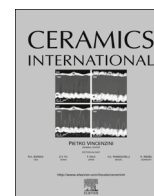




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Synthesis, processing, characterization, and applications of red mud/carbon nanotube composites



Saloumeh Mesgari Abbasi ^{a,*}, Alimorad Rashidi ^b, Azam Ghorbani ^c, Gholamreza Khalaj ^a

^a Department of Materials Engineering, Saveh Branch, Islamic Azad University, Saveh, Iran

^b Nanotechnology Research Center, Research Institute of Petroleum Industry (RIPI), Tehran, Iran

^c Department of Chemistry, Saveh Branch, Islamic Azad University, Saveh, Iran

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ABSTRACT

In the present study, red mud/carbon nanotube (RM/CNT) composites were synthesized by decomposition of hydrocarbon gas using the chemical vapor deposition (CVD) method. Red mud, a by-product of the Bayer process of alumina production, was used as a catalyst in this process. Synthesis was done at different growth temperatures (600, 700, 750 and 850 °C). The microstructure and morphology of the synthesized nanocomposite was characterized by transmission electron microscopy (TEM), field emission scanning electron microscopy (FESEM), energy-dispersive X-ray spectroscopy (EDX), X-ray diffraction (XRD), Brunauer–Emmett–Teller (BET), Simultaneous Thermal Analysis (STA), and Raman spectra analysis.

The prepared nanocomposite, which was prepared at 750 °C, was used for Pb (II) adsorption. The adsorption characteristics of the nanocomposite for Pb (II) removal were investigated as a function of pH, adsorbent dose, and contact time. The best result was obtained with 0.05 g of adsorbent at a pH=5 with 40 min of contact time. The results proved that RM/CNT composites are a good adsorbent for lead ions in comparison with unprocessed RM or pristine CNTs.

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1. Introduction

Red mud is a solid waste residue formed after the caustic digestion of bauxite ores during the production of alumina [1]. Red mud is a highly alkaline waste material with a pH of 10–13, because of the sodium hydroxide solution used in the refining process. Red mud is primarily composed of fine particles containing aluminum, iron, silicon, titanium oxides, and hydroxides [1]. Each year, about 90 million tons of red mud is produced globally. For every ton of alumina produced, the process can leave behind a third of a ton to more than two tons of red mud [1,2]. Because of its alkaline nature, and the chemical and mineralogical species present in red mud, this solid waste causes a significant impact on the environment, and proper disposal of waste red mud presents a huge challenge where alumina industries are installed [1,2].

Many attempts have been made in past years to find some practical applications for red mud. Some of the successful applications have included construction materials and ceramics [3], surface treatment for carbon steel [4], and a low-cost adsorbent for

removal of pollutants from aqueous solutions or the gas phase [5,6]. In addition, red mud can also be employed as a catalyst for hydrogenation, hydro dechlorination, and hydrocarbon oxidation [7,8]. Of these processes, the most promising efficient technique has been identified as adsorption with a suitable adsorbent.

Carbon nanotubes (CNTs) are unique and versatile adsorbents because of the availability of extensive surface area, its micro porous structure, and high adsorption capacity; but the high cost restricts their use [9]. Hence, the production of carbon nanotubes from cheaper materials is a need, and has gained significant attention for waste water treatment in developing countries. Various researchers have developed carbon nanotubes from red mud by fluidized bed chemical vapor deposition (CVD) [10]. Red mud has thermally stable oxides, e.g., Al₂O₃, SiO₂, and TiO₂, which can play two important roles: to disperse the Fe phases for the CVD reaction, and to contribute to relating to the amphiphilic character of the product with their hydrophilic surfaces [11].

CVD is a relatively simple and economical technique for the synthesis of CNTs as compared with arc-discharge, and laser ablation methods [12]. It can be done at low temperatures and ambient pressures. This method offers higher purity, a larger yield, and better control of the growth parameters and structure. To date, CVD is considered a cost-effective method for the production of good-quality CNTs. It has the potential to scale up the

* Corresponding author.

