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Nanotubes in a Flash—Ignition and Reconstruction

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Single-walled carbon nanotubes (SWNTs) exhibit a range of unusual mechanical and electronic properties because of their unique structure and dimensions. Here we report another unusual property. We accidentally discovered that SWNTs ignite when exposed to a conventional photographic flash. This photoeffect occurs for SWNTs prepared by the carbon arc, laser ablation, or chemical vapor deposition upon exposure to a camera flash at close range (several cm away from the sample). Ignition did not occur for multiwalled nanotubes, graphite powder, fluffy carbon soot, or C_{60}.

Frames taken from a real-time video recording of burning SWNTs after the application of the photoflash (Fig. 1, A and B) show red hot spots immediately after the flash (Fig. 1B). The sample burns down in air, generating CO and CO_2 and leaving behind oxidized catalyst particles (such as Ni-Y or Fe, used for SWNT synthesis) and traces of disordered carbonaceous materials. The effect we describe occurs only on dry, "fluffy," as-prepared nanotube samples (see below). After flashing SWNTs, we observed an associated large photoacoustic effect caused by the absorption of incident light on the SWNT samples, in which acoustic waves are caused by the expansion and contraction of trapped gases (*1*).

Ignition and burning occur when local increases in temperature are sufficient to initiate the oxidation of the carbon and propagate as more heat is released by this exothermic reaction. Flashlight consists of a combination of various wavelengths, which intend to emulate sunlight (without the ultraviolet light). In air, the average light power needed to ignite SWNTs was found to be 100 mW cm^{-2} ($\pm$20 mW) for a sample density of about 0.2 g cm^{-3} (pulse rise time: 50 μs; decay time: 1.2 ms). When the sample is compacted to higher densities, larger power is needed to ignite SWNTs; for densities >1 g cm^{-3}, ignition occurs at about 300 mW cm^{-2}. At higher densities, the tubes are less prone to catching fire because of the lack of oxygen access and loss of heat into the bulk of the sample.

Thermal conductivity of nanotubes along the tube axes is expected to be very high (*2–4*). Bulk SWNTs form bundles that crisscross each other in the pristine samples (Fig. 1C). The heat pulse generated by the absorption of flashlight will initially be confined to the tubes within a bundle, especially along their axes. The high trapped thermal energy densities, necessary for ignition, are most easily attained when the bundles are separated and surrounded by oxygen, and the heat wave is locally confined in the nanotube structures. As the material is compacted, more and more bundles are in contact with each other, and the heat is rapidly dissipated into the bulk.

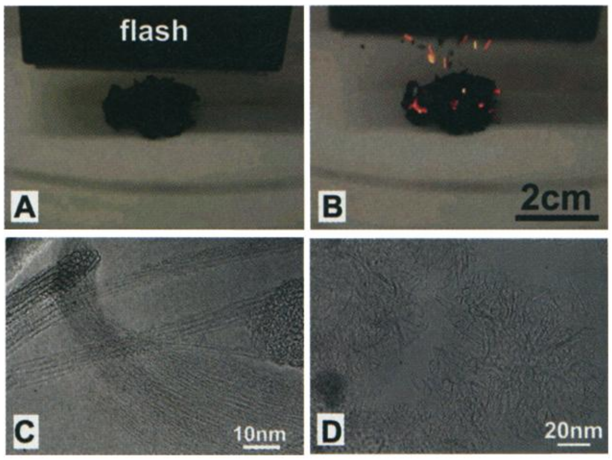

Fig. 1. (**A** and **B**) Sequence of burning of SWNT: (A) original sample (about 2 cm outer diameter) showing the flash on top; (B) sample soon after flashing exhibiting the ignited SWNT material with burning red and yellow spots. (**C**) High-resolution transmission electron microscopy (HRTEM) image of pristine SWNT, in which a cross section of an individual bundle is clearly observed. (**D**) Typical HRTEM image of remaining carbonaceous material obtained after flashing SWNTs in air; the presence of reconstructed single-walled structures such as nanohorns is noteworthy. See the supplