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ACTIVATED CARBON FIBERS FROM A NATURAL SOURCE AND THEIR USE FOR THE COMPETITIVE ADSORPTION OF PHARMACEUTICALS

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Introduction

Pharmaceutically active compounds are emerging contaminants that represent a real risk to the environment, as well as a potential risk to human health. Sulfamethoxazole (SMZ) and metronidazole (MNZ) are among the most prescribed antibiotics and are frequently detected in surface water ecosystems since its natural biodegradation is very slow (1). Unfortunately, traces of these drugs reach the watercourses due to the ineffectiveness of the effluent treatments. One aspect to take into consideration is the presence of different species in the same environment affecting each other adsorption. Therefore it is imperative to find new technologies that allow the removal of these contaminants before they reach the watercourses. In this context, the use of activated carbon fibers (ACFs) obtained from agroindustrial residues is an interesting approach as it involves a dual environmental impact towards a circular economy via recycling of residues for obtaining a high added value product with applications in water treatment (2). Taking this into account, the objective of this work is to study the performance of ACFs obtained from sheep wool residues in the competitive adsorption of SMZ and MNZ from solution.

Materials and Methods

Sheep Merino wool provided by S.U.L. (Uruguay) was used as precursor material, with 29 µm of diameter and already clean and spun. The material was stabilized by oxidation under air atmosphere at 300 °C, during two hours, carbonized in inert atmosphere up to 800 °C and kept at this temperature for 30 minutes. Activated carbon fibers were obtained by physical activation of the carbonized material with CO_2 at 950 °C during 30 minutes, and left cooling under nitrogen flow until room temperature. The entire process took place in a Carbolite oven. The chemical composition (C, H, N, and S) of the samples was measured by elemental analysis (LECO CHNS-932), and the oxygen content was determined by direct quantification (LECO-VTF-900). The ACFs were characterized by N_2 adsorption at -196 ºC (3Flex, Micromeritics) and the main textural parameters were determined (e.g., S_{BET}, micropore and total pore volumes). The adsorption studies of MNZ and SMZ were carried out at 22 °C, both in single component solutions and mixtures of varied concentrations (SMZ:MNZ molar ratio of 1:1, 2:1 and 1:2) to assess the competitive adsorption.

Results and Discussion

Through elemental analysis it was found that the main changes in composition occur during carbonization. The carbon content increased from 61 to 83 wt.%, at the expenses of nitrogen, oxygen and hydrogen reduction. The activation rendered an activated carbon fibre with a surface

area of ca. 955 m^2g^{-1}, a total pore volume of 0.414 cm^3g^{-1}, accounting for a large fraction of micropores (ca. 90 %). The kinetic studies for the adsorption of SMZ and MNZ showed that equilibrium was obtained after 48 hours in both cases. The parameters associated to Langmuir, Freundlich and Temkin models calculated from the fitting of the experimental equilibrium adsorption isotherms of single component solutions are presented in Table 1. Data was best fitted to a Langmuir model in both cases, pointing out to similar adsorption sites on the ACF.

Table 1. Adsorption parameters obtained from fitting experimental data to various models.

	Langmuir			Freundlich			Temkin		
	q_m (mmolg^{-1})	B (Lmmol^{-1})	R^2	K_F	1/n	R^2	K_T (Lmmol^{-1})	B	R^2
MNZ	0.63	25.5	0.995	1.054	0.28	0.981	1232	0.086	0.982
SMZ	0.55	603	0.998	0.136	0.38	0.873	10708	0.092	0.988

The competitive adsorption is presented in Figures 1, corresponding to experiments of a mixture 1:1 of both pollutants, for initial concentrations of the drugs ranging from 10 to 40 ppm. As seen, the uptake of each compound decreased slightly compared to single component solution. However, the decrease is more pronounced for SMZ than for MNZ, indicating the stronger affinity of the latter and its predominance in the competitive adsorption.

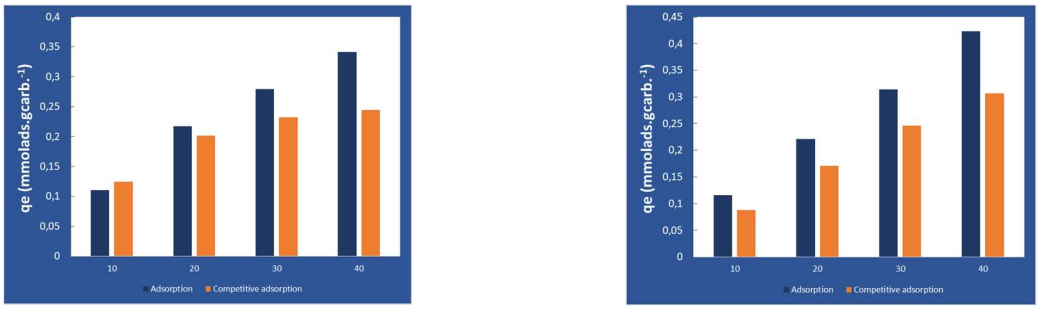

Figure 1. Adsorption capacities of MNZ (left) and SMZ (right) on the ACF from single component (blue bars) and 1:1 binary solutions (orange bars) of different initial concentrations of the pollutants.

Conclusions

Activated carbon fibers from wool showed good adsorption capacity and fast adsorption rates for the removal of the studied drugs (MNZ and SMZ), close to those reported for activated carbons with similar porous features. Co-adsorption studies showed the preferential adsorption of MNZ over SMZ, rendering slightly lower adsorption capacities for the latter, since they both compete for the adsorption sites in the ACF.

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