JOURNAL OF CRITICAL REVIEWS

ISSN- 2394-5125 VOL 7, ISSUE 19, 2020

ADSORPTION OF PHARMACEUTICALS COMPOUND RIFABUTIN IN AQUEOUS SOLUTIONS USING MAGNETIC NANOCOMPOSITES AS AN ADSORBENT IN AN AQUEOUS SOLUTION

Ramesh Auchat and Swati Goyal*

Department of Chemistry, Dr. A. P. J. Abdul Kalam University, Indore, Madhya Pradesh

*Corresponding Author Email: swatipc2011@gmail.com

Received : 06.10.2020 Revised : 09.11.2020 Accepted : 11.12.2020

Abstract

In the present investigation an adsorption method using an adsorbent MNC (Magnetic Nanocomposites) was designed for the rapid removal of rifabutin antibiotics from water. The batch reactor was utilized to create the best operating conditions by experimenting with various elements such as pH, duration time, drug concentration, and adsorbent dose. The following operating parameters were used in the batch reactor: pH 10.9, 100 minutes, 0.35 mg/mL rifabutin, and 1.66 g/L MNC as adsorbent. Meanwhile, the isothermal Langmuir, Freundlich and Tempkin models were used to investigate the kinetic constants using adsorption equilibrium. The Freundlich isotherm produced the best results, and the first pseudo order was better for removing rifabutin via adsorption activation of MNC (Magnetic Nanocomposites). The adsorption efficiency could reach 95%, suggesting that MNC is an excellent adsorbent for Rifabutin removal from water. The adsorption kinetics fitted the pseudo-first-order model perfectly. The adsorption isotherms study showed the maximum sorption capacity. Thermodynamic parameters for the adsorption were estimated, and the ΔH° and ΔG° values indicated the endothermic and spontaneous nature of the sorption process.

Keywords: Rifabutin, Wastewater, Adsorbent, Adsorption, Isotherm

1. INTRODUCTION

Traditionally, the effect of chemical pollution has focused almost entirely on the traditional priority contaminants. However, the growing use of pharmaceuticals worldwide, known as the so-called emerging contaminants, has become a new environmental issue, which has raised great concern among scientists in the last few years [1]. In human and veterinary medicine, over 3,000 chemical products are used [2]. Antibiotics and growth hormones used in human and veterinary medicine have a major impact on surface and groundwater quality [3]. Some other studies also showed the presence of different mixtures of pharmaceuticals in wastewaters and surface waters [4-

ISSN- 2394-5125 VOL 7, ISSUE 19, 2020

5]. Anti-inflammatory medications, antibiotics, sulfa drugs, antifungal drugs, antidiabetics, barbiturates, -blockers, diuretics, antihypertensive drugs, hormones, lipid regulators, anti neoplastics, psychiatric drugs, histamine-blockers, topical products, and antiseptics have all been found in wastewater and surface waters [6-7]. In all over India, an estimated 90% of waste-water is discharged directly into rivers and streams without treatment. The main cause of wastewater production is the manufacturing or chemical processes in industries. Industrial waste-water generally contains specific and readily identifiable chemical compounds. But water pollution is concentrated within a few sub sectors, mainly in the form of toxic wastes and organic pollutants. Out of this, a large portion can be traced to the processing of industrial chemicals and to the food product industries. The textile industry, a major consumer of water for several of its wet processing operations, is also a major producer of effluent waste-water containing organic surfactants, salts, acids, alkalis, solvents and drugs as some of its main constituents. Drugs though present in only small amounts are highly detectable and thereby are capable of causing serious problems of an aesthetic nature in the receiving water bodies. Major sources of water pollution are showed in figure 1.

Antibiotics are widely used as growth promoters in human and animal medicine and livestock and aquaculture operations. There are many new drugs and their derivatives on the market. However, although lipophilic drug candidates may have potential pharmacodynamic activities, only around 40% of them make it to market due to poor bioavailability. As a result, it is important to improve the solubility of poorly water-soluble drugs. Rifabutin is first-line therapy for tuberculosis with high permeability and low solubility [8]. Rifabutin was discovered to have efficacy against mycobacterium avium complex disease, a bacterial infection that is most often seen in AIDS patients in late stages. Despite this, Rifabutin can cause serious side effects such as extreme skin rash or scratching, pale skin, fatigue, easy bruising or bleeding, fever, chills, body aches, flu symptoms, or eye pain or redness, as well as vision loss.

The Liquisolid method is a revolutionary technique in which water-insoluble drugs are dissolved in a non-volatile solvent and then converted into a free-flowing, non-adherent, and compressible powder using a carrier and coating content. Absorption and adsorption occur when a substance is dissolved in a non-volatile solvent and combined with carrier materials with a porous surface. The coating material is applied after the liquid adsorption process on the external and internal surfaces of the porous carrier particle [9].

The presence of Rifabutin and other antibiotics in natural environments can cause bacteria to acquire and transmit antibiotic-resistant genes, which potentially threatens ecosystem functions and human health [10, 11]. Therefore, it is of great importance to developing efficient and cost-effective treatment technologies for the removal of Rifabutin from contaminated waters to minimize its ecological risks.

