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ORIGINAL ARTICLE

Applicability comparison of different models for ammonium ion adsorption by multi-walled carbon nanotube

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Abstract The aim of the present research was to remove ammonium ions (NH_4^+) from aqueous solution using multi-walled carbon nanotubes (MWCNTs). The adsorption behavior of ammonium ions (NH_4^+) from aqueous solution by multi-walled carbon nanotubes (MWCNTs) was investigated as a function of some parameters such as contact time, initial ammonium ion concentration, and temperature. Adsorption equilibrium data for the removal of ammonium ions were examined by fitting the experimental data to various models. Six isotherm models were used to describe the isotherm experiment data. Langmuir, Freundlich, and Halsey isotherm models provided much better isotherm data fitting than the other isotherm models (Temkin, D–R, and Harkins–Jura). The Langmuir equation was found to best represent the equilibrium data for ammonium ion-MWCNT system. The average relative errors (ARE) value demonstrated that the Langmuir provided the best model for these data.

1. Introduction

Excess aqueous ammonia could contribute to the eutrophication of lakes and rivers, depletion of dissolved oxygen and toxicity to fish and other aquatic organisms. With comparing traditional methods (biological nitrification/denitrification, chemical coagulation, selective ion exchange, air stripping, chemical precipitation, etc.) (Rostron et al., 2001; Jorgensen and Weatherley, 2003; Bae et al., 2002) to removing aqueous ammonia from wastewater, the adsorption method is more economically feasible and environmentally friendly. Most studies on ammonium adsorbed from aqueous solution have been focused on mineral materials as adsorbents, such as zeolites (Huang et al., 2010; Zhao et al., 2010; Yusof et al., 2010; Lei et al., 2008) and limestone (Hussain et al., 2006). Plant materials are a kind of abundant reproducible natural resource and a type of waste. Therefore, they were studied extensively as low-cost adsorbents to remove a variety of pollutants from aqueous solution, e.g. heavy metals (Majumdar et al., 2010), dyes (Ho et al., 2005), nitrate (Orlando et al., 2002), phosphate (Eberhardt and Min, 2008), phenol (Cherifia et al., 2009) and

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ammonium ions (Liu et al., 2010a,b; Karadag et al., 2008; Baker and Fraij, 2010; Mara~nón et al., 2006; Karadag et al., 2006; Wahab et al., 2010). However, no information is available on the removal of aqueous ammonia by carbon nanotubes as an adsorbent from aqueous solution.

Carbon nanotubes (CNTs) as new adsorbents have gained increasing attention of many researchers, since Long and Yang (2001) first reported that CNTs were more efficient for the removal of dioxins than activated carbon. According to the grapheme layer, CNTs can be classified into single-walled CNTs (SWCNTs) and multi-walled CNTs (MWCNTs) (Trojanowicz, 2006). Due to their large specific surface area, small size, and hollow and layered structures, CNTs have been proven to possess great potential as superior adsorbents for removing many kinds of organic and inorganic contaminants including 1,2-dichlorobenzene (Peng et al., 2003), trihalomethanes (Lu et al., 2005), microcystins (Yan et al., 2006), fluoride (Li et al., 2001), lead (Li et al., 2002), nickel (Chen and Wang, 2006) and arsenate (Peng et al., 2005). Recently, Wu (2007) has investigated the adsorption equilibrium, kinetics and thermodynamic parameters of CNTs for the removal of reactive dye. Although the adsorption capacity is increased using CNTs as adsorbents, it might suffer from the inconvenience of tedious centrifugation separation process. To overcome this problem, Fugetsu et al. (2004) have encapsulated MWCNTs in Ba^{2+}-alginate matrix to constitute a cage and used it as the adsorbent for the elimination of ionic dyes.

