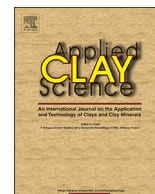




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Research paper

Adsorption of methylene blue from aqueous solution onto porous cellulose-derived carbon/montmorillonite nanocomposites

Dong Shen Tong^a, Cheng Wen Wu^a, Moses O. Adebajo^b, Gui Chen Jin^a, Wei Hua Yu^a, Sheng Fu Ji^c, Chun Hui Zhou^{a,d,*}

^a Research Group for Advanced Materials & Sustainable Catalysis (AMSC), State Key Laboratory Breeding Base of Green Chemistry Synthesis Technology, Discipline of Industrial Catalysis, College of Chemical Engineering, Zhejiang University of Technology, Hangzhou 310032, China

^b Queensland University of Technology, GPO Box 2434, Brisbane, QLD 4001, Australia

^c State Key Laboratory of Chemical Resource Engineering, Beijing University of Chemical Technology, Beijing 100029, China

^d Centre for Future Materials, University of Southern Queensland, Toowoomba, Queensland 4350, Australia

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ABSTRACT

Biomass-derived amorphous carbon has received considerable attention in recent decades. In this paper, carbon/montmorillonite (CMt) nanocomposites were synthesized by a hydrothermal carbonization of cellulose and montmorillonite (Mt), and the adsorption properties were evaluated by the removal of methylene blue (MB) from aqueous solutions. The CMt was characterized by X-ray diffraction, Fourier transform infrared spectroscopy, Nitrogen adsorption-desorption isotherm, Scanning electron microscopy, and Thermal analysis. The pseudo-first-order and pseudo-second-order kinetic models were used to describe the kinetic data and the Langmuir, Freundlich, Redlich-Peterson and Sips models were applied to describe the adsorption isotherms. The results showed that the equilibrium adsorption data was found to fit better to the Redlich-Peterson adsorption model, and the adsorption capacity for the removal of MB on CMt was $138.1 \text{ mg} \cdot \text{g}^{-1}$. The kinetic process of adsorption could be described by the pseudo-second-order model. The porous structure of CMt was responsible for the adsorption of MB. But the adsorption ability could be improved by increasing pH. Lastly, the adsorption mechanism was obtained.

1. Introduction

Dyes are used widely as coloring agents in many industries, such as textile, printing, leather, and paints, and so on (Tünay et al., 1996; Cassano et al., 2001; Yin et al., 2015). Dye effluents contain the trace amounts of organic dyes and toxic compounds. Nowadays, dyes in water are becoming one of the main sources of water pollution and represents a major environmental concern (Tong et al., 2010; Zhou et al., 2012; Smith et al., 2016). Methylene blue, a cationic dye, is one kind of the water-soluble dyes with high chromaticity, which is extensively used in chemical indicators, dyes, biological stains and drugs. It is quite visible and stable in water at room temperature (Deng et al., 2012; Liu et al., 2016b; Russo et al., 2016). To date, various methods and strategies have been used for the treatment of wastewater dyes, including adsorption, coagulation/flocculation, biodegradation, photodegradation, membrane filtration, chemical oxidation and electrochemical treatment (Qiu et al., 2013, 2014a, 2015a; Gadekar and Ahammed, 2016; Jia et al., 2016; Zeng et al., 2016). Among these

techniques, adsorption is regarded as a more suitable method for the removal of dyes due to its efficiency and economy. Many adsorbents have been studied, such as activated carbon, biological substances and polymer materials, and so on (Ghosh and Bhattacharyya, 2002; Hajjaji and El Arfaoui, 2009; Gil et al., 2011; Cao et al., 2016, 2017; Silva et al., 2016; Huang et al., 2017; Ma et al., 2017; Yu et al., 2018). Among them, activated carbon is widely used because of its high surface area, chemical inertness and high adsorption capacity (Amin, 2009; Subramani et al., 2016). However, due to its microporous structure ($< 2.0 \text{ nm}$), the high price and regeneration cost, activated carbon limits bulky dye molecules into the inner surface of the pores and is infeasible as the adsorbent in the future (Gil et al., 2011; Silva et al., 2016). Consequently, cheaper and more economic alternative adsorbents are still developing.