Pharmaceutical compounds-related hazards must be avoided and minimized by treating effluent-containing drugs with the necessary technique. Traditional techniques (biological

ISSN- 2394-5125 VOL 7, ISSUE 19, 2020

process, filtration, coagulation, flocculation, and sedimentation), advanced oxidation process (AOPS), membrane treatment, and adsorption have all been tested to extract antibiotics from aqueous solutions [12-14]. Each approach has its own removal performance, capital costs, benefits, drawbacks, and operating rates. Physical techniques, especially adsorption, have been found to be more appropriate and efficient than chemical processes for the removal of recalcitrant and organic compounds in previous studies [15]. Furthermore, the adsorption method will create a high-quality effluent free of harmful substances [16, 17]. Bamboo charcoal [18], montmorillonite [19], bio-char [20], graphene oxide [21], soil and sediment [22], activated carbon [23], multiwalled carbon nanotubes (MWCNTs) [24], and single-walled carbon nanotubes (SWCNTs) [25], Magnetic nano adsorbent (MNA) have all been tested as adsorbents for Rifabutin. MNA, due to their relatively large specific surface areas, unique hollow and porous structure, high mechanical strength, small size, and remarkable electrical conductivities have been utilized as a new and promising adsorbent to remove many kinds of organic and inorganic contaminants.

2. EXPERIMENTAL

2.1 REAGENTS AND SOLUTIONS

The Rifabutin (Molecular weight 444.43, Molecular formula $C_{46}H_{62}N_4O_{11}$ was purchased from Sigma-Aldrich, USA. The chemical structure of Rifabutin is presented in Table 1. The distilled water was used to prepare the stock solution of Rifabutin Other chemicals used in this study were prepared. All chemicals applied in this study were of analytical grade or higher. Sulphuric acid, acetic acid and boric acid were obtained from Merck, Germany. Surfactant MNA was purchased from Sigma-Aldrich, USA. Deionized (DI) water (18.25 MΩ·cm) was come into being from a water purification system (EMD Millipore Corp., Merck KGaA, Darmstadt, Germany).

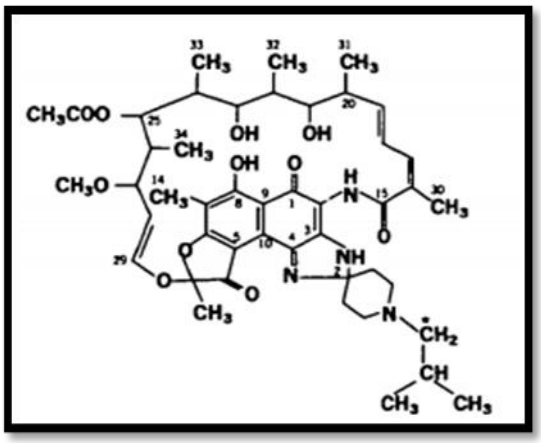

Figure 1: Structure of Rifabutine

ISSN- 2394-5125 VOL 7, ISSUE 19, 2020

2.2 APPARATUS

For pH measurements decibel DB 1011 digital pH meter was used and a spectrophotometer with a 1.0 cm light path quartz cells (systronic spectrophotometer 166 over the wavelength range 325 – 900 nm) was used for all degradation analysis at λ max of 370 nm. SEM photograph of UPR reveals surface texture and porosity. SEM was performed using a Zeiss EVO 50 instrument. Powder X-ray diffraction (XRD) measurements were performed on Diffractometer system XPERT-PRO X-ray powder diffract meter using a graphite monochromatic with Cu Ka radiation (k = 1.5406 Å).

2.3 PROCEDURE

All the adsorption experiments were carried out in dark using brown glass vials (total volume = 150 ml) with 50 ml Rifabutin solution and MNA on an HZQ-120H heating oscillator (Yiheng Scientific Instrument Co., Ltd., Shanghai, China) with a speed of 160 rpm. The pH was adjusted using dilute HCl and NaOH aqueous solution (aq.). Rifabutin adsorption kinetics studies were conducted at pH = 7 and temperature = 20 °C with an initial concentration of Rifabutin of 80 mg/L and adsorbent dosage of 20 g/L. At predetermined times (5–600 min), the vials were sacrificially sampled. Besides, to investigate adsorption thermodynamics, the adsorption kinetics assays were carried out at 30 °C and 40 °C as well. For the Rifabutin adsorption isotherm experiment, the initial concentration of Rifabutin was varied from 2 to 80 mg/L with a fixed adsorbent dosage of 20 g/L, and the mixture (pH = 7) was shaken for 24 h at 20 °C to reach the adsorption equilibrium. To explore the effect of adsorbent dosage on adsorption, different doses of adsorbent (5–50 mg/L) were added into Rifabutin solution (80 mg/L), and the mixture (pH = 7) was shaken for 24 h at 20 °C. To probe the effect of pH on adsorption, the equilibrium tests were conducted with an initial Rifabutin concentration of 80 mg/L, an adsorbent dosage of 20 g/L, and a final solution pH 2–10 at 20 °C for 24 h. To examine the effect of ionic strength, 0–0.25 mol/L NaCl or CaCl2 were added into Rifabutin solution (80 mg/L) with an adsorbent dosage of 20 g/L at pH = 7, the temperature of 20 °C, and shaking for 24 h. After adsorption is completed, the solution was filtered through a 0.22 μm microfiltration membrane. The concentrations of Rifabutin were detected via SP-756P Ultraviolet-Visible Spectrophotometer at 370 nm.

The adsorption amount at predetermined time t (qt, mg/g) and equilibrium adsorption amount (qe, mg/g) of Rifabutin on materials and removal efficiency (R, %) were calculated via:

$$q_t = \frac{(C0 - CT)V}{m} \quad (1)$$

$$q_e = \frac{(C0 - Ce)V}{m} \quad (2)$$

$$R = \frac{(C0 - Ce)}{Co} \times 100\ \% \quad (3)$$

Where *Ct* (mg/L) is the residual concentration in the liquid phase at sampling time *t* (min); *C*0 and *Ce* (mg/L) are the initial and equilibrium concentrations of Rifabutin, respectively; *V* (L) is the total volume of the solution; and *m* (g) is the mass of adsorbent.

