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Langmuir, Freundlich, Temkin and Dubinin–radushkevich Isotherms Studies of Equilibrium Sorption of Ampicilin unto Montmorillonite Nanoparticles

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Authors' contributions

This work was carried out in collaboration between all authors. All authors read and approved the final manuscript.

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ABSTRACT

Present study was accomplished to prospect the viability of using the montmorillonite (Mon) nanoparticles as an adsorbent to remove the Ampicillin under various experimental conditions. The Physico-chemical characteristics of the studied adsorbent were surveyed. Langmuir, Freundlich, Temkin and Dubinin–Radushkevich isotherms were applied to portray the data obtained from the adsorption studies. The findings showed that the highest R^2 values were related to Langmuir and Dubinin–Radushkevich isotherm models. The greatest adsorption capacity (qe) for Langmuir and Dubinin–Radushkevich isotherm models were recognized to be 134.48 mg/g and 141.22 mg/g, respectively; and the separation factor was calculated to be 0.113 which is indicative of a favorable

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sorption. Temkin Isotherm model clarified that the heat of sorption process was 34.61 J/mol; and the mean free energy calculated by Dubinin–Radushkevich isotherm model was anticipated to be 2.56 Kj/mol which these undoubtedly demonstrate the physisorption process for Ampicillin adsorption experiments.

Keywords: Montmorillonite nanoparticles; ampicilin; isotherm.

1. INTRODUCTION

Antibiotics are the most extensively used drugs to prevent or treat bacterial infections in humans, animals and plants [1,2]. Antibiotics discharged from wastewater treatment plants (WWTPs) and as through effluents from pharmaceutical manufacturing plants to the environment have received a growing concern because of their potential toxic effect on the aquatic biota as well as human [3,4]. Ampicillin is a common antibiotic used in human and animal medicine, and is an amphoteric, beta-lactam of the penicillin class [5]. The Ampicilin is one of the most commonly used antibiotics at Iran and worldwide, which finds wide applications in human therapy and the livestock industry, and is difficult to be metabolized [6-8].

The presence of antibiotics in the environment has, therefore, received considerable attention [9]. It has been found that antibiotics are, in general, poorly absorbed by the human body and, thus, are excreted either unchanged or as their metabolites via urine and feces [10,11]. Furthermore, most antibiotics are highly toxic, soluble in water and of low biodegradability [12].

Removing antibiotics is difficult and requires expensive process. In recent years, there has been an increasing interest to the treatment of pollution generated by drug residues, including antibiotics [13,14]. Physical and Physico-chemical and biological techniques have proved their efficiency for this purpose [15,16]. The Physical and Physico-chemical techniques used for antibiotics removal from wastewater include ozonation, flocculation, electro-flocculation, reverse osmosis, ultrafiltration, coagulation and photodegradation process [17,18]. These most conventional methods are non-destructive and merely transfer pollutants from one phase to the other, which always result in secondary pollution [19,20]. Also Biological processes, the most cost-effective for wastewater treatment, which are destructive and have been extensively studied do not always, appear relevant for the removal of recalcitrant compounds, owing to their low biodegradability [21]. Of these methods, sorption has proved to be an attractive process because of its low cost and ease of operation [22,23]. Activated carbon has been widely used for this purpose because of its high adsorption capacity [24]. However, its high cost sometimes tends to limit its use. Clays are widely applied in many fields such as polymer nano-composites, adsorbents for pollutant ions, catalysts, photochemical reaction fields, sensors and biosensors, due to their high specific surface area, chemical and mechanical stabilities, and a variety of surface and structural properties [25-27].

Clay is a potentially good adsorptive material because of its large surface area, chemical and mechanical stability, and layered structure [27]. The adsorption of pollutant into natural clays has recently been studied by various researchers. Among natural clays, montmorillonite acts as a adsorbent for pollutant due to its low-cost, high abundance, easy manipulation, and harmlessness to the environment [28]. The most-used clays as nano-adsorbents are montmorillonite group and kaolinite group clays [29]. In this paper, the possibility of using montmorillonite nanoparticles to remove Ampicilin was investigated using batch adsorption studies. The equilibrium data were analyzed so that we can understand the adsorption mechanism and different models (Langmuir, Freundlich, and Temkin and Dubinin–Radushkevich) were applied to fit the experimental data.

