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Time-Resolved Measurements of Carbon Nanotube and Nanohorn Growth

DAVID GEOHEGAN, Oak Ridge National Laboratory

Mechanisms for carbon nanotube growth have been investigated for both laser vaporization (LV) and chemical vapor deposition (CVD) synthesis techniques through the use of time-resolved, in situ laser-based diagnostics for the measurement of absolute growth rates. Optimization of both the production of loose single-wall carbon nanotubes (SWNTs) by LV and the sustained growth of mm-long, vertically-aligned carbon nanotube arrays (VANTAs) by CVD are described. For SWNT growth by laser co-vaporization of carbon and trace metal catalysts at high (1200 $^\circ$C) temperatures, nanotubes are found to grow at $\sim$ 1–5 microns/second to lengths of only several microns, as determined by gated-ICCD imaging and laser spectroscopy of the plume of ejected material. Efforts to scale the LV production of SWNTs utilizing an industrial Nd:YAG laser (600 W average power, 1-500 Hz repetition rate, 0.5-10ms pulse width) are described. In addition to vaporizing material at much higher rates, the high-power laser irradiation provides sufficient plasma plume density and temperature to enable the growth of novel single-wall carbon nanohorn (SWNH) structures without the need for metal catalysts in the target. Applications of these SWNH structures as metal catalyst supports will be discussed. Through the application of time-resolved reflectivity and direct imaging, CVD growth of VANTAs from hydrocarbon gases at sustained rates of $0.2 - 0.5$ microns/second have been directly measured over millimeters of length at lower ($\sim$ 700 $^\circ$C) temperatures. Now, through a new laser-CVD setup at the ALPS (Advanced Laser Processing and Synthesis) facility at ORNL, high-power laser heating is being employed for the fast and position-controlled growth of carbon nanotubes on substrates. In situ fast optical pyrometry is employed to record the rapid thermal processing of metal-catalyst-prepared substrates to investigate the nucleation and early growth behavior of CVD-grown nanotubes. New nanotube growth and tunable Raman spectroscopy facilities at the Center for Nanophase Materials Sciences at ORNL will be outlined. In collaboration with Alex Puretzky, Zuqin Liu, David Styers-Barnett, Christopher M. Rouleau, Hongtao Cui, Ilia Ivanov, Bin Zhao, and Hui Hu, Oak Ridge National Laboratory and the Center for Nanophase Material Sciences.