ISSN- 2394-5125 VOL 7, ISSUE 19, 2020

Rifabutin concentration in the solution phase (*Cd*, mg/L) was determined upon centrifugation and filtration, and the percent of Rifabutin desorbed as calculated via:

$$D = \frac{Cd}{(C0 - Ce)} \times 100\% \quad (4)$$

3. RESULTS AND DISCUSSION

3.1 EFFECT OF ADSORBENT DOSE

The aim of this study was to see how pH affects Rifabutin adsorption. To assess the optimum pH for maximum adsorption, the initial pH of the solutions (50 mL, initial concentration 50 mg L 1) was adjusted from 3 to 11 using 0.1mol/L HCl or NaOH. The adsorbent dose was 0.5 g L-1, with a 4 hour agitation period. Furthermore, to see how the pH of the adsorbate solution influenced Rifabutin adsorption. Preparing an adsorbent–adsorbate solution with various amounts of adsorbents applied to a set initial drug concentration and shaking it can be used to test the effect of adsorbent dose on the adsorption method. In general, as the adsorbent dose is increased, the amount of drug removed increases. The rate of increase in percent drug removal was found to be rapid at first, but it slowed as the dosage was increased. This phenomenon can be explained, based on the fact that at a lower adsorbent dose the adsorbate (drug) is more easily accessible and because of this, removal per unit weight of adsorbent is higher.

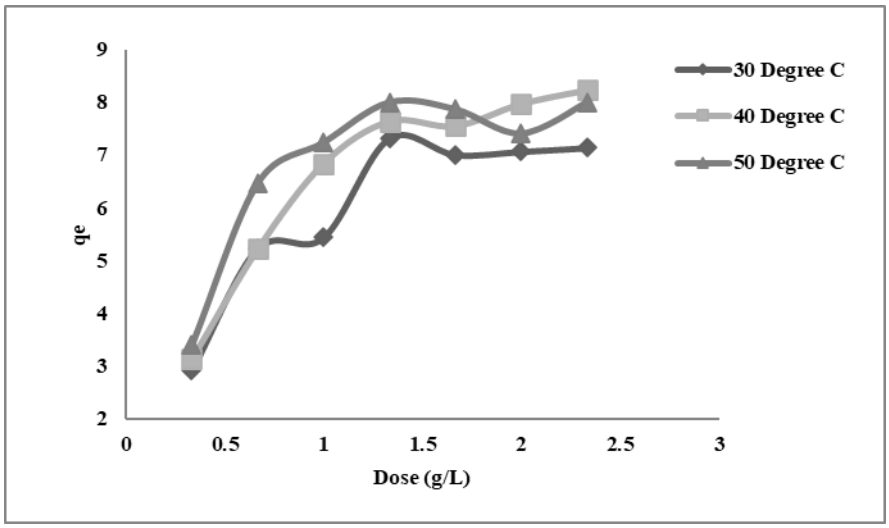

Figure 2: Effect of amount of adsorbent for the removal of rifabutin at 0.35mg/mL at pH 10.9 and different temperatures

3.2 EFFECT OF ADSORBATE CONCENTRATION

Over a broad range of Rifabutin concentrations, the effect of initial concentration on MNA removal efficiency was investigated. Figure depicts the findings. As shown in the figure, Rifabutin uptake was rapid at lower concentrations (10-15 mg/L) but decreased as concentrations increased, indicating that the amount of Rifabutin adsorbed decreased. The amount of Rifabutin adsorbed per unit mass of adsorbent increased (from 0.5 to 1.49 mg/g) as the Rifabutin concentration increased from 10 to 30ppm, but the percentage

ISSN- 2394-5125 VOL 7, ISSUE 19, 2020

sorption decreased (from 99.959 to 99.128). As all of the sites are occupied, on the other hand, the adsorbed concentration becomes nearly constant. This means that a monolayer is forming on the carbon surface. In fact, the more concentrated the solution, the better the adsorption capacity of MNA. Owing to the high porosity of the adsorbent, the carbon impregnated with iron has a higher Rifabutin sorption potential than the other two active carbons.

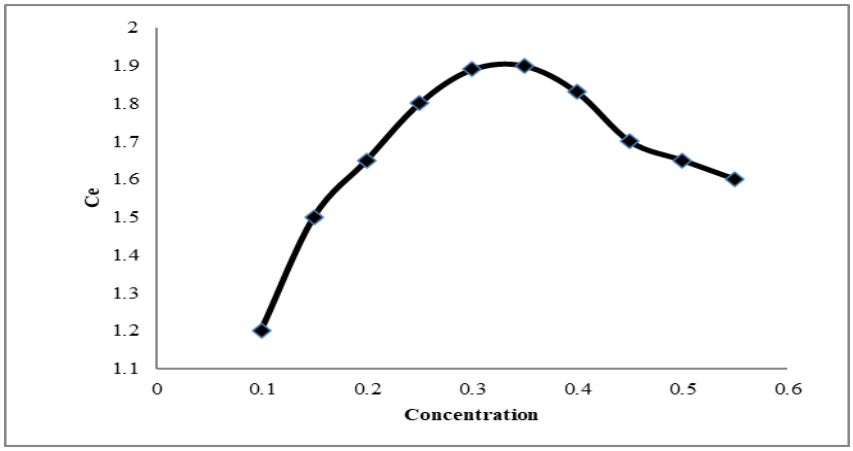

Figure 3: Effect of concentration for the removal of rifabutin over MNA at 1.66 g/L at pH 10.9 and 30°C temperature

3.3 EFFECT OF PH

This research was to look at how pH affects Rifabutin adsorption. The initial pH of the solutions (50 mL, initial concentration 50 mg L 1) was changed from 3 to 11 using 0.1mol/L acetic acid or boric acid to determine the optimal pH for maximal adsorption. The adsorbent dose was 0.5 g L-1, and the agitation time was 4 hours. Furthermore, a collection of brown flasks containing the same concentrations of Rifabutin but without the adsorbent were used as blanks to see how the pH of the adsorbate solution affected Rifabutin adsorption.

