British Journal of Pharmaceutical Research
9(5): 1-10, 2016, Article no.BJPR.22820
ISSN: 2231-2919, NLM ID: 101631759

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Experimental and Kinetic Studies on *Penicillin G* Adsorption by *Lemna minor*

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Authors' contributions

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

Article Information

DOI: 10.9734/BJPR/2016/22820
Editor(s):
(1) Abdelwahab Omri, Department of Chemistry and Biochemistry and Departments of Biomolecular Sciences, Laurentian University, Canada.
Reviewers:
(1) Alejandro Córdova Izquierdo, Universidad Autónoma Metropolitana Unidad Xochimilco, Mexico.
(2) M. Angeles calvo torras, Autonomous University of Barcelona, Spain.
Complete Peer review History: http://sciencedomain.org/review-history/12620

Original Research Article

Received 29th October 2015
Accepted 21st November 2015
Published 10th December 2015

ABSTRACT

This study examined the ability of dried duckweed (*Lemna minor*) to remove soluble *Penicillin G* from aqueous solution. Batch experiments were conducted to investigate the effects of pH, contact time, initial *Penicillin G* concentration, biomass dose and temperature on penicillin G adsorption. Maximum adsorption capacity of the duckweed was 36.18 mg g^{-1} when 94.6% of the *Penicillin G* was removed. The adsorption equilibriums were analyzed by Langmuir, Freundlich, Temkin and BET isotherm models. It was found that he data fitted to Langmuir better than isotherm other models. Batch kinetic experiments showed that the adsorption followed pseudo-second-order kinetic model with correlation coefficients greater than 0.99. The adsorption capacity of *penicillin G* increased from 31.11 to 41.82 mg/g with increasing the temperature from 20 to 50℃, indicating that the process is endothermic. According to achieved results, it was defined that *Lemna minor* not only was an inexpensive absorbent, but also a quite effective factor in removal of *Penicillin G* from water and wastewater.

Keywords: Adsorption; Lemna minor; Penicillin G; antibiotic; kinetic.

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1. INTRODUCTION

Pharmaceuticals are not environmentally different from other chemicals, such as pesticides and herbicides, but they have not aroused attention as potential pollutants until fairly recently [1,2]. Antibiotics are designed specifically as a drug to treat or prevent infective diseases in human or animal body [3,4]. The use of antibiotics has become an indispensable in human life and the global market consumption of these drugs increase steadily every year [5,6]. For preventing or treating infections in humans or animals, only some parts of the antibiotics given dose are metabolized and the rest are excreted still as active compound [7,8]. *Penicillin* is an extremely important antibiotic, and *penicillin G* is the raw material of the semi synthetic penicillin's [9]. Over 20 countries manufacture over 11,000 tones of *penicillin* per year. Its wide usage is a result of its lack of toxicity and irritancy [10,11]. *Penicillin G* is commonly produced by submerged aerobic fermentation using strains of Penicillium chryogenum. Penicillin G is a weak monocarboxylic acid (pKa = 2.75) [12]. For several decades now antibiotics have been used in veterinary and human medicine, yet these compounds when released into the environment have potential risks for aquatic and terrestrial organisms [13]. High concentrations of antibiotics are now detected in drinking waters, reducing their quality [14]. They generally have a low biodegradability and high toxicity, and some are reported to have mutagenic and carcinogenic characteristics [15]. The removal of antibiotics from pharmaceutical wastewater is quite expensive, however this wastewater must be treated properly prior to the release into environment [16]. Among the available process for the treatment of wastewater containing antibiotics, the adsorption process is considered as the most effective and efficient method [17,18]. The main drawback of adsorption process for wastewater treatment is the cost of adsorbent [19]. Commercially available adsorbents such as activated carbon are expensive [20]. If inexpensive and abundantly available materials are found in nature as the alternative adsorbents, the adsorption process offers a very attractive method for environmental remediation [21,22]. Recently, various materials such as Azolla, wheat straw, fungus, orange peels, Rice straw, Wheat straw and Sawdust been applied to develop as low-cost and effective adsorbent [23,24]. The *Lemna minor* is one of wide-spread aquatic plant which is belonged to duckweed species with special characteristics including rapid growth, high nutritional value, and high water purification capabilities have been used to remove the pollutant from water and wastewater [25,26]. Therefore, the purpose of present study was to assess the *Lemna minor* ability as adsorbent for remove the *Penicillin G* antibiotic. Also, the effect of several parameters including contact time, pH, adsorbent dose and initial *Penicillin G* concentration was investigated.

