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Adsorption of dyes from aqueous solutions by microwave modified bamboo charcoal

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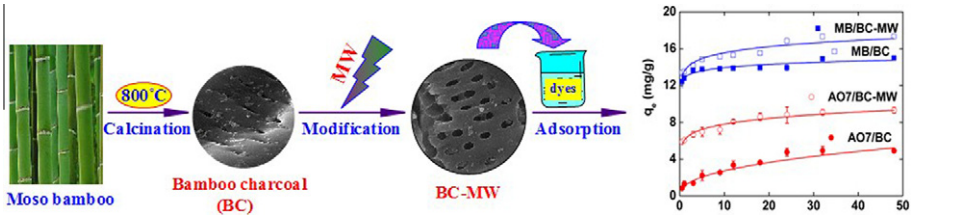
HIGHLIGHTS

- A novel cost-effective biosorbent was investigated for dyes adsorption.
- Microwave modified bamboo charcoal could be used as a potential adsorbent for dye removal.
- The mechanism regarding bamboo charcoal adsorption of dyes was explored.

GRAPHICAL ABSTRACT



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ABSTRACT

The performance and mechanism of bamboo charcoal (BC) and microwave modified BC (BC-MW) for the adsorption of methylene blue (MB) and acid orange 7 (AO7) from aqueous solutions were investigated. MB and AO7 adsorption was stronger on BC-MW than on BC due to the greater pore diameter, higher hydrophobicity as well as more surface charge. Kinetic results showed that the surface and intraparticle diffusion were the rate-controlling steps for the adsorption. All adsorption isotherms were highly nonlinear and fitted well by Freundlich and Dubinin–Radushkevich models. Hydrophobic, π–π and electrostatic interactions are mainly responsible for the adsorption of dyes, while the surface area and pore volume of BCs in conjunction with H-bond interaction had minute contribution. The adsorption was pH-dependent with more adsorption at pH below 5 for MB and below 3 for AO7, respectively. Higher ionic strength led to higher adsorption capacity. The thermodynamic analysis showed that the adsorption was spontaneous and endothermic. The distributions of the adsorption site energy on BCs were calculated to be heterogeneous. Furthermore, the fixed-bed columns packed with BC and BC-MW resulted in an excellent adsorption of both dyes.

1. Introduction

Environmental contamination by dyes has gained widespread public attention as a pervasive problem [1–3]. The removal of dyes from wastewaters is an important environmental issue. There are several strategies for the treatment of dye wastewater, such as advanced oxidation processes (AOPs) [4–6], biological treatment [7], ion exchange [8] and adsorption [9]. Adsorption has been proven to be an effective process for dye removal due to its low cost, high adsorption capacity and environmental friendliness [9,10]. Porous materials, particularly activated carbon (AC), have been commonly used as adsorbents for dye removal from wastewaters [9]. Unfortunately, AC has some shortcomings such as slow desorption kinetics and difficulties in regeneration, arising primarily from the closed, irregular-shaped, and highly porous micropores with wide pore size distributions [11]. Additionally, AC is mainly made from the non-renewable source of coal, which implies decreasing application in the future [12,13]. Therefore, it is of great importance to

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develop cost-effective and environmentally friendly adsorbents for dye removal.

Numerous novel adsorbents, such as natural minerals (e.g., clays, zeolites and other siliceous materials, etc.), waste materials (e.g., by-product from agriculture and industry, etc.) and bioadsorbents [14], have been developed for the treatment of dye effluents. Among these materials, bioadsorbents attract much interest because they can be produced from a variety of natural materials (i.e. chitosan, peat, biomass, etc.). In recent years, a new-style bioresource, bamboo charcoal (BC), has been recognized as a promising adsorbent because it is cost-effective, renewable and environmentally friendly [13,15]. BC can be produced from the widespread fast-growing speed and short growth period moso bamboo plants in China. The bamboo and bamboo residues can be transformed to BC at a high temperature under nitrogen atmosphere, which is a mature technology used in China. Currently, the area of bamboo forest is 4.84 million hm^2 with an average increase of 126,000 hm^2 per year in China, according to the statistics of the Sixth National Forest Resources Survey. Additionally, the cost of BC is much less compared with AC in China [16]. BC, as a newly emerged bioresource with porous structure and large surface area, has attracted extensive attention for their potential use in wastewater treatment. Several researchers revealed that BC could effectively adsorb many contaminants, such as antibiotics, dibenzothiophene, nitrate-nitrogen, N-vinylpyrrolidone and so on, in wastewaters [13,17–20]. Nonetheless, to our knowledge, the adsorption of dyes by BC has been seldom investigated, and the mechanism for the adsorption has not been adequately interpreted.

