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Adsorption of Ciprofloxacin by Sugarcane Bagasse Activated Biochar in Aqueous Solution

UBIÑA L.J.B.[1,*], GUIA R.J.A.[1], AQUINO C.L.E.[1], SOMERA L.R.A.[1], BALELA M.D.L.[2], BELTRAN A.B.[3], CORNISTA J.C.[4], BAUTISTA-PATACSIL L.[1]

[1]Department of Engineering Science, University of the Philippines Los Baños, Los Baños, Philippines

[2]Department of Mining, Metallurgical, and Materials Engineering, University of the Philippines Diliman, Quezon City, Philippines

[3]Department of Chemical Engineering, De La Salle University, Manila, Philippines

[4]Department of Biology, University of the Philippines Manila, Manila, Philippines

*corresponding author:
e-mail: lbubina@up.edu.ph

Abstract Antibiotics such as ciprofloxacin (CIP) in wastewater pose a significant and pressing concern as an emerging contaminant. Conventional wastewater treatments are not optimized to effectively remove antibiotics, resulting in their persistence in bodies of water and significantly contributing to a global health threat, antimicrobial resistance (AMR). This study investigated the potential for removing CIP from aqueous solution through adsorption using phosphoric acid (H_3PO_4) activated biochar derived from sugarcane bagasse (SBAB). The removal percentage of CIP reached 45.37% at an equilibrium time of 60 minutes with a 0.8 g L^{-1} SBAB and an initial CIP concentration of 25 mg L^{-1}. The experimental equilibrium capacity, q_e is 14.45 mg g^{-1}. The adsorption kinetics followed the pseudo-second order (PSO) model, which primarily assumes chemisorption, with a q_e of 14.68 mg g^{-1}. The Freundlich isotherm best describe the adsorbent-adsorbate relationship, suggesting a a multilayer adsorption process. The SBAB will be used to produce 3D-printed adsorbents, which will be installed in a laboratory-scale packed-bed adsorption column.

Keywords: Emerging Contaminants of Concern, Ciprofloxacin, Antimicrobial Resistance, Activated Biochar, Adsorption

1. Introduction

Antimicrobial resistance (AMR) is a significant global health challenge, with microorganisms, such as bacteria, starting to develop resistance to medications like antibiotics, reducing treatment effectiveness and increasing disease risks. The overuse and misuse of antibiotics, combined with contamination from hospital and pharmaceutical wastewater effluents, as well as agricultural runoff, exacerbate this issue (Peñafiel et al., 2020). Conventional wastewater treatment plants (WWTPs) are inadequate for removing antibiotics, highlighting the need for improved treatment methods.

Ciprofloxacin (CIP), a widely used fluoroquinolone antibiotic, is commonly found in wastewater and surface water. While various treatment methods exist, including biological, chemical, and physical treatments, adsorption is particularly cost-effective and sustainable (Movasaghi et al., 2019). Different adsorbents, such as clay minerals, natural polymers, metal-organic frameworks, and activated carbons, have been studied. Activating biochar enhances its adsorptive properties by increasing surface area and pore size and introducing functional groups. Activation methods include chemical processes using acids or alkalis, as well as physical processes involving heating in the presence of oxidizing agents (Panwar & Pawar, 2020).

Biochar from agricultural waste, such as coconut shell, rice hull, and sugarcane bagasse, has been studied, with sugarcane bagasse showing promise due to its high adsorptive capacity over the other biomass.

2. Methods

A >98.0% (T)(HPLC) purity of ciprofloxacin (CIP) (with a chemical formula of $C_{17}H_{18}FN_3O_3$ and molecular weight of 331.25 mg $mmol^{-1}$) was used in the experiments. CIP was dissolved with a bare minimum amount of 0.1 M hydrochloric acid (HCl) and diluted with distilled water.

Sugarcane bagasse (SB) was collected at ProGreen Distillery in Balayan, Batangas. The SB samples were size-reduced, washed with distilled water, and oven-dried. The pyrolysis of the dried SB was performed in a tube furnace at 600 °C for 1 h, with a ramp-up temperature of 10 °C/min and continuous nitrogen gas purging to prevent oxidation. It was chemically activated by impregnating it with 85% phosphoric acid (H_3PO_4) at a 1:5 biochar-to-acid ratio. The mixture was oven-dried to remove moisture and was then subjected to heat treatment with parameters similar to pyrolysis, inducing thermochemical reactions to enhance the surface chemistry and structure of the biochar. The sugarcane bagasse activated biochar (SBAB) was washed with distilled water to remove excess H_3PO_4.

