JCAMECH

Vol. 51, No. 1, June 2020, pp 247-252
DOI: 10.22059/jcamech.2020.304476.517

A Brief Review on the Influences of Nanotubes' Entanglement and Waviness on the Mechanical Behaviors of CNTR Polymer Nanocomposites

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ARTICLE INFO

Article history:
Received: 14 June 2020
Accepted: 11 July 2020

Keywords:
Carbon nanotube (CNT)
Agglomeration
Waviness
Polymer nanocomposites (PNCs)

ABSTRACT

Invention of carbon nanotube (CNT) in the 1990s introduced a new class of materials whose extraordinary mechanical, thermal, and electrical properties seemed appealing enough to the research community to devote their time and effort for the purpose of analyzing composite materials reinforced by CNTs. Particularly, the marvelous stiffness of CNTs has made it possible to reach a high-modulus composite once such a nanomaterial is dispersed into various types of matrices. Among all of these products, CNT-reinforced (CNTR) polymer nanocomposites (PNCs) are used more than the others due to their incredible specific stiffness and fracture toughness. Although PNCs can bring a lot for the designer due to their inherent merits, it must be pointed out that some practical phenomena take place in the microstructure of such advanced materials whose neglecting can be resulted in negative outcomes. Motivated by this reality and based upon the authors broad researches in this area, present review is organized to show how can the mechanical behaviors of PNCs be affected by entanglement of the CNTs inside the inclusions and their wavy shape.

1. Introduction

Since introduction of carbon nanotubes (CNTs) in 1990s [1], remarkable attention of the research community was attracted by materials and structures with at least one dimension in the range of nanometer. Generally, carbonic nanomaterials are capable of showing a wide range of improved mechanical, electrical, and thermal properties from themselves [2-5]. Hence, many of the attempts were conducted by scientists and engineers in order to understand the accurate properties of advanced composite materials reinforced via dispersion of CNTs in the matrix. It is reported that CNTs are capable of showing a marvelous Young's modulus of even larger than 1 TPa [6]. This reality is one of the most important reasons led to the appearance of CNT-reinforced materials, also known as "CNTR nanocomposites". Poly(vinyl alcohol) was used as the host matrix of CNTs for the purpose of manufacturing and characterizing CNTR nanocomposites in 1999 [7]. In this experimental study, catalytically-grown CNTs were implemented as the starting material due to the unavailability of Arc-grown ones.

Implementation of CNTs for the goal of fabricating CNTR polymer nanocomposites (PNCs) results in observing a low percolation threshold of about 0.1-2 vol. % in addition to the enhancement of the equivalent stiffness of the material [8]. The influence of using different types of CNTs (e.g., single-walled CNT (SWCNT), double-walled CNT (DWCNT), and multi-walled CNT (MWCNT)) on the improvement of the polymers' material properties were explored within the framework of an experimental study [9]. It was shown that the fracture toughness of PNCs reinforced via amino-functionalized DWCNTs can be enhanced by up to 40% once a 0.5 wt.% of CNTs is inserted into the epoxy [9]. In another experimental investigation, it was reported that in the case of manufacturing PNCs with poorly-dispersed CNTs, greater rheological characteristic responses can be obtained [10]. Indeed, the time-dependent behaviors of PNCs containing poorly-dispersed CNTs were closer to solids once compared with PNCs manufactured via a well dispersion of CNTs [10]. A combination of experimental and theoretical frameworks was utilized by another group of researchers in order to survey the mechanics of PNCs. In this endeavor [11], MWCNTs were synthetized using the low-cost, controllable chemical vapor deposition (CVD) method and added to a thermoset polymer. A remarkable increase in the tensile strength and failure strain of the phenolic matrix was appeared once a limited content of CNTs is added. Furthermore, an experiment-based study proved that SWCNTs cannot enhance the mechanical behaviors of polymers as efficient as MWCNTs [12]. It is shown that adding the content

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of SWCNTs cannot be resulted in the improvement of the material properties as a general trend.

