REVIEW ARTICLE

Adsorption of Amoxicillin onto Graphene Oxide Poly (carboxymethyl cellulose -co-acrylic acid) Hydrogel: Isotherm and thermodynamic studies

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ABSTRACT

In the present study, graphene oxide/Poly (carboxymethyl cellulose -co-acrylic acid), GO / P (CMC-co-AA) Hydrogel is synthesized by a free radical polymerization and is applied for the adsorption of Amoxicillin (AMX) from aqueous media. To investigate the structure of GO /Poly(CMC-co-AA), Hydrogel, FTIR, FESEM analysis are applied. The effects of pH and contact time on AMX adsorption are studied. The adsorption isotherms of amoxicillin on the GO/P(CMC-co-AA) composite could be illustrated well by the Freundlich and Langmuir model. According to the results of equilibrium data, it was shown that a maximum adsorption capacity of 12.486 mg/g with Langmuir isotherm was the predominant model, and adsorption was a monolayer. Thermodynamic studies also reveal that the adsorption process is exothermic.

Keywords: adsorption, Beta-Lactam, Amoxicillin, Graphene oxide, Hydrogels, Water pollution.

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INTRODUCTION

Water pollution has worsened in numerous areas worldwide, particularly in areas with threats of high pollution, such as Europe, Asia, Africa, and America.[1] One of the vital contaminants was categorized as emerging micropollutants (MP). The MPs were primarily synthetic organic chemicals that have been recently found in environments.[2] Examples of emerging micropollutants were drugs, which can enter wastewater networks after use in industries and households.[3] Drugs were commonly found in different environmental compartments. The growing use of drugs has raised questions regarding their potential risk to the environment, human health, and water quality.[2] Antibiotic Amoxicillin (AMX) (Figure 1) was a ß-lactam antibiotic and the only phenolic penicillin. ß-lactam antibiotics (AMX) present a structure based on a ß-lactam ring which was responsible for variable side chains and antibacterial activity.[5] It was usually chosen because it was better absorbed than other ß-lactam antibiotics.[6] Antibiotics Amoxicillin has low concentrations; (ng/L to µg/L), which makes them hardly degradable, remains as an active compound within feces and urine.[7] Their presence in the environment can be linked to some potential problems in the future. In cases of such long lifetimes, the damage to the health risk for humans and the environment can undoubtedly be severe. Therefore, this study investigates the removal of amoxicillin (AMX) drug from aqueous water.[8] There were many methods for AMX removal from wastewater, such as ultrasonication, ozonation, biodegradation, and adsorption.[9-11] Adsorption process has been proven to be an effective process for removal of different antibiotic contaminants from aqueous solutions because of its flexibility, simple design, easy operation, suitability for continuous and batch processes, low capital cost, the possibility of reuse and regeneration, and capability to remove a wide range of contaminating concentrations.[12] Different adsorbents have been utilized for the removal of antibiotics (AMX), where low adsorption capacity has been observed on bentonite and activated carbon (AC).[13,14], etc. On the other hand, the adsorption of the widely used amoxicillin (AMX) onto GO/P(CMC-co-AA) is studied and evaluated the equilibrium isotherms and kinetics and concluded that GO/P(CMC-co-AA) showed a better performance.

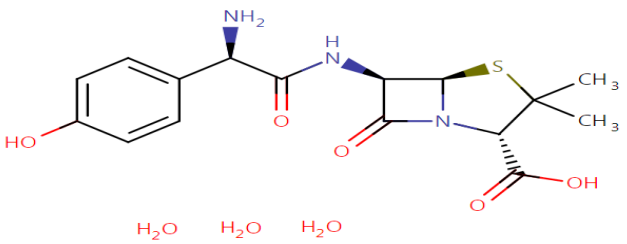

Figure 1: Structure of amoxicillin

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Experimental

Chemicals

All chemicals were used in this study containing acrylic acid (AA) and Graphite powder are purchased from Sigma-Aldrich. Carboxymethyl cellulose sodium salt (CMC), Potassium permanganate ($KMnO_4$), and a sulfuric acid (H_2SO_4) (95–98%) solution from Scharlau, N,N-methylene-bisacrylamide (MBA) and potassium persulfate (KPS) from Kemiou Chemical Reagent Co, Ltd, China. Amoxicillin and Sodium hydroxide was purchased from Merck. Graphite powder was obtained from Fluka. All the materials used were analytical grade pure and used without further purification.