2. MATERIALS AND METHODS

2.1 Chemicals

The *Penicillin G* used in the experiments was supplied by Aldrich Chemicals. Stock solutions of *Penicillin G* were prepared in distilled water. All chemicals used in this study were purchased from Merck Co. All solutions used in this study were diluted with distilled water as required.

In this study, the *Lemna minor* was used for *Penicillin G* removal from aqueous solutions. The *Lemna minor* were collected from *Anzali* wetland. The collected materials were then washed several times with distilled water to remove all dirt particles. The dried materials were then ground, using steel mill. The crushed particles were then treated with 0.5 M H_2SO_4 for 2 h followed by washing with distilled water and then kept for shaded dry [15].

The BET surface area of *Lemna minor* was measured using surface area analyzer (Micromeritics ASAP 2020 V3.04 H). Treated *Lemna minor* biomass before and after use were examined using an environmental scanning electron microscopy (ESEM) instrument (Philips XL30).

2.2 Adsorption experiments of synthetic *Penicillin G* solution

The Initial *Penicillin G* solutions with different concentrations were prepared by diluting a *Penicillin G* stock standard solution of 1000 mg/L with distilled water. The solution pH was adjusted using either diluted 0.1 M H_2SO_4 or 0.1 M NaOH solution. 100 ml of *Penicillin G* solution and 4 g biomass was placed in a Pyrex glass and stirred at constant speed for 90 hours using a magnetic stirrer (Remi Make, India). The equilibrium times at different temperatures (30℃, 40℃ and 50℃) were estimated by testing the samples collected at different time intervals until *Penicillin G* content in the sample was constant. At the end of the

equilibration time, the solution was centrifuged (EBA21, Hettich) at 3600 rpm for 10 min. The equilibrium concentration of *Penicillin G* in solution was determined by *HPLC* (kenaver, Germany). In the HPLC analysis, the mobile phase was a mixture of 50 μm phosphoric acid and methanol (40:60,v/v). The flow rate was 1.0 ml/min, the detection wavelength was 215 nm, and the injection volume was 10 ml [12]. All batch experiments were carried out in triplicate.

The amount of *Penicillin G* adsorbed per was calculated by following mass-balance equation [27,28]:

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

Where C_0 and C_e are the initial and the equilibrium *Penicillin G* concentrations in liquid phase (mg/L) respectively, V is the volume of solution and m is the mass of adsorbent.

2.3 Adsorption Isotherms

The equilibrium adsorption isotherm is importance in the design of adsorption systems. Although several isotherm equations are available, but four important isotherms including Langmuir, Freundlich, Tekmin and BET isotherms were selected.

The Langmuir equation is given as follows [29,30]:

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

Where q_e (mg/g) and C_e (mg/L) are the amount of adsorbed *Penicillin G* per unit weight of adsorbent and unabsorbed *Penicillin G* concentration in solution at equilibrium, respectively. The constant K_L is the Langmuir equilibrium constant.

The Freundlich isotherm model equation is expressed as [12]:

$$\text{Ln } q_e = \ln K_F + \frac{1}{n} \ln Ce$$

Where q_e is the equilibrium *Penicillin G* concentration on the adsorbent (mg g^{-1}); Ce, the equilibrium *Penicillin G* concentration in solution (mg L^{-1}); and K_F is the Freundlich constant.

The Tempkin isotherm model equation is expressed as [29]:

$$q_e = B \ln A + B \ln Ce$$

Where B = RT/b, T is the absolute temperature in K, R the universal gas constant (8.314 JK^{-1} mol^{-1}), A the equilibrium binding constant and the constant B is related to the heat of adsorption.

The BET isotherm model in the linear form as used is represented as [20]:

$$\frac{c_e}{(C_i - C_e)q} = \left(\frac{1}{q_m\, A}\right) + \left(\frac{A-1}{q_m\, A}\right)$$

C_e is the equilibrium concentration (mg/L), C_i the adsorbate monolayer saturation concentration (mg/ L), A the BET adsorption isotherm relating to the energy of surface interaction (l/mg).

2.4 Adsorption Kinetics

The study of kinetic models was performed in contact time between 10-180 min with *Penicillin G* concentration of 25 and 200 mg/L and optimum amount of pH and adsorbent dose. To evaluate the differences in the biosorption rates and uptakes, the kinetic data were described with Elovich, Intraparticle diffusion, pseudo first, pseudo second order models.

