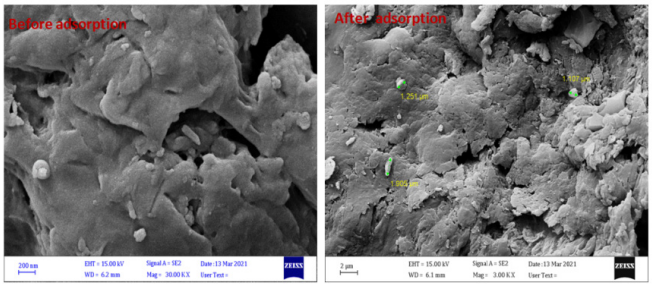

Figure 3: FESEM image of hydrogel a) before Adsorption, b) after adsorption

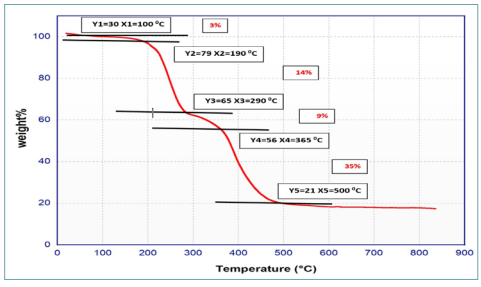

Figure 4: Thermo gravimetric analysis of hydrogel

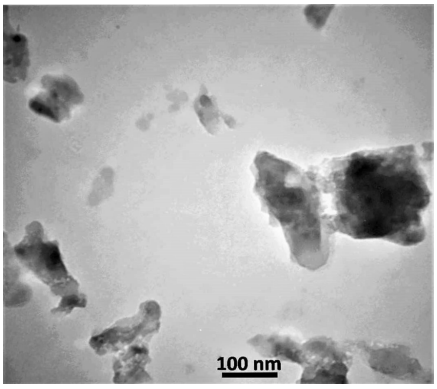

Figure 5: TEM image of hydrogel

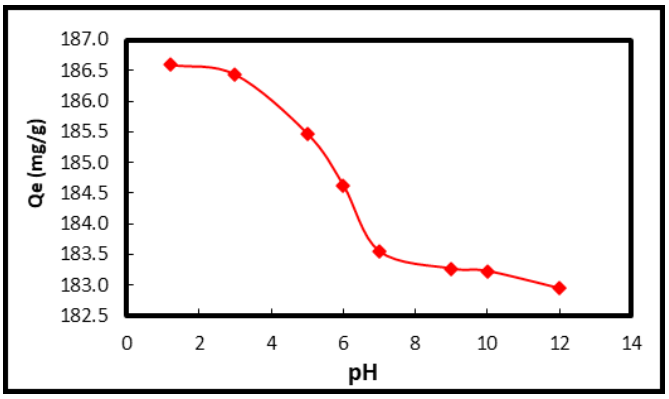

Figure 6: Effect of solution pH of the adsorption drug onto hydrogel

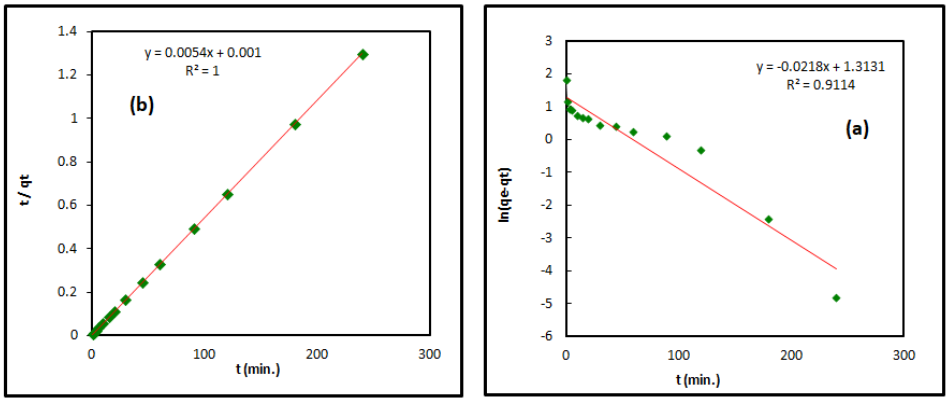

Figure 7: Effect of kinetic adsorption of first order (a) Second order (b) Equilibrium time

Table 1: Parameters of the kinetic adsorption of drug

First order	*Second order*
k_1 q_e R^2	k_2 q_e R^2
0.0045 1.0882	0.0114 0.1514 48.754 1

TEM

TEM technology mostly contributes to estimation the morphology of the prepared hydrogel surface where the macroscopic features were observed or affected by the interface or defects. TEM images are shown in Figure 5 of hydrogel. It consists of collection and dark spots, i.e. from a dark background in color attributed to the hydrogel.[22,23]

Effect of pH Solution

The effect of solution pH on the adsorption of a drug from aqueous solutions was studied. (10 mL) of drug solution was added at a concentration (1000 mg/L) taking different pH (1–12) and used water bath shaker at room temperature for an hour and at a speed (120 rpm) until reaching the equilibrium, and the absorbance was measured after Separation of the solution by centrifuge at speed (6000 rpm) about (10 minutes) as show in Figure 6.[24] That found the best removal of drug at acid medium at pH 1 and the adsorption capacity increase from (186.5 to 183,5 mg /g).[25]

Adsorption Kinetic

The adsorption kinetics of a drug on the hydrogel surface were studied, and the kinetic models of the adsorption process were determined, which describe the experimental data of the adsorption process in an accurate way for drug adsorption on the hydrogel.[26] What is the model first-order and model second-order.[27,28] as shown in the Figure 7. The kinetic constants and the value of the correlation coefficient (R^2) were calculated as shown in the Table 1.

Through the results, it was found that the correlation coefficient (R2) of the second-order model is high compared to the first-order model, so the adsorption process follows the second-order model.[29,30]

CONCLUSION

- The hydrogel surface is characterized by a high surface area and a high adsorption efficiency to removal drug from aqueous solutions.
- The drug equilibrium time on the hydrogel surface is one hour, and the adsorption efficiency decreases with increasing weight of hydrogel, and the removal percentage increases with increasing surface weight.
- The study proved that the adsorption kinetics of a drug on the hydrogel surface apply to the second-order model.

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