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Comparison of Ceftriaxone, Ofloxacin, and Metformin Adsorption onto Activated Carbon from Aqueous Solutions

HANGANU D.[1*], FAVIER L.[2], LUTIC D.[3], HARJA M.[1*]

[1] "Gheorghe Asachi" Technical University of Iasi-Romania, Faculty of Chemical Engineering and Environmental Protection, 73 Prof. D. Mangeron Blvd., 700050, Iasi, Romania

[2] Univ Rennes, Ecole Nationale Supérieure de Chimie de Rennes, CNRS, ISCR-UMR 6226, F-35000 Rennes, France

[3]" Alexandru Ioan Cuza" University of Iasi, Faculty of Chemistry, Bvld. Carol I No 11, 700506, Iasi, Romania

*corresponding author:
e-mail: maria.harja@academic.tuiasi.ro:

Abstract Pharmaceutical compounds have been widely identified as organic pollutants in wastewater with adverse effects on the environment. This paper focuses on three active substances: ceftriaxone, ofloxacin, and metformin. The main reason for the interest is their ability to be excreted 80-90% unmetabolized, contributing to their high concentration in wastewater. One of the most frequently used removal methods is adsorption, while activated carbon is an etalon for adoptive materials. The batch experiments were performed with an initial concentration of 50 mg/L, and the concentrations of ceftriaxone, ofloxacin, and metformin were determined using spectrophotometric methods. Ceftriaxone and ofloxacin exhibited over 90% removal efficiency at neutral pH, while metformin required an acidic environment for the adsorption to take place. This study aims to compare the distinct way in which the substances respond within the adsorption onto activated carbon, including the influence of adsorbent mass, initial concentration and potential mechanism for every situation. The toxic effect generated of pharmaceutical compounds makes it primary to investigate the optimal parameters for their removal.

Keywords: Activated carbon, adsorption, ceftriaxone, pharmaceutical compounds

1. Introduction

The pharmaceutical industry has grown significantly in recent years, leading to increased drug consumption. Pharmaceutical compounds are increasingly detected in the environment through direct discharge or excretion. The active substances selected for this study were chosen because of their unique pharmacokinetic properties. According to the official patient information leaflet, ceftriaxone is excreted unmetabolized 50-60% in urine and 40-50% in bile (European Medicine Agency -EMA, 2015), ofloxacin is eliminated unmodified 75-80% through urine and 4-8% in fecal (EMA, 2023), in contrast for metformin no metabolites have been identified in humans (EMA, 2024). Their removal is crucial due to their toxic effects on ecosystems. The metformin has embryotoxic effects, and inhibits the mitochondrial complex I, inducing an oxidative stress mechanism. Metformin also induces alterations in the zebrafish embryos, craniofacial malformations and pericardial edema. Ceftriaxone inhibits the photosystem II by the nitro group substituent and can affect digestion in daphnia magna. Ceftriaxone can cause pigment formation and malfunctions in the cardiovascular system and the cranial nerve. Multiple studies have been conducted on the ecotoxicology of ofloxacin showing deterioration in Daphnia magna crustacean species causing effects on fertility, lipid peroxidation, and ulcerative erosion.

One of the most appealing approaches for removing active pharmaceutical compounds from aquatic solutions is adsorption due to its simple equipment and ease of implementation (Hanganu et al., 2024). Activated carbon (AC) is frequently used as an adsorbent because of its large surface area and porous structure. Kong et al. (2017) studied the efficacy of a luffa sponge-derived AC adsorbent focusing on ofloxacin, and achieved maximum adsorption of 132 mg/g. Wang et al., (2017) obtained an adsorption capacity of 134.9 mg/g. Alibrahim, (2023) studied ceftriaxone adsorption on tea waste-activated carbon, showing a high removal capacity (787.5 mg/g). This article aims to investigate the adsorption behavior of ofloxacin, ceftriaxone, and metformin on activated carbon powder and propose possible adsorption mechanisms.