The CIP adsorption experiments using SBAB were conducted using a batch adsorption method. The effect of adsorbent dosage (0.2 to 1.0 g L^{-1}), initial adsorbate concentration (1 to 25 mg L^{-1}), and contact time (1 to 180 min) were investigated. The samples were filtered, and the concentrations were measured using a UV-Vis Spectrophotometer with a wavelength of 277 nm. The equilibrium adsorption capacity (q_e, mg g^{-1}) and percent removal of CIP were calculated and fitted in kinetic and isotherm models.

3. Results

The effect of activated adsorbent dosage on the removal of Ciprofloxacin (CIP) was evaluated using an initial concentration of 25 mg L^{-1}. In Figure 1, the optimal dosage of 0.8 g L^{-1} achieved a 40.60% removal. Increasing the adsorbent dosage enhances removal efficiency with more adsorption sites present and declined at 1 g L^{-1} dosage.

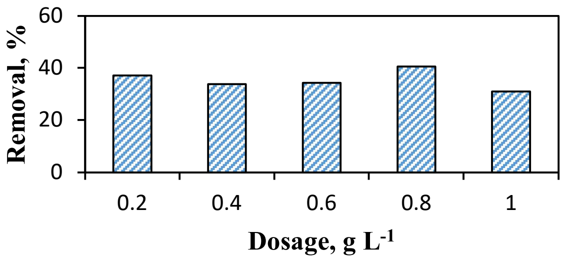

Figure 1. Removal (%) of CIP with varying dosage.

As shown in Figure 2, a rapid initial CIP uptake occurred within the first minutes and reached equilibrium at 60 minutes, at a 45.37% removal. Other studies have reported variable equilibrium times based on initial concentrations and the type of adsorbent material. The pseudo-second order model (PSO), with an R^2 of 0.99, provides a better fit to the data compared to the pseudo-first order model (PFO), with an R^2 of 0.055, indicating that chemisorption is the governing mechanism. Additionally, the PSO equilibrium capacity, q_e, is 14.68 mg g^{-1}, while the experimental q_e value is 14.58 mg g^{-1}.

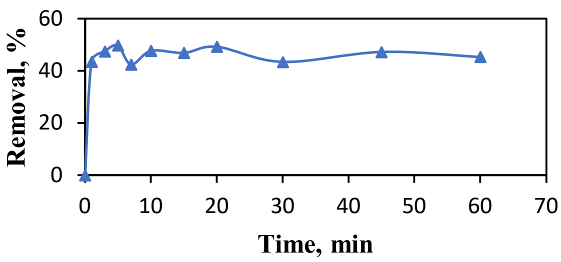

Figure 2. Removal (%) of CIP with 0.8 g L^{-1} dosage.

The effect of varying the initial concentration was also investigated with constant adsorbent dosage of 0.8 g L^{-1}. Shown in Figure 3, the adsorbate became saturated at 15 to 25 mg L^{-1} initial concentration reaching constant capacity. The adsorption isotherms were analyzed using Langmuir and Freundlich models, achieving high R^2 values of 0.95 and 0.98, respectively. The maximum adsorption capacity (q_{max}) for the Langmuir model was 16.34 mg L^{-1} while the Freundlich constant, K_F is 1.99. While Langmuir suggests monolayer adsorption on a homogeneous surface, Freundlich indicates multilayer adsorption on a heterogeneous surface.

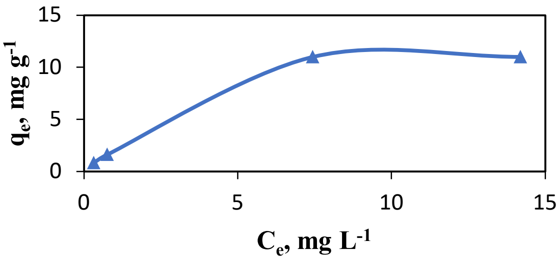

Figure 3. Equilibrium concentration vs equilibrium capacity at varying initial concentration of CIP.

4. Conclusion

The adsorption process follows the Pseudo-second order kinetic model while it follows the Freundlich isotherm models. The modeling results suggest that CIP's adsorption process using SBAB involves chemisorption, with multilayer adsorption. SBAB showed promising results in removing Ciprofloxacin in the simulated wastewater. Characterization, other operating conditions, and antibiotics should be investigated to validate further.

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