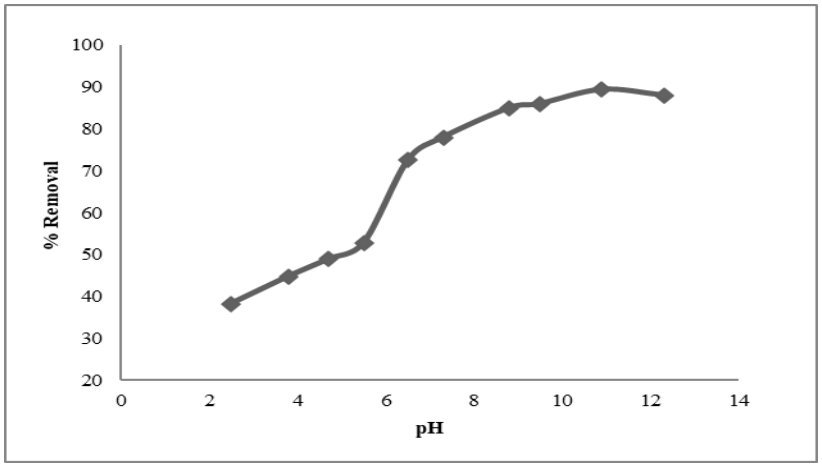

Figure 4: Effect of pH for the removal of rifabutin over MNA at 1.66 g/L at 30°C temperature and different pH

ISSN- 2394-5125 VOL 7, ISSUE 19, 2020

3.4 EFFECT OF CONTACT TIME

The effect of contact time on the percentage removal of Rifabutin from aqueous solution using MNA was investigated keeping initially constant the adsorbent dosage (0.1 g), drug concentrations (25 mg/L and 50 mg/L), the volume of solution (40 mL), and stirring speed (120 rmp) at room temperature (Figure) Results indicate that the uptake of Rifabutin drug was very fast at the beginning (first 3 h) and then continued to increase until reaching equilibrium after 22–24 h. This is due to the presence of several adsorption sites on the adsorbent surface in the early stages of the reaction, which eventually become saturated with the drug as contact time increase. The observed moderate rates of adsorption after the first 2–3 hours could be due to repulsive forces between solute molecules on the solid and bulk phases.

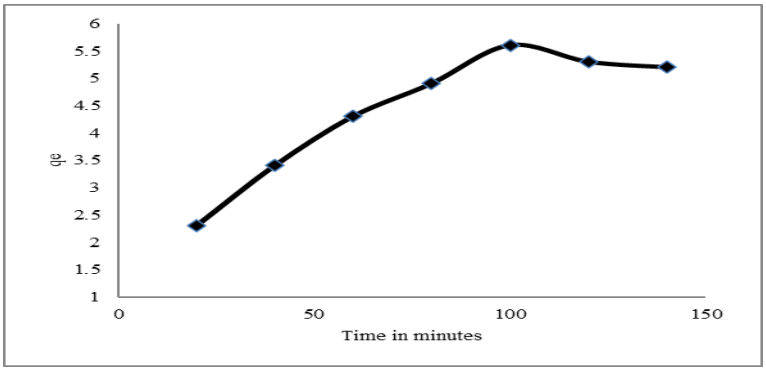

Figure 5: Effect of contact time for the removal of rifabutin over MNA at 1.66 g/L at pH 10.9 and 30°C temperature

3.5 EFFECT OF TEMPERATURE

Temperature is another key factor to consider when using an adsorbent to remove the dye. The adsorption of malachite green by UPR at various temperatures is shown in Figure 5.7. The dye sorption potency is totally affected by temperature, implying that as the temperature of the system rises, the fraction of dye removed increases. This is because the chemical reactions between the malachite green dye and the adsorbent are endothermic.

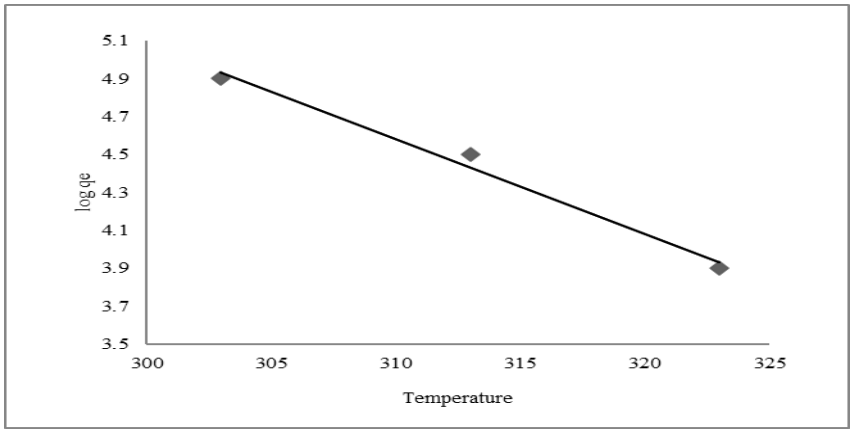

Figure 6: Effect of temperature for the removal of rifabutin over MNA at 1.66 g/L at pH 10.9 and different temperatures

ISSN- 2394-5125 VOL 7, ISSUE 19, 2020

4. ADSORPTION ISOTHERM MODELING

Only a few experimental interpretations of adsorption isotherms were in use prior to 1914. Following that, a number of isotherm equations were suggested by various researchers. The following are some that are often used:

• Langmuir isotherm
• Freundlich isotherm
• Temkin isotherm

4.1 LANGMUIR ISOTHERM

The Langmuir biosorption isotherm [26] was based on the following assumptions:

• Fixed number of biosorption sites: at equilibrium, at any temperature, a fraction of the biosorbent surface sites (θ) is occupied by adsorbed molecules and the rest (1-θ) is free.

