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Enhanced adsorption of phenols from liquids by aluminum oxide/carbon nanotubes: Comprehensive study from synthesis to surface properties

Ihsanullah [a], Hamza A. Asmaly [a], Tawfik A. Saleh [b], Tahar Laoui [e], Vinod Kumar Gupta [c,f], Muataz Ali Atieh [d,*]

[a] Department of Chemical Engineering, King Fahd University of Petroleum & Minerals, Dhahran 31261, Saudi Arabia
[b] Chemistry Department, King Fahd University of Petroleum & Minerals, Dhahran 31261, Saudi Arabia
[c] Department of Chemistry, Indian Institute of Technology Roorkee, Roorkee 247667, India
[d] Qatar Environment and Energy Research Institute, Qatar Foundation, PO Box 5825, Doha, Qatar
[e] Department of Mechanical Engineering, King Fahd University of Petroleum & Minerals, Dhahran 31261, Saudi Arabia
[f] Center for Environment and Water, Research Institute, King Fahd University of Petroleum and Minerals, Dhahran 31261, Saudi Arabia

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ABSTRACT

In the present work, alumina/carbon nanotube nanocomposite was synthesized and characterized using field emission scanning electron microscopy (SEM), thermogravimetric analysis (TGA) and Brunauer–Emmett–Teller (BET) surface analyzer. The nanocomposite was evaluated for the 4-chlorophenol and phenol removal from aqueous solution with comparison to the pristine carbon nanotubes. Batch adsorption experiments were carried out to evaluate the effect of pH, agitation speed, contact time, adsorbent dosage, and initial concentration on the 4-chlorophenol and phenol removal efficiency. Experimental result showed the adsorption of 4-chlorophenol and phenol by nanocomposite to be pH dependent with the highest removal achieved at pH 6. CNTs showed better adsorption efficiency than CNT–Al_2O_3 which could be assigned to the increase in the surface area from 155.5 m^2/g of CNT to 227.5 m^2/g of CNT–Al_2O_3. Langmuir and Freundlich isotherm models were applied for analyzing adsorption equilibrium data of 4-chlorophenol removal on the as-prepared nanocomposite and pristine carbon nanotubes, which suggested that the Langmuir model provides better correlation of the experimental data with R^2 of 0.994 and 0.999 respectively. While for phenol, correlation coefficients of Langmuir adsorption isotherm model were 95% and 99.4% for CNT and CNT–Al_2O_3 respectively.

1. Introduction

Phenolic compounds have found applications in the production of wide varieties of phenolic resins, used in the construction of automobiles and appliances, adhesives and epoxy resins, as well as other various applications [1]. Phenolic compounds, generated in petrochemical and petroleum, coal conversion and phenol-producing industries, were identified as common contaminants in industrial wastewaters. They are highly toxic in nature and cause severe health effect on organisms, even at low concentrations, hence this aspect had attracted their classification as priority pollutants, and several forms of phenols have been classified as hazardous pollutants. It is classified by US Environmental Protection Agency (EPA) as toxic organic chemicals [2]. Chlorophenols are formed from the addition of chlorines, from one to five, to the original phenol molecule. This group of chemicals possesses weak acid characteristics and is capable of penetrating the human skin. Following human exposure, they pose acute toxicity due to ready absorption into the gastrointestinal system, with symptoms including vomiting, nausea and increased respiratory rate [3]. Chlorophenols in the environment have the potential to serve as precursor of dioxin, a characteristic which has attracted considerable interest. In addition, they produce noticeable effects in drinking water at concentrations as low as 0.1 mg/L [4].

Several techniques like electrochemical oxidation and sensors [5–13], sorption, chemical coagulation, solvent extraction, bioremediation, photo-catalytic degradation and adsorption were reported for the removal of noxious impurities from wastewater, with varying levels of success achieved and each technique was also characterized by its inherent limitations [14–19]. Adsorption is considered as the most promising technique for the removal of impurities from wastewater, even at low concentrations. Several previously developed adsorbents such as MWCNTs [20,21], nanoparticles and nanocomposites [22–25], rubber tire [26,27], and other low cost adsorbents [28–33] have been extensively used for instantaneous removal, maximum adsorption of dyes and other noxious impurities.

Carbon-based adsorbent materials, which are hydrophobic and nonpolar, have good potential for pollutant removal in wastewater. Their large surface area, porosity and functional groups are features enhancing their adsorption efficiency. In this study, carbon nanotubes (CNTs) and CNT–alumina nanocomposites were studied for 4-chlorophenol and phenol removal from aqueous solutions.

* Corresponding author.
E-mail address: motazali@kfupm.edu.sa (M.A. Atieh).

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2. Material and methods

2.1. Adsorbent synthesis

Carbon nanotubes (CNTs) used in this study were purchased from Nanostructured and Amorphous Materials, Inc., USA. The CNTs had 95% purity, outside diameter of 10–20 nm and length ranging from 1 to 10 μm. Aluminum oxide (Al_2O_3) from aluminum nitrate was impregnated onto 5 g of the CNT in ethanol (98% purity), followed by sonication (110 V at 40% amplitude) and calcination at 350 °C for 3 h.

