MASS TRANSFER AND ADSORPTION OF AMOXICILLIN FROM WASTEWATER USING WHEAT GRAIN

BOUKHELKHAL A., BENKORTBI O., HAMADACHE M., GHALEM N. and HANINI S.

Biomaterials and transport phenomena laboratory, Faculty of Science and Technology, Chemical Engineering and Environment Department, University of Medea , Ain D'heb 26000, Medea Algeria
E-mail: benkortbi_oth@yahoo.fr

ABSTRACT

Wheat grains are a natural product which grows in the north of Algeria, was used as adsorbent to remove amoxicillin antibiotic produced by SAIDAL, antibiotical company from wastewater. Adsorption isotherm of amoxicillin on both crude and traited wheat grain was determined in batch tests. A model was developed regarding the kinetic partitioning of amoxicillin and regarding the mechanism governing the forward transfer of amoxicillin. Results were interpreted in terms of a two-film theory for flat interface. Different experimental parameters such as effect of contact time, temperature.The results showed that the maximum % removal of amoxicillin was found to be 84 % on wheat grain treated with 20% tartaric acid for the following optimum conditions: contact time of 5min and temperature of 25°C, particle size of 150 μmg.

Keywords: Wastewater, Amoxicillin, Wheat grain, Adsorption, Kinetic, Mass transfer, Modelling.

1. Introduction

Pharmaceutical ingredients are actually found as residues in water and have been recognized as part of the hazardous chemical substances able to alter the natural equilibrium system of the surrounding environment ((Chatterjee, 2008, ElSayed *et al*., 2013).

Recently, antibiotics were quantified in hospital sewage water and wastewater (Ghauch *et al*., 2009), in rivers and in wastewater treatment plants.Various techniques as well as coagulation, biodegradation and chemical oxidation etc., have been used for the removal of antibiotics from wastewater. Adsorption processes have proved to be an effective technique because of major advantages such as applicability over a large concentration range of sorbate, effective removal efficiency, low instrumentation cost, and the presence of many rate-controllable parameters (Bajpai, *et al*., 2012, Zha, *et al*., 2013).

Many researches in this area have been conducted to develop a process that can fight effectively the water contamination problem. Amongst these, adsorption is considered to be the most potential one due to its flexibility, simplicity of design, high efficiency and ability to separate wide range of chemical compounds. (Padmavathy, 2008,).

However, only a few reports have been published on the removal of antibiotics from wastewater using alternative adsorbents. Researchers currently tried to find new adsorbents biomaterials such as wheat grain, available, inexpensive and can be competitive with the activated carbon and possessing good adsorptive properties.

2. Material and method

2.1. Chemicals and analysis

The antibiotic drug amoxicillin antibiotic was kindly furnished by the pharmaceutical company Saidal of Medea (Algeria) and used as model adsorbate. Tartaric acid was purchased from FLUKA. The amoxicillin concentration in the solution was analyzed using a UV spectrophotometer (Shimadzu UV Mini-1240) by monitoring the absorbance changes at a wavelength of maximum absorbance of 532 nm.

2.2. Adsorption studies

To study the adsorption capacity of wheat grain, the adsorbent (2 g) was placed in a 1000 mL Erlenmeyer flask containing an aqueous solution of 0,24 g/L of amoxicillin (500 mL) and the solution was shaken at 100 rpm at 25°C. Various experimental conditions, including contact time, temperature, agitation speed and adsorbent dose were tested for their impact on amoxicillin adsorption.The amount adsorbed of amoxicillin onto wheat grain was calculated using the following relationship :

$$Q(mg/g) = (C_0 - C_e) * V / m \quad (1)$$

where C_0 and C_e are the initial and equilibrium amoxicillin concentrations(mg/L) respectively, V the volume of solution (L) and m the adsorbent dose (g).
All experiments were carried out in duplicate.