The pseudo first order kinetics model in the linear form as used is represented as [31,32]:

$$\text{Log } (q_e - q_t) = \log q_e - \frac{K_1}{2.303} t$$

Where q_e and q_t are the amounts of *Penicillin G* (mg g^{-1}) adsorbed at equilibrium and time t, respectively, and k_1 is the rate constant of adsorption (min^{-1}) biosorption.

The pseudo-second-order kinetic model is expressed as [12]:

$$\frac{t}{q_t} = \frac{1}{k_2 q e^2} + \frac{1}{qe\, t}$$

Where q_e is the biosorbed *Penicillin G* amount at equilibrium (mg g^{-1}) for the pseudo-second-order biosorption, q_t is the amount of *Penicillin G* biosorbed at time t (mg g^{-1}) and k_2 is the pseudo-second-order kinetic rate constant (g mg^{-1} min^{-1}).

The intraparticle diffusion equation can be written as follows [13]:

$$q_t = k_{dif}\, t^{0.5} + c$$

where C is the intercept, and kdif is the intraparticle diffusion rate constant (mg g^{-1} min^{-1}).

The Elovich model equation is generally expressed as [20]:

$$q_e = (\frac{1}{\beta})\ln(\alpha\beta) + (\frac{1}{\beta})\ln t$$

where α is the initial adsorption rate (mg/g.min) and β is the adsorption constant (g/mg) during any experiment.

3. RESULTS AND DISCUSSION

The specific surface area is related to the number of active adsorption sites of dried *Lemna minor*. The adsorption was increased with the specific surface area and pore volume of the sorbent. The specific surface area of the modified *Lemna minor* was determined in the size of 32.5 m^2/g.

Dried *Lemna minor* was also examined before and after use using environmental scanning electron microscopy. Fig. 1(a) clearly shows the pore textural structure of dried *Lemna minor* before use. However, as shown in Fig. 1(b), clear pore textural structure is not observed on the surface of dried *Lemna minor* after use which could be due to either agglomeration on the surface or the incursion of *penicillin G* into the pores of dried *Lemna minor* biomass.

3.1 Effect of Contact Time and Initial *Penicillin G* Concentration

The adsorption of *penicillin G* onto *Lemna minor* was studied as a function of contact time in order to decide whether the equilibrium was reached. For, 10-200 mgL^{-1} of *penicillin G* solutions at pH 5 were contacted with 4 gL^{-1} of *Lemna minor* suspensions. The samples were taken at different periods of time and analyzed for their *penicillin G* concentration (Fig. 2). The *penicillin G* adsorption rate is high at the beginning of the experiment because initially the adsorption sites are more available and *penicillin G* ions are easily adsorbed on these sites [33]. The equilibrium can be reached within 75 min, and thus, further adsorption experiments were carried out for a contact time of 75 min.

3.2 Effect of pH

The surface charge of the adsorbent and the ionization degree of the adsorbate are strongly affected by the pH of the aqueous solutions, hence the uptake of *penicillin G* by the adsorbent depends on the solution pH. In order to evaluate the effect of pH on the adsorption of *penicillin G* onto *Lemna minor*, the adsorption experiments were carried out with initial *penicillin G* concentration of 50 mg L^{-1} and biomass dose of 4 gL^{-1} by varying the pH of the solutions over a range of 2–10 (Fig. 3). The uptake of *penicillin G* by the *Lemna minor* is almost constant in the pH range of 2–5. However when the pH value exceeds 5, the adsorption of *penicillin G* decreases abruptly. *Penicillin G* as a acid compound with p*K*a value of 2.8 is dissociated at pH>p*K*a [34]. At higher pH values, the ionization degree of *penicillin G* and the quantity of OH^- ions increase thereby the diffusion of *penicillin G* are hindered, and the electrostatic repulsion between the negatively charged surface sites of the adsorbent and *penicillin G* ions increases [35]. As a result, the removal of *penicillin G* is greater at lower pH compared to the higher pH. Similar results were reported by *Malakotian* [12]. From the experimental results, pH 5 was selected as an optimum pH value.