E-mail addresses: sm.mesgari@iau-saveh.ac.ir, mesgari.s@gmail.com (S. Mesgari Abbasi).

production of CNTs to the commercial level [13].

In recent years, metal oxide nanoparticles supported on carbon nanotubes have been extensively studied and found to be excellent catalysts, as well as an effective adsorbent for the removal of heavy metal ions and hazardous organic chemicals from water [14].

Poinern et al. [15] synthesized a CNT-Ferrihydrite nanocomposite and reported that CNT-Ferrihydrite proved to be a good adsorbent for arsenic. Gupta et al. [16] synthesized alumina-coated multiwall CNTs. They showed that this material was found to be an effective adsorbent for the removal of lead ions from aqueous solutions. However, RM /CNT composites have not been previously investigated for the remediation of lead ions from water.

The objective of this study is to investigate the ability of RM/CNT composites to remove lead ions from contaminated water by adsorption. The potential of CNT/RM composites for lead removal was determined by carrying out batch adsorption studies. In this work, we report the synthesis of RM/CNT composites by catalytic decomposition of methane (CH_4), as a hydrocarbon gas, using the CVD method and red mud as a catalyst.

2. Materials and methods

The red mud waste was obtained from the Jajarm Alumina Plant (Iranian Alumina Co.). The red mud was washed with distilled water several times until it reached a neutral pH, and was then dried to a constant weight before use. Then, it was sieved through 250 mesh steel and divided for chemical and mineralogical analysis. RM/CNT composites were synthesized by the CVD method at different temperatures (600, 700, 750 and 850 °C). For each run, 10 g of as-prepared red mud (as a catalyst) was put into a quartz boat and then placed in a horizontal tubular quartz reactor under atmospheric pressure. Subsequently, a mixture of methane and a carrier gas ($\text{CH}_4/\text{H}_2:1/10$) was introduced into the quartz tube, which was maintained at the reaction temperature for 45 min before the furnace was cooled down to room temperature under N_2 protection. The total flow rate was 500 sccm, and from room temperature to the desired temperature, the heating rate was $10\text{ }^\circ\text{C min}^{-1}$.

2.1. Characterization methods

To identify the chemical composition of the red mud, X-ray fluorescence (XRF) was employed. The morphologies and particle sizes of the as-received and RM/CNT composites were characterized by transmission electron microscopy (TEM) (FEG, Philips CM200) at an accelerating voltage of 120 kV, and by field emission scanning electron microscopy (FESEM) (Hitachi, S-4160, Vacc 15 kV), with electron beam energy in the 10–30 kV range. Energy-dispersive X-ray spectroscopy (EDS) was used to identify the catalyst present in the methane-treated red mud using an INCA, from Oxford Instruments. X-ray diffraction (XRD) and Raman spectra analysis were used to confirm the catalyst responsible for CNT formation. Raman spectra analysis and XRD measurements were carried out with the Thermo Nicolet (Almega, USA), and the PAN analytical X'pert XRD System (Philips, Netherlands), with $\text{Cu } k_\alpha = 1.5406\text{ \AA}$, 40 kV, and 30 mA, respectively. The thermal stability of the RM/CNT composite was determined by STA 409 PC Luxx (Netzsch, Germany).

2.2. Adsorption experiments

The adsorption capacity of the RM/CNT composite was investigated. For this, a Pb solution was prepared by diluting the appropriate volume of a stock solution. The pH of the solutions was adjusted by adding 0.1 M HNO_3 or 0.1 M NaOH as required.

The adsorption studies were carried out by placing known volumes of the Pb solutions in contact with predetermined amounts of the adsorbents in tightly closed bottles.