- All sorption processes are homogeneous.
- A monolayer surface phase is formed.
- One sorbate molecule reacts with only one active site.
- No interaction between the sorbate species.
- There is only one sorbate

The equation proposed by Langmuir was universally applicable to chemisorption with some restrictions involving physical adsorption. This equation can be used to describe physical or chemical adsorption on a solid surface with a single form of adsorption active core. The Langmuir equation can be used to describe equilibrium conditions for sorption activity in various adsorbate-adsorbent systems or for various conditions within any given system as long as its limitations and restrictions are clearly understood. The Langmuir equation is given by:

$$q = \frac{qmax\ Kaeq}{1{=}Kaeq} \tag{5}$$

Where *Qmax* indicates the monolayer adsorption capacity of adsorbent (mg/g) and the Langmuir constant *b* (L/mg) is related to the energy of adsorption. For fitting the experimental data, the Langmuir model was linearized as

$$\frac{1}{q} = \frac{1}{qmax} + \frac{1}{Ka\ qmax\ Ceq} \tag{6}$$

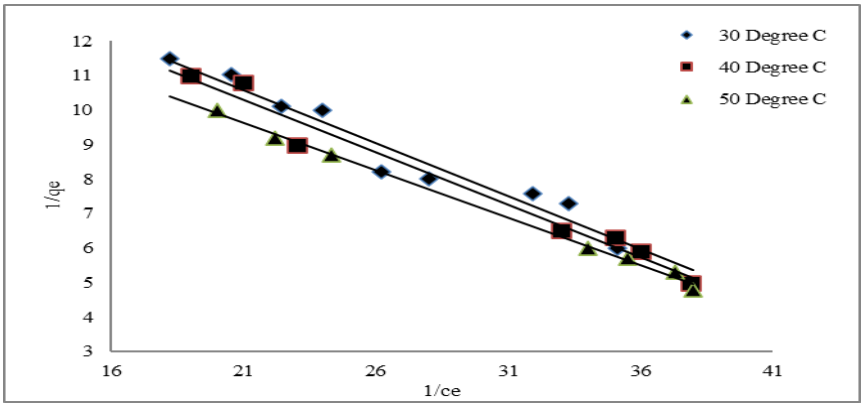

Figure 7: **Langmuir adsorption isotherm for the adsorption of rifabutin over MNA**

ISSN- 2394-5125 VOL 7, ISSUE 19, 2020

Table 1: Langmuir Constants for rifabutin over MNA

Temp.	b($molg^{-1}$)	Q^o($Lmol^{-1}$)	bQ^o	R^2	%RSD#
30 °C	7.396	1.468	0.235	0.919	1.18
40 °C	5.763	1.723	0.528	0.937	1.52
50 °C	4.025	1.938	0.738	0.926	1.89
#average of three replicate measurements					

4.2 FREUNDLICH ISOTHERM

Freundlich adsorption isotherm was proposed by Boedecker in 1895 as an empirical equation. Later Freundlich [27] made some usable modifications as a result of which, it considered great importance. The Freundlich adsorption equation can be written as:

$$q = KC_{eq}^{1/n} \quad (7)$$

Taking the ln of both sides,

$$\ln q = \ln K + \frac{1}{n} \ln C_{eq} \quad (8)$$

Where 'q' is equilibrium adsorption capacity (mg/g), 'Ce' is the equilibrium concentration of the adsorbate in solution, 'K', and 'n' are constants related to the adsorption process such as adsorption capacity and intensity respectively.

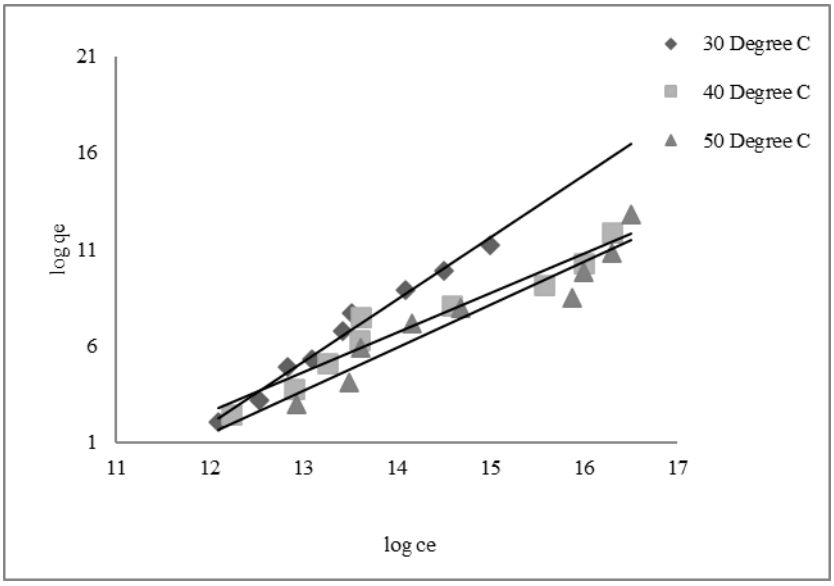

Figure 8: Freundlich adsorption isotherms for adsorption of rifabutin over MNA

ISSN- 2394-5125 VOL 7, ISSUE 19, 2020

Table 2:**Freundlich constants for the rifabutin over MNA**

Temp.(^oC)	K_f	N	R^2	%RSD#
30	1.897	1.936	0.984	0.931
40	2.763	1.362	0.936	1.132
50	3.571	1.028	0.928	1.062
#average of three replicate measurements				

4.3 TEMKIN ISOTHERM

Temkin and Pyzhev suggested that due to the indirect adsorbate/biosorbent interaction, the heat of adsorption of all the molecules in the layer would decrease linearly with coverage [28]. The linear form of Temkin isotherm can be written as:

$$q = \frac{RT}{b} \ln (A_T C_{eq}) \quad (9)$$

Where *AT* (L/mg) and *bT* are Temkin isotherm constants, 'T' is absolute temperature in Kelvin and 'R' is the universal gas constant (J/mol.K). Ceq is the equilibrium concentration of the adsorbate.