2.2. Preparation of stock solution

Stock solutions of 4-chlorophenol and phenol with initial concentrations of 2 ppm were prepared by serial dilution of 1000 ppm solution made from dissolved 1000 mg of 4-chlorophenol or phenol in 1 L deionized water. The pH of the stock solution was adjusted using 1.0 M nitric acid and 1.0 M sodium hydroxide and buffer solutions were added to maintain constant pH during the experiments.

2.3. Batch adsorption experiment

Batch mode adsorption experiments were performed at room temperature to study the effects of pH, adsorbent dosage, contact time and agitation speed on the 4-chlorophenol and phenol adsorption efficiency of the CNT impregnated with aluminum oxide. Each experiment was conducted in a volumetric flask and the initial and final concentrations of 4-chlorophenol and phenol were obtained using UV–VIS spectrophotometer with a wavelength in the range of 200–350 nm. The adsorption capacity (q) was calculated using the following equations;

$$\%\,\text{removal} = \frac{C_i - C_e}{C_i} \times 100 \tag{1}$$

$$\text{Adsorption Capacity } q_e\ (\text{mg/g}) = \frac{C_i - C_e}{M_s} \times V \tag{2}$$

where C_i is the initial concentration of adsorbate ion in the solution (mg/L), C_e is the equilibrium concentration of adsorbate ion in solution (mg/L), V is the total volume of solution (L), and M_S is the carbon dosage (g).

2.4. Adsorption isotherm models

Adsorption isotherms are mathematical models that describe the distribution of the adsorbate between the liquid and adsorbent based on assumptions mainly related to homogeneity/heterogeneity of adsorbents, type of coverage, and possibility of interaction. The absence of interaction between adsorbate molecules is the main assumption of the Langmuir model, implying that adsorption is localized in a monolayer.

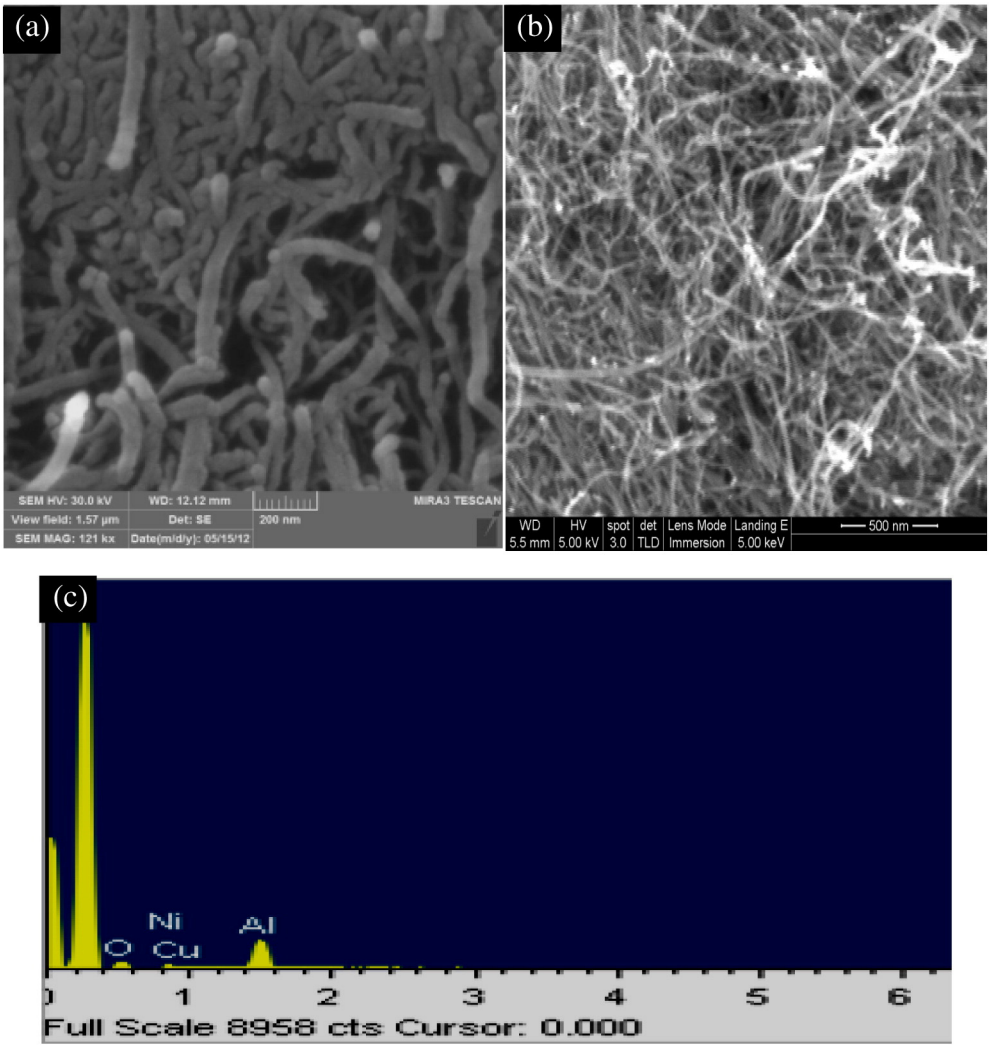

Fig. 1. SEM micrographs of raw CNTs (a) and CNT–Al_2O_3 (b); and EDX spectrum of CNT–Al_2O_3 (c).