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Adsorption of malachite green from aqueous solution by carboxylate group functionalized multi-walled carbon nanotubes: Determination of equilibrium and kinetics parameters

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ABSTRACT

MWCNTs functionalized with the carboxylate group are used as efficient adsorbent for the rapid removal and fast adsorption of malachite green from the aqueous solutions. The optimized contact time and pH were 10 min and 9, respectively. The effect of temperature revealed that the adsorption capacity of malachite green (MG) dye increased with increasing contact time, temperature and pH of the working solution. The adsorption equilibrium and kinetic data was well fitted and found to be in good agreement with the Langmuir isotherm model and pseudo second order kinetic model respectively with high correlation coefficient.
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Introduction

In the recent world, water refinement is considered as a large research area because of the deficiency in the reserve of fresh drinking water and that is at ubiquitous an important concern universally. The obviation of contaminants is becoming a principal environmental subject matter contain to their broad applications in industrial activities of many industrial plants, including textile companies, dye factory, food producers and electroplating factories, release wastewater containing dyes and thereby contaminate water resources [1]. Additionally, some dyes are proved to be noxious and carcinogenic agents to the environment. Textile dyeing effluents are composed of complex mixtures of dyes auxiliary chemicals, salts, acids, bases, organ chlorinated, compounds and occasionally heavy metals [2]. Noxious dyes cause severe detrimental and harmful effects on the human health it includes allergic reactions, dermatitis, skin irritation and also provoke carcinogenic and mutagenic reactions in biological creatures [3]. To eliminate, eradicate or to reduce the danger of contamination produced with as wastewater; this wastewater should be filtered before evacuated within the environment. Removal of dye is, howsoever, one of the important difficulties at the treatment of this wastewater [4]. For the removal of dyes 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 polluted aquatic source, but among all the adsorption was proved to be a most

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economical and efficient method for the removal of noxious dyes from aqueous solution, it has been extensively applied because it is a simple and cost effective technique and low cost adsorbents such as bottom ash and deoiled soya were used as adsorbents for the adsorption of basic fuchsine and azo dye acid orange. Activated carbon and activated rice husks were used for the adsorption of safranin-T from wastewater [14,15]. Several other previously developed adsorbents such as carbon nanotubes [16–23], MWCNTs [24,25], nanoparticles and nanocomposites [26–30], rubber tire [31,32], and other low cost adsorbents [33–38] etc. are extensively used for the rapid removal of noxious impurities from the aqueous solution. As regards, the dye removal process has the important difficulty of needing access of the terminating slush remainder [39]. Carbon nanotubes (CNTs) have attracted keen attention in multidisciplinary areas due to their unique hollow tube structure and their many outstanding mechanical, electronic and optical properties [40].

In comparison with classical adsorbents such as activated carbon and clay, CNTs is more attractive because of its favorable physicochemical stability, high selectivity, and structural diversity and in the present work we used the MWCNT-COOH as adsorbent for removal of malachite green from equates solution and several influential parameters were well studied and investigated.

Experimental

Materials

Malachite green (MG; $C_{23}H_{25}N_2$; 329.02 g/mol) used in this work was from the commercial manufacturing company Dy Star Co. (Germany) at 90% purity. The MG dye has one amide group (Fig. 1). The dye possesses positive charge even in highly acidic solutions due to the functional groups present in the dye [41]. The stock solution was prepared by dissolving the MG dye in de-ionized water to a concentration of 50.00 g/L. The working solutions of dyes were prepared by diluting the dye stock solutions to the demanded concentrations of MG dye. To regulate the pH of the dye solutions NaOH and HCl was used.

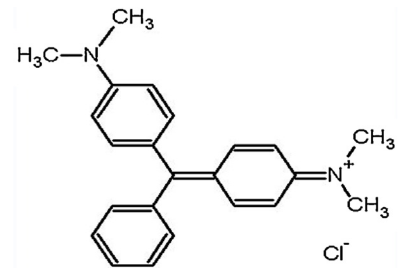

Fig. 1. Structure of malachite green (MG).

