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Development of new phosphated cellulose for application as an efficient biomaterial for the incorporation/release of amitriptyline

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ABSTRACT

In the last years has increased the study about the using of natural biopolymers and theirs derivatives in the removal (adsorption/incorporation) of contaminats of medium aqueous, and theirs utilization in the desorption (release) de drugs. However, there not in the literature studies about the utilization of the cellulose and cellulose phosphate in the adsorption (incorporation)/desorption (release) of the drug amitriptyline (AMI). Therefore, in this study was accomplished the synthesized of the phosphated cellulose (PC) through the reaction of pure cellulose (C) with sodium trimetaphosphate (P) under-reflux, for 4 h and at 393 K. The efficiency of the reaction was observed by XRD, TG/DTG, ^{31}P NMR and EDS. The adsorption study for the AMI in aqueous medium was carried out by varying the time, pH, concentration, temperature and ionic strength. The results showed that the PC showed a greater adsorption capacity of AMI than pure cellulose, presenting an increase of about 102.72% in the adsorption capacity of the drug by cellulose after the phosphating reaction. In desorption of drug from the surface of biomaterials was performed by varying the pH and time, where it was observed that PC showed a maximum release of 40.98% ± 0.31% at pH 7.

1. Introduction

In recent years, it is growing a social concern in relation to mental health. According to the World Health Organization (WHO), mental disorder affects about 450 million people worldwide, and 20% are children and adolescents, what makes this disease a very important issue for today [1]. Due to these factors, many psychiatric medicines (anxiolytics, sedatives and antidepressants) are among the most prescribed medications in the world, thus increasing the release of these drugs in water bodies, where are found in concentrations of ng L^{-1} and µg L^{-1} [2,3].

The amitriptyline (AMI) (Fig. 1(a)) is a tricyclic antidepressant widely used in the treatment of anxiety and depression [3]. This drug is considered a threat to environmental stability, due to their persistence and biological activity [1]. The presence of amitriptyline was detected in surface waters in the United Kingdom at concentrations from 0.5 to 21 ng L^{-1} and 1.5 ng L^{-1} in drinking water in France. Studies performed on fish embryos of *Danio Rerio* species observed the effects of amitriptyline exposure in the embryos at sublethal concentrations: 1, 10 and 100 ng L^{-1}. The studies showed that the drug significantly reduced the incubation time and the length of the embryos bodies after exposure to varying concentrations, and a coordinated modulation of physiological and biochemical parameters such as the level changes of adrenocorticotropic hormone (ACTH), oxidative stress and antioxidant parameters [4]. So many techniques for the removal of this contaminant has drawn the attention of researchers, such as membrane distillation [5] membrane bioreactors [6], advanced oxidation processes (UV radiation and Fenton's reagent) [7] adsorption [1–3] among others.

Among the various options mentioned for the drug removal, adsorption is a very effective method that can be used for this purpose. Adsorption may be defined as the enrichment of one or more components in the interfacial layer and may occur either as chemisorption and physisorption [8]. Different materials have been used as adsorbents applied in the amitriptyline removal in the aqueous medium, such as activated carbon [1], kaolinite [2] and montmorillonite [3]. Typically, adsorbents are chemically functionalized to increase its adsorptive capacity [8]. However, there are no reports in the literature using natural biopolymers and their derivatives as adsorbents for the removal of this drug.

Cellulose is the most abundant natural biopolymer in the world and therefore has attracted considerable attention due to its low cost. In addition, cellulose tends to have a high adsorption capacity