2.3. Model development

The extraction of amoxicillin is governed by the transfer of antibiotic molecules from the aqueous to the organic phase. In fact, the two phases involved in this study are the aqueous phase (phase 1 with a volume V_1 and an amoxicillin concentration at time t of $C_1(t)$) and the organic phase (phase 2 with a constant volume V_2 and an amoxicillin concentration $C_2(t)$). At the start of the extraction experiments, all the effluent reside in the aqueous phase, and thus the concentration of amoxicillin in the two phases is $C_1(t) = C_1(0)$ and $C_2(t) = 0$. So the mass balance of amoxicillin in the system at any given time is:

$$V_1 C_1(0) = V_1 C_1(t) + V_2 C_2(t) \quad (2)$$

Since a two-film model is used to describe the solute transfer, the mass transfer rate is given by: $J = KA(C_1(t) - C_1^*)$
where K is the overall mass transfer coefficient, A is the total interfacial area between the two phases: $V_1 \frac{dC_1}{dt} = -KA(C_1(t) - mC_2(t))$ after integrating between the limits $C_1(0)$ and $C_1(t)$ and rearranging gives:

$$C_1(t) = C_0\left((1-\beta)\exp(-\alpha t) + \beta\right) \quad (3)$$

Where,

$$\alpha = \frac{KA}{V_1}(1 + kV_r) \quad (4)$$

and

$$\beta = \frac{kV_r}{1 + kV_r} \quad (5)$$

The overall resistance to antibiotics transfer, as represented by KA, can be described as the sum of the resistances of both aqueous and organic boundary films; and then the overall combined mass transfer coefficient can be represented as:

$$\frac{1}{KA} = \frac{1}{K_{aq}A} + \frac{k}{K_m A} \quad (6)$$

It should then be possible to estimate the individual aqueous phase and organic phase mass transfer coefficients, since a plot of $1/KA$ against k should yield a straight line of slope$1/k_m A$ and intercept $1/k_{aq}A$.

3. Results

3.1. Effect of the contact time

A serie of flasks containing each 0,24 g/L of amoxicillin was used to examine the effect of the contact time. Flasks were stirred at a speed of 100 rpm at home temperature and pH of 7. Samples were taken at various time intervals. From fig 1, It can be seen that the maximum adsorbed amount for different particle size is obtained for a contact time of : 40 mn for 500 µm , 30 mn for 400µm, and 20 mn for 150 µm. In addition, the amount of amoxicillin adsorbed increases with a decrease in particle size of the wheat grain. When the particle size decreases, the surface area increases.

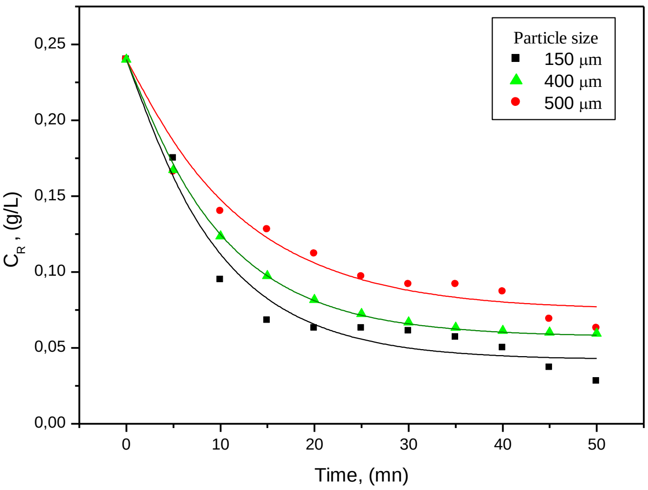

Figure 1: Time-course of amoxicillin adsorption on crude wheat grain for different particle size (T= 25°C ; C_0= 0,24g/L ; m = 4 g ; pH7; w = 350 rpm)

To improve the adsorption efficiency, a treatment of the wheat grain has been carried out for different percentage of tartaric acid. the contact time was found 5 minutes with 20% of tartaric acid and percentage removal of R = 84% and no remarkable changes was observed beyond this time, therefore considered for the rest of the experiments (Fig. 2).

