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Selected topics on the synthesis, properties and applications of multiwalled carbon nanotubes

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1. Introduction

Carbon nanotubes (CNTs) have an astounding array of properties that make them interesting candidates for numerous applications. The geometric and chemical variations found in CNTs provide a rich area of study for both science and applications [1,2]. These variations are created by the unique bonding configurations of carbon that make it a ubiquitous part of our environment. The one dimensional nature of the basic CNT structure enabling ultra-high surface area, the ability to act as a semiconductor or a metal, the existence of multiple direct bandgaps, the relative ease of attachment for numerous chemical functional groups and ability to decorate CNTs with nanoparticles, all drive an array of scientific and technology issues that have been studied by research groups across the globe in recent years [3,4]. A variant of the standard nanotube can be found by integrating the CNT structure with another unique and valuable property of carbon; its anisotropy. The well-known difference between the basal plane and z-direction properties of graphite for example translates directly into anisotropy between the longitudinal and transverse properties of carbon nanotubes. This large anisotropy in structure and properties is the basis of research exploiting edge vs. basal plane properties of graphite and graphene and increases the variety of properties and applications for CNT systems [5]. In particular, graphene edges are expected to be more reactive, hold a higher charge density and concentrate electric fields as in the case of nanowalls or nanosheets [6,7].

With the recent increased understanding of the formation of graphene and its nanostructural compatibility with CNTs, an opportunity exists to enhance CNT properties by integrating the edge properties of graphene with the CNT one dimensional framework. Graphenated carbon nanotubes (g-CNTs) are one way to achieve this hybrid structure [8,9]. This enables an optimization of charge and reactivity per unit volume not previously possible at the nanoscale. It is an engineered network with the concentration of high charge density, high reactivity edges arranged in three dimensional nanoscale space. This enables ultra-high surface area coupled with high charge density. To put these structures into context, we have previously introduced a diagram that classifies the nanostructures by their edge density; the EDGE Triangle [10]. Perhaps the most promising nanostructure involves the use of these g-CNTs in an aerogel network which has yet to be explored [11].

Enhanced charge density and reactivity are expected to improve performance for a variety of applications, including; energy storage (e.g., batteries, supercapacitors), energy conversion (e.g., fuel cells), electrochemical sensors, electrodes for neural stimulation, field emission sources and electrodes for industrial processes (e.g., materials synthesis, purification). Similarly, one-dimensional nanostructures, such as oxide or metal nanoparticles, can be integrated with CNTs to provide enhanced functionality for many of these same applications. These decorated CNTs can be fabricated using post deposition processing or during growth [12,13]. Many fundamental issues of CNT growth translate to the growth of integrated graphene–CNT or nanoparticle–CNT composites; for example, the use of bulk synthesis in the gas phase vs. synthesis on a substrate, the role of the catalyst to enhance growth, the gas phase precursor ratios or the presence of additional reactive species in the gas phase and post deposition processing. Polymer–CNT composites formed by post deposition processing also provide enhanced functionality in these applications. This review will cover the broad area of conventional CNTs growth as well as the formation of hybrid nanostructures, such as g-CNTs.

Due to the remarkable variation in carbon nanotube properties, (e.g., from semiconducting to metallic and inert to reactive), numerous applications are of potential interest. In this article we will focus on properties related to electrode applications addressed by multi-walled carbon nanotubes (MWCNTs). Integrated graphene–CNT materials are expected to be candidates to improve these applications even beyond the already exciting CNT results. Similarly nanoparticle–CNT hybrid structures are also able to combine the best properties of CNTs and functional nanoparticles. We will not address semiconducting, single wall CNT applications. Although of significant interest, these applications are likely farther from commercialization and simply present a breadth of variety which would be difficult to cover in this limited review.

2. Structure and properties

2.1. Structural considerations

With the growing importance of energy applications in particular, a need has emerged to characterize the various forms of activated and nanostructured carbons in the context of their electrochemical charge-storage capacity. However, the use of gravimetric or area specific capacitance can be uninformative or even misleading due to the critical role of exposed linear edge density on charge-transfer processes; edges exhibit a 20× greater specific capacitance than basal planes [14]. For example, edge plane sites, either at the tips or along the length of CNTs can dominate both the chemical and electronic properties of the system, thus, a more robust normalization is needed for materials that possess this extreme anisotropy. By classifying nanostructures based on linear edge density, a deeper understanding of material performance can be obtained and a more informed comparison of nanostructures is enabled. Recently, we proposed a classification for carbon nanostructures based on the dimensional organization of their edge structures (Fig. 1) called the EDGE (Electron Density of Graphene Edges) Triangle [10]. This classification is organized based on the dimensional structure of sp^2 bonded carbon edges (i.e., edges of 2 dimensional carbon sheets that occur in graphite, graphene and even the tips of CNTs). Morphological benchmarks of the classification are provided, including the unique ability of CNTs to deposit in a vertically aligned forest and a recently reported [8] graphenated CNT (g-CNT) hybrid which increases the linear edge density of nanostructured carbons by an order of magnitude. As more and more applications draw upon the combined superlative properties of CNTs, graphene, and other nanostructured carbons, this dimensional classification provides new insights into both the optimization of existing materials as well the development of new hybrid nanostructures.

2.2. Electronic properties

MWCNTs have been shown to exhibit ballistic transport of charge carriers and possess exceptional electrical conductivity (1.85×10^3 S cm^{-1}) [15]. Current densities approaching 10^9 A cm^{-2} have also been reported [16]. Coupled with their robust mechanical properties, this makes them a strong candidate as interconnects for microelectronic devices and electrodes in numerous applications. Although MWCNTs exhibit metallic conductivity, their band structure varies considerably from the model of a free electron in a metal. Interesting and complex effects arise from the concentric walls of the MWCNT as well as from quantization of the electron wave function around the nanotube circumference with varying diameters. The multiple concentric walls of the MWCNT allow modification of the outer walls with adsorbates and functional groups while maintaining mechanical integrity and electrical conductivity. For example, the formation of graphene–CNT and nanoparticle–CNT hybrid structures

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