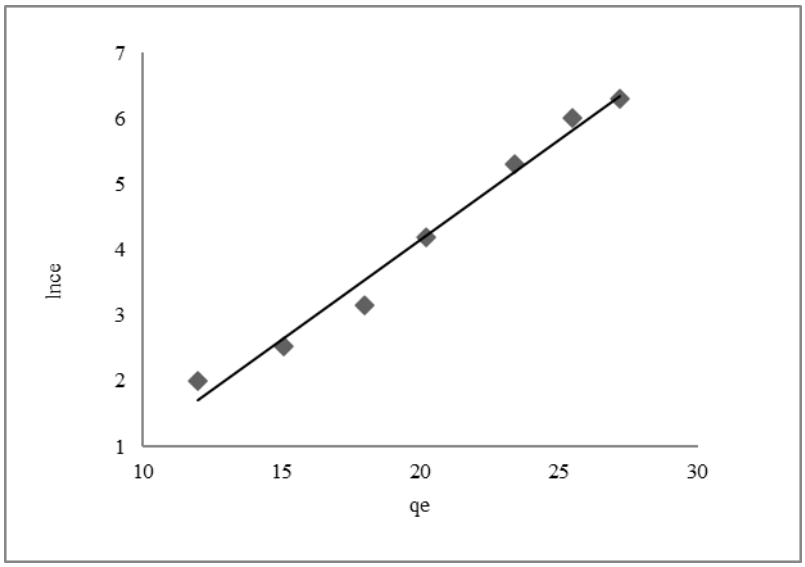

Figure 9: Tempkin adsorption isotherms for adsorption of rifabutin over MNA

Table 3: **Tempkin constants for the Rifabutin over MNA**

Temp.(^oC)	B ($Jmol^{-1}$)	A (Lg^{-1})	B	R^2
30	2.298	31.456	31732	0.937

ISSN- 2394-5125 VOL 7, ISSUE 19, 2020

5. THERMODYNAMIC STUDY

Temperature is a well-known factor that influences adsorption processes. At 298 K, 318 K, and 338 K, the adsorption of Rifabutin onto MNA was investigated. The adsorption potential of MNA increases with increasing temperature against the Rifabutin adsorption mechanism, indicating that the process is endothermic, as shown in Fig. The following van't Hoff equation is used to derive the thermodynamic parameters related to the adsorption process.

$$\ln K_c = \frac{\Delta Ho}{RT} + \frac{\Delta So}{R} \quad (10)$$

$$K_c = \frac{Qe}{Ce} \quad (11)$$

Where T is the solution temperature (K), R is the universal gas constant, Kc is the thermodynamic equilibrium constant, ΔH^o and ΔS^o are changes in the enthalpy and entropy, respectively. A plot of lnKc against 1/T is shown in Fig. gives a straight line and the values of

ΔS^o and ΔH^o can be found from the intercept and slope. The calculated thermodynamic parameters at three different temperatures are given in Table.

From Table 1, it is clear that the adsorption process is endothermic for Rifabutin adsorption because of the positive ΔH^o value, which is identified by the increase in adsorption capacity at high temperatures for the Rifabutin adsorption process. Also, the Kc values are increasing with the temperature indicating the endothermic nature of the adsorption process in the case of Rifabutin adsorption onto MNA. The negative value of ΔG^o implies a decrease in randomness by the adsorbed species and indicates the stability of the adsorption process with no structural change at the solid-liquid interface.

ΔG^o, Gibb's free energy change is the fundamental criterion of the spontaneity of a reaction. The negative values of ΔG^o indicate that the reaction occurs spontaneously at a given temperature. The free energy change was calculated from the following equation

$$\Delta G^o = -RT \ln K_c \quad (12)$$

The negative ΔG^o values confirm that the adsorption process is feasible thermodynamically as well as the spontaneous nature of the sorption having a high affinity of Rifabutin, by the adsorbent. Furthermore, the ΔG^o values decrease with the increasing temperature for Rifabutin adsorption onto MNA indicating the sorption process is more desirable at a higher temperature. Generally, the ΔG^o values for physical and chemical sorption are in the range of 0 to 20 KJ mol-1 and 80 to 400 KJ mol-1. In this study, the ΔG^o values are in the range of 0.053 to 2.20, indicating the adsorption process is generally physical in nature.

ISSN- 2394-5125 VOL 7, ISSUE 19, 2020

Table 4: **Thermodynamic parameters of rifabutin over MNA at pH 10.5 at different temperatures**

Adsorbent		ΔG° kJmol^{-1})		ΔH° (kJ mol^{-1})	ΔS°(Jk^{-1} mol^{-1})
	30°C	40°C	50°C	30°C	30°C
MNA	9.3651×10^3	8.9472×10^3	7.4639×10^3	-72.248×10^3	75.823

6. ADSORPTION KINETICS MODELS

The rate of Rifabutin adsorption on the modified zeolite was investigated using various kinetic models (Lagergren, pseudo-second-order, Elovich, and diffusion model). Figure 8 depicts the effect of contact time on adsorption performance. Adsorption kinetics is studied using first and second-order equations. The following is the pseudo-first-order rate equation:

$$\frac{dq}{dt} = k_1 (q_e - q) \quad (13)$$

Integrating Eq. (3) results in:

$$\frac{\ln qe - q}{qe} = -k_1 t \quad (14)$$

The pseudo-second order equation can be expressed as:

$$\frac{dq}{dt} = k_2 (q_e - q)^2 \quad (15)$$

Integrating Eq. (5) gives:

$$\frac{t}{q} = \frac{1}{k2qe2} + \frac{1}{qe} t \quad (16)$$

By plotting ln qe –q/ qe for the first order and t/q for second order versus t, the lines with slopes of $-k_1$ and 1/qe are obtained.

