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## Review

## Stabilization and dispersion of carbon nanomaterials in aqueous solutions: A review

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## ABSTRACT

Carbon nanomaterials (CNMs) have become candidates for numerous applications in nanocomposites, microelectric devices, sensors, energy storage, microelectronics, biomedicines, and mechanical resonators. However, a key challenge is how to enhance the dispersion and stabilization of CNMs in water. Thus, this review summarizes recent progress on the stabilization and dispersion of various CNMs (such as carbon nanotubes, graphene (oxides), and fullerenes) in aqueous solutions. The stabilization and dispersion of target CNMs is reviewed to address the effects of water quality conditions (pH, ionic strength, temperature), natural/synthetic dispersing agents, and the effects of ultrasonication, acidification, and/or UV irradiation on dispersion and stabilization. Applications of CNMs are also briefly addressed regarding removing micropollutants by adsorption in water and wastewater treatment. The outlook for future research challenges on CNMs is also discussed.

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## 1. Introduction

Carbon nanomaterials (CNMs) are defined as carbon materials that have physical features with at least one dimension of 100 nm or less, and include carbon nanofilms and carbon nanocoatings (<100 nm in one dimension), carbon nanotubes and carbon nanowires (<100 nm in two dimensions), and CNMs (<100 nm in three dimensions) [1]. Among the three main groups (natural, incidental, and engineered) into which CNMs are classified, engineered CNMs are produced intentionally and are fabricated from the “bottom up” [2]. CNMs, including carbon nanotubes (CNTs), graphene oxides (GOs), and fullerenes, are widely used in various applications such as electro-optical devices, plastics, catalysts, and components in composites [2].

In particular, since their discovery in 1991, CNTs have shown such unusual mechanical, electrical, thermal, and chemical properties that they have become candidates for many applications, including nanocomposites, energy storage, microelectronics, and medical devices [3,4]. Several studies anticipated the production of CNTs at millions of tons in 2010, and a \$1 trillion worldwide market for nanoproducts by 2015 [5,6]. Additionally, other CNMs, such as graphene and fullerenes, have drawn intense research interest and large-scale applications in various areas due to their unique physicochemical properties [7–10]. However, to maximize the advantage of CNMs (e.g., as effective adsorbents in water), they should not form aggregates and must be well dispersed, so that dispersed and stabilized CNMs in solution can greatly increase the interaction of CNMs with, for example, contaminants in solutions.

Difficulties in the stabilization and dispersion of CNMs originate from their propensity to aggregate, particularly in aqueous systems, due to substantial van der Waals attractions and specific hydrophobic interaction between CNMs [11–14]. Researchers have used novel and unique approaches to address such challenges in the aggregation or bundling of CNMs, including covalent and non-covalent functionalization schemes [15–21]. Generally, chemical functionalization is based on the covalent linkage of functional entities onto the carbon support of CNTs [13]. For example, for CNTs, direct covalent sidewall functionalization is associated with a change in hybridization from  $sp^2$  to  $sp^3$ , leading to a partial loss of their optical, electrical, and thermal properties [22,23]. Defect functionalization is another method for the covalent functionalization of CNMs. Acids, such as  $HNO_3$ ,  $H_2SO_4$ , or a mixture thereof [24,25], and strong oxidants, such as  $KMnO_4$  [26], ozone [27], and reactive plasma [28], can create defect functionalization on CNMs.

While covalent methods can provide valuable functional groups on the CNM surface, these methods have two main disadvantages [13]: (i) during the functionalization reaction, particularly along with damaging ultrasonication and/or oxidation processes, many defects are inevitably created on the CNM surfaces, and in some extreme cases, CNMs may be fragmented into smaller pieces and (ii) concentrated acids or strong oxidants are frequently used for CNM functionalization, which are environmentally unfriendly. Thus, non-covalent functionalization is an alternative method for changing the interfacial properties of CNMs. The suspension of CNMs (a typical example of non-covalent treatment) in the presence of various polymers causes the wrapping of polymer around the CNMs to form super-molecular complexes of CNMs [29–32]. The polymer wrapping process is achieved through van der Waals interactions and  $\pi$ - $\pi$  stacking between CNMs and polymer chains containing aromatic rings [13].

