Journal of Alloys and Compounds 643 (2015) 74–78

Contents lists available at ScienceDirect

Journal of Alloys and Compounds

journal homepage: www.elsevier.com/locate/jalcom

A facile one-step method of coating aluminum on multiwall carbon nanotubes

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ARTICLE INFO

Article history:
Received 19 January 2015
Received in revised form 15 March 2015
Accepted 23 March 2015
Available online 25 April 2015

Keywords:
Nano-structures
Wettability
Thermal analysis
X-ray diffraction

ABSTRACT

Molten aluminum has very low tendency to wet carbon nanotubes (CNTs) due to the large difference of surface tension forces, which is the major issue in the liquid phase processing of Al-CNTs composite. Here in this study, a simple approach is presented to coat aluminum on CNTs using potassium hexafluorotitanate. During the process, titanium carbides were formed on the surface of the nanotubes providing wetting sites to coat aluminum in the liquid phase. These coated nanotubes could be used as a precursor, for not only liquid phase process but also for solid phase processing, to fabricate Al-CNTs composites.

1. Introduction

Aluminum based carbon nanotubes (Al-CNTs) composite is a promising material for many conventional and contemporary applications. However, wetting and dispersion of carbon nanotubes (CNTs) in molten aluminum (Al) is quite difficult owing to their large differences of surface tension forces i.e. 45 mN/m and 955 mN/m, respectively [1]. Many researchers addressed the issue by coating nickel on CNTs [2,3], which reduced the surface forces and improved the wetting and dispersion. However, their work mainly encompassed electroless plating approach, which in turn was a lengthy process with various processing steps.

Limited work is available for the coating of Al on CNTs. So et al. [1], coated Al on substrate supported vertically grown CNTs using electrolysis of aluminum chloride. There process involved two steps: in first step they decorated the nanotubes with aluminum nanoparticles using electroless coating technique and in second step they applied aluminum powder on aluminum plated nanotubes, followed by high temperature annealing to achieve complete wetting. In 2013, in the succession of their previous work, So et al. [4] used silicon carbide to increase wetting between CNTs and aluminum. This time they introduced a three-step process: in first step they crushed silicon powder with CNTs using mechanical milling, then they coated the silicon on the walls of nanotubes through high energy ball milling and finally silicon carbide was formed on the walls of the nanotubes by high temperature annealing (i.e. 1300 C). The Al-CNT composite prepared by using above mentioned CNT precursor demonstrated up to 15% increase in tensile strength.

The present work is a part of our project to fabricate Al-CNT composite, where we have coated CNTs with Al using potassium hexafluorotitanate (K_2TiF_6). In 2010, Baumli et al. [4] used multifunctional fluxes (alkali chloride-K_2TiF_6) to attain a perfect wettability of carbon particles by liquid Al. We have extended their work by using K_2TiF_6 (without alkali chloride) to coat Al on multiwall carbon nanotubes (MWCNTs) in a simple and facile way.

1.1. Hypothesis and approach

It is established so far that aluminum has partial or negligible wetting with CNTs, which is the primary factor of partial augmentation in the mechanical properties of Al-CNTs composite. As described in foregoing discussion that limited work has been reported on the wetting of aluminum with CNTs, however, Baumli et al. demonstrated improved wetting of aluminum with graphite [5,6] and carbon fibers [7] using multifunctional flux. They used a mixture of chloride flux and potassium hexafluorotitanate ($MCl + K_2TiF_6$) during melting of aluminum having graphite powder or carbon fiber at 750 °C. The multifunctional flux effectuated: dissolution of the oxide layer present on aluminum and formation of titanium carbide by an exchange reaction between titanium present in molten aluminum and carbon.

We took the inspiration from Baumli's work and developed the hypothesis that using potassium hexafluorotitanate during

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http://dx.doi.org/10.1016/j.jallcom.2015.03.262
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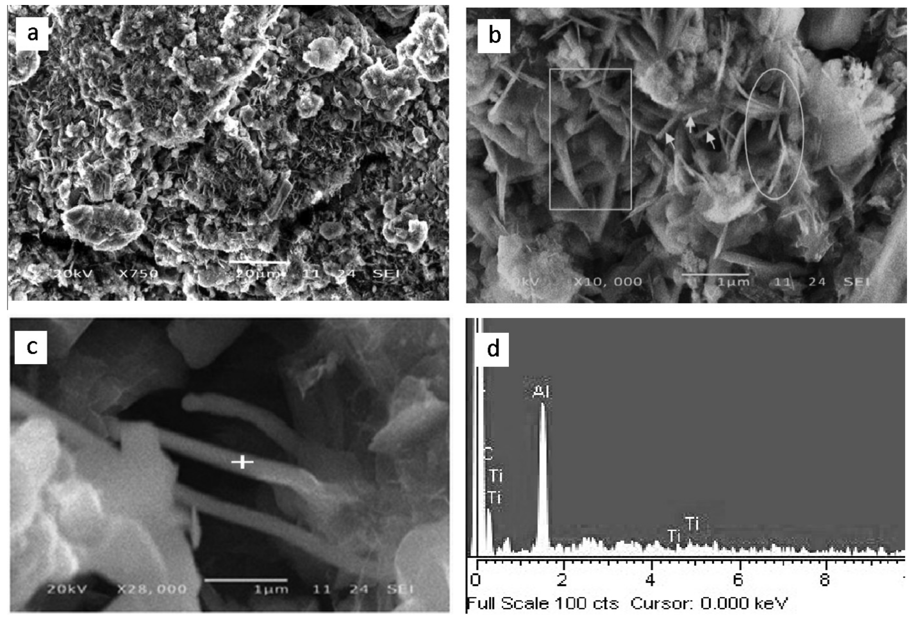