Three measures in succession may be used to explain the overall rate of adsorption in a solid-liquid system: (1) adsorbent transfer from the bulk of the liquid to the solid surface; (2) adsorbate diffusion from the surface via the pores of the adsorbent; (3) adsorption on the adsorbent's empty sites.

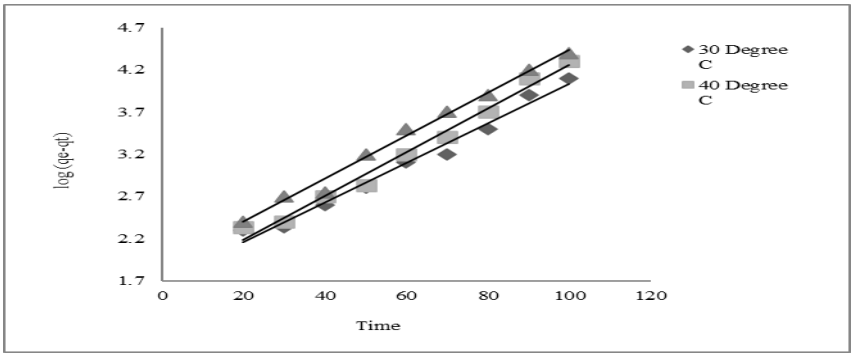

Figure 10: Lagergren pseudo first order plots for adsorption of rifabutin over MNA at pH 10.9 at different temperatures

ISSN- 2394-5125 VOL 7, ISSUE 19, 2020

Table 5: Rate constant k_{ad} for rifabutin over MNA

Temp.(^oC)	k_{ad}	%RSD[#]
30	0.169	0.989
40	0.192	0.923
50	0.208	0.967
#average of three replicate measurements		

7. CONCLUSION

In a batch adsorption experiment, Rifabutin was tested and evaluated as a potential adsorbent for the removal of Rifabutin drug from its aqueous solution. The adsorption experiments were carried out over a broad range of conditions, including pH of the solution, adsorbent dosage, temperature, initial concentration, and contact time. The optimum pH for Rifabutin adsorption was in the range of 5–7, and it took about 120 minutes for the adsorption to achieve equilibrium. The results suggest that none of the investigated cations or anions had a substantial impact on the adsorption of Rifabutin onto the MNA. As it suspended in water, the Rifabutin antibiotic interacted with the oxidized MNA at the solid/liquid interface. The experimental data were fitted to nonlinear kinetic models, and the second-order kinetic model best described the kinetic of Rifabutin adsorption. However, the intra-particle diffusion model gave multiple linear regions suggesting that the adsorption could also follow multiple adsorption rates. Equilibrium data were fitted to three known isotherm models, and the Freundlich model gave the best fit with the maximum adsorption capacity of 253.38 mg g-1. The results of the present study indicate that oxidized MNA can be a good alternative adsorbent for the removal of Rifabutin and other pollutants from aqueous solutions.

REFERENCES:

[1]. Z. Zhang, D. P. Grover and J. L. Zhou, "Monitoring Pharmaceuticals residues in sewage effluents, In: Handbook of water purity and quality," Ed.: Satinder Ahuja, Academic Press – Elsevier, Amsterdam, Page 315-342, 2009.

[2]. T. A. Ternes, A. Joss and H. Siegrist, "Scrutinizing pharmaceuticals and personal care products in wastewater treatment," Environmental Science and Technology, Vol. 38, Issue 20, Page 392A–399A, 2004.

[3]. P. Chang, J. S. Jean, W. T. Jiang and Z. Li, "Mechanism of tetracycline sorption on rectorite," Colloids and Surface A-Physicochemical Engineering Aspects, Vol. 339, Issue 1-9, Page 94–99, 2009.

[4]. M. Oosterhuis, F. Sacher and T. L. T. Laak, "Prediction of concentration levels of metformin and other high consumption pharmaceuticals in wastewater and regional surface water based on sales data," Science of the Total Environment, Vol. 442, Page 380-388, 2013.

ISSN- 2394-5125 VOL 7, ISSUE 19, 2020

[5]. F. Sacher, M. Ehmann, S. Gabriel, C. Graf and H. J. Brauch, "Pharmaceuticals have become of increasing environmental concern," *Journal of Environmental Monit*oring, Vol. 10, Issue 5, Page 664-670, 2008.

[6]. B. Kasprzyk-Hordern, R. M. Dinsdale and A. J. Guwy, "The occurrence of pharmaceuticals, personal care products," *Water Res*earch, Vol. 43, Issue 2, Page 363-380, 2009.

[7]. P. Verlicchi, M. Al-Aukidy and E. Zambello, *Sci. Total Environ*., Vol. 429, Page 123, 2012.

[8]. S. D. Seth and V. Seth, Textbook of Pharmacology. Elsevier New Delhi, Third Edition, 2009

[9]. S. Panda, R. Varaprasad, K. Priyanka and R. P. Swain, "Liquisolid technique: a novel approach for dosage form design," International Journal of Applied Pharmaceutics, Vol. 9, Page 8-14, 2017.