Preparation of Graphene Oxide (GO) and GO/(CMC-co-AA) Hydrogel

The GO nanosheets are synthesized using a modified Hummer's method.[15]. GO/P(CMC-co-AA) hydrogel was synthesized via free radical polymerization of GO and acrylic acid monomers in deionized water. Briefly, GO (0.5 g) was dissolved with deionized water (30 mL) in a four-neck flask equipped with a mechanical stirrer, condenser, nitrogen inlet, and thermometer. Then, CMC (3g) and acrylic acid (12 g) solution. In the next step, the cross-linker (MBA, 0.15 g) and initiator (KPS, 0.1 g) are charged to the reaction flask upon stirring till a homogenous mixture is prepared. The flask is heated up to 60°C in a water bath, and the reaction is constantly stirred until the polymerization process completion. The resulting composite polymer is immersed in deionized water which is replaced with fresh deionized water repeatedly for 24 hours. Subsequently, the purified hydrogel is dried in a vacuum oven at 60°C and the prepared hydrogel are named as GO/P(CMC-co-AA). and stored at ambient temperature for further usage.[16]

Characterization

The presence of amoxicillin on the surface of GO/P(CMC-co-AA) was proved by Fourier transform infrared spectrometry (FT-IR, 8400 S, Shimadzu, Japan) in the wavenumbers range of (400–4000 cm^{-1}). Surface morphologies of GO/P(CMC-co-AA) were analyzed by Field emission scanning electron microscopic (FE-SEM, JEOL, JSM-6701F, Japan). Amoxicillin concentrations were measured by quantitative analysis at (UV–Vis). spectrophotometer (1600, PC Instruments). amoxicillin solutions are analyzed at 272 nm, and these values were the wavelengths which the solutions give the maximum absorbance.

Batch Adsorption Experiments

All adsorption experiments are carried out in a series of 100 ml flasks containing 50 mg GO/P (CMC-co-AA) and 10 mL AMX solutions. To obtain adsorption isotherms, 50 mg GO/P(CMC-co-AA) are contacted with 10 ml of different concentrations of AMX solutions (10–100 mg/L). The mixtures are then shaken at 150 r/min for 2 hours at 298 K. When the process is reaching adsorption equilibrium; the appropriate solution is gathered to filter using a syringe with 0.4 mL filter and then determine by (UV–Vis) spectrophotometer (1600 UV-VIS) at 272 nm. Simultaneously conducting blank experiments is AMX solution without adsorbent to eliminate the effects caused by agents. Furthermore, additional experiments are also carried out to detect the influence of pH on the adsorption properties. The effect of pH on AMX sorption by GO/P(CMC-co-AA) is determined by adjusting the pH values of the solutions to the designated values. Used in the experiment is 0.01 M NaOH or 0.01 M HCl with the initial concentration of 80 mg/L of AMX. All adsorption experiments are performed to ensure a single variable. The equilibrium adsorption amount of AMX is calculated as follows:[17]

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

where q_e (mg/g) was the adsorption capacity of AMX at equilibrium time, C_e, and C_0 (mg/L) were AMX solution concentrations at equilibrium time and initial time, M and V were volume of mass of adsorbent GO/P(CMC-co-AA) (g) and AMX solution (L) and, respectively.