Fig. 1. (a) Bulk mass of the treated mixture showing agglomeration, (b) a higher magnification SEM micrograph showing Al coated nanotubes (arrows), needle like chemical phases (circle) and clusters of Al and the nanotubes (rectangle), (c) at some isolated regions, individual coated CNTs are also observable and (d) EDS spectrum of the marked region in (c), the coated CNT consisted of small amount of titanium along with aluminum and carbon.

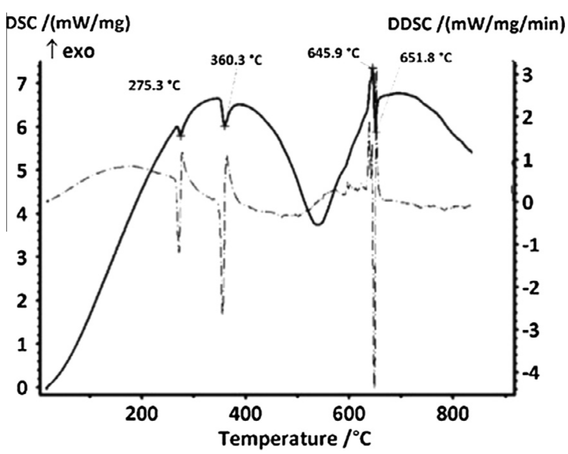

Fig. 2. DSC spectrum of the mixture along with differential curve, showing occurrence temperatures of various thermal changes.

fabrication of Al-CNT composite may increase the wettability of aluminum with CNTs due to the following assumptions:

- CNTs are basically rolled sheets of graphite. Therefore, the surface crystalline structure or precisely speaking the unit mesh on the surface of both the materials is comparable with graphite structure.
- Outer surface of the CNTs is always accompanied by certain amount of amorphous carbon, which may promote the direct formation of titanium carbides prior to the exchange reaction. Hence, it could be expected that titanium carbides may form at even lower temperatures than reported by Baumli (i.e. <750 °C).
- Additionally, surface imperfections (discontinuities, voids, contours, bends, etc.) of the nanotubes may provide easy nucleation/formation sites for titanium carbides.

To validate the above hypothesis, an experimental regime was planned, where substantial quantities of CNTs and potassium hexafluorotitanate were used along with aluminum.

2. Experimental

The nanotubes used for the present work were multi wall carbon nanotubes (MWCNTs), synthesized by chemical vapor deposition. The nominal diameter and length of the nanotubes was 10 nm and 1.5 μm, respectively. Detailed synthesis of the nanotubes is discussed elsewhere [8]. The flux used was potassium hexafluorotitanate (K_2TiF_6), having purity more than 98%, supplied by Sigma–Aldrich. The Al powder, having 125 μm particle size and more than 99.5% purity, was purchased from GoodFellow-UK. 1.3 g of MWCNTs and flux mixture (titanium to carbon ratio = 5) and 1.25 g of aluminum powder was placed in an alumina crucible and heated to 790 °C in an argon atmosphere furnace. A heating rate of 10 °C/min was used to achieve the temperature and the mixture, was heated for 60 min. Subsequently, the furnace was cooled to room temperature and the crucible was removed.

The treated mass (agglomerated powder) was subjected to scanning electron microscopic (SEM) using secondary electron imaging mode at 20 kV accelerating potential. The powder was slightly ground in mortar and pestle and sprinkled on a metallic stub.

To evaluate various reactions in the mixture at different temperatures, differential scanning calorimetric (DSC) studies were carried out. For the purpose, 10 mg of the mixture, having same composition, was heated to 850 °C in argon using a Netzsch STA 409C system. A heating rate of 10 °C/min and a constant flow of argon were maintained throughout the heating cycle. DSC scan exhibited four distinguishable peaks at various temperatures, therefore, the mixture was heated to those temperatures separately and treated masses were subjected to XRD for phase analyses.

To find the phases present in the mixtures, treated at various temperatures, a Siemens D-500 X-ray diffractometer (XRD) was used. A copper X-ray source with Cu Kα radiation having wavelength of 1.54173 Å was used. The diffractometer was operated at 40 kV and 30 mA tube potential and current, respectively, with a scan rate of 0.1° per minute and a step size of 0.02° 2θ.

3. Results and discussion

3.1. SEM

An SEM micrograph at low magnification, Fig. 1a, shows the bulk mass obtained after the treatment, where agglomerates can be seen. At relatively higher magnification (Fig. 1b), the agglomerates revealed into Al coated nanotubes (arrows), needle like chemical phases (circle) and clusters of Al and the nanotubes (rectangle). Fig. 1c shows the isolated coated nanotubes having around 100 nm diameter; the increase in diameter occurred due to thick Al coating. The nanotubes' surfaces were fully covered with Al and demonstrated appreciable wetting. An EDS spectrum is shown in