Clay minerals are frequently used as adsorbents to remove dyes, heavy metals, and surfactants from aqueous solutions owing to their efficiency and low-cost (Rida et al., 2013; Li et al., 2014; Gao et al., 2016). Montmorillonite (Mt) is one of the most abundant clay minerals

* Corresponding author at: Research Group for Advanced Materials & Sustainable Catalysis (AMSC), State Key Laboratory Breeding Base of Green Chemistry Synthesis Technology, Discipline of Industrial Catalysis, College of Chemical Engineering, Zhejiang University of Technology, Hangzhou 310032, China.

E-mail addresses: Chun.Zhou@usq.edu.au, clay@zjut.edu.cn (C.H. Zhou).

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with 2D sheet-like morphology. The montmorillonite sheets consist of three layer structural units, which are composed of a central $\text{AlO}_4(\text{OH})_2$ octahedral sheet and two SiO_4 tetrahedral sheets (Wu et al., 2016). Substitutions within the lattice structure of Al^{3+} for Si^{4+} in the tetrahedral sheet and of ions of lower valence, particularly Mg^{2+} , for Al^{3+} in the octahedral sheet result in a net negative charge on the clay surfaces. The charge imbalance is offset by the exchangeable cations such as Na^+ or Ca^{2+} in the interlayer. So, Mt has the property of adsorption of cations by ion-exchange (Zhang et al., 2010; Tong et al., 2014; Zhou et al., 2016). Furthermore, Al^{3+} and Mg^{2+} cations of the octahedral sheet can be leached after acid treatment and both surface area and pore diameter further increased. In recent, Mt has received increasing attention as low-cost adsorbents due to its outstanding properties of ion-exchange ability and high surface area. Several kinds of modified Mt have been synthesized and used for adsorption of toxic materials (Mu et al., 2015; Jalalia et al., 2016; Ngwabebhoh et al., 2016). However, unfortunately the process of ion exchange would introduce another cation ion in the system and lead to the secondary pollution.

It's well known that carbon materials have been widely used as catalysts (Song et al., 2017; Lin et al., 2018), functional composites (Cheng et al., 2017; Wang et al., 2017), sensing (Liu et al., 2016a,b; Liu et al., 2017), and so on. Recently, it was reported that carbonaceous materials with abundant oxygen-containing groups and hydrogen-containing functional groups anchored on the surface of solid adsorbents have emerged as a new class of adsorbents for removing contaminant from aqueous solutions (Zhou et al., 2012; Ai and Li, 2013; Zhang et al., 2015b). Cellulose is the most abundant non-food biomass resource produced via photosynthesis. Cellulose composed of glucose monomers linked by β -1,4 glycosidic bonds, can be used as a carbon former in the nanocomposite materials (Tong et al., 2013; Zhou et al., 2013; Qiu et al., 2014b, 2015b; Wu et al., 2014a,c; Góes et al., 2016; Mohammed et al., 2016). Therefore, if the carbonaceous adsorbent is directly prepared from cellulose, it will reduce the cost with the high surface area and encourage it to be used in more fields (Bakandritsos et al., 2004; Sun et al., 2017; Wu et al., 2017; Zhang et al., 2017a,b).

In this study, we used two cost-efficient and environmental friendly materials including Mt and cellulose to prepare carbon/montmorillonite (CMt) by hydrothermal process. The CMt was used as adsorbent to remove MB from aqueous solutions and CMt exhibited excellent performance by electrostatic and adsorption in porous materials. The Langmuir, Freundlich, Redlich-Peterson and Sips isotherm equations were used to fit the equilibrium isotherm. The adsorption properties were studied by changing the influencing parameters such as initial solution concentration, solution pH, and contact time. The adsorption rates were simulated by the pseudo-first-order and pseudo-second-order equations and also tested for validity of the equations. Finally, a mechanism of the adsorption was proposed.

2. Material and methods

2.1. Material

Cellulose and methylene blue (MB) were purchased from Sinopharm Chemical Reagent Co., Ltd., China and used without further purification. The M.W. of cellulose was 36,000 and the purity was over 99%. The montmorillonite was obtained from Zhejiang Changan Rengheng Science and Technology Co., Ltd., China. The cation exchange capacity (CEC) of the montmorillonite was 50 mmol/100 g.