In a theoretical study, the influence of the bonding condition between CNT and polymer on the constitutive behaviors of PNCs was captured. In this work, the well-known Lennard-Jones (LJ) van der Waals (vdW) potential was selected for the goal of considering the adhesive interface between the CNT and polymer. The results of this study indicate on the fact that better mechanical behavior can be enriched once (6,6) armchairs are utilized in the PNC instead of (18,18) ones [13]. It is reported that the interfacial bonding between the CNT and matrix can possess high impacts on the fracture toughness, yield strength, and other mechanical properties of PNCs [14]. In the cases that debonding of CNTs and matrix happens, the probability of appearance of voids in the nanocomposite will grow and also the stress concentration will be increased [14]. On the other hand, the load-carrying capacity and stiffness of the PNC will be lessened in the cases of which a weak bonding between nanofiller and matrix exists in the PNC [14]. The bonding issue is of great importance types of NCs, either polymeric (PNC) or metallic (MNC); more data about the perfect- and imperfect-type bonding condition between the CNT and matrix in PNCs/MNCs can be found in Refs. [15-20]. Also, the volunteers are addressed to take a brief look at the authors former review about various types of NCs [21] and the critical review conducted by Norouzi, Barati, Noroozi [22] dealing with the numerical investigation of NCs.

2. Agglomeration phenomenon

One of the phenomena whose destroying effect on the material properties of the CNTR-PNCs must be considered is the agglomeration of the CNTs. Simply, agglomeration is the accumulation of a large number of CNTs in a limited inclusion inside the PNC. This phenomenon affects the mechanical behaviors of the PNC negatively because it does avoid from the uniform distribution of the CNTs in any desired point which is not preferable at all. The reason of occurrence of such a phenomenon is that CNTs will be entangled in the vdW potential of other CNTs' and this will be resulted in the local construction of inclusions filled with a large number of CNTs [23]. Indeed, this is due to the outer surface of the CNTs which acts as a bridge for the transfer of stress between matrix and CNT in general; however, sometimes it will cause an entanglement of a group of CNTs in each other's attractive vdW potential [9]. It is reported that this phenomenon will make several problems in the PNC, e.g., it will affect the efficiency of adding CNTs in polymeric matrix. In other words, agglomeration results in observing that the mechanical behavior of the PNC will improve once a 0.1 wt. % of CNTs is utilized and in the cases that greater amounts of CNTs are employed, the improvement will be replaced with negative growth [9]. In 2010s, CNTs were utilized by researchers for the purpose of analyzing the effect of adding PNC coatings on glass fibers on the tensile strength of the coated glass fibers [24]. It was experimentally proven that addition of the content of the available CNTs cannot certify the reinforcement phenomenon in general. Indeed, aggregation of CNTs makes it hard to enrich stiffer PNCs by gradual increase of the content of the CNTs. It is mentioned that in addition to the approach of functionalization, one other key method to reduce the entanglement of the CNTs is to avoid using the so-called CVD for producing and synthesizing CNTs [24]. Similar outcomes were obtained experimentally in Ref. [25]. In fact, this work was concerned with the investigation of the fracture toughness properties of PNCs reinforced via MWCNTs and it was observed that the fracture mechanism of PNCs is dramatically influenced by the existence of aggregations containing a group of MWCNTs [25]. Such observation can be similarly found in Ref. [15]. In this study, the fracture behaviors of cured PNCs were explored via an experimental viewpoint and it was reported that the existence of CNTs' agglomerates can deeply affect the fracture behaviors of the CNTR PNCs. In one of researches dealing with the appropriate distribution of CNTs in polymeric matrices, it is shown that using functionalized CNTs after a plasma-polymerization will avoid from the entanglement of CNTs inside inclusions and results in observing a better performance [26]. This reality was proven once the scanning electron microscopy (SEM) micrographs of PNCs manufactured from both raw and plasma-polymerized CNTs were compared. Using a wholly different viewpoint, the effects of aggregation of CNTs on the Young's modulus and Poisson's ratio of PNCs were included within the framework of a multi-scale modeling [27]. It was numerically shown that the existence of inclusions including a large number of CNTs inside themselves can dramatically affect the Young's modulus of the PNC in negative manner. Furthermore, it is reported that there exists a difference between the size of the inclusions containing agglomerated CNTs in a PNC and it must be tried to reduce the size of the agglomerates in a PNC for the purpose of reaching a better performance [28]. Indeed, once large agglomerates are broken into smaller ones, the stiffness-enhancement will be lesser affected by the presence of aggregation. In a numerical investigation, a multi-scale stochastic modeling was introduced in order to study the mechanical behaviors of CNTR PNCs addressing the agglomerates of CNTs using the irregular tessellation method [29]. In this work, a modified micromechanical scheme is introduced which is able to take into consider the agglomeration phenomenon by defining an agglomeration factor i.e. a function of the volume fraction of the CNTs inside the PNC.