Adsorbent

The MWCNTs functionalized with carboxylate group (content of Carboxylate, 6 wt%; with purity >95%; average diameter 1–2 nm; length 530 nm and SSA ~400 m^2/g) was prepared by catalytic chemical vapor deposition (CVD). The developed adsorbent was further characterized using FTIR Spectrometer, (BOMEM, made in 85 Canada, 100 spectra accumulation, 2 m^{-1} resolution) Spectra were acquired in the 4000–400 cm^{-1} range (Fig. 2) and the surface textural and morphological properties was characterized using scanning electron microscopy (SEM) in a KYKY microscope, model EM 3200)Fig. 3).

Adsorption studies

In the present work, for the study of removal capacity of MG dye by MWCNT-COOH adsorbent, 20.0 mg of MWCNT-COOH was added into 20.0 mL of the MG dye solution (10.0–50.0 mg/L), samples were fretted at a appropriate time (2 min to 22 min in the kinetic experiments; and 10 min in the equilibrium isotherms) at different temperatures (298–338 K). The pH of the dye solutions ranged from 2.0 to 10.0. The MG dye and adsorbent samples were filtered through a 0.2 μm membrane filter and the suspensions containing of MG dye and MWCNT-COOH were centrifuged at 4500 rpm for 5 min using a centrifuge model; 5702R (Pendorf, Germany). The concentrations of remaining MG dye were analyzed using the UV–vis spectrophotometer furnished by Varian (Cary 100 Bio) (London—England) at maximum wavelength of 615 nm. Then, the amount of the MG dye removal by the MWCNT-COOH was determined by the difference between the initial and residual concentration of MG dye solution. The removal capacity of MG dye by MWCNT-COOH was calculated using the following equations, respectively [42]:

$$q = \left(\frac{C_0 - C_e}{w}\right) \times V \quad (1)$$

$$\%\text{Removal} = 100 \times \frac{(C_0 - C_e)}{C_0} \quad (2)$$

where q was the amount of MG dye taken up by the MWCNT-COOH (mg/g), C_0 was the initial MG dye concentration put in contact with the MWCNT-COOH (mg/L), C_e is the MG dye concentration (mg/L) after the adsorption procedure, w is MWCNT-COOH mass (g) and V is the volume of the MG dye solution (L). The Adsorptions time curve for removal of MG dye by MWCNT-COOH adsorbent shown in Fig. 4. To evaluate the fitness of kinetic and isotherm equations to the experimental data, the average relative error (ARE) is

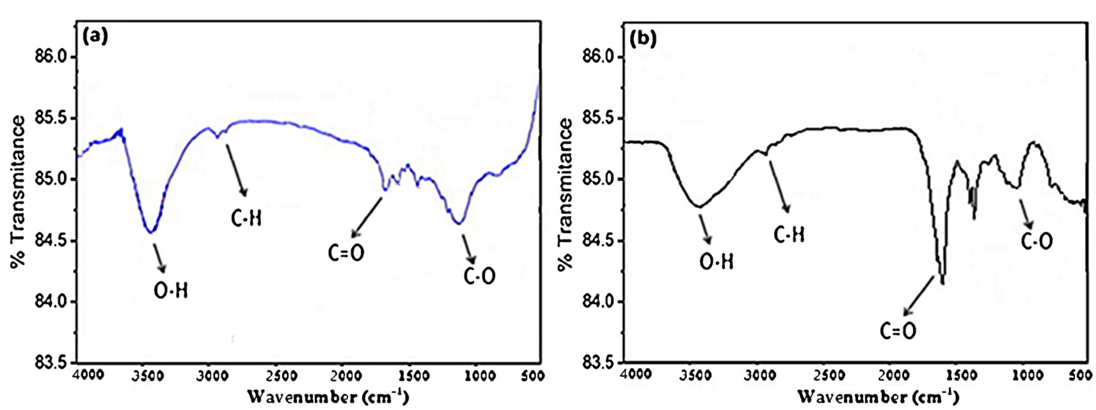

Fig. 2. Infrared spectra of adsorbent: (a) MWCNT-COOH; (b) MWCNT + MG.

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