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Solvothermal growth of the bimetal organic framework (NiFe-MOF) on sugarcane bagasse hydrochar for the removal of dye and antibiotic

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ABSTRACT

In this study, the bimetal organic framework incorporated on the sugarcane bagasse hydrochar (AHC) surface (NiFe-MOF@AHC) composite was prepared using a solvothermal method. The preparation parameters (concentrations of $Ni(NO_3)_2$, $FeCl_3$, H_4BTeC, and reaction time) were optimized via the response surface methodology-central composite design (RSM-CCD) for the high adsorption removals of the large-sized pollutants (crystal violet (CV) dye and tetracycline (TC)). The optimized NiFe-MOF@AHC ($NiFe_{op}AHC$) was characterized with dual three-dimensional (3-D) structures of intertwined nanosheets porous networks and hexagonal spindle-shaped crystals with mesopore structures 2.3 times higher than the surface area of AHC, ample carboxylic and metal-carboxylate groups, and high thermal stability at a wide temperature range (0–800 °C). The $NiFe_{op}AHC$ composite adsorption removal of both CV dye and TC demonstrated a fast removal rate, primarily by chemisorption. The maximum adsorption removal (Q_{max}) of $NiFe_{op}AHC$ for CV dye (395.9 mg g^{-1}) was 1.5 times lower than that of AHC, but the removal of TC (568.1 mg g^{-1}) was 2.7 times higher than that of AHC. The hydrogen bonding, π-π or π-EDA interactions, surface complexation, and acid-base interactions might play roles as the dominant mechanism for the removal of the CV dye/ TC. The regeneration studies showed that the removal of both CV dye and TC was reduced after the 1st cycle and maintained until the 4th cycle, demonstrating that the solvothermal growth of NiFe-MOF on AHC succeeded in producing a stable and recyclable adsorbent.

1. Introduction

Globally, water containing dyes and antibiotics caused adverse effects on human health and the environment. Dyes and antibiotics are generally low in biodegradability, containing large and complex structures that are difficult to be removed by conventional treatment methods, such as coagulation and precipitation [1,2]. In particular, the crystal violet (CV) dye and tetracycline (TC) are commonly detected in our environment. The CV dye is a triphenylmethane dye with brilliant color and is widely used in the textile, dyeing, leather manufacturing, printing, and food industries. It is carcinogenic, teratogenic, and mutagenic even at a very low concentration [3]. On the other hand, TC is one of the most widely used antibiotics worldwide to prevent human and animal infections [4]. However, TC is not completely absorbed by living bodies and is excreted through urine and feces. Its presence in water can increase microbial resistance of pathogenic bacteria, and hence it is important to treat water containing TC [5].

Meanwhile, adsorption technology is one of the most promising and cost-effective methods for the removal of a wide variety of pollutants, dyes and antibiotics included. In this respect, the hydrochar prepared by the hydrothermal carbonization method would be a good choice of adsorbent. Although hydrochar has a low surface area, it contains numerous functional groups on its surfaces [6]. Moreover, the surface area of hydrochar could be enhanced via chemical modifications to improve its efficiency for the removal of pollutants [7,8]. The iron-loaded sludge biochar [9], polyaminocarboxylated modified hydrochar [10], hybrid silicate-hydrochar composite [11], and TiO_2(B) nanosheets@hydrochar composites [12] are among the many modified hydrochars that have been developed for dye or antibiotic removals.

MOFs were known for their high surface area and porosity, which allowed them to adsorb dyes and antibiotics efficiently [13–17]. MOFs are metal ions or metal clusters containing an organic linker to form crystalline materials with periodic network structures [18]. In particular, the Fe-based and Ni-based MOFs have a large surface area, high

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