On the other hand, some of the Chinese researchers introduced a novel bioinspired method for the purpose of wet synthesizing of CNTs in order to lessen the dimensions of the agglomerates in PNCs by the means of using acetone solvent [30]. It was experimentally shown that using this technique can be resulted in fabricating PNCs with performances a hundred percent better than those manufactured using aggregated nanotubes. In another experimental attempt, researchers employed functionalized MWCNTs in the framework of an electrochemically influenced *in situ* polymerization to fabricate an enzymatic glucose biosensor that the aggregation of CNTs is controlled in [31]. A new theoretical method was developed based upon experimental data for the goal of analyzing the fracture behaviors of PNCs reinforced with CNTs regarding for the agglomeration phenomenon [32]. This newly developed method included an agglomeration factor whose determination was depended on two empirical constants which can be obtained comparing the experimental data and theoretical ones [32]. Moreover, a sample natural rubber PNC was manufactured by Medupin, Abubakre, Abdulkareem, Muriana, Abdulrahman [33] to be utilized as an anthropomorphic prosthetic foot. They observed a sudden drop-down in the stress-strain curve of the CNTR PNCs which was attributed to the accumulation of the CNTs inside the inclusions. In one of the recent works, nitrogen plasma treated CNTs were implemented in order to enhance the thermo-mechanical stability of the PNCs by avoiding from the occurrence of the agglomeration phenomenon [34].

On the other hand, there exists some other researches concerned with the mechanical responses of continuous systems manufactured from PNCs. In some of these attempts, the influence of existence of agglomerated CNTs on both static and dynamic responses of the NC system was taken into consideration within the framework of micromechanical or numerical modeling-based studies. One can find the detail of the abovementioned procedure by referring to Refs. [23, 35-45].

3. Waviness phenomenon

Another crucial issue whose missing during the design procedure of PNCs can be resulted in unpredicted outcomes is the existence of a wave in the structure of the utilized CNTs [46-49]. In other words, CNTs in PNCs can be rarely found in straight shape due to their very high flexibility [7, 50]. Recalling the CNTs' micron- and nanometer-order length and diameter, respectively, it can be simply understood that such nanostructures can bend easily and manifest in any desired shape except straight one. It is reported that one of the main reasons of occurrence of such a wavy nature for the CNTs is the fact that some defects (i.e., pentagon-heptagon topological defect pair) will appear in the CNTs' lattice during the fabrication procedure [51]. In such condition, the CNT will bend because of its inherent weight and also the tendency to another CNTs through the existing vdW potential [51]. Also, the wavy nature of the CNTs can be also attributed to the limited space of which the CNTs were grown [51]. It is worth mentioning that another category of curvy CNTs is known from 1994 whose shape can be described as a helical path [51, 52]. Indeed, such CNTs are named coiled CNTs (CCNTs) which are majorly grown with different helix angles, diameters, and also various pitches [51].

