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Muscovite-MWCNT hybrid as a potential filler for layered silicate nanocomposite

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ABSTRACT

Hybridizing multiwalled carbon nanotube (MWCNT) with clay created a new class of functional materials especially for producing polymer layered silicate nanocomposite. The hybrid MWCNT/muscovite compound was produced using chemical vapor deposition (CVD) technique. It is rare to be found that the clay material such as muscovite to be used as a supporting material for MWCNT synthesis. X-ray Diffraction (XRD) and high resolution transmission electron microscope (HRTEM) were used to characterize the hybrid compound. XRD analysis revealed the existence of carbon element among several clay elements of muscovite clay confirming the deposition of CNT. Microstructural analysis using HRTEM further showed that MWCNT was successfully grown on the surface of muscovite particles.

1. Introduction

The world of hybrid compounds provides exciting challenges and opportunities for chemists, physicists, biologists, and material scientists. The use of hybrid compounds not only help to improve functionality of the conventional materials for building block of devices, but also offer interesting new characteristics and potentials of the newly developed materials in science and technological applications, together with cheaper and more environmentally friendly production route. Among hybrid compounds, carbon nanotubes (CNTs)–inorganic hybrids are new promising class of functional materials, which combine the multi-characteristic and functionality of organic–inorganic frameworks. For example, the works on CNTs–ceramics recently had shown exceptional performance in several applications such as improving the efficiency in photovoltaic (CNT–ZnO) [1], superior activities in photo catalysts (CNT–TiO_2) [2,3], increased sensitivity in gas sensors (CNT–SnO_2) [4], and enhanced capacities in super capacitors (CNT–MnO_2) [5]. There is a tendency of utilizing hybrid compound as filler in PMC. In polymer matrix composite (PMC) study, almost all single compound materials for composite reinforcement had been utilized such as CNT in epoxy [6,7], $CaCO_3$ in polypropylene [8], talc and kaolin in nylon [9], alumina in epoxy [10], mica in poly(dimethylsiloxane) [11] etc. For example, CNTs–ceramic such as CNTs–Al_2O_3 had been used as reinforcement instead of using either alumina or CNTs alone, which improved the mechanical properties of the composite [12,13]. As far as hybrid compound in PMC is concerned, most of the works reported were dealing with mineral fillers and in most cases, only physical blending was employed in achieving hybridization effect. To date, polymer layered silicate nanocomposites (PLSN) with hybrid compound had not been reported before especially when hybrid compound was concerned. It had been demonstrated that reinforcement effect of muscovite could be improved by applying ion exchange treatment [14]. CNT-muscovite hybrid could be regarded as a new approach to improve the reinforcement effect of muscovite in PMC thus giving enhancement to the composite performance. CNT-muscovite hybrid showed that the muscovite particles were physically covered by CNT network which modified the surface of flake-shape muscovite. The amount of CNT network that was attached to the surface of muscovite could be controlled during the synthesis process. CNT-muscovite hybrid supposedly possessed combination properties of muscovite and excellent properties of CNT. In long term, this new class of reinforcement material may have a great potential to further improve composite performance by using hybrid filler in place of single filler system. We have demonstrated previously that it was possible to grow CNT on inorganic particles in order to produce hybrid filler for polymer composite [15]. In this paper, we had used similar method to incorporate CNT on muscovite particles which later will be used to prepare CNT-muscovite nanocomposite. Through the same method, multiwalled carbon nanotubes (MWCNT)-muscovite had been successfully synthesized via chemical vapor deposition (CVD) method using nickel catalyst. The morphology of MWCNT-muscovite was being investigated.

2. Experiment

2.1. Materials and method

MWCNT was directly grown on muscovite particles by using nickel as the metal catalyst. The metal catalyst was attached on muscovite particles by precipitating nickel nitrate hexahydrate ($Ni(NO_3).H_2O$)

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and muscovite powder in distilled water with the presence of sodium hydroxide, NaOH. After 24 h, the precipitated catalyst paste was dried and calcined at 900 °C for 10 h. Then the catalyst underwent reduction process under the presence of hydrogen gas and followed by methane decomposition in a custom-made horizontal tube furnace similar to earlier reports from elsewhere [15], under the presence of methane and nitrogen atmosphere at 800 °C for 30 min. The CVD synthesized was then characterized accordingly.

2.2. Characterization of MWCNT-muscovite

MWCNT-muscovite hybrid was analyzed using X-ray Diffraction (XRD) method. The intensity was measured by step scanning in the 2θ range of 10°–80°. The morphology of MWCNT-muscovite hybrid was analyzed using Leo Supra-35VP Field Emission Scanning Electron Microscope (FESEM) and high Resolution Transmission Electron Microscope (HRTEM — Model Philip TECNAI 20 (200 kV)). Energy Dispersive X-ray (EDX) spectroscopy was used to analyzed the compositions of carbon (C) and muscovite ($KAl_2(AlSi_3O_{10})(OH)_2$) elements.

