

Not all Carbon Nanotubes are Created Equal

Gyula Eres

Oak Ridge National Laboratory, Oak Ridge, TN 37831

The outstanding properties of carbon nanotubes (CNTs) that stem from their unique molecular structure have created enormous excitement promising a flood of new applications. However, after nearly two decades of research even applications that appeared easily feasible are slow to materialize. This situation developed because the intrinsic structure, purity and perfection of CNTs turned out to be much harder to control than it was anticipated initially. Progress is slow primarily because the molecular level growth mechanisms are poorly understood. In this talk I discuss the effect of synthesis methods on the product distribution and the purity of CNTs from the perspective of current understanding of the synthesis science of carbon network formation.

The extreme reaction conditions used in most CNT synthesis methods wipe out all specificity toward a particular precursor and broaden the distribution of the final products. Chemical vapor deposition (CVD) of CNTs in a form of vertically aligned nanotube arrays (VANTA) is an important advance. VANTA growth occurs under milder conditions and most importantly it is compatible with real-time monitoring that enables growth kinetic studies. Detailed mechanistic studies of CNT formation are made difficult by the extremely complex CVD reaction environment. However, identifying the reaction type can provide important clues about the products and their distribution that is necessary to make the first step toward meaningful control of the reaction outcome.

We combine a molecular beam growth environment with real-time characterization techniques to search for characteristic kinetic features of CNT growth that help in identifying the overall reaction type. This work identified acetylene as the primary precursor that must be supplied directly or formed as an intermediate reaction product for CNT growth to occur. Real-time monitoring shows that the CNT growth rate changes throughout the entire process without ever settling at a steady level. Other kinetic features that help narrowing down the possible reaction type include a threshold for growth and acetylene flux dependence of not just the growth rate but the diameter of the CNTs. After an initial induction period the growth process enters into an autoaccelerating stage. The autoacceleration stage transitions into a protracted deceleration stage that is universally recognized in termination of CNT growth. These kinetic features are at odds with the prevailing picture according to which CNT growth occurs by precipitation of carbon from a metallic particle of certain size that determines both the diameter and the chirality of the CNT. Instead, I present a general framework in which carbon species catalyze rapid C-C bond formation and the incorporation of new carbon into a growing sp^2 carbon network, and discuss the implication of such a process on the product distribution in CNT synthesis that is relevant for practical applications.