In order to increase the adsorption capacity of carbon-based adsorbents for dyes, attention has been paid on the structure and surface modification. Because most dyes have large-size and charges [21], a large pore size, high surface charge as well as hydrophobic characteristic are theoretically required for the adsorbents. Microwave (MW) is a part of electromagnetic spectrum occurring in the frequency range of 300 MHz to 300 GHz [5], with the properties of molecular-level heating, which leads to homogeneous and quick thermal reactions [22,23]. The advantages of MW radiation over other types of radiation, such as plasma irradiation, for the modification or regeneration of adsorbent are in terms of energy savings and better performance with ulterior adsorption capacity as well as the rate of adsorption [24,25]. Modification of adsorbents by MW radiation results in an increase in the average pore size and hydrophobicity [26]. Moreover, BC is a good MW absorber receiving MW energy directly through dipole rotation and ionic conduction [26]. Thus, it is possible to increase the adsorption of dyes on BC modified by MW radiation (BC-MW).

In the present study, the adsorption of dyes onto BC and BC-MW were investigated. Two commonly used dyes with opposite charge, methylene blue (MB) and acid orange 7 (AO7), were examined. The main objectives are (1) to identify the key process controlling the rate of dye adsorption by BCs, (2) to provide direct evidence regarding the relative importance of multiple mechanisms for the adsorption of dyes, and (3) to evaluate the performance for dye removal from wastewaters.

2. Materials and methods

2.1. Chemicals

MB (>99%) and AO7 (>99%) were purchased from Mecicine group chemical reagent Co. Ltd. and Shanghai cystal pure reagent Co. Ltd., respectively (see Table S1 in the Supplementary materials (SM) for their physicochemical properties). DI water (18.0 mΩ cm) obtained from a Millipore Milli-Q system was used. All the other reagents were above analytical grade.

2.2. BC production and modification

The BC was prepared according to the following procedure. Moso bamboo was firstly heated to evaporate water at 100–150 °C for 2–3 days, followed by pre-carbonization at 150–270 °C for 2–3 days. Then, it was carbonized at 270–450 °C for 1–2 days, calcined between 450 and 800 °C for 0.5–1 day, and finally kept at 800 °C for a short time. All the processes were carried out under nitrogen atmosphere.

The obtained massive BC was ground and sieved through a 100-mesh screen. After washed with DI water, the BC particles were dried at 105 °C. One portion of BC particles were modified by a household 2450MHz MW oven (MM72lAAU, China). The schematic diagram of the experimental apparatus is shown in Fig. S1 in the Supplementary materials (SM). A quartz crucible was installed on the bottom of MW oven, where the sample was exposed to MW irradiation. BC-MW was obtained by the radiation of BC at 550 W for 5 min, which was proved as an effective means of modification.

2.3. Batch experiments

The adsorption was carried out in a 30-mL serum bottle. For the adsorption kinetics, 20 mL of aqueous solution containing 50 mg/L MB and AO7, respectively, were mixed with 0.05 g of adsorbents. Preliminary experiments showed that adsorption on this dosage gave a straightforward comparison of adsorption data. The solution contained 5 mM $CaCl_2$ as background electrolyte and 200 mg/L of NaN_3 as biocides. The bottles were sealed and placed in a shaking table at 150 rpm in dark (293 ± 1 K). At regular time intervals, the bottles were taken out for dye analysis. With respect to adsorption isotherms, the equilibrium time was set at 48 h with different initial MB and AO7 concentrations at 5–100 mg/L and different temperatures at 293, 303, and 313 K. The initial pH was set at 7.0 and the variation was less than 0.2 during the whole process. Control experiments without BCs resulted in less than 5% loss of dyes. To investigate the effect of pH and ionic strength, the adsorption at fixed dye concentration of 50 mg/L were conducted at different pHs (1–12) and ionic strengths (0–1 M) at 293 K. The pH was adjusted by 1 M HNO_3 solution and 1 M NaOH, and the ionic strength was provided by the addition of $NaNO_3$. All the experiments were conducted in duplicate.

2.4. Column experiments

Dynamic adsorption experiments were carried out in a laboratory-scale column (Fig. S2). A glass column of 50 cm length and 20 mm internal diameter was filled with 12 g of BC and BC-MW, respectively. Experiments were carried out with an influent solution of 200 mg/L MB and 100 mg/L AO7, respetively, at 293 K and pH of 7.0. An upward flow was controlled at constant rate of 6.7 mL/min using a peristaltic pump. Samples of the effluent were collected and analyzed for dye concentration.

2.5. Analysis and characterization

After filtration through 0.45 μm membrane, the concentrations of MB and AO7 in the filtrate were measured at 663 and 484 nm, respectively, on a Cary 50 ultraviolet visible (UV) spectrophotometer (Varian, USA). For the characterization of BC and BC-MW, elemental analysis was performed using a Vario Micro cube elementar analyzer (Elementar Company, Germany). The Brunauer–Emmett–Teller (BET) surface area, pore volume and pore size distribution were determined using a Micromeritics ASAP 2020 nitrogen adsorption apparatus. The surface binding and elemental