



## Review

# Directional alignment of carbon nanotubes in polymer matrices: Contemporary approaches and future advances



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## ABSTRACT

The emergence of carbon nanotubes (CNTs) has created new opportunities for the fabrication of polymer composites that possess strong potential for a wide spectrum of applications. Numerous significant advances have been attained to date, and more technological challenges await the optimization of a system to fill the gap between expectations and practical performance. Despite this tremendous progress, challenging issues related to directional alignment and CNT assembly within a polymer matrix still remain. This review presents the development of contemporary approaches in the directional alignment of CNTs in polymer matrices, with particular emphasis being placed on the recent progress, exciting breakthroughs and active pursuit in improving CNT alignments through different approaches.

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## 1. Introduction

Carbon nanotubes have been recognized as one of the most versatile materials of the new century. This class of material has generated intense interest in the scientific community for a host of applications, ranging from large scale structures to automobiles to nanometer-scale electronics [1]. Consistent research efforts have proven that CNTs are not only useful and interesting systems in and of themselves but can also be used in the fabrication of nanocomposite materials. Their unique properties have spurred a great deal of interest in using CNTs as nanofillers. CNTs exhibit many superior properties that are promising for use as filler particles, and the introduction of CNTs in polymer matrices represents a new direction for the development of composite materials in wider applications. The increased interest in incorporating CNTs into polymer matrices most likely originates from their potential to reach thermal, electrical and rheological percolations at relatively low concentrations [2–6]. Because the combined exceptional properties exhibited by the composites cannot be found in the individual constituent materials, nanocomposites incorporating CNTs are highly desirable. Thus, efficient fabrication and processing are major goals in the current state of modern nanotechnology [7].

Despite these high expectations, the ultimate practical application seeks a CNT/polymer composite with physical properties that approach the theoretical maximum values of an individual nanotube or polymer matrix [8]. The initial interest in the alignments of CNTs in polymer matrices was introduced by Ajayan et al. [9], who conceptually proved that the remarkable properties of CNTs could be realistically transferred to a polymer matrix. These findings paved the way for the development of various approaches essential for attaining the desired composite properties through incorporating and aligning CNTs in a host matrix. The focus of subsequent research has addressed the aforementioned issues. There have been a surge of reports and literature regarding this new frontier of materials that is open to researchers. From 1991 to 2013 (since their discovery by Iijima), CNT research has gradually and steadily shifted from synthesis and surface modifications towards practical applications as filler in various types of composites. While many of the early studies on CNT/polymer composites primarily focused on the incorporation and characteristics of CNTs in polymer matrices, recent efforts undertaken by materials scientists and engineers have converged on the applications of nanotube properties in the development of multifunctional composites [10]. Few effective routes have been identified and proposed to utilize the excellent properties of CNTs on a macroscopic scale. Recent breakthroughs and achievements have spurred significant interest in using CNTs as structural and functional materials in the fabrication of nanocomposite materials. It is believed that, through a fundamental understanding of their processing–structural–performance relationships, the creation of multifunctional composites with controlled hierarchical structures may offer a wide range of future applications.

Nevertheless, these findings show that very few nanocomposite architectures have been developed to date that can approach their ideal performance; a vast portion of research is still struggling to meet targeted expectations. Despite the tremendous discovery of the substantial and immediate potential applications of CNT-embedded polymer composites, the challenges in translating CNT properties into a polymer matrix and the difficulties in addressing the low reproducibility of CNT properties with different preparation methods must be resolved. Incorporating an organized CNT architecture into a carefully selected polymer matrix and engineering the interfaces between the two constituents is crucial [11]. When considering a CNT/polymer composite, the two critical issues related to the nanotube and the CNT–polymer interphase

chemistry are the dispersion and the alignment of the nanotubes in the polymer matrix. While the former is highly desired to obtain a uniform system and a higher nanotube density per volume unit for more capillary distribution in the polymer matrix [12], the latter is of particular importance for applications that draw on the anisotropic thermal and electrical properties, as well as the mechanical strength and fluid transport characteristics. Macroscopic materials comprised of CNTs with highly anisotropic properties are expected to possess vast potential applications [13]. However, to enhance the anisotropic nature of the CNTs and attain optimal performance, it is essential to develop directionally aligned CNT structures in the composite [14] and obtain the highest possible fraction to exploit the excellent intrinsic axial properties of CNTs in an actual material [15]. The resulting aligned CNT/polymer composites could offer immense potential and open doors for diverse technological possibilities in a broad spectrum of applications in various materials and engineering fields. In general, the alignment of CNTs in a polymer matrix can be performed (i) during the formation of the composite through techniques such as infiltration, fiber drawing and magnetic/electrical fields or (ii) after the incorporation of the CNTs through approaches such as mechanical stretching and polymer compression [16].

At present, numerous approaches such as blending and solution mixing have been successfully established to incorporate CNTs in polymer matrices [17,18]. Despite the high value of aligned CNT/polymer composites for many device-based applications, the preparation of CNT/polymer composites with simple and straightforward solutions or melt blending approaches has compromised the dispersion and alignment of CNTs. Even under experimental conditions, a well aligned array of CNTs tends to become isotropic and cluster upon incorporation into the composite, eventually dispersing into random orientations [19,20]. Therefore, feasible alignment approaches that are integratable to the device fabrication process and applicable to a wide range of polymer matrices are highly desirable [21]. While dispersion issues have been well addressed in existing literature [22–26], few reviews have been devoted to topics related to the directional alignment of CNTs in a polymer matrix [27,28]. The synthesis methods and applications of aligned multi-walled carbon nanotubes (MWCNTs) have been critically reviewed by Sun et al. [29]. However, the major highlights were focused on their potential utilization in organic optoelectronic materials and devices. Ahir et al. [27] also presented a review of the approaches developed for the CNT alignment in polymer composites, with particular attention paid to ultrasonic cavitation and shear mixing. While numerous experimental studies have been recently performed, there has been no systematic and comprehensive literature review on the classifications of the approaches used for the horizontal and vertical alignment of CNTs in a polymer matrix. From a scientific standpoint, it is necessary to fully comprehend the macrostructural properties of the aligned CNTs and their nanocomposite behavior. Therefore, in concert with previously reviewed literature, this paper reviews the state-of-the-art concerning aligned CNTs in polymer composites. The factors motivating current efforts in the development of CNT/polymer composites are discussed. The major highlights of this review concern the development of contemporary approaches, as well as the potential applications of the resultant CNT/polymer composites. It is hoped that this contribution can serve as a general reference to guide feasible processing methods for the fabrication of aligned CNT/polymer nanocomposites.

## 2. Aligned carbon nanotube/polymer nanocomposites: rationales and motivations

The physicochemical properties of a nanocomposite are primarily dictated by the macro- and microstructures produced during

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