RESULTS AND DISCUSSION

The FTIR spectra of GO, P(CMC-AA), and GO/P(CMC-co-AA) are examined to verify that amoxicillin is bonded on the GO/P(CMC-co-AA) surface, as shown in Fig. 2 a. In the FT-IR spectra of graphene oxide (GO), the absorption peaks appearing at 3444cm^{-1}, 1224.84 cm^{-1}, 1724.42 cm^{-1}, and 1049.31 cm^{-1} were attributed to OH stretching, epoxy C-O, C=O in the carboxyl group, alkoxy C-O, vibrations, respectively [18]. In the FT-IR spectra of (CMC-AA), Figure 2b show the FT-IR spectra determined for Poly (CMC-co-AA) hydrogel in the spectral range of 4000–400 cm^{-1}, the band at 1732.13 cm^{-1}, attributed to C=O stretching from esters, is present in (AA) powder because the (AA) chains were crosslinked through the esterification among the AA carboxylic acid groups. In comparison to the band at 1516.10 cm-^{1}, which was attributed to the asymmetric axial deformation of carboxylate groups from CMC, the intensity of the band at 1732.13 cm^{-1} is the strongest. The bands at 1338.64 cm^{-1} and 1450.52 cm^{-1} are attributed to the stretching of carboxylate groups in CMC and C=O of esters. The signals in the spectral region between 1209.41 and 875.71 cm^{-1} are attributed to (C–O) and (C–C) stretching vibrations of the CMC glucopyranose ring.[19,20] The FTIR spectra of the GO/P(CMC-co-AA) composite is studied the (O-H) stretching vibration carboxylate group of CMC, AA, and GO overlapping of bands at 3417.63 cm^{-1} and observed the (C-N)bending at 1319.22 cm^{-1}, but the FT-IR for GO/P(CMC-co-AA) is shown the shift of the group's bands (C=O) reveals the interactions between the carboxylic groups of (CMC)with O-H groups on (AA) and GO platelets is given at 1650.95cm^{-1}.[21]

To examine the effect of graphene oxide (GO) sheets on the surface morphology of the resultant GO/P(CMC-co-AA), FE-SEM observation of GO/P(CMC-co-AA) (0.5 wt% GO) is conducted in Figure 3. It could be seen that the fracture surface morphology of the GO/P(CMC-co-AA) (Figures 3a, b) was obviously different from that of the polymer composite after

adsorption by drug (Figures 3c, d). the surface of GO/P(CMC-co-AA) sheet looks coarse carpet which can due to the link of residual (water molecules), hydroxyl and carboxyl groups with the sheets. However, the polymer composite containing the GO sheets provides a rough and coarse surface (Figures 3b and c). Furthermore, the pores in polymer composite become more uniform and smaller.[22]

Adsorption Study

Effect of Contact Time

To study the equilibrium state, AMX adsorption on GO/P(CMC-co-AA) is investigated during 140 minutes contact times. As Figure 4 appears, for AMX concentration and in the first 15–20 minutes of adsorption experimental, the adsorption process happens rapidly. The adsorption process is completed from 40 to 60 minutes of contact time, and an equilibrium state is achieved. So, the excess contact time up to 140 minutes had no effect on AMX adsorption, and 120 minutes contact time is considered an equilibrium time and used for all the rest experiments.[23]

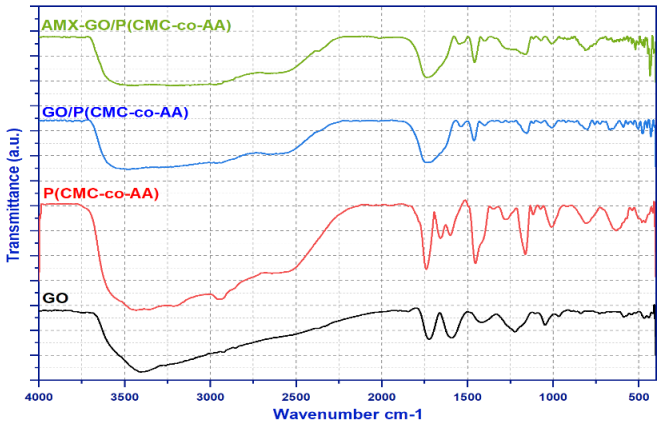


Figure 2: (A to D): FTIR spectra of (a) GO (b) P(CMC-AA), (c) GO/P(CMC-co-AA), (d) AMX-(GO/P(CMC-co-AA).