3.3 Effect of *Lemna minor* Concentration

The effects of *Lemna minor* dose on the removal of *penicillin G* from aqueous solutions were investigated by using different *Lemna minor* dose in the range of 0.5–8 g L^{-1} and initial *penicillin G* concentration of 100 mg L^{-1} at pH 5. As the *Lemna minor* dose was increased from 1 to 8 g L^{-1}, the equilibrium adsorption capacity of *Lemna minor* (q_e), decreased from 6.0 to 2.6 mg g^{-1}, whereas, the *penicillin G* removal efficiency increased from 5.5% to 58.5% (Fig. 4). The increase in adsorption percentage of *penicillin G* was probably due to the increased more availability of active adsorption sites with the increase in *Lemna minor* dose [36]. The decrease in equilibrium adsorption capacity of *Lemna minor* for *penicillin G* uptake could be attributed to two reasons. First, the *Lemna minor* particles aggregated with increasing the adsorbent dose hence total surface area of the adsorbent decreased and diffusion path length of *penicillin G* increased [37]. Secondly, the increase in *Lemna minor* dose at constant concentration and volume of *penicillin G* lead to unsaturation adsorption sites, so the equilibrium adsorption capacity of *Lemna minor* decreased [12].

The adsorption kinetics is one the most important data in order to understand the mechanism of the

adsorption and to assess the performance of the adsorbents. For evaluating the kinetics of *penicillin G-Lemna minor* interactions, the pseudo-first-order, pseudo-second-order and intraparticle diffusion models were used to fit the experimental data. The pseudo-first-order rate constant k_1 and the value of q_e cal were calculated and the results are given in Table 1. The correlation coefficient (R^2) is relatively low which may be indicative of a bad correlation. In addition, q_e cal determined from the model is not in a good agreement with the experimental value of q_e exp. Therefore, the adsorption of *penicillin G* onto *Lemna minor* is not suitable for the first-order reaction.

From Table 1, the value of C obtained from intraparticle diffusion model is not zero, and the correlation coefficient is not satisfactory thereby intraparticle diffusion may not be the controlling factor in determining the kinetics of the process. The pseudo-second-order rate constant k_2 and the value of q_e cal were determined from the model and the results are presented in Table 1. The value of correlation coefficient is very high (R^2 >0.994) and the calculated q_e cal value is closer to the experimental q_e exp value. In the view of these results, the pseudo-second-order kinetic model provided a good correlation for the adsorption of *penicillin G* onto *Lemna minor* in contrast to the pseudo-first-order and intraparticle diffusion model.