2. MATERIALS AND METHODS

Ampicillin (CAS Number 69-53-4; chemical formula, $C_{16}H_{19}N_3O_4S$; MW, 349.40 g/mol) was used as the adsorbate in this study obtained from Sigma Aldrich Co; it was used without further purification. The chemical structure of Ampicillin, shown in Fig. 1.

Ampicillin stock solution was prepared by dissolving an accurately weighed amount of Ampicillin in distilled water to achieve a concentration of 200 mg/L, and subsequently diluted to the required concentrations. All chemicals used in this study were of analytical-laboratory grade, being purchased from Merck.

A montmorillonite nanoparticle was obtained from the Iranian nanosany Corporation and was used without further purification. Chemical analysis of the clay was carried out using an X-ray fluorescence spectrometer (XRF-Philips PW2400). BET surface area and total pore volume of the clay were measured using Autosorb-1 (Quantachrome Corporation, New York), with N_2 gas as the adsorbate.

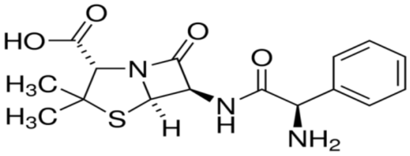


Fig. 1. The chemical structure of Ampicillin

2.1 Adsorption Experiments

Adsorption experiments were carried out by batch method at room temperature. The time-dependent behavior of Ampicillin adsorption was studied by varying the contact time between the adsorbate and adsorbent in the range 10-120 min. The initial concentration of Ampicillin was kept at 25 mg/L, while the dose of Mon-nanoparticles was 0.5 g/L. At the end of each adsorption experiment, the solution and solid phase were separated by centrifugation at 4000 rpm for 15 min. Ampicillin concentration at samples was determined by a High Performance Liquid Chromatography (HPLC) (Agilent 1100 Series) at wavelength 204 nm. The mobile phase was 60% 0.025 M KH_2PO_4 buffer solution in ultra pure water and 40% acetonitrile at a flow rate of 0.50 mL/min.

The adsorption capacity of Ampicillin on adsorbent was calculated using the following equation [30,31]:

$$q_e = \frac{(Co - Ce) \times V}{M}$$

Where C_0 and C_e are the Ampicillin concentrations in mg/L initially and at a given time t, respectively, V is the volume of Ampicillin solution in L, and M is the weight of sorbent in g.

3. RESULTS AND DISCUSSION

The Tables 1 and 2 represented the chemical composition and physicochemical properties of the Mon-nanoparticles, respectively. As it can be seen in Table 1, Montmorillonite has considerable levels of SiO_2 (57.7%) and Al_2O_3 (18.1%) and the quantity of other metal oxides is observed to be lesser than 20%. The investigation of the surface physical morphology of Montmorillonite was fulfilled using the scanning electron microscopy (SEM) technique. Fig. 2 shows the SEM images of the Montmorillonite. Furthermore, it was found that the surface area of Montmorillonite is 245.5 m^2/g.

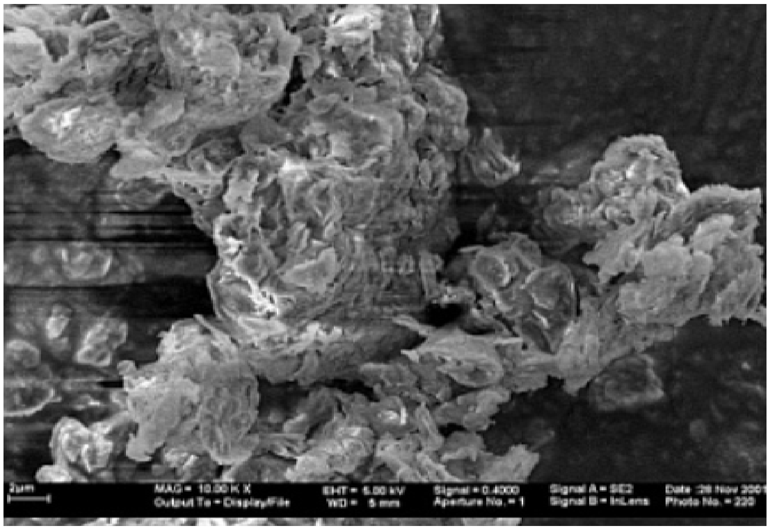


Fig. 2. SEM micrographs of the Montmorillonite samples

Table 1. Chemical compositions of Mon-nanoparticles

Component	%
SiO_2	57.7
Al_2O_3	18.1
Fe_2O_3	7.25
MgO	3.11
CaO	1.64
Na_2O	1.22
TiO_2	0.68
MnO	0.29
LOI	9.25

