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Research article

Effects of aspect ratio of multi-walled carbon nanotubes on coal washery waste water treatment

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ABSTRACT

The dependency of adsorption behaviour on the aspect ratio of multi-walled carbon nanotubes (MWCNTs) has been explored. In this study, effect of growth temperature on yield and aspect ratio of MWCNTs by catalytic chemical vapour deposition (CCVD) method is reported. The result revealed that yield and aspect ratio of synthesised MWCNTs strongly depend on the growth temperature during CCVD operation. The resulting MWCNTs were characterized by High Resolution Transmission Electron Microscope (HRTEM), Dynamic Light Scattering (DLS) and X-ray diffraction (XRD) techniques to determine it diameter, hydrodynamic diameter and crystallinity respectively. Aspect ratio and length of the grown MWCNTs were determined from the HRTEM images with the hydrodynamic diameter using the modified Navier-Stokes and Stokes-Einstein equations. The effect of the prepared MWCNTs dosage were investigated on the Turbidity, Iron (Fe) and Lead (Pb) removal efficiency of coal washery effluent. The MWCNTs with higher length (58.17 μm) and diameter (71 nm) tend to show high turbidity and Fe removal, while MWCNTs with lower length (38.87 μm) and diameter (45 nm) tend to show high removal of Pb. Hence, the growth temperature during CCVD operation shows a great effluence on the aspect ratio of MWCNTs which determines it area of applications.

1. Introduction

Water resources pollution has been an increasing global challenge for scientific community. Over the last few decades, this pollution by effluent of coal washery, hydrocarbons and heavy toxic metals as a result of human activities have been under serious investigation in a bid to protect and manage the environmental side effects tied to the prevalence of these pollutants. It is however not strange that continuous exposure to heavy metals with relatively high density and atomic weight such as iron, lead, chromium, cadmium and arsenic even at low level over time may resulted in bioaccumulation and resulting health consequences in humans ((WHO), 2011; Othman et al., 2017). Due to increase in demand for good drinking and industrial water has necessitated the enforcement of law in protecting the use and processing of water resources.

For the removal of heavy metals from polluted effluent, synthesis of high efficient adsorbent is of paramount importance. The discovery of carbon nanotubes (CNTs) in 1991 (Iijima, 1991) is an important stepping stone for the nano technological progress considering it wield range of applications in areas of water treatment, composites, energy storage and electronic device (Kariim et al., 2017; Shah and Tali, 2016; Su et al., 2016; Tasis et al., 2006; Yang et al., 2013). Unfortunately, the physicochemical properties and cost of production as hinders some of these applications. Basically, CNTs can be synthesised by arc discharge, laser ablation and chemical vapour deposition (CVD) method (Yang et al., 2016; Maria and Mieno, 2015; Voelskow et al., 2014; Hutchison et al., 2001; Tombros et al., 2008; Tsoufis et al., 2008). Among these methods, CVD appears to be the most promising method for industrial scale production of CNTs due to its simplicity, local cost and good morphology properties (Aliyu et al., 2017; Hayder et al., 2017; Iijima and Ichihashi, 1993).

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In the CVD method, hydrocarbons are decomposed on a metal catalyst at a higher temperature of ≥500 °C to ≤1000 °C for a certain period of time in a control system. During this process, the precipitation of carbon on the catalyst particles resulted in the formation of CNTs. Reaction temperature, hydrocarbon flow rate, reaction time, catalyst particle size and carrier gas flow rate are the crucial parameters which affect the morphological structure and also determine the type of CNTs produced (Hayder et al., 2017; Tsoufis et al., 2008). These CNTs produced can be either single–walled (SWCNTs) or multi-walled (MWCNTs) as the case may be (Shah and Tali, 2016; Iijima and Ichihashi, 1993; Iijima, 1991). MWCNTs have been used in a great number of applications and its effectiveness depends on their structural parameters such as diameter, number of walls, length and aspect ratio (Abdulrahaman et al., 2017; Iijima and Ichihashi, 1993). These aspect ratios pose the major property of MWCNTs which subsequently determine or limit their area of application (Abdulrahaman et al., 2017).

In the present study, the effect of growth temperature on the diameter, length and aspect ratio of MWCNTs produced with Fe-Ni catalyst on kaolin support by CVD method was studied. The diameter and hydrodynamic diameter were investigated using HRTEM and Dynamic Light Scattering technique, while the length and aspect ratio of the MWCNTs were determined by relating the diameter from the HRTEM images with the hydrodynamic diameter from Dynamic Light Scattering using the modified Navier-Stokes and Stokes-Einstein equations. Based on our knowledge, there are limited or no literature reports on the effect of aspect ratio of MWCNTs and it application in coal washery waste water treatment. In view of this, the effect of dosage of MWCNTs at different aspect ratio were investigated on the Turbidity, Iron (Fe) and Lead (Pb) removal efficiency from the coal washery effluent in order to improve the quality of water supply and protect the environment.