In a micromechanical study, the wavy shape of the CNTs was assumed to be a light curvature similar to a sinusoidal shape [14]. In this article, the wavy CNT was assumed to be identical with its projections in axial and transverse directions and the theory of fiber-reinforced composites was extended for the PNC containing wavy CNTs. Within the framework of an stochastic multi-scale modeling, the influence of the wavy shape of the CNTs on the mechanical properties of CNTR PNCs was observed following the assumption that the material properties of a wavy CNTs are forced to be within a bounded range [27, 53]. According to the obtained results, it is reported that the effect of the wavy shape of the CNTs on the material properties of the PNC is greater than that of their entanglements [27]. Similar to the method utilized in [14], another theoretical investigation was carried out for the purpose of considering the effect of the bow-shaped wavy structure of the CNTs on the mechanical properties of shape memory PNCs while the negative influence of the nanofillers' agglomeration is included, too [54]. The comparison of the extracted results with experimental ones reported in [55], certified the validity of the presented methodology. The stochastic consideration of the effect of CNTs' wavy shape on the equivalent elasticity modulus of CNTR PNCs was fulfilled by an Iranian researcher [56] in conjunction with the author's previous contributions in this area. It seems that the proposed methodology was one of the efficient numerical methods because of its acceptable agreement with data obtained via experimental investigations. Moreover, another group of researchers in the Georgia Institute of Technology reported the wavy nature of the CNTs as the most important mechanism which affects the material properties of such nanostructures within the framework of both experimental and numerical approaches [57, 58]. Emphasizing on the crucial impact of the CNTs' non-straight shape on the variation of the coefficient of thermal expansion (CTE) of a fuzzy fiber-reinforced composite (FFRC), it is revealed that the CTE of the advanced composite can be even improved once the wavy shape of the CNTs is governed in a way that the amplitude of the sinusoidal shape of the CNTs is parallel with the longitudinal axis of the fiber [59]. More data on the issue that how can the mechanical characteristics of PNC materials and structures can be influenced by the existence of waviness phenomenon can be simply found referring to complementary Refs. [60-66]. It must be declared that more explanation about the above references will not be presented because of the fact that their main concepts are reviewed in the aforementioned discussions and it is assumed that the readers are now familiar enough with them.

4. Concluding remarks

The major objective of this review was to put emphasize on the effects of presence of CNTs' entanglement and their non-straight shape on the material properties of PNCs manufactured using various types of CNTs, namely SWCNT, DWCNT, and MWCNT. To this purpose, a general framework was presented in the first part insisting on the general concepts of invention of PNCs and the reasons of which such nanomaterials are able to show extraordinary behavior from themselves once compared with conventional materials. Afterward, the physical reasons and various types of viewpoints of which agglomeration and waviness phenomena could be justified on their basis were discussed in detail within the frameworks of sections 2 and 3, respectively.

According to the above brief literature review, it can be claimed that it is approximately impossible to avoid from the agglomeration of the CNTs in PNCs. In fact, the vdW potential of each individual CNT can attract several CNTs in itself which is the main mechanism of appearance of agglomeration phenomenon in the CNTR PNCs. However, following some approaches can be resulted in an improvement in the state of the NC in the presence of aggregation of the CNTs. First of all, it is proven that utilization of functionalized CNTs can be resulted in a better resistance of the CNTs against the aggregation phenomenon. The second approach whose efficiency in reducing the entanglement probability is proven is to employ plasma-polymerization technique to manufacture the PNCs. It must be considered that lowering the amount of the aggregated CNTs in CNTR PNC is a key factor to improve their mechanical performance because in the case of having agglomerated CNTs in the PNC, adding the content of the available CNTs cannot be resulted in the enhancement of the system's mechanical response.

On the other hand, it was mentioned that the wavy nature of the CNTs in the PNC can play a negative role in the determination of the equivalent stiffness of the PNCs because it decreases the reinforcing efficiency of the CNT. From materials' point of view, this phenomenon takes place due to the thousands-order length-to-diameter ratio of the CNTs in general. In other words, CNTs possess a flexible lattice that its bending due to its inherent mass is not out of mind at all. In addition to this reason, it must be mentioned that during the synthesizing procedure, some topological defects can be appeared in the CNTs which lead to have wavy CNTs in the PNCs.

Based on this brief review, it can be claimed that the main mechanism of decreasing the performances of CNTR PNCs are the abovementioned phenomena and their accurate consideration can be resulted in generation of more realistic data about the mechanical behaviors of NC systems. It is noteworthy that to the best of the authors' knowledge the best way of analyzing the mechanical behaviors of PNCs is to analyze them via numerical simulation-based methods if experimental nanoidentation is not reachable at the moment.

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