2.2. Preparation of CMt

A typical synthesis process was performed as follows. In order to obtain the composite, acid activated Mt was firstly synthesized. In brief, 50.0 g of 20 wt% H_2SO_4 and 10.0 g of Mt were refluxed at 80 °C in a 250 mL three-necked round bottom flask equipped with a condenser, a

magnetic stirring bar and a thermometer for 4 h. Then the solid product was centrifuged, washed three times and dried at 120 °C. Finally, the product was designated as HMt.

50 mL of distilled water, 1.0 g of cellulose, and 6.0 g of HMt were sonicated for 10 min and then introduced into a Teflon-lined stainless steel autoclave (100 mL). The mixture was heated at 220 °C for 24 h. After the treatment, the catalyst and the unreacted cellulose were removed by filtration. The black product was centrifuged, washed with distilled water and ethanol several times, and then dried in an oven at 60 °C for 12 h. The final products were ground and kept for further use. The black product was named as CMt.

2.3. Material characterization

The X-ray diffraction (XRD) measurements were collected using a Thermo ARL SCINTAG X'TRA diffractometer between 2° and 80° (2 θ) with a scanning rate of 0.1°/s, employing Cu K α radiation ($\lambda = 1.54056 \text{ \AA}$).

The nature of surface species was determined by Fourier transform-infrared spectroscopy (FT-IR). The FT-IR spectra were recorded using a Nicolet AVATAR-370 spectrometer in KBr pellets. The samples were dried at 110 °C, mixed with KBr, and exposed to infrared light. The pellets were immediately measured after preparation under ambient conditions in the mid-infrared region. The spectra were the result of averaging of 32 scans at wavelengths ranging from 4000 to 400 cm^{-1} .

A nitrogen adsorption system (Micromeritics ASAP2010) was employed to record the adsorption-desorption isotherms at the liquid-nitrogen temperature of 77 K. Thermogravimetric analysis (TGA) of nanocomposites was carried out using thermogravimetric (TG) and derivative thermogravimetric (DTG) methods on a Mettler Toledo thermobalance. TG/DTG curves were recorded with a 10 °C/min heating rate under air atmosphere (65 $\text{mL}\cdot\text{min}^{-1}$) between 30 and 900 °C. Scanning electron microscopy (SEM) analysis was conducted using an Hitachi S-4700(II) electron microscope. For the SEM experiments, each sample was fixed on an aluminum stub and coated with gold.

2.4. Adsorption of methylene blue (MB)

The adsorption of MB in aqueous solution on the as-prepared CMt nanocomposites was performed in a batch experiment which was carried out in a thermostated shaker with a shaking speed of 150 rpm using 100 mL Erlenmeyer flasks. In a typical adsorption procedure, 0.05 g of CMt was added into 50 mL of MB solutions of varying initial concentrations (60–160 $\text{mg}\cdot\text{L}^{-1}$) under oscillation at room temperature and each set of the experiments was repeated three times. At predetermined time intervals, the samples were separated from the mixture by centrifugation with a speed of 4000 rpm for 4 min. The effect of pH on adsorption of MB on CMt was studied over a pH range of 1.8–10.5 with an initial concentration of 100 $\text{mg}\cdot\text{g}^{-1}$ for 240 min. The pH was adjusted by adding aqueous solutions of 0.1 $\text{mol}\cdot\text{L}^{-1}$ HCl or 0.1 $\text{mol}\cdot\text{L}^{-1}$ NaOH. The concentration of dye left in the supernatant solution was determined from the calibration curve prepared by measuring the absorbance of different predetermined concentrations of MB solutions at $\lambda_{\text{max}} = 664 \text{ nm}$ using a Shimadzu UV-2550 UV-vis spectrophotometer. The amount of MB adsorbed per unit mass of the adsorbent was evaluated by using the mass balance equation:

$$q = \frac{(C_0 - C_e)V}{m} \quad (1)$$

where q ($\text{mg}\cdot\text{g}^{-1}$) is the amount adsorbed per gram of adsorbent, C_0 and C_e are the initial and equilibrium concentrations of MB in the solution ($\text{mg}\cdot\text{L}^{-1}$), respectively, m is the mass of CMt (g), and V (L) is the initial volume of the MB solution.

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