While the stabilization and dispersion of CNMs in aqueous solutions have been studied widely [33–36], a broader analysis of CNM stabilization and dispersion in water is essential, because the

unique properties of CNMs in terms of size, shape, surface area, volume, morphology, functional groups, and functionalization of CNMs affect their stabilization and degree of dispersion in water. Thus, this review provides a comprehensive analysis of the aqueous stabilization and dispersion of CNMs, particularly CNTs, GOs, and fullerenes, under various water quality conditions, natural/synthetic dispersing agents, and ultrasonication, acidification, and/or UV irradiation conditions. Additionally, this review briefly surveys recent publications on the synthesis and potential applications of CNMs for micropollutant removal in water and wastewater.

## 2. CNM synthesis

Kroto et al. developed the chemistry of fullerenes in the mid 1980s [37]. Fullerenes are geometric cage-like structures of carbon atoms that are composed of hexagonal and pentagonal faces [38].  $C_{60}$  was the first closed and convex structure. A few years later, CNTs that are now one of the widely known CNMs were discovered by Iijima, who described a multi-walled CNTs (MWNTs) preparation process after a random event during the testing of a new arc evaporation method for  $C_{60}$  carbon molecule fabrication in 1991 [39]. While MWNTs that have diameters from 2 to 100 nm and lengths of tens of microns consist of an array of such cylinders, formed concentrically and separated by 0.35 nm, similar to the basal plane separation in graphite [39], single-walled nanotubes (SWNTs) consist of a single sheet of graphene rolled seamlessly to form a cylinder with diameter of the order of 1 nm and lengths of up to centimeters [40,41].

CNTs can be fabricated by various methods. For example, high-temperature preparation techniques, such as arc discharge, were originally used to fabricate CNTs [42]. The arc discharge synthesis of CNTs is simple in circumstances where all the growth conditions are ensured for MWNTs [43]. It has been reported that MWNTs can be synthesized using an arc discharge technique under He, ethanol, acetone, and hexane atmospheres at various pressures (150–500 Torr) [44], and arc discharges in the three organic atmospheres (ethanol, acetone, hexane) produced more MWNTs, at least double, than those in the He atmosphere. Additionally, arc discharge is commonly used for the deposition of some non-standard CNTs. Unlike standard MWNTs deposition using a gas atmosphere, a previous study showed a high-yield synthesis of MWNTs by arc discharge in liquid nitrogen, suggesting that this technique may be a practical option for the large-scale synthesis of MWNTs with high purity [45]. Unlike MWNTs, SWNTs are synthesized with a transition metal catalyst in the arc discharge deposition process, typically in a hydrogen or argon atmosphere, using a composite anode (a composition of graphite and a metal, such as Ni, Fe, Co, Pd, Ag, or Pt, or mixtures of Co, Fe, or Ni with other elements like Co–Ni, Fe–Ni, Fe–No, Co–Cu, Ni–Cu, or Ni–Ti) [43].

High-temperature preparation methods, such as arc discharge, are currently being replaced by laser ablation [46–48] and low-temperature chemical vapor deposition (e.g., <800 °C) [49–51], because the orientation, alignment, nanotube length, diameter, purity, and density of CNTs can be controlled precisely with these new techniques [52]. However, for most of those techniques, supporting gases and a vacuum are required. Because gas-phase methods are volumetric, they are appropriate for applications such as composite materials that require large quantities of nanotubes and industrial-scale fabrication methods to make them economically practicable [43]. However, the disadvantages of gas-phase synthesis methods are low catalyst production, where only a small percentage of the catalysts form nanotubes, short catalyst lifetimes, and low catalyst number density [53]. While many

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