Available online at www.sciencedirect.com

SciVerse ScienceDirect

journal homepage: www.elsevier.com/locate/he

Experimental study of oxytetracycline retention by adsorption onto polyaniline coated peanut shells

Fouzia Belaib, Mokri Azzedine, Boukellal Boubeker, Meniai Abdeslam-Hassen*

Laboratoire de l'Ingénierie des Procédés de l'Environnement (LIPE), Université Constantine 3, Constantine, Algeria

ARTICLE INFO

Article history:
Received 28 January 2013
Received in revised form
1 May 2013
Accepted 3 May 2013
Available online 12 June 2013

Keywords:
Oxytetracycline
Adsorption
Coating
Peanut shells
Kinetic

ABSTRACT

The release of drugs and their metabolites into the environment is of a great concern, as far as the water pollution is concerned. One can cite oxytetracycline which is used in large quantities and it is essential to reduce its presence in aquatic environments.

The present study concerns the adsorption process of oxytetracycline onto peanut shells coated with polyaniline as well as the influence of different physico-chemical parameters such as contact time, concentration, solution pH, ionic strength, and temperature on the retention capacity.

The effect of coating on the performance of the adsorption was clearly put into evidence and a thermodynamic study showed that the reaction was exothermic.

The results confirmed the effectiveness of the adsorption process for the decontamination of aquatic environments loaded with oxytetracycline.

1. Introduction

Nowadays, drug substances are essential for the health of humans and animals and their consumption has a positive effect on life expectancy. However, residues of these substances are transferred in various ways into the aquatic environment and even into drinking water [1]. Drug residues are present in trace amounts and may have consequences on health and environment. Various physico-chemical treatments can be used to degrade drugs which are contaminating waters for environmental and health protection and also for a possible reuse of non-conventional water [2].

Recently, there has been much concern about the presence of antibiotics in the aquatic and soil environments because of their toxicity, disturbance to the ecosystem function and poor biodegradability. The continuous release of antibiotics into aquatic environment increases the potential for antibiotic resistance among microbial populations [3], and their degradation have often led to even more toxic compounds [4], hence the need to develop new processes for more effective treatment for these pollutants such as Oxytetracycline (OTC) which is one of the most common antibiotic compound.

In fact there are many available techniques for oxytetracycline removal including adsorption [5], enzymatic degradation [6], oxidation [7], and photochemical degradation [8]. Among these methods, adsorption technology provides a practical method for the in situ removal of pollutants from wastewater [9]. The objective of the present work consists in the elimination of this antibiotic from water to elucidate its interactions with a biosorbent such as peanut shells in terms of solution pH, ionic strength and temperature and to investigate the adsorption kinetics, isotherms, and thermodynamic properties involved in the surface reactions.

* *Corresponding author*. Tel./fax: +213 818880.
E-mail address: belaibfouzia@yahoo.fr (F. Belaib).

0360-3199/$ – see front matter
http://dx.doi.org/10.1016/j.ijhydene.2013.05.015

2. Materials and methods

2.1. Adsorbent

The used peanut shells are agricultural by-products, which were well washed and dried in an oven brand Binder at a temperature of 105 °C for 48 h then ground-by a mortar to obtain fine heterogeneous particles which were passed through an electrical brand RETSCH sieve in order to obtain particles of homogeneous sizes, characterized by a mean diameter varying between 0.315 and 0.355 mm. The obtained solid mass was kept in a desiccator ready for use.

2.2. Adsorbate

OTC of 99% purity was provided by the pharmaceutical Saidal group and was used without further purification. The stock solution of OTC (approximately 1000 mg/L) was prepared by dissolving OTC in bi-distilled water and further diluted to the concentrations required for the experiments Fig. 1.

The chemical formula of OTC is $C_{22}H_{25}ClN_2O9$ and its molecular weight is 496.9 g.