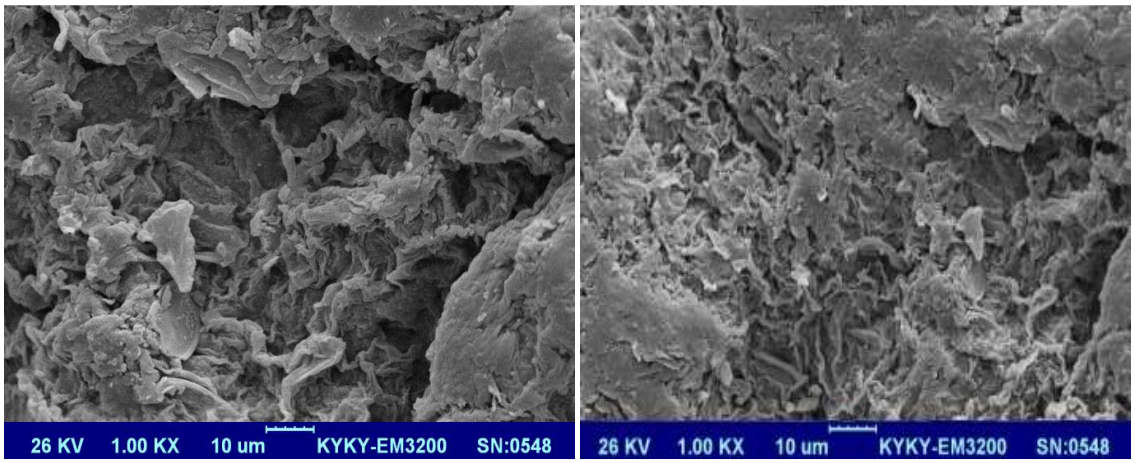

Fig. 1. The SEM image of modified *Lemna minor* before and after used

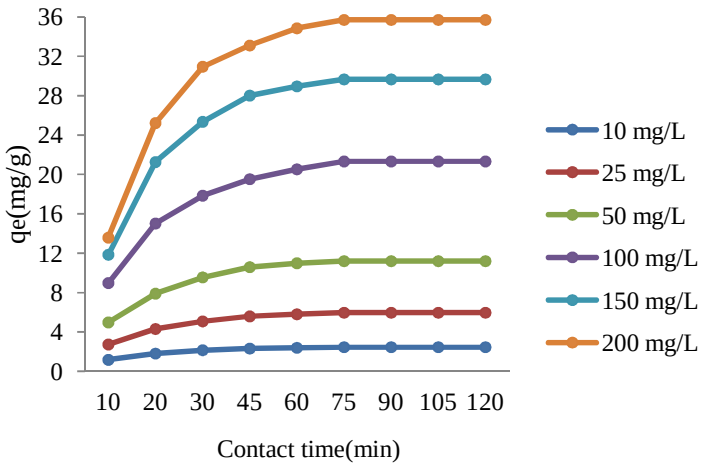

Fig. 2. Effect of contact time and initial *penicillin G* concentration (pH = 3, dose: 4 g/L and temp 30℃)

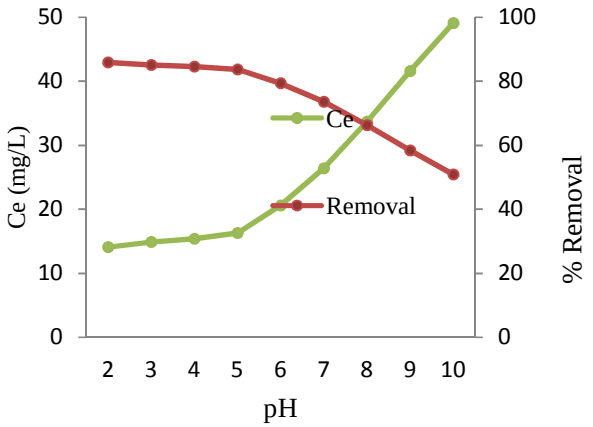

Fig. 3. Effect of pH on adsorption (Con = 100 mg/L, Contact time = 75 min, dose: 4 g/L and temp 30℃)

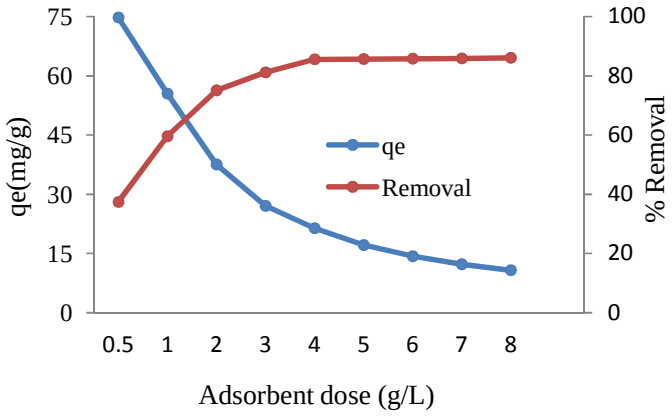

Fig. 4. Effect of adsorbent dose (Contact time = 75 min, pH = 5, Con: 100 mg/L and temp 30℃)

Table 3. Parameters of pseudo-first-order, pseudo-second-order and intraparticle diffusion models

C_0(mg/L)	q_e (mg/L)	Pseudo-first-order			Pseudo-second-order			Intraparticle diffusion		
		k_1	q_e (mg/g)	R^2	q_e (mg/g)	k_2	R^2	k	C (mg/g)	R^2
25	5.96	0.014	3.41	0.901	5.68	0. 32	0.994	0.128	2.17	0.845
50	11.45	0.0 22	7.65	0.916	11.39	0. 49	0.996	0.156	2.95	0.869
100	21.32	0.036	17.39	0.887	21.94	0. 67	0.999	0.194	3.64	0.837
200	35.71	0.044	29.76	0.877	36.42	0. 86	0.998	0.223	4.11	0.912

A study of the temperature dependence of adsorption reactions gave valuable knowledge about the enthalpy and entropy changes during adsorption. The removal of *penicillin G* onto *Lemna minor* was studied at 20, 30, 40 and 50℃ to determine the adsorption isotherms and thermodynamic parameters. As seen in Fig. 5, adsorption ability of *Lemna minor* for *penicillin G* increased with increasing temperature. The adsorption capacity of *penicillin G* increased from 31.11 to 41.82 mg/g by increasing the temperature of the solution from 20 to 50℃, indicating that the process is endothermic.