The bottles were subjected to shaking for different periods of time by placing them in a Perth Scientific shaking water bath at 75 rpm and 25 °C. After shaking, the suspension was filtered twice using a 0.22 μm Millipore syringe filter unit; then the filtrate was analyzed for residual lead by flame atomic absorption spectroscopy.

3. Results and discussion

3.1. Characterization of red mud (RM)

The chemical composition of the red mud that was determined by the X-ray Fluorescence (XRF) analyzer is given in Table 1.

It can be seen from Table 1 that the primary chemical compositions of red mud are Fe_2O_3 , Al_2O_3 , SiO_2 , CaO, Na_2O , TiO_2 , K_2O , and MgO. Red mud, a by-product of the aluminum industry, consists of a mixture of metal oxides. The X-ray diffraction pattern (XRD) of the red mud is shown in Fig. 1.

Some of the combinations identified in the sample are hematite (Fe_2O_3), calcite (CaCO_3), sodium calcium silicon oxide ($\text{Na}_{1.8}\text{Ca}_{1.1}(\text{Si}_6\text{O}_{14})$), kaolinite ($\text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4$), katoite ($\text{Ca}_3\text{Al}_2(\text{SiO}_4)(\text{OH})_8$), anatase and rutile (TiO_2), and cancrinite ($\text{Na}_6\text{Ca}_2\text{Al}_6\text{Si}_6\text{O}_{24}(\text{CO}_3)_2 \cdot 2\text{H}_2\text{O}$).

The particle size distribution of the red mud is shown in Fig. 2.

It can be seen that the red mud particles are mostly in the range of 2–80 μm with a mean value of 30 μm .

3.2. RM/CNT composite characterization

XRD analysis is an important tool used for clarifying the as-synthesized CNTs in the catalysts [13]. The XRD patterns of the RM/CNT composite synthesized at different temperatures are shown in Fig. 3. As can be seen in the pattern in Fig. 3(a), no diffraction peaks attributed to the CNTs were observed at 600 °C. In Fig. 3(a), the peaks at $2\theta = 52^\circ$ are relevant to iron (Fe), the diffraction peaks at $2\theta = 38^\circ$, 42° , and 50° are reflections of magnetite (Fe_3O_4) and the cancrinite [$\text{Na}_6\text{Ca}_2\text{Al}_6\text{Si}_6\text{O}_{24}(\text{CO}_3)_2 \cdot 2\text{H}_2\text{O}$] has many diffraction peaks at $2\theta = 32^\circ$, 38° , and 62° . There are no peaks characteristics of carbon.

It is obvious from the XRD pattern in Fig. 3(b) that the diffraction peaks at $2\theta = 30^\circ$ and 50° correspond to the (002) and (100) planes of the graphite, which is attributed to the graphitic structure of CNTs [12,13], while other peaks are attributed to the red mud support. For instance, in pattern in Fig. 3(b), the diffraction peaks at $2\theta = 28^\circ$, 33° , 36° , 41° , and 61° are related to gehlenite ($\text{Ca}_2\text{Al}_2\text{SiO}_7$), while the peaks at $2\theta = 52^\circ$ and 57° are relevant to cohenite (Fe_3C) and those at $2\theta = 38^\circ$ and 46° are related to tricalcium aluminate ($3\text{CaO} \cdot \text{Al}_2\text{O}_3$).

The XRD confirmed the conversion of hematite (Fe_2O_3) to magnetite (Fe_3O_4) after reaction with methane at 600 °C; after an increase in temperature to 750 °C, magnetite was converted to iron carbide (Fe_3C), and then carbon was formed from the decomposition of iron carbide [17]. Based on these data and the observed microstructure, a synthesized RM/CNT composite is possible at 750 °C. The EDS spectrum of the as-received RM and RM/CNT composite is shown in Fig. 4(a) and (b).

It can be seen that after the growth of the CNTs, it shows a distinct C peak, indicating the higher content of deposited carbon on this catalyst (Fig. 4(b)).

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