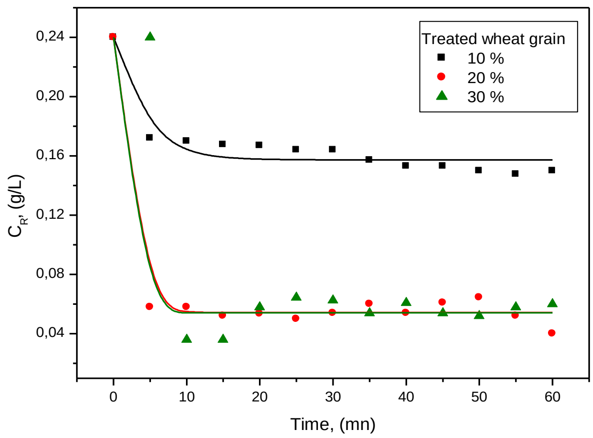

Figure 2: Time-course of amoxicillin adsorption on treated wheat grain for different percentage of tartaric acid ((T= 25°C ; C_0= 0,24g/L ; m = 4 g ; pH7; w = 350 rpm; particle size of 150 µm)

Effect of temperature

Impacts of different temperature conditions in the range from 20 to 50 on the adsorption capacity of amoxicillin onto treated wheat grain with tartaric acid have been tested in order to explain the adsorption thermodynamics. In the light of the result, Increasing temperature values caused significant decrease in amoxicillin adsorption demonstrating both of nature and characteristics of the adsorption process (Fig. 3).

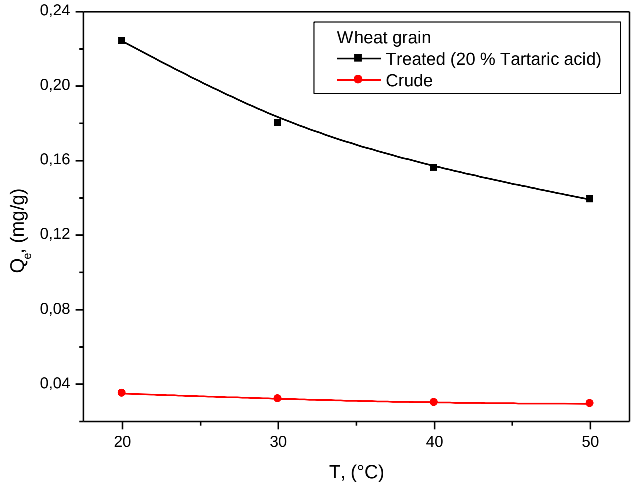

Figure 3: Effect of temperature on Amoxicillin adsorption onto wheat grain (C_0=0,24g/L, m=4g, contact time 5 min, w= 300 rpm, and pH 7).

3.2. Model development

In order to verify the model that described the amoxicillin transfer from aqueous phase to the organic phase according to the two-film model, experimental results ($C_1(t)$ versus t data) were modelled using a microcomputer program, written in MATLAB language and involving the "fminsearch" function to optimize the model coefficients according to the non-linear least-squares method. The theoretical and experimental results are compared in Fig 4 in which the solid line represents the result of the two-film model, while the symbols correspond to the experimental results (Siti *et al.,* 2012). It was found that the model fitted accurately experimental data with a correlation coefficient R^2 of 0.999.

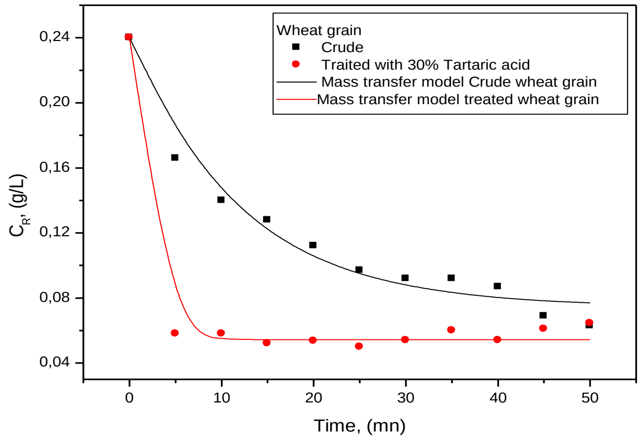

Figure 4: Time-course of amoxicillin adsorption on wheat grain (C_0 = 0,24g/L; m= 4 g; pH7; w = 300 rpm; T= 25°C.

The values of the model coefficients α and β given by the program were then inserted into Eqs. (9) and (10) to calculate the combined mass transfer coefficient (KA) as shown in Table 1.