Effects of Solution pH

The solution (pH) that can affect the physicochemical properties of the adsorbate (AMX) was a major factor controlling the AMX molecule adsorption process. Furthermore, changes in pH can alter the surface electric charge of adsorbate (AMX) and adsorbent GO/P(CMC-co-AA)] which affects the electrostatic interactions between the adsorbate (AMX) and adsorbent [GO/P (CMC-co-AA)]. Thereby changing the adsorption process behavior. The pH is adjusted in the range of 1.2–10, and the effects of pH on the adsorption amount capacity of AMX onto GO/P(CMC-co-AA) are explored. The results are showed in Figure 5. It is proved the fact that the AMX adsorption strongly depended on the pH of the solution.[24]

As shown in Fig. 5, the maximum adsorption capacity of 11.9429 mg/g for AMX is obtained at pH = 1.2. AMX has three pKa values of 2.4 (carboxyl), 7.4 (amine), 9.6(phenol), respectively. When the pH value is 2.4, AMX in the cationic form of being generated electrostatic attraction with negatively charged GO/P (CMC-co-AA) on the surface simplifies the

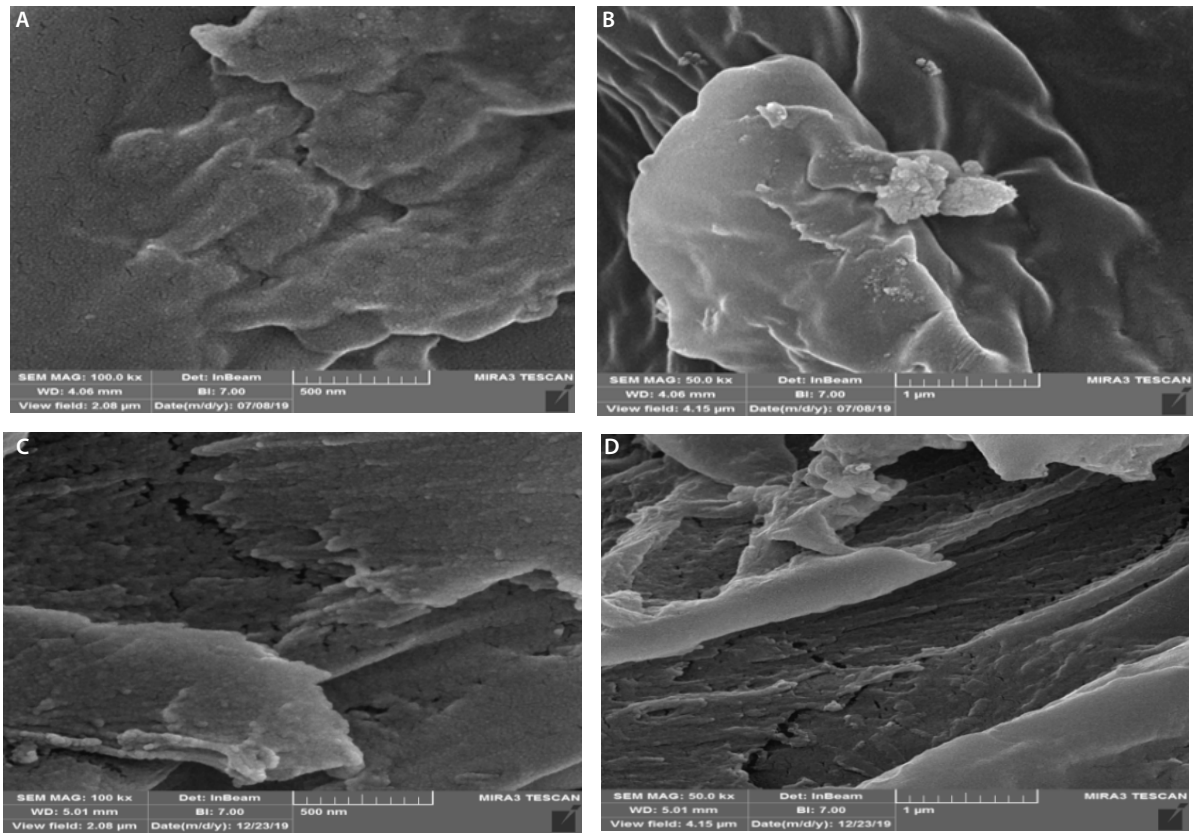


Figure 3: FE-SEM images of (a,b) (GO/P(CMC-co-AA) and (c,d) AMX-(GO/P(CMC-co-AA

