



On the influence of the processing conditions on the performance of electrically conductive carbon nanotube/polymer nanocomposites

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ARTICLE INFO

Article history:

Received 15 February 2008

Received in revised form 14 April 2008

Accepted 15 April 2008

Available online 24 April 2008

Keywords:

Nanocomposites

Conductivity

Processing conditions

ABSTRACT

We prepared multiwalled carbon nanotube/polystyrene (MWCNT/PS) nanocomposites using a latex-based process, the main step of which consists of directly mixing an aqueous suspension of exfoliated MWCNTs and a PS latex, both stabilized by an anionic surfactant. After freeze drying and compression molding homogeneous polymer films with well-dispersed carbon nanotubes were produced as evidenced by scanning electron microscopy. Conductivity measurements performed on our nanocomposite films show that they have a low percolation threshold and exhibit high levels of electrical conductivity above this threshold. We observe that both these properties are influenced by the applied processing conditions, i.e., temperature and time, and provide a plausible explanation based on the diffusive motion of the MWNTs in the polymer melt during the compression molding stage.

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1. Introduction

Recently, much attention has been given to the use of carbon nanotubes (CNTs) as filler in conductive nanocomposites in order to harness their exceptional electrical properties [1,2]. CNTs are used as a dispersed conductive phase in an insulating polymer matrix. In the case of conductive nanocomposites, the target is to obtain a network of connected filler particles, which allow electrical current flow through the sample. In fact, the actual aim is to combine the “advantages” of both types of materials, i.e., the high conductivity of the CNTs with the good processability, low density, and so on, of polymeric materials. In the long term, these “conductive plastics” are expected to be able to replace metals or semiconductors for applications in which the latter are currently still preferred. Examples of such applications are electrostatic dissipation [3], electromagnetic interference shielding [4], multilayer printed circuits [5], and transparent conductive or antistatic coatings [6,7].

The electrical conductivity of composites consisting of a polymer matrix filled with conductive filler particles is usually discussed in terms of percolation theory [8–10]. The electrical

conductivity of composites made of conductive filler particles dispersed in an insulating matrix strongly depends on the filler loading. At low filler concentrations the conductivity remains close to that of the electrically insulating matrix polymer because the filler particles are individually dispersed or grouped into small clusters. Above a critical filler volume fraction, the conductivity increases by many orders of magnitude over a small range in filler loading. This so-called percolation threshold coincides with the formation of a system spanning, conduction network of filler particles in the continuous polymer phase. Far above the percolation threshold, the conductivity of the nanocomposite levels off and does not increase significantly with the further addition of CNTs.

Interestingly, there is a considerable body of evidence for the presence of an insulating layer between the CNTs even above the percolation threshold, as in fact also seems to be the case for other types of conductive fillers including carbon black [11–19]. This then implies that the percolating filler particles are not in actual contact with each other, and that conductivity must occur via some tunneling or hopping process through the insulating layer that separates them. This can only happen if the shortest distance between two neighboring particles is below a certain value, estimated to be in the order of a couple of nanometers [20–22]. The type of electron transport involved should be strongly dependent on the CNT/polymer system in hand, and depends in particular on the statistics of inter-particle separations. It follows that the percolation