The equilibrium adsorption isotherm is of importance in the design of adsorption systems. In general, the adsorption isotherm describes

how adsorbate interacts with adsorbents and thus is critical in optimizing the use of adsorbents. Several isotherm equations are available, and four important isotherms were selected for this study: The Langmuir, Freundlich, Tekmin and BET isotherms. The Langmuir adsorption isotherm assumes that adsorption takes place at specific homogeneous sites within the adsorbent, and it has been used successfully for many adsorption processes of monolayer adsorption.

The values for q_m, K_L, k_F , B, k_t , A and n are summarized in Table 2. The isotherm data were calculated from the least square method and the related correlation coefficients (R^2 values) are given in the same table. As seen from Table 2, the Langmuir equation represents the adsorption process very well; the R^2 values were all higher than 0.99, indicating a very good mathematical fit. The fact that the Langmuir isotherm fits the experimental data very well may be due to the homogeneous distribution of active sites onto the *Lemna minor* surface, since the Langmuir equation assumes that the surface is homogenous. As seen in Table 4, the maximum adsorption capacities for *penicillin G* onto *Lemna minor* at 20, 30, 40 and 50℃ were found to be 32.36, 36.18, 38.45 and 42.27 mg/g, respectively. Maximum adsorption capacities of *Lemna minor* increased with increasing temperature.

It is generally stated that the values of n in the range of 1 to 10 represent good adsorption. In the present work, the exponent was 1 <n <10, indicating favorable adsorption.

The essential features of the Langmuir isotherm can be expressed in terms of a dimensionless constant separation factor (R_L), which is defined by the following relationship [38].

$$R_L = \frac{1}{1+ K_L C_0}$$

The value of R_L indicates the type of the isotherm; which is unfavorable (R_L >1), linear (R_L =1), favorable (0<R_L <1) or irreversible (R_L=0). The results given in Table 2 show that the adsorption of *penicillin G* onto *Lemna minor* is favorable.

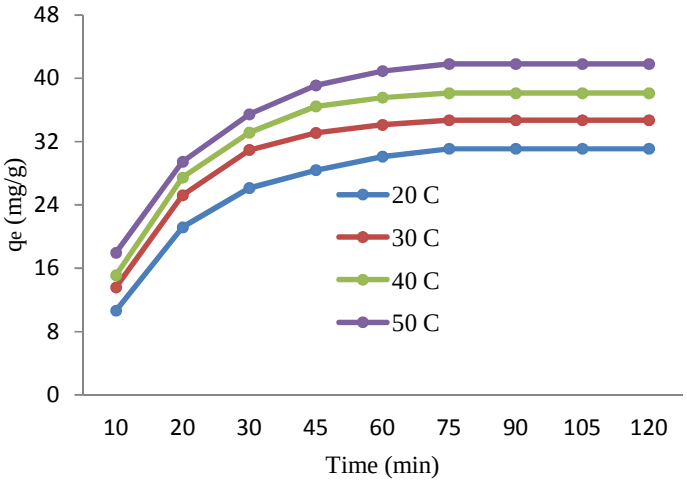

Fig. 5. Effect of contact time on the adsorption at different temperature

Table 4. The adsorption isotherms constants for the removal *penicillin G* onto *Lemna minor*

Tem	Langmuir model			Freundlich model			Temkin model			BET model		
(℃)	qm	KL	R2	n	KF	R2	B	A	R2	kt	B	R2
20	22.45	0.0014	0.994	2.65	6.14	0.812	31.71	7.41	0.804	14.4	0. 041	0.914
30	23.95	0.0039	0.995	2.17	7.39	0.845	37.45	6.52	0.781	17.7	0. 073	0.943
40	25.29	0.0063	0.997	3.09	7.85	0.791	41.12	4.36	0.768	21.2	0.095	0.904
50	26.17	0.0098	0.998	3.44	8.13	0.826	44.69	2.79	0.819	23.4	0.104	0.935

4. CONCLUSIONS

1. The present study shows that *Lemna minor*, can be used as an adsorbent for the removal of antibiotics from aqueous solutions.
2. The amount of *penicillin G* adsorbed was found to vary with adsorbent dosage, initial *penicillin G* concentration, pH, and temperature.
3. The amount of *penicillin G* uptake (mg/g) was found to increase with increase in contact time, temperature, and in initial *penicillin G* concentration, but decreased with an increase in adsorbent dosage.
4. The rate of adsorption was found to conform to pseudo-second-order kinetics with a good correlation.
5. Equilibrium data fitted very well in the Langmuir isotherm equation, confirming the monolayer adsorption capacity of *penicillin G* onto *Lemna minor* with a monolayer adsorption capacity of 36.18 mg/g at room temperature.
6. The dimensionless separation factor (R_L) showed that *Lemna minor* can be used for removal of *Lemna minor* from aqueous solutions.
7. *Lemna minor*, an inexpensive and easily available material, can be an alternative for more costly adsorbents used for antibiotics removal in wastewater treatment processes.