adsorption process. While at high pH, the competitive inhibition of excessive hydroxide ions and another coexistence anion will reduce the adsorption effect. Therefore, the adsorption capacity of AMX at pH = 1.2 is higher than that at pH = 7.4 and 9.6. AMX is an anionic and zwitterionic form. In both cases, part of the anionic in the former can arise electrostatic repulsion with negatively charged GO/P(CMC-co-AA) so that the adsorption process capacity continued to decrease.[25,26]

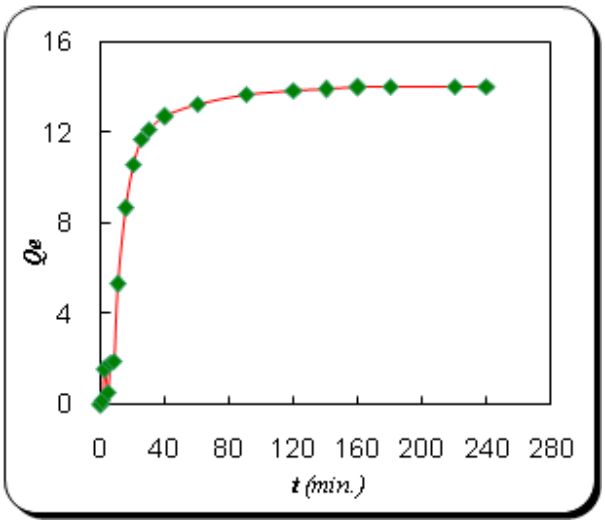


Figure 4: The effect of contact time on the AMX adsorption (AMX initial concentration of 80 ppm, adsorbent dosage of 0.05, and T = 25°C)

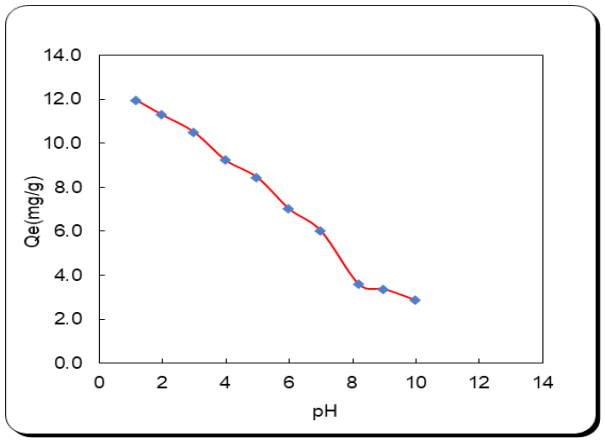


Figure 5: Effect of pH on AMX adsorption by GO/P(CMC-co-AA)

Adsorption Isotherms

Isotherm models exhibit the form of adsorbate (AMX) adsorption at a fixed temperature. The adsorption experimental was performed according to the arrangement of adsorbate (AMX) molecules on the surface of adsorbent GO/P(CMC-co-AA) based on multi-layer and single-layer adsorption. In the present work, to investigate the equilibrium condition of adsorbate (AMX) in solid-phase GO/P(CMC-co-AA) and liquid phase (AMX solution), Two types of isotherm models containing Freundlich and Langmuir are evaluated.