2.3. Synthesis of coated peanut shells

The used solid support (peanut shells coated with the polyaniline) was obtained by the in situ polymerization of aniline. The used peanut shells shape and size varied between ($0.315 \leq d < 0.355$ mm). Ammonium persulfate $(NH4)_2S2O_{8,}$, hydrochloric acid HCl and then aniline were added and the reaction mixture was stirred for 2 h, giving green colloidal particles that were filtered and dried. Green balls were obtained ultimately, ready for use [10].

3. Adsorption procedure

The effects of time, solution pH, temperature, and ionic strength were investigated in batch adsorption experiments. 30 mg was weighed into 50-mL conical flask and suspended into 30 mL of OTC solution of different initial concentration. The initial solution pH (2.0–12.0) was adjusted to the required value by adding concentrated HNO_3 or NaOH solutions. The flasks were sealed and placed in temperature-controlled water bath oscillator at an agitation speed of 600 rpm until equilibrium was reached. OTC Solution samples were used to determine adsorption kinetics. Finally, the supernatant liquids were filtered using filter paper for analysis by a UV–vis spectrophotometer (SHIMADZU UV-160A) at the maximum absorption wavelength of 275.8 nm. To control the experiments, the OTC solutions without adsorbents were prepared under the same condition.

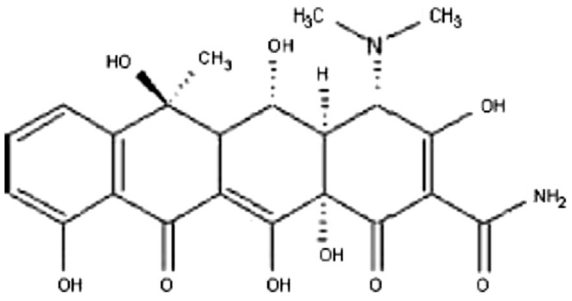

Fig. 1 – Structure of OTC.

3.1. Adsorption kinetic study

An adsorbent mass of 30 mg was mixed with 30 ml of OTC solution at a fixed initial concentration, at 20 °C, in conical flasks. Sorption experiments were carried out in batchwise and samples from the solution were withdrawn at given time intervals and separated from the sorbent by filtration. The amount of OTC adsorbed was obtained from a mass balance before and after sorption. To ensure reproducibility of the results, the experiments were repeated twice at an initial pH and OTC concentration of 6 10 mg/l, respectively.

3.2. Equilibrium studies

Adsorption experiments were carried out by adding a fixed amount of adsorbent (0.3 g) to a series of conical flasks filled with 30 ml diluted OTC solutions. The experiments were carried out at initial pH solution of 6. Removal efficiency R of OTC on peanut shells coated with polyaniline and sorption capacity (q) were calculated from the following Eqns. (1) and (2):

$$R(\%) = 100 \times (Co - C)/Co \quad (1)$$

$$q = V(Co - C)/m \quad (2)$$

with Co and C the initial and final OTC concentrations (mg/l) in aqueous solutions, respectively, V is the volume of the solution and m the mass of adsorbent (g).

4. Results and discussion

4.1. Effect of contact time on adsorption

The effect of initial contact time on Peanut shells coated with polyaniline adsorption is shown in Fig. 2 which indicates that the sorption increased for increasing contact time and remained constant.

Equilibrium was reached after about 15 min of contact time and the curves show that the adsorption occurred in two stages: a first rapid step followed by a much slower one leading to a high OTC adsorption percentage of 79.11%.

4.2. Effect of initial concentration

The effect of initial OTC concentration on adsorption onto peanut shells coated with polyaniline (PS PANI) in the range of (5–80 mg/l) was investigated. The curves corresponding to concentrations of 5, 10, 20, 40, 60 and 80 are clearly far apart, implying that they are free sites on the surface of the adsorbent, with a high driving force for migration of OTC. These results are consistent with that of Ref. [11] and confirm that there will be more molecules that will diffuse to the surface