2. Materials and methods

2.1. Materials

The acetylene and Argon gases were sourced from BOC Nigeria and they were of analytical grade with percentage purity of 99.99%. Also, the effluent used was obtained from coal washery pond of coal mine at Akwuke, Enugu State, Nigeria.

2.2. Methods

2.2.1. Production of MWCNTs

The effect of synthetic temperature on aspect ratio of MWCNTs growth was performed in a CVD reactor consisting of a glass quartz tube, furnace and temperature controller. The catalyst used, which was thoroughly characterized in earlier studies (Aliyu et al., 2017), had a BET surface area of 3.76 m^2/g, pore volume of 1.9×10^{-3} cm^3/g and pore size of 0.5986 nm. The HRTEM of the catalyst also show lattice fringes of 0.3 nm with estimated catalyst particle size of 29–31 nm (Aliyu et al., 2017). About 1.0 g of the prepared calcined catalyst ($NiFe_2O_4$/kaolin) were weighed and spread in a quartz boat (11 cm × 2.6 cm) and placed at the centre of the glass quartz tube as shown in Fig. 1. The furnace was heated at 10 °C/min while argon (Ar) was flowing over the system at 30 mL/min to create an inert environment, removal of contaminants and prevent oxidation of the catalyst during the experiment. After the purging of the system from room temperature to 700 °C, the Ar flow rate was adjusted to 230 mL/min and C_2H_2 was then introduced at flow rate of 150 mL/min for reaction period of 45 min.

The growth experiment was stopped after 45 min by switching off the C_2H_2 gas, while the furnace was allowed to cool down to room temperature under a continuous flow of argon at a flow rate of 30 mL/min. The quartz tube was disconnected from the setup and the quartz boat was removed and the CNTs deposit was weighed.

The growth reactions were performed at the following temperatures: 750, 800, 850 and 900 °C. To ensured comparability, the experiments were conducted using the same mass of catalyst (1.0 g), constant flow rate of argon and acetylene. The investigation of the aspect ratio at different growth temperature was conducted after the characterization of the prepared MWCNTs using HRTEM and Dynamic Light Scattering technique to determine its morphology and hydrodynamic diameter respectively.

2.2.2. Characterization of MWCNT

The grown MWCNTs samples' surface morphology was examined using high resolution transmission electron microscope (Philips CM20FEG), while the hydrodynamic diameter were determined using Dynamic Light Scattering; Zetasizer (Nano S). For HRTEM studies, the samples were prepared by sonication of MWCNTs in isopropanol and few drops of the resultant suspension was put onto holey copper grid and dried for the analysis. Also, for the Dynamic Light Scattering technique, 1 mg of the samples were dispersed in 10 mL of an organic solvent (methanol) by means of a sonication (Misonix 3000; power: 1 W/mL, pulse on: 3 s, pulse off: 3 s, time: 1 min) and a centrifugation (5 min @ 3000 rpm). The supernatant was then transferred into a polystyrene quartz cuvette using a syringe with 0.22 μm filters and placed in the Zetasizer Nano S cuvette holder immediately for analysis. The crystal phase identification of MWCNTs was performed using Bruker AXS D8 X-ray diffractometer system coupled with Cu-Kα radiation of 40 kV and a current of 40 mA. The λ for Kα was 0.1541 nm, scanning rate was 1.5°/min, while a step width of 0.05° was used over the 2θ range value of 20–80°.

2.2.3. Coal washery effluent and treatment

The effluent samples from coal washery pond of coal mine at Akwuke, Enugu State, Nigeria were collected and stored in a tight container and it turbidity, Fe and Pb contents were pre-determined before treatment. Jar test analysis was carried out by adding 200, 400, 600, 800 and 1000 mg of the prepared MWCNTs on 1000 mL of coal washery waste water sample and it turbidity were determine using turbidity meter. The turbidity removal percentage of each of the prepared MWCNTs was calculated using Equation (1). While in another set of experiment, 1000 mL of the effluent were treated with 200, 400, 600, 800, and 1000 mg at 1000 rpm stirring speed for 45 min and allowed to settle and filter. The filtrates were analysed using the AAS machine to measure the Fe and Pb concentration.

$$\text{Turbidity Removal Efficiency } (\%) = \frac{N_O - N}{N_O} \times 100 \quad (1)$$

where N_O and N are the initial and final turbidity in NTU.

3. Results and discussion

The effect of the growth temperatures on the yield of MWCNTs produced was studied under the same reaction conditions: growth times (45 min); C_2H_2 flow rate (150 mL/min) and Ar (280 mL/min); and the results obtained were presented in Fig. 2. The result shows that as the reaction temperature increased from 700 °C to 750 °C, the mass quantities of MWCNTs produced increased after which