Freundlich isotherm

The Freundlich isotherm was based on heterogeneous and multi-layer adsorption. The linear equation was shown below.[27]

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

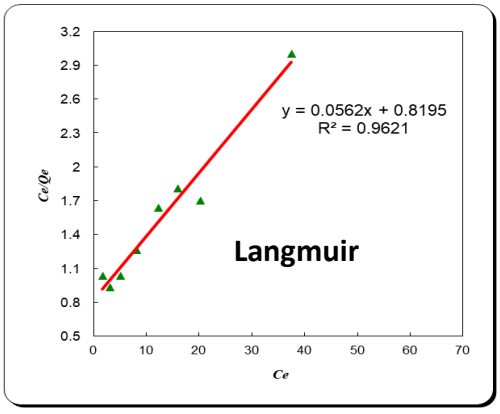
where n and k_f can be evaluated from the linear plot of ($\ln q_e$ versus $\ln C_e$). N was a deviation from the linearity of adsorption and 1/n was a heterogeneous factor. k_f (L/g) as Freundlich constant was related to the bond energy.

Langmuir Isotherm

Langmuir's theory is based on two assumptions that adsorption energy was constant during the process, and the adsorption of adsorbates occurs on a homogeneous surface by monolayer adsorption. The linear equation was appeared below.[28]

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

Qe (mg/g) was equilibrium adsorption capacity, Ce (mg/L) was the equilibrium concentration after adsorption experimental , qm and kL were Langmuir constants constant of heat adsorption maximum adsorption capacity of the solid phase, respectively. The linear form of the Freundlich and Langmuir isotherm models is applied, and the results are shown in Figure 6 and Table 1. As can be seen, the first linear type of Langmuir model



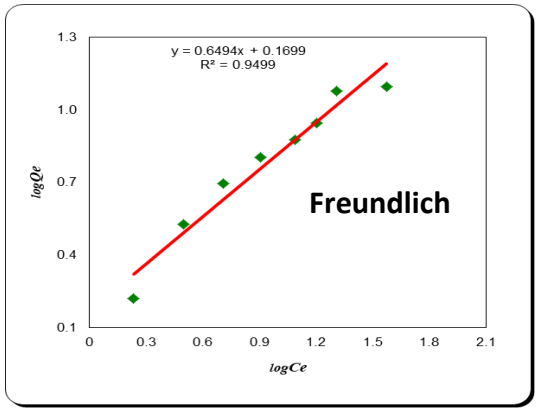


Figure 6: Adsorption isotherms of the Amoxicillin adsorption at 25°C.

Table 1: Different isotherm parameters for adsorption of AMX on GO/P(CMC-co-AA).

Isotherm models' parameters	*Freundlich isotherm parameters*			*Langmuir isotherm parameters*		
	n	*Kf*	*R2*	*qm*	*KL*	*R2*
Amoxicillin	1.53988	1.47877	0.9499	17.79356	0.06858	0.9621

Table 2: Thermodynamic parameters obtained for adsorption process of AMX using GO/P(CMC-co-AA) material

Concentration of drug (mg/L)	*Keq*	*ΔS° (J/mol)*	*ΔH° (kJ/mol)*	*-ΔG° (kJ/mol)*			
				10 0 C	*15 0C*	*20 0C*	*25 °C*
Amoxicillin	2.8519	-28.3147	-14226.1	6.5951	6.0714	5.9299	5.8283.

was more compatible with the experimental adsorption results, which approves that monolayer adsorption of AMX happened on GO/P(CMC-co-AA) and acceptor groups of adsorbate (AMX) are distributed homogeneously and uniformly on the surface of adsorbent GO/P(CMC-co-AA).[29] From the results of Table 1 according to the correlation coefficient (R^2)values of all the isotherm models.