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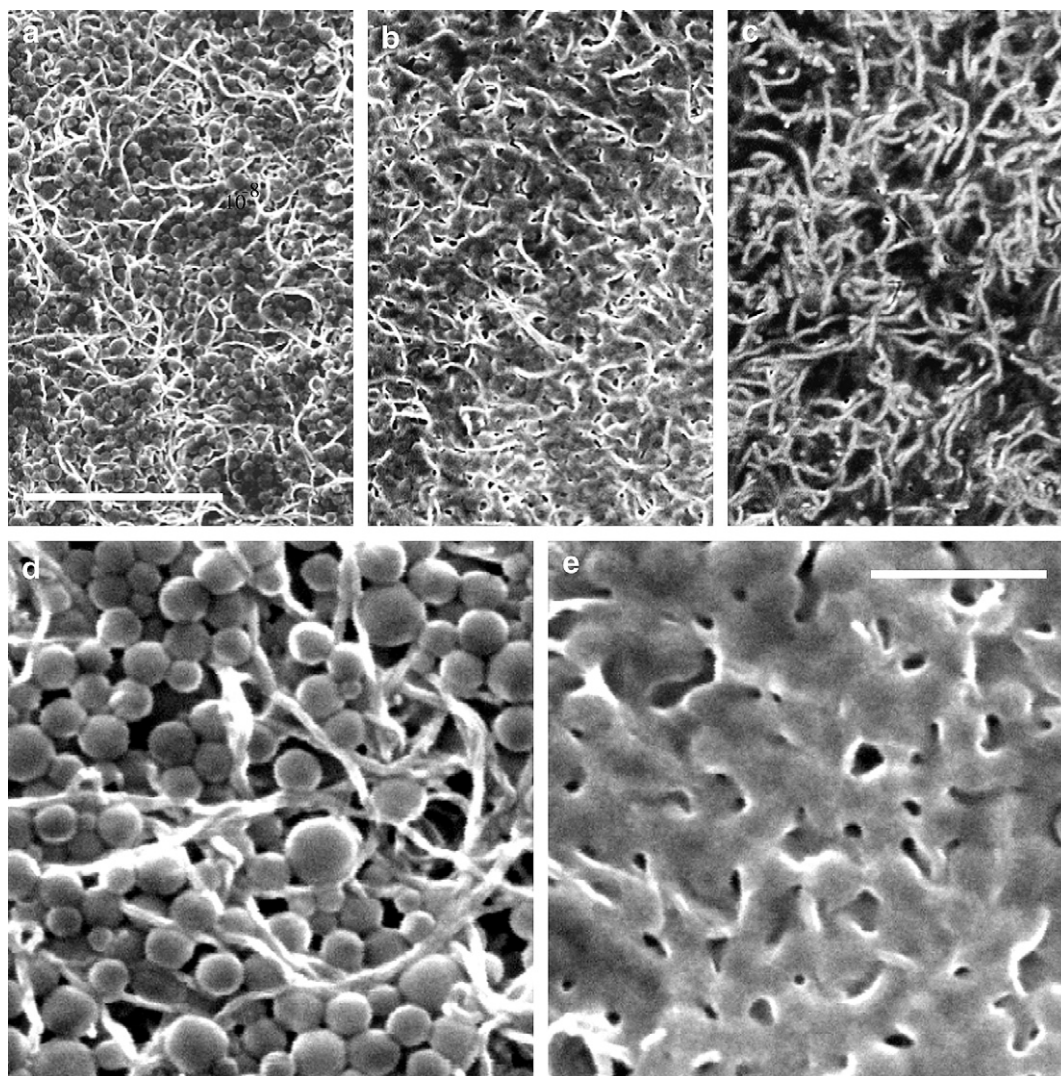


Fig. 1. SEM image of a freeze dried MWCNT/PS powder containing 7.8 wt% of MWCNTs (a) after heating at 60 °C, i.e., at a temperature at which no flow of the polymer takes place. The polymer particles are in a close-packed configuration and the CNTs are located in the interstitial space between them (b) after heating at 100 °C, the glass transition temperature of polystyrene. The polymer particles are deformed due to the polymer 'flow'; and (c) SEM surface image of the nanocomposite after processing at 180 °C, i.e., at a higher temperature than the flow temperature of the polymer. Scale bar for the three images (a–c) 1 μm; (d) detail of Fig. 1a. Scale bar: 200 nm; (e) detail of Fig. 1b. Scale bar for (d and e) 200 nm.

threshold is not only purely geometrically defined, as usually tacitly implied, but also physically defined, exactly because the particles need not quite touch for conduction to take place.

It is now clear that the percolation and conductivity of conductive nanocomposites are a sensitive function of the aspect ratio of the CNT filler particles [23,24], the degree of bundling and the polydispersity in size and shape [25–28], the interaction between the matrix and the filler and between the filler particles themselves [29], their average degree of orientation [30,31] and, interestingly, the conditions under which the composites are processed [32–34]. This last aspect is important because it means (i) that the measured percolation thresholds might not be the lowest possible for a given material system and (ii) that one should be weary comparing theoretical predictions that are often based on equilibrium percolation arguments [35,36]. The question of if and how equilibrium percolation is achieved in practice in nanocomposites seems to be a key one.

Here, we systematically address the problem of processing conditions on the percolation of electrically conductive CNT/polymer nanocomposites, produced by means of a latex-based technology [37,38]. The procedure that we follow comprises several steps. First, the CNTs are exfoliated or debundled in a surfactant

solution by means of sonication [39,40]. The resulting aqueous surfactant–CNT dispersion is then mixed with an aqueous polymer latex. This is the key step of the process, because it determines how effective the incorporation of the CNTs into the polymer matrix is going to be. At this point, an equilibrium distribution is presumably regulated by the relative amounts of the rods and the latex particles and the way in which they interact. Then, the mixture of the two types of colloidal particles, i.e., the CNTs and latex particles, is freeze dried. In principle, the sublimation of the water induced by freeze drying is not expected to significantly modify the aggregated state of the CNTs nor the quality of mixing of the CNTs and the polymer latex particles. It seems that the CNT structure and quality are also not affected by this treatment [41]. Of course, freeze drying does induce a compaction of the CNT network that becomes denser because of the water removal.

After compaction of the powder consisting of submicron polymer particles and CNTs, the filler particles are forced into the interstitial space between the polymer latex particles [42,43]. The size distribution of the polymer particles governs the structure of the space where the fillers are confined, see Fig. 1a and d. Solvent removal and compaction do lead to an out-of-equilibrium

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