CONSENT

It is not applicable.

ETHICAL APPROVAL

It is not applicable.

COMPETING INTERESTS

Authors have declared that no competing interests exist.

REFERENCES

1. Hirsch R, Ternes T, Haberer K, Kratz KL. Occurrence of antibiotics in the aquatic environment. Sci Total Environ. 1999;225: 109–18.
2. Gulkowsk A, Leung HW, So MK, Taniyasu S, Yamashita N. Removal of antibiotics from wastewater by sewage treatment facilities in Hong Kong and Shenzhen, China. Water Reserch. 2008;42:395-403.
3. Adriano WS, Veredas V, Santana CC, Goncalves LRB. Adsorption of amoxicillin on chitosan beads: Kinetics, equilibrium and validation of finite bath models. Biochemical Engineering Journal. 2005;27: 132-7.
4. Polo M, Ferro-García MA, Medellín-Castillo NA. Kinetic study of the adsorption of nitroimidazole antibiotics on activated carbons in aqueous phase. J. Colloid Interf. Sci. 2010;345:481-490.
5. Zhao Y, Geng J, Wang X, Gu X, Gao S. Adsorption of tetracycline onto goethite in the presence of metal cations and humic substances. J. Colloid Interf. Sci. 2011; 361:247–251.
6. Yu A, Lin C, Yu T-H, Lateef SK. Removal of pharmaceuticals in secondary wastewater treatment processes in Taiwan. J. Hazard Mater. 2009;167:1163–9.
7. Kim SH, Shon HK, Ngo HH. Adsorption characteristics of antibiotics trimethoprim on powered and granular activated carbon. J. Ind. Eng. Chem. 2010;16:344-349.
8. Putra EK, Pranowo R, Sunarso J, Indraswati N. Performance of activated carbon and bentonite for adsorption of amoxicillin from wastewater: Mechanism, isotherms and kinetics. Water Res. 2009; 43:2419e2430.
9. Bi PY, Dong HR, Guo QZ. Separation and Purification of *Penicillin G* from fermentation broth by solvent sublation. Separation and Purification Technology. 2009;65:228–31.
10. Jonathan W, Peterson A, Laura J, Petrasky B. Adsorption and breakdown of penicillin antibiotic in the presence of titanium oxide nanoparticles in water. Chemosphere. 2012;87:911–7.
11. Yaghmaeian K, Moussavi Gh, Alahabadi A. Removal of amoxicillin from contaminated water using NH4Cl-activated carbon: Continuous flow fixed-bed adsorption and catalytic ozonation regeneration. Chemical Engineering Journal. 2014;236:538–544.
12. Malakootian M, balarak D, Mahdavi Y, Sadeghi SH, Amirmahani N. Removal of antibiotics from wastewater by azolla filiculoides: Kinetic and equilibrium studies. IJAPBS. 2015;4(7):105-113.
13. Aksu Z, Tunc O. Application of biosorption for *Penicillin G* removal: Comparison with