Table 2. Physicochemical properties of Mon-nanoparticles

Parameter	Value
BET surface area	245.5 m^2/g
Total pore volume	2.14×10^{-3} cm^3/g
Average pore diameter	51 (Å)
Particle size	1-2 nm
Moisture	1-2%

3.1 Adsorption Isotherms

The adsorption data were analyzed to see whether the isotherm obeyed the Langmuir, Freundlich, Dubinin-Radushkevich (D-R) and Temkin isotherm models equations [32-36]:

Langmuir equation= $\frac{1}{q_e} = \frac{1}{q_{max}} + \frac{1}{q_{max}\,K_L} \times \frac{1}{C_e}$

Freundlich equation= Log $q_e = \frac{1}{n}\log Ce + \log K_F$

D-R equation: $\text{Ln}\ q_e = \text{Ln}\ q_m - \beta\varepsilon^2$

Temkin equation: $q_e = \frac{RT}{b}\ln A + \frac{RT}{b}\ln C_e$ B= $\frac{RT}{b}$

Where q_{max}, the monolayer capacity of the adsorbent (mg/g); K_L, the Langmuir constant (L/mg) and related to the free energy of adsorption; q_m, the theoretical saturation capacity (mg/g); and ε, the Polanyi potential, which is equal to RT Ln (1 + (1/Ce)), where R (J/mol K) is the gas constant and T (K) is the absolute temperature; β, a constant related to the mean free energy of adsorption per mole of the adsorbate (mol^2/kJ^2). For Freundlich isotherm, K_F is the adsorption capacity at the unit concentration (L/g) and $\frac{1}{n}$ is adsorption intensity. $\frac{1}{n}$ Values specify whether isotherm is irreversible ($\frac{1}{n} = 0$), favorable ($0 < \frac{1}{n} < 1$) or unfavorable ($\frac{1}{n} > 1$). For Temkin isotherm, the A, b, R, T are equilibrium binding constant (L/g) Temkin isotherm constant, universal gas constant (8.314 J/mol K) and temperature at 298 K, respectively. B is constant related to the heat of sorption (J/mol).

The linear plot of 1/Ce versus $1/q_e$ (Fig. 3a) is utilized to calculate the q_{max} and K_L in Langmuir equation. The calculation of q_m and β of D-R equation is performed by plotting Ln q_e versus ε^2 (Fig. 3b). K_F and 1/n of Freundlich equation are achieved from the slope and intercepts of the linear plot of log q_e versus log C_e (Fig. 3C).

Temkin isotherm contains a factor that is explicitly entered into the adsorbent–adsorbate interactions. By ignoring the extremely low and large value of concentrations, the model assumes that heat of adsorption (function of temperature) of all molecules in the layer would decrease linearly rather than logarithmic with coverage. As implied in the equation, its derivation is characterized by a uniform distribution of binding energies (up to some maximum binding energy) was carried out by plotting the quantity sorbed q_e against Ln Ce and the constants were determined from the slope and intercept (Fig. 3d).

The Langmuir, Freundlich, Temkin and D-R parameters for the adsorption of Ampicillin onto Mon-nanoparticles were listed in Table 3. The comparison of R^2 represented in Table 3 advocates that D-R and Langmuir model have more facility to portray the data compared to Temkin and Freundlich models.

The constant β is related to the mean free energy E (kJ/mol) of adsorption per molecule of the adsorbate when it is moved to the surface of the solid from the solution. It is calculated by the following relationship [37-40]:

$$E = \frac{1}{(2\beta)^{1/2}}$$

Where *β* is represented as the isotherm constant. Based on the linear plot obtained from D-R model, the q_m was determined to be 141.22 mg/g; the mean free energy, E, was observed to 2.56 Kj/mol which this value indicates the adsorption of Ampicillin is a physisorption process. The R^2 value for this isotherm model was calculated to be 0.991.