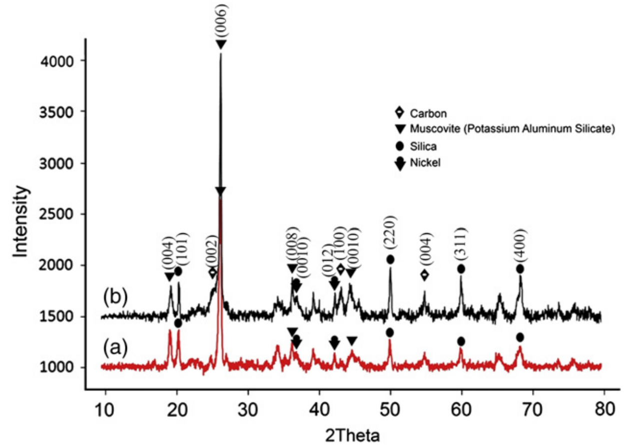
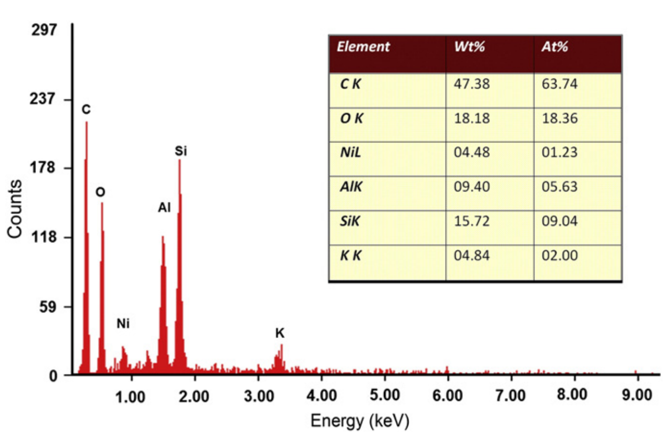


Fig. 2. XRD analysis of muscovite; (a) before methane decomposition, and (b) after methane decomposition.

3. Result and discussion

Multiwalled CNTs were grown onto muscovite particles via CVD process using a complex of nickel-muscovite as the catalyst precursor and methane gas as the carbon source. The aim of the work is to deposit MWCNT on muscovite particle in order to produce MWCNT-muscovite hybrid compound. The catalyst precursor was prepared with regards to the amount of metal catalyst, calcination time, calcination temperature, reduction time, and reduction temperature. The calcination time and calcination temperature were used based on previously reported works [15]. From the previously reported work, it was found that the optimum growth of MWCNT occurred at 900 °C calcination temperature. In a similar work it had also been reported that the calcination temperature lower than 900 °C caused poor attachment of the metal catalyst onto the alumina support which then led to failure of MWCNT growth during methane decomposition process. On the other hand, higher calcination temperature recorded an excessive interaction between metal catalyst and the alumina support which render the catalyst inactive. Another study of CNT synthesis had also reported similar observation when higher calcination temperature was used where catalyst particles strongly diffused into the support material lattices that hinder the accessibility of the active catalyst species to the reactants [16]. By using the same procedures, the MWCNT-muscovite hybrid compound was successfully synthesized. The yield of MWCNT-muscovite obtained at the end of the process was analyzed using EDX analysis and the result is depicted in Fig. 1. As can be seen in Fig. 1, based on weight percentage, 47.38% of carbon was recorded which was attributed to CNT deposition on muscovite. This analysis confirmed that the methane decomposition process had successfully performed as expected which could be described by equation $CH_4 \rightarrow 2H_2 + C$, where carbon was the highest element that could be detected using EDX analysis. In Fig. 2, the corresponding peak associated with carbon (2θ = 26.1°) which was believed to be CNT was highly visible. The presence of another peaks associated in XRD could be attributed to the nickel catalyst and potassium aluminum silicate from muscovite.



Element	Wt%	At%
C K	47.38	63.74
O K	18.18	18.36
NiL	04.48	01.23
AlK	09.40	05.63
SiK	15.72	09.04
K K	04.84	02.00

Fig. 1. EDX spectrum of MWCNT-muscovite hybrid yield.

Further investigation on the morphological aspect of the compound was done using FESEM. Fig. 3(a–d) shows the images of MWCNT-muscovite hybrid compound at different magnifications ranging from 5× to 50× respectively. It was clear that the MWCNT had been successfully deposited on the flake-shape muscovite which covered almost the entire surface area of the particle. It was interesting to note that the deposition of MWCNT was reasonably homogenous at this stage. The HRTEM images shown in Fig. 3(e–h) confirm that the morphology of the MWCNT-muscovite was in the form of wire-like hollow structures. The multiwall CNT was clearly seen as presented in Fig. 3(g–h). From these HRTEM images, a close observation on CNT structure using HRTEM revealed that there were some small particles on the each tip of MWCNT which was believed to be nickel particles. This observation seemed to be paralleled to several reports [15,17,18], where the small particles on the tips of CNT were the metal catalyst particles. In CVD reaction, the nickel particles acted as metal catalyst to grow the CNTs in a suitable condition. At elevated temperature (900 °C), the nickel particles which were attached to the surface of the muscovite particles were melted and methane started to decompose. By the reaction of catalyst particle, the carbon atom underwent rearrangement to form graphite tube and the growth mechanism is believed to be followed the tip growth of CNT. The nickel particles catalyzed the growth of the MWCNT until the desired reaction time was achieved. The HRTEM images also showed that the diameter of MWCNT was about the same size of the nickel catalyst particles, which meant that the catalyst size had a strong influence on the size of MWCNT. The development of this MWCNT-muscovite compound was to contribute in developing new filler/reinforcement for polymer composite applications.

4. Conclusion

The MWCNT-muscovite hybrid was successfully deposited using this approach. This work proved that rather than metal oxides, minerals were also capable to be used as supported material to metal catalyst in the synthesized CNT. Moreover, the synthesized MWCNT-muscovite could be used in polymer composite as filler/reinforcement to improve some properties. This hybrid particle can be used as