2. Materials and method

All chemicals used in this study were procured from Sigma Aldrich. The concentration was measured using a spectrophotometric method, with Jasco V550 device: 233 nm for metformin, 287 nm for ofloxacin and 240 nm for ceftriaxone. The batch studies were performed by mixing 50 mL of a 50 mg/L solution with activated carbon powder. The mixture was shaken for one hour at 450 rpm speed and let in contact for 24 hours to achieve equilibrium. Adsorption isotherms were studied using initial concentrations of 10, 20, 40, 60, 80, and 100 mg/L for ceftriaxone, with 0.2 g/L activated carbon at pH 7 and 296 K. For ofloxacin, experiments were performed at initial concentrations of 10, 20, 30, 40, and 50 mg/L, with 0.1 g/L activated carbon under the same conditions as ceftriaxone.

3. Results and discussions

The adsorption rate of ceftriaxone, ofloxacin and metformin is presented in Fig. 1. Experimental results show that ofloxacin removal was slightly more effective than ceftriaxone. At 0.1 g/L AC 56.5 % of ceftriaxone was removed, while the removal rate for ofloxacin was 61.23%. Increasing the AC dose to 0.2 g/L, the rates reached 80.06%, respectively 98.69%. Ofloxacin, at pH neutral, exists in a zwitterionic form and its adsorption mechanism is based mostly on electrostatic interactions, hydrogen bonding, π–π EDA interactions, Van der Waals forces (Ashraf et al., 2023). The adsorption model was Langmuir for both antibiotics with R^2 values of 0.9835 for ceftriaxone and 0.9862 for ofloxacin, indicating monolayer adsorption. For metformin, adsorption at an initial concentration of 50 mg/L and pH 7 was negligible. Therefore, additional experiments were conducted at pH 5.5, where adsorption remained below 18%. The experiments were replicated with an initial concentration of 10 mg/L at pH 5.5 and 3.8 with a maximum removal rate of 13.8 mg/g. This can be explained by the structural dynamics of metformin at different pH values. Briones and Sarmah (2018) studied metformin's structural behavior on different values of pH, reporting pKa1 = 3.1, pKa 2= 13.8. At the pH 3.25, the fraction of metformin bi-protonated achieved 41%. Therefore, at pH 3.8 the fraction of bi-protonated metformin is increased compared to pH 5.5. Gaussian calculations were performed on the metformin model optimized using the DFT method at the B3LYP/3-21G level of theory to ensure a minimum on the potential energy surface, it can be observed that the difference in dipole moment for those two forms is not very big. The calculated dipole moments were 4.11 D for the +1 charge state and 4.42 D for the +2 charge state. Therefore, the mechanism of metformin absorption is influenced by a dipole-dipole interaction on a lower scale so the electrostatic interaction can have a bigger impact.

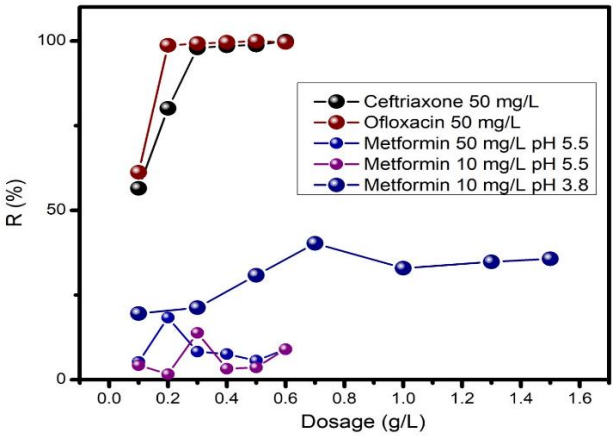

Figure 1. Effect of adsorbent mass for the removal of active substances on activated carbon

4. Conclusion

In this study, activated carbon was investigated as an adsorbent for ceftriaxone, ofloxacin, and metformin. The influence of adsorbent mass and initial concentration on the adsorption efficiency was analyzed. The results showed high adsorption capacities for both ofloxacin and ceftriaxone, while metformin removal requires further investigation to optimize the process. Ceftriaxone and ofloxacin higher removal efficiency (over 95%) at neutral pH, while metformin required an acidic environment. The removal of pharmaceutical compounds is essential to mitigate their toxic effects.

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