activated carbon. Process Biochemistry. 2005;40(2):831-47.
14. Ding R, Zhang P, Seredych M, Bandosz TJ. Removal of antibiotics from water using sewage sludge- and waste oil sludge-derived adsorbents. Water Research. 2012;90:40-6.
15. Bui TX, Choi H. Adsorptive removal of selected pharmaceuticals by mesoporous silica SBA-15. J. Hazard Mater. 2009; 168:602–8.
16. Choi K-J, Kima S-G, Kimb S-H. Removal of antibiotics by coagulation and granular activated carbon filtration. Journal of Hazardous Materials. 2008;151:38–43.
17. Ding R, Zhang P, Seredych M, Bandosz TJ. Removal of antibiotics from water using sewage sludge- and waste oil sludge-derived adsorbents. Water Research. 2012;90:40-6.
18. Crisafully R, Milhome MAL, Cavalcante RM, Silveira ER. Removal of some polycyclic aromatic hydrocarbons from petrochemical wastewater using low-cost adsorbents of natural origin. Bioresour. Technol. 2008;99:4515-4519.
19. Ali I. New generation adsorbents for water treatment. Chem. Revs. 2012;112:5073-509.
20. Zazouli MA, Mahvi AH, Mahdavi Y, Balarakd D. Isothermic and kinetic modeling of fluoride removal from water by means of the natural biosorbents sorghum and canola. Fluoride. 2015;48(1):15-22.
21. Zazouli MA, Balarak D, Mahdavi Y. Effect of *Azolla filiculoides* on removal of reactive red 198 in aqueous solution. J Adv Environ Health Res. 2013;1(1):1-7.
22. Zazouli MA, Yazdani J, Balarak D, Ebrahimi M, Mahdavi Y. Removal acid blue 113 from aqueous solution by canola. J Mazand Uni Med S. 2013;23(2):73-81.
23. Diyanati RA, Balarak D, Ghasemi SM. Survey of efficiency agricultural weast in removal of acid orang 7(AO7) dyes from aqueous solution: Kinetic and equilibrium study: Iranian Journal of Health Sciences. 2013;2(1):35-40.
24. Balarak D, Mahdavi Y. Survey of efficiency agricultural waste as adsorbent for removal of P-Cresol from aqueous solution. International Research Journal of Pure & Applied Chemistry. 2016;10(2):1-11.
25. Balarak D, Pirdadeh F, Mahdavi Y. Biosorption of acid red 88 dyes using dried *Lemna minor* biomass. Journal of Science, Technology & Environment Informatics. 2015;1(02):81–90.
26. Diyanati RA, Yousefi Z, Cherati JY, Balarak D. The ability of *Azolla* and *Lemna minor* biomass for adsorption of phenol from aqueous solutions. J Mazand Uni Med S. 2013;23(106):21-8.
27. Zazouli MA, Mahvi AH, Dobaradaran S, Barafrashtehpour M, Mahdavi Y, Balarak D. Adsorption of fluoride from aqueous solution by modified *Azolla filiculoides*. Fluoride. 2014;47(4):349-58.
28. Zazouli MA, Balarak D, Karimnezhad F, Khosravi F. Removal of fluoride from aqueoussolution by using of adsorption onto modified *Lemna minor*: Adsorption isotherm and kinetics study. J Mazand Uni Med S. 2014;23(109):208-17.
29. Ali I. The quest for active carbon adsorbent substitutes: Inexpensive adsorbents for toxic metal ions removal from wastewater. Sepn. & Purfn. Rev. 2010;39(3-4):91-171.
30. Ali I, Asim M, Khan TA. Low cost adsorbents for the removal of organic pollutants from wastewater. J. Environ. Manag. 2012;113:170-183.
31. Gupta V, Ali I, Mohan D. Equilibrium uptake and sorption dynamics for the removal of a basic dye (basic red) using low-cost adsorbents. J. of Colloid Interface Sci. 2003;265:257-264.
32. Gupta VK, Ali I, Saini VK, Gerven TV, Bruggen BV, Vandecasteele C. Removal of dyes from wastewater using bottom ash. J. Ind. & Eng. Chem. 2005;44:3655-64.
33. Zhang L, Song X, Liu X, Yang L, Pan F. Studies on the removal of tetracycline by multi-walled carbon nanotubes. Chem. Eng. J. 2011;178:26-33.
34. Mohammadi AS, Sardar M. The removal of *Penicillin G* from aqueous solutions using chestnut shell modified with H2SO4: Isotherm and kinetic study. Journal of Health & Environmental. 2012;6(1):497-508.
35. Rahardjo A, Susanto M, Kurniawan A, Indraswati N. Modified Ponorogo bentonite for the removal of ampicillin from wastewater. J. Hazard Mater. 2011;190: 1001–8.
36. Zhang CL, Qiao GL, Zhao F, Wang Y. Thermodynamic and kinetic parameters of ciprofloxacin adsorption onto modified coal fly ash from aqueous solution. Journal of Molecular Liquids. 2011;163:53–6.

37. Rivera-Utrilla J, Prados-Joya G, Sánchez-Polo M. Removal of nitroimidazole antibiotics from aqueous solution by adsorption/bioadsorption on activated carbon. J. Hazard Mater. 2009;170:298–305.

38. Balarak D, Bazrafshan E, Kord Mostafapour F. Equilibrium, kinetic studies on the adsorption of acid green 3 (AG3) dye onto *Azolla filiculoides* as adosorbent. American Chemical Science Journal. 2016;10(2):1-11.

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