In this account applicability of carbon nanotubes (CNTs) as an adsorbent to remove ammonium ions from aqueous solution is investigated and reported. The aim of the present study was to evaluate the potential and effectiveness of CNTs for NH_4^+ the removal from aqueous solution. It also provides information about the effect of various parameters such as contact time (t) and initial ammonium concentration (C) on the adsorption process. The adsorption capacity of the adsorbent has been studied by using the adsorption isotherm technique. Various isotherm equations were used to assess the best isotherm equation which represents the experimental data adequately and satisfactorily.

2. Materials and methods

2.1. Raw materials

MWCNTs (Armchair (6,6), Young's modulus (0.94T TPa), tensile strength (GPa 126.2T), purity, >95; diameter 1–2 nm; length, 5–30 nm; surface area, ~400 m^2/g; and manufacturing method, catalytic chemical vapor deposition (CVD) were purchased from NanoAmor Nanostructured & Amorphous Materials, Inc., USA. Doubly distilled water was used and all adsorbents were washed before using. Fig. 1 presents SEM image of carbon nanotubes. Ammonium chloride salt (NH_4Cl) (molecular weight, 53.16 g/mol) was supplied by Merck, Germany (maximum purity available). Doubly distilled deionized water (HPLC grade 99.99% purity) was obtained from Sigma Aldrich Co. (Germany).

2.2. Adsorption studies

For the purpose of the study of the adsorption process, firstly a 1000 mg/L stock solution of ammonium was prepared by dissolving 3.819 g ammonium chloride (NH_4Cl) in deionized water and adding water to 1000 mL. The solutions to be used were prepared by diluting the stock solution with deionized water when necessary.

Adsorption thermodynamic and kinetic experiments were conducted by using a 100 mL glass flask containing 0.05 g of the adsorbent and 10 mL of the ammonium ion solution with the initial concentration, 140 mg/L. The glass flask was sealed with a glass stopper. These samples were then mounted onto an Ultrasonic Bath for 5–45 min at various temperatures (288,

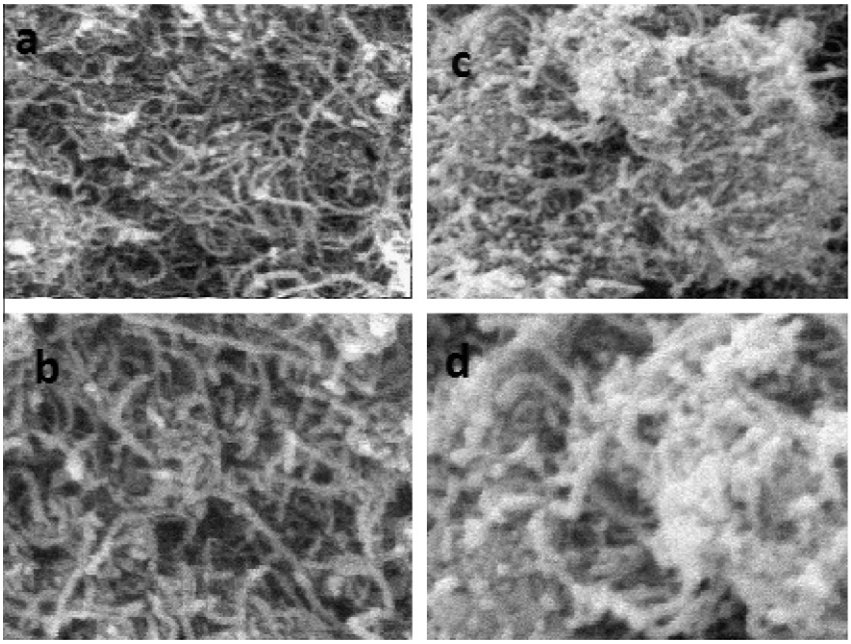

Figure 1 SEM images of carbon nanotubes (a) before adsorption 7500×, (b) before adsorption 15,000×, (c) after adsorption 7500×, and (d) after adsorption 15,000×.

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