



## Review

## A review on carbon nanotubes and graphene as fillers in reinforced polymer nanocomposites

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## ABSTRACT

Recently, carbonaceous nanofillers such as graphene and carbon nanotubes (CNTs) play a promising role due to their better structural, functional properties and broad range of applications in every field. This paper reviews the synthesis and properties of CNTs along with the use of graphene as a novel substitution to the nanotubes as fillers. This review also focuses on the issues related to the processing, dispersion and alignment of CNT within nanocomposites. Furthermore a comparative analysis has been carried out between the importance of graphene and carbon nanotubes as fillers and their substantial changes in mechanical and electrical properties of matrix.

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## 1. Introduction

Materials play key role in every field of technology such as medical, sensors, computing etc. to make our lives more comfortable. There is always a critical need to use new materials with improved properties so that the demand of betterment can be satisfied. To meet the necessity and to create novel materials with meliorated properties it is essential to understand the relationship between structures and the properties of existing materials. Thus the properties can be tailored by combining two or more materials according to the requirements. Polymers are the most desired materials due to their low cost, reproducibility, easy processing etc. In past few decades there has been a great expansion in research related to the polymer nanocomposites due to development of advanced materials for potential applications [1–5]. Polymer nanocomposites is a combination of polymer matrix and with a large range of filler materials both, organic or inorganic which have at least one dimension of nanometer range. These fillers can be one dimensional, two- or three-dimensional. Carbonaceous nanofillers such as graphene, carbon nanotubes (CNTs) play a very promising role due to their better structural and functional properties such as high aspect ratio, high mechanical strength high electrical properties etc. than others [6–10]. In this review we focus on CNT and graphene based polymer nanocomposites in which CNTs and graphene are dispersed into the polymer matrix. Matrix guards reinforcements from atmospheric conditions, provides color and textures, offers load transfer, etc. Development of these polymer nanocomposites depends upon properties of nanofillers *i.e.*, CNTs or graphene, their dispersion within the polymer matrix as reinforcements, interaction between reinforcement and polymer matrix and alignment of nanofillers into the matrix [11]. In consequence of simple processing of polymer matrix as compared to ceramics and metals, it is seen that carbon fibers reinforced polymer matrix is of great interest [12]. CNTs are considered as an excellent filler due to their unique properties such as high aspect ratio, high tensile strength, high conductivity etc. [9,10]. Though CNT reinforced polymer nanocomposites have good attracting properties but they vary with the process of fabrication of nanocomposites. Fig. 1 highlights the importance of factors like CNT shape, size, interaction, dispersion, alignment etc. which plays an important role in the fabrication of CNT reinforced polymer composites. By using various approaches, results can be optimized. A combination of these excellent properties of the filler and the composite synthesis route, these two provides an excellent source of composite materials for various engineering applications. These composites could be used as conductive glue, gas storage devices, sensors, energy storage devices, light weight aircraft applications, defense applications, etc. [1,13–15].

In this review we are focusing on structural properties of CNTs, routes of synthesis of CNTs and mechanical, electrical and thermal properties of CNTs. The later sections will discuss about processing of CNT/polymer nanocomposites by different routes such as solution mixing, melt processing and *in situ* polymerization. Due to small dimensions of CNTs and exhibiting strong Van der Waals forces they have tendency to stick together. In consequence, inefficient load transfer from matrix to fillers. To avert these

problems, functionalization techniques are used *i.e.* covalent and non-covalent functionalization. Incorporation of CNTs into the polymer matrix, results in substantial changes in the mechanical, electrical and thermal properties of polymer composite [16–18]. Furthermore the review in short detail also discusses the importance and use of CNTs or graphene as fillers in the polymer matrix along with their applications.

## 2. Carbon nanotubes

The discovery of carbon nanotubes in 1991 by Iijima [19], brought the revolutionary changes into the field of polymer nanocomposites. Ajayan et al. reported first carbon nanotubes reinforced polymer nanocomposites, after the discovery of the same [20]. As reported by Thostenson et al. [21], the orientation and magnitude of chiral vector in graphene sheet defines the morphology of carbon nanotubes. Carbon nanotubes are basically rolled up graphene sheets (hexagonal structures) into cylindrical form and capped with half shape of fullerene structure. There are two types of carbon nanotubes: (1) Single walled nanotubes (SWNTs), can be considered as single graphene sheet rolled into a cylinder and (2) Multi walled nanotubes (MWNTs), it can be considered as stacking of concentric layers of several graphene layers in the form of cylinders with an interspacing of 0.34 nm. On the basis of atomic arrangement there are three types of structures zigzag, armchair and chiral structures (Fig. 2). Properties of carbon nanotubes are highly dependent on morphology, size and diameter. Carbon nanotubes can be metallic or semiconducting; it depends on the atomic arrangement of carbon nanotubes. Initially, MWNTs were reported by Iijima by arc evaporation method in 1991 [5,19,23] and later on he reported the discovery of SWNT in 1993 by arc evaporation of metal impregnated (Fe-graphite) electrodes in the presence of methane-argon [23].

### 2.1. Synthesis

A large numbers of carbon nanotubes are produced via various methods such as arc evaporation method, laser ablation, chemical vapor deposition, electrolysis, flame synthesis etc. every year. Arc evaporation method, laser ablation and chemical vapor deposition are the techniques which are used broadly for synthesis of CNTs [24,25].

#### 2.1.1. Arc discharge method

This was the method used by Iijima for the preparation of carbon nanotubes. In this method, CNTs are formed by creating a hot plasma discharge between two graphite electrodes which are connected to the power supply (100 A; 20 V) in the presence of Helium (He) gas. According to the Ebbesen and Ajayan, as the pressure of He in the chamber is increased up to a certain value, the yield of nanotubes also increases, but after that value, further increase in He pressure leads to the fall in CNT yields [26]. Better quality of the nanotubes depends upon the lowering of the current [27]. Various gases like, N<sub>2</sub> [28], CF<sub>4</sub> [29] were used in place of He gas. Shimotani et al. reported the increased yield of nanotubes with the use of organic vapors [30].

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