Figure 5 shows that the plot of $1/KA$ versus k yielded a straight line, allowing to estimate the combined film mass transfer coefficients for both crude and treated wheat grain K_CA and K_TA which were 0,86×10^{-7} m^3/s and 1,56×10^{-6} m^3/s, respectively.

Table 1: Values of model coefficients (*α* and *β*) and combined mass transfer coefficient (*KA*)

Parameters	***α***	***β***	**KA (s/m^3)**	**R^2**
Crude wheat grain	0,24	0,78	$5.71.10^{-7}$	0,992
Treated wheat grain 20 Tartaric acid	0,57	0,229	$2.94.10^{-6}$	0,997

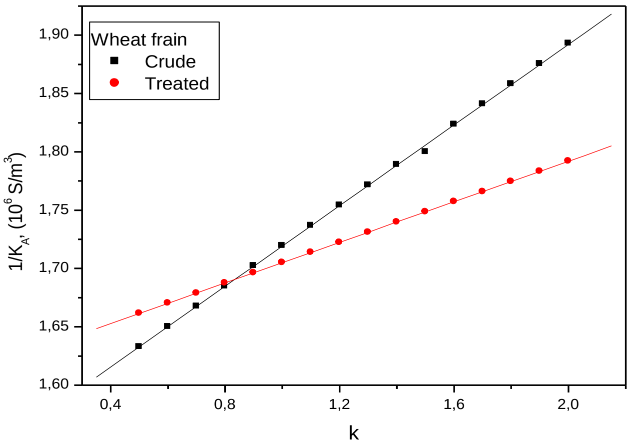

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Figure 5: Plot of the combined mass transfer coefficient (1/*KA*) against the equilibrium partition coefficient (k) for amoxicillin adsorption on wheat grain

4. Conclusions

In this study, the ability of wheat grain treated with 20% of tartaric acid to remove amoxicillin antibiotics was tested using kinetic aspects.The results indicated that, adsorption capacity of the sorbent was affected by, particle size and temperature.

The study permitted to establish the following optimum conditions: particle size,150 μm, and temperature of 25◦C. The effects of several parameters on the transfer of amoxicillin antibiotic between the aqueous phase and the organic phase were studied; and it was shown that the kinetic transfer process was successfully modeled using the general two-film theory of mass transfer to flat interface.

REFERENCES

1. Bajpai, S.K., Bajpai, M. and Rai, N. (2012) Sorptive removal of ciprofloxacin hydrochloride from simulated wastewater using sawdust: Kinetic study and effect of pH Afr. j, **38** N° 5, 673-682.
2. Chatterjee, R. (2008) Fresh produce from wastewater. Environ. Sci. Technol, **42** (21), 7732-2008.
3. ElSayed, E.M., Prasher, S.O. and Patel, R.M. (2013) Effect of nonionic surfactant Brij 35 on the fate and transport of oxytetracycline antibiotic in soil, J. Environ. Manage, **116,** 125-134.
4. Ghauch, A., Tuqan., A., and Abou Assi, H. (2009) Antibiotic removal from water: Elimination of amoxicillin and ampicillin by microscale and nanoscale iron particles Environ. Pollut, **157,** pp.1626–1635.
5. Ozbay, I., Ozdemir, U., Ozbay, B and Veli, S. (2013) Kinetic, thermodynamic, and equilibrium studies for adsorption of azo reactive dye onto a novel waste adsorbent: charcoal ash, Desalination and Water Treat, **51,** 6091–6100
6. Padmavathy, V. (2008) Biosorption of nickel (II) ions by baker's yeast: kinetic, thermodynamic and desorption studies, Biores. Technol. **99,** 3100–3109.
7. Siti, H., Mohd-Setapar, H.S., Hanapi, M. and Siti, N.M. (2012) Kinetic study of antibiotic by reverse micelle extraction technique, J. Taiwan. Inst. Chem E, **43,** Issue 5, 685-695.
8. Zha, X., Zhou, Y., Jin, X. and Chen, Z. (2013) The removal of amoxicillin from wastewater using organobentonite , J. Environ. Manage, **129,** 569-576.