Thermodynamic Studies

The work is the investigation of thermodynamic aspects of the adsorption experiment. For this purpose, calculations of the free energy (ΔG), entropy (ΔS), and enthalpy (ΔH) are performed using Eq. (4)-(6).[30]

$$\Delta G^\circ = -RT \ln \ln K_{eq} \quad (4)$$

$$\Delta G^\circ = \Delta H^\circ - T\Delta S^\circ \quad (5)$$

$$\ln K_{eq} = -\frac{\Delta H^\circ}{RT} + \frac{\Delta S^\circ}{R} \quad (6)$$

where ΔG (kJ/mol), ΔH (kJ/mol), ΔS (J/mol K) are the changes in the free energy of the system, the free enthalpy, and the free entropy, respectively, T (K) is the temperature. R (8.314 J/mol K) is the gas constant, and K_{eq} is the constant reaction equilibrium; experimental are carried out for one concentration of AMX solution (80 mg/L) at 10, 15, 20, 25°C. Changes in entropy and enthalpy are determined based on the plot of $\ln K_{(Xm)}$ versus 1000/T (see Figure 7). The results are presented in Table 2. The results have appeared that the adsorption of AMX is exothermic (negative values of ΔH for study AMX drug). The adsorption experimental is also characterized by negative values of ΔS, indicating complex interactions between the adsorbate (AMX) and the surface of the sorbent GO/P(CMC-co-AA). In addition, the negative value of ΔG has suggested a reduction of random effects at the solid-liquid interface, reduction of the degree of freedom of drug molecules during adsorption experimental, and the fact that the adsorption of AMX on the GO/P(CMC-co-AA) material is a spontaneous process.[31] There was a correlation between the mechanism of adsorption process and the values of the (ΔH). A value of (ΔH) in the range from 80 to 400 kJ/mol was taken to indicate chemisorption, while a value from (ΔH) less than −20 kJ/mol indicates physisorption. Thus, the results determined suggest that one of the mechanisms of the adsorption process of AMX on the GO/P(CMC-co-AA)

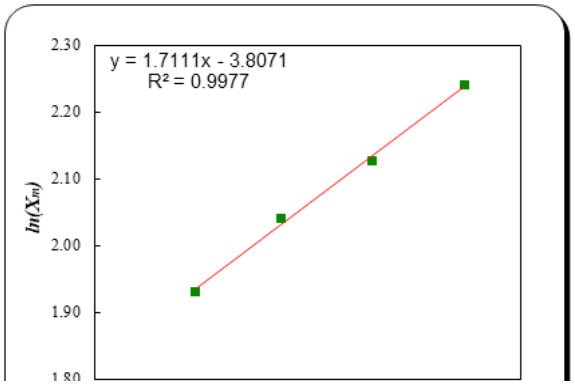


Figure 7: Plot of ln Xm against reciprocal absolute temperature for adsorption of AMX onto GO/P(CMC-co-AA) polymer composite

surface is physical interactions between molecules of AMX and the surface of the sorbent GO/P(CMC-co-AA). These results suggest that GO/P(CMC-co-AA) has a better affinity to amoxicillin.[32]

CONCLUSION

In this study, a successful hybrid hydrogel is prepared for the removal of the Amoxicillin drug from aqueous water. The maximum adsorption capacities for the Amoxicillin drug are estimated at 298 K as 12.486 mg drug/g adsorbent. Thermodynamic studies appeared that the adsorption of the drug is more favorable at lower temperatures; the negative value of enthalpy for the adsorption of the drug indicates an exothermic process, suggesting that the adsorption mechanism would be of physical nature. This adsorption experimental is found to be better fitted by the Langmuir model. The adsorption of the drug onto GO/P(CMC-co-AA) hybrid hydrogel is a pH-dependent process with maximum removal efficiency at the initial pH =1.2.

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