[10]. M. A. Gilliver, M. Bennett, M. Begon, S. M. Hazel and C. A. Hart, "Antibiotic resistance found in wild rodents," *Nature*, Vol. 401, Issue 6750, Page 233–234, 1999.

[11]. B. Halling-Sørensen, S. Nors Nielsen, P. F. Lanzky, Ingerslev, F. Holten, H. C. L¨utzhøft and S. E. Jørgensen, "Occurrence, fate and effects of pharmaceutical substances in the environment-A review," *Chemosphere*, Vol. 36, Issue 2, Page 357–393, 1998.

[12]. V. Homem, and L. Santos, "Degradation and removal methods of antibiotics from aqueous matrices – a review," Journal of Environmental Management, Vol. 92, Issue 10, Page 2304–2347, 2011.

[13]. N. Jaafarzadeh, B. Kakavandi, A. Takdastan, R. R. Kalantary, M. Azizi, and S. Jorfi, "Powder activated carbon/Fe3O4 hybrid composite as a highly efficient heterogeneous catalyst for Fenton oxidation of tetracycline: degradation mechanism and kinetic," RSC Advances, Vol. 5, Issue 103, Page 84718–84728, 2015.

[14]. B. Kakavandi, A. Takdastan, N. Jaafarzadeh, M. Azizi, A. Mirzaei, and A. Azari, "Application of Fe3O4@C catalyzing heterogeneous UV-Fenton system for tetracycline removal with a focus on optimization by a response surface method," Journal of Photochemistry and Photobiology A: Chemistry, Vol. 314, Page 178–188, 2016.

[15]. B. Kakavandi, A. Esrafili, A. Mohseni-Bandpi, A. Jonidi Jafari, and R. Rezaei Kalantary, "Magnetic Fe3O4@C nanoparticles as adsorbents for removal of amoxicillin from aqueous solution," Water Science & Technology, Vol. 69, Issue 1, Page 147– 155, 2014.

[16]. M. Shirmardi, A. H. Mahvi, A. Mesdaghinia, S. Nasseri, and R. Nabizadeh, "Adsorption of acid red 18 dye from aqueous solution using single-wall carbon nanotubes: kinetic and equilibrium," Desalination and Water Treatment, Vol. 51, Issue 34–36, Page 6507–6516, 2013.

[17]. M. Vosoughi Niri, M. Shirmardi, A. Asadi, H. Golestani, A. Naeimabadi, M. J. Mohammadi, and M. Heidari Farsani, "Reactive red 120 dye removal from aqueous solution by adsorption on nano-alumina," Journal of Water Chemistry and Technology, Vol. 36, Issue 3, Page 125–133, 2014.

[18]. P. Liao, Z. Zhan, J. Dai, X. Wu, W. Zhang, K. Wang, and S. Yuan, "Adsorption of tetracycline and chloramphenicol in aqueous solutions by bamboo charcoal: a batch and fixedbed column study," Chemical Engineering Journal, Vol. 228, Page 496–505, 2013.

[19]. Y. Zhao, X. Gu, S. Gao, J. Geng, and X. Wang, "Adsorption of onto montmorillonite: cations and humic acid effects," Geoderma, Vol. 183–184, Page 12–18, 2012.

ISSN- 2394-5125 VOL 7, ISSUE 19, 2020

[20]. P. Liu, W. J. Liu, H. Jiang, J. J. Chen, W. W. Li, and H. Q. Yu, "Modification of bio-char derived from fast pyrolysis of biomass and its application in removal of tetracycline from aqueous solution," Bioresource Technology, Vol. 121, Page 235–240, 2012.

[21]. Y. Gao, Y. Li, L. Zhang, H. Huang, J. Hu, S. M. Shah, and X. Su, "Adsorption and removal of tetracycline antibiotics from aqueous solution by graphene oxide," Journal of Colloid and Interface Science, Vol. 368, Issue 1, Page 540–546, 2012.

[22]. Z. Zhang, K. Sun, B. Gao, G. Zhang, X. Liu, and Y. Zhao, "Adsorption of tetracycline on soil and sediment: effects of pH and the presence of Cu(II)," Journal of Hazardous Materials, Vol. 190, Issue 1–3, Page 856–862, 2011.

[23]. A. C. Martins, O. Pezoti, A. L. Cazetta, K. C. Bedin, D. A. S. Yamazaki, G. F. G. Bandoch, T. Asefa, J. V. Visentainer, and V. C. Almeida, "Removal of tetracycline by NaOHactivated carbon produced from macadamia nut shells: kinetic and equilibrium studies," Chemical Engineering Journal, Vol. 260, Page 291–299, 2015.

[24]. L. Zhang, X. Song, X. Liu, L. Yang, F. Pan, and J. Lv, "Studies on the removal of tetracycline by multi-walled carbon nanotubes," Chemical Engineering Journal, Vol. 178, Page 26-33, 2011.

[25]. L. Ji, W. Chen, L. Duan, and D. Zhu, "Mechanisms for strong nadsorption of tetracycline to carbon nanotubes: a comparative study using activated carbon and graphite as adsorbents," Environmental Science & Technology, Vol. 43, Issue 7, Page 2322–2327, 2009.

[26]. I. Langmuir, "The adsorption gasses on plane surface of glass, mica and platinum," Journal of the American Chemical society, Vol. 40, Issue 9,Page 1361-1368, 1916.

[27]. H. M. F. Freundlich, "over the adsorption in solution," Zeitschrift für physikalische Chemie, Vol. 57, Issue 1, Page 385-470, 1906.

[28]. C. aharoni and M. Ungarish, "Kinetics of activated chemisorption Part 2. The oriental models," Journal of the Chemical Society, Faraday Transactions, Vol. 73, Page 456-464, 1977.