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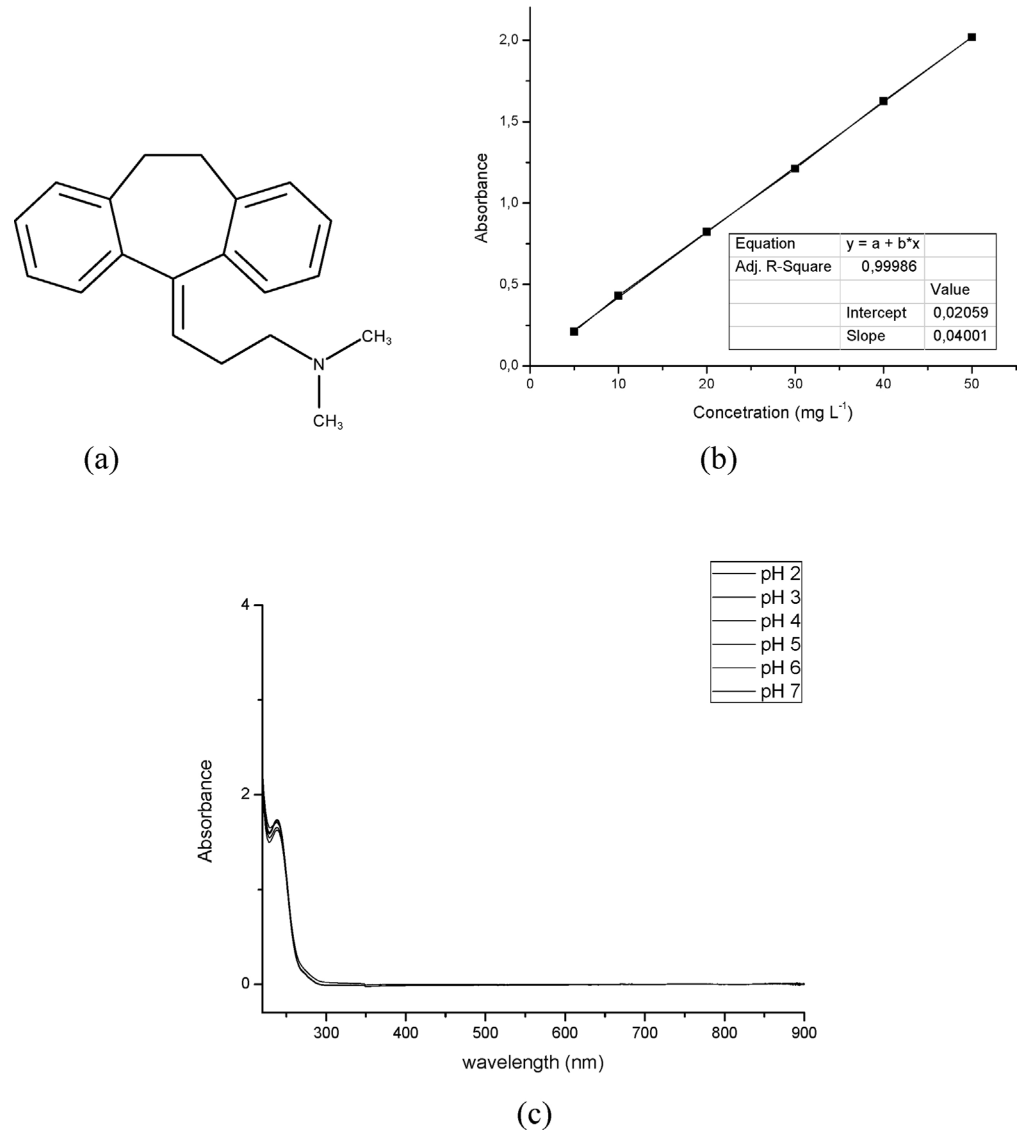

Fig. 1. (a) Structure of amitriptyline. (b) AMI calibration curve. (c) Scan of the AMI solution at pHs in study.

of pollutant species, especially after an appropriate chemical modification of its surface, in order to incorporate molecules into its structure. The new binding molecules on the surface of the solid material confer new properties, more advantageous, differing from those initial [9].

The increase in studies related to cellulose derivatives enabled the emergence of new biomaterials such as those containing phosphate. The chemical incorporation of phosphate in the cellulose structure changes its properties and, consequently, facilitates to the new synthesized material, the characteristics associated with immobilized group.

Adsorption of heavy metals (Fe^{3+}, Cu^{2+}, Mn^{2+}, Zn^{2+}, Co^{2+} and La^{3+}) and macromolecules (lysozyme, myoglobin, hemoglobin and albumin) are among the main applications of cellulose phosphate [10]. Regarding the application of pure cellulose and its phosphate derivative, in the adsorption of drugs, there are still few related researches. An example of this application is that researchers have shown that phosphorylation of the cellulose caused a significant increase in the absorption of the drug ranitidine compared to pure cellulose [10,11].

Like this, due the characteristics shown by cellulose biopolymer and, mainly, his phosphate derivate (cellulose phosphate) in relation the adsorption of heavy metals, macromolecules and drug, it's necessary one study about the adsorption (incorporation) and desorption (release) of the drug amitriptyline using as support the cellulose and cellulose phosphate, in view of that not there reports in the literature about this study.

This paper aims to synthesize the phosphate cellulose, obtained by reaction with sodium trimetaphosphate in reflux, characterized them by XRD, TG/DTG, ^{13}P NMR and EDS, and apply them in adsorption (ranging time, pH, concentration, temperature and ionic strength) and release (desorption) (varying pH and time) of the drug amitriptyline. Finally, the experimental data were adjusted to different kinetics, isotherms and thermodynamic physicochemical models.

2. Materials and methods

2.1. Materials

Pure microcrystalline cellulose (C) (Fagron), sodium trimetaphosphate (P) (Aldrich), Sodium hydroxide (Synth), hydrochloric acid (Synth), amitriptyline HCl (Pharma nostra Com-