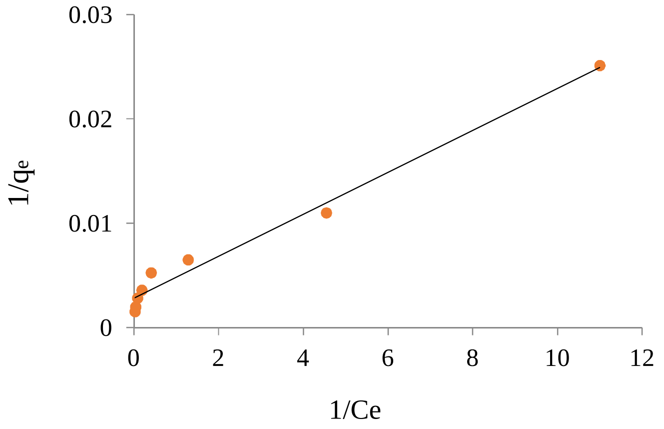
Also considering the Table 3, the values of 1/n = 0.425 and n=2.35 indicate the favorable sorption of Ampicillin unto Mon-nanoparticles; the R^2 value is achieved to be 0.82.

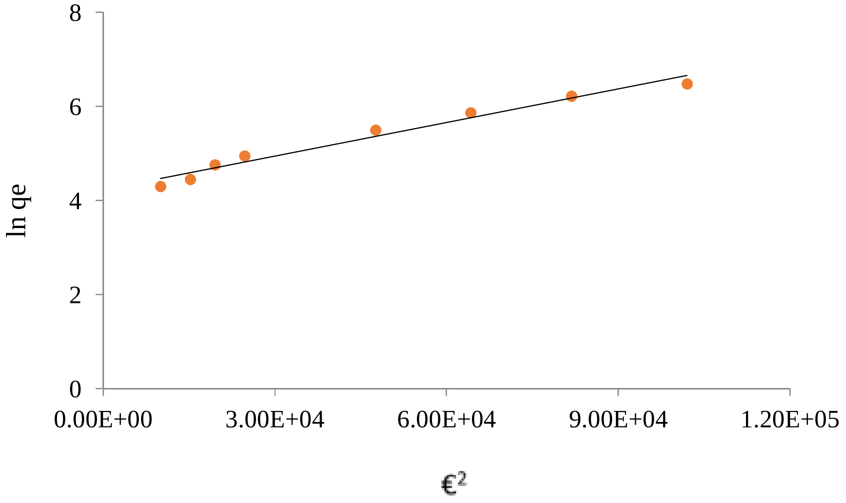
Moreover, Table 3 shows that the values of A and B is 1.85 L/g and 34.61 J/mol, respectively which is indicative of the adsorption heat and a physical adsorption process; the R^2 was found to be 0.84.

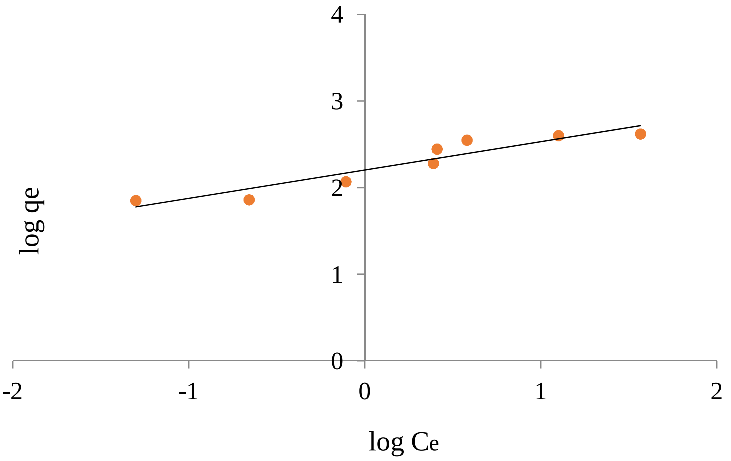
The critical properties of the Langmuir isotherm may be affirmed from viewpoint of equilibrium parameter R_L, which is a dimensionless constant related to separation factor or equilibrium parameter [41-44]:

$$R_L = \frac{1}{1 + (1 + K_L\,C_e)}$$

K_L is considered as the constant referred to the adsorption energy (Langmuir Constant). R_L is a value that reveals the adsorption nature so that the adsorption could be unfavorable (R_L>1), linear (R_L=1), favorable (0<R_L<1) and irreversible (R_L=0). According to the data represented in Table 3, the favorable Langmuir isotherm is observed for RL values greater than 0 and lesser than 1. Furthermore, the maximum monolayer coverage capacity (q_{max}), which is calculated by the Langmuir Isotherm model, was obtained to be 134.48 mg/g and K_L (Langmuir isotherm constant) is 0.047 L/mg. R_L (the separation factor) is 0.254 and it shows that the equilibrium sorption was favorable; and the R^2 value is 0.989 and it is confirmed that the sorption data was appropriately fitted to Langmuir Isotherm model. Table 4 depicts the maximum adsorption capacities (q_{max}) of some adsorbents for the better understanding of the absorbent performance.







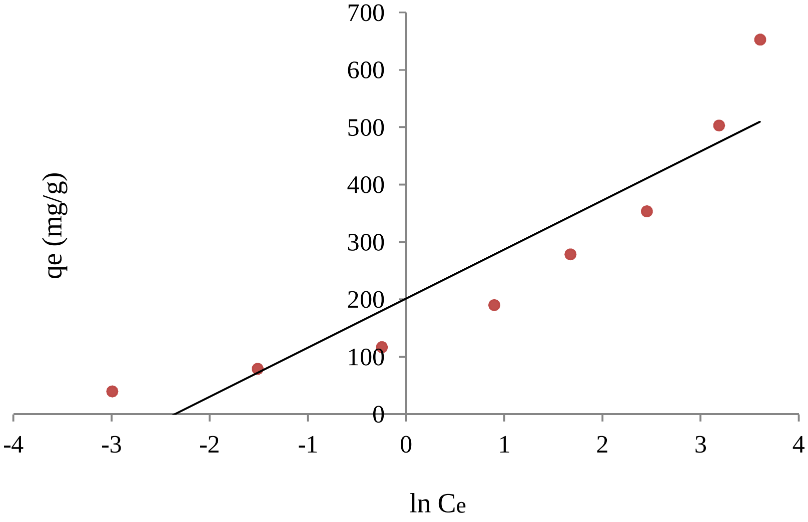


Fig. 3. a: Langmuir, b: D-R C: Freundlich and d: Temkin adsorption isotherms of Ampicillin Mon-nanoparticles

Table 3. Isotherms constants for the removal AMX onto Pumice

Langmuir model				Freundlich model		
q_m (mg/g)	**R_L**	**K_L**	**R^2**	**n**	**K_F**	**R^2**
134.48	0.254	0.0047	0.989	2.35	4.571	0.821
D-R model				**Temkin model**		
q_m (mg/g)	**β**	**E**	**R^2**	**B**	**A**	**R^2**
141.22	0.0062	2.56	0.991	34.61	1.85	0.842

Table 4. Maximum adsorption capacities (q_{max}) of some adsorbents for different antibiotics

Adsorbent	Antibiotic	q_{max} (mg/g)	Ref
Mon-nanoparticles	Ampicilin	134.48	present work
Azolla filiculoides	Sulfamethoxazole	17.25	[1]
Activated Carbon	Ciprofloxacin	242.8	[2]
Clay	Sulfonamide	46.29	[3]
MWCNS	Tetracycline	196.4	[10]

Adsorbent	Antibiotic	q_{max} (mg/g)	Ref
MWCNS	Metronidazole	266.7	[11]
MWCNS	Amoxicillin	159.4	[12]
Lemna minor	Penicillin G	26.17	[18]
Alumina-coated-MWCNT	Tetracycline	347.9	[20]
MgO nanoparticles	Cephalexin	500	[45]
Organobentonite	Amoxicillin	30.12	[46]

4. CONCLUSION

The present study shows that the Mon-nanoparticles were successfully used as an adsorbent for the quantitative removal of Ampicillin from aqueous solutions. The experimental adsorption equilibrium data of Ampicillin on Mon-nanoparticles were compared with the Langmuir, Freundlich, Temkin and Dubinin–Radushkevich models and adsorption capacities were determined. These results showed that Langmuir and D–R isotherm model was better than Freundlich and Temkin model.

CONSENT

It is not applicable.

ETHICAL APPROVAL

It is not applicable.

COMPETING INTERESTS

Authors have declared that no competing interests exist.

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