Time-Resolved Diagnostics of Single-Wall Carbon Nanotube Synthesis by Laser Vaporization

A. A. Puretzky,
Department of Materials Science and Engineering, University of Tennessee, Knoxville, TN
E-mail:1ap@ornl.gov

D. B. Geohegan, H. Schittenhelm, X. Fan, M. A. Guillorn, P. F. Britt, and S. J. Pennycook
Oak Ridge National Laboratory, Oak Ridge, TN

The laser vaporization of carbon targets which contain metal catalysts is one of the most versatile techniques for the synthesis of single wall carbon nanotubes (SWNT) for electronic and structural applications. Unfortunately, there is very little agreement on the mechanisms or rates for SWNT growth, which are key elements for the production of SWNT with controlled properties or approaches to scale their production.

In our study several different time-resolved spectroscopic techniques are applied in situ to form a comprehensive picture of the SWNT growth process by laser vaporization, including intensified CCD-array imaging of Rayleigh scattered laser light and laser-induced luminescence, plasma and laser-induced emission spectroscopy, absorption spectroscopy, and pyrometry. The diagnostics demonstrated that SWNT grow in a propagating vortex ring microreactor over extended periods of time by condensed phase conversion from a feedstock of aggregated nanoparticles. The general picture of SWNT growth based on these diagnostics will be discussed using time, space and temperature coordinates.

The imaging diagnostics are applied, along with ex situ TEM and FESEM analysis of collected deposits for: 1) synthesis and characterization of short SWNTs to control their size and estimate the growth times and rates; 2) condensed phase growth of SWNT by annealing treatments to develop a new synthesis approach for electronic and structural materials; 3) for synthesis of SWNT by long pulse laser vaporization to modify the growth conditions, increase the yield and produce raw material which can be purified easier.

The SWNT growth rates were estimated by limiting the growth time at elevated temperatures. Two different cases of laser vaporization (LV) were investigated: 1) short-pulse LV at 1000°C (8 ns, 1.06 μm); 2) long-pulse LVat 1000°C (a train of ~80, 200ns-pulses, total length 200 μs, 1.06 μm).

Raw material collected from different locations in the chamber (indicated by the in situ diagnostics) was characterized by Raman spectroscopy, TEM, and FESEM, and thermogravimetric analysis (prior to and following various chemical etches and oxidative purification treatments) to investigate the SWNT growth mechanism and to optimize the processing conditions.

This research was sponsored by the U.S. Department of Energy and the Laboratory-Directed Research and Development Program at ORNL under contract DE-AC05-00OR22725 with the Oak Ridge National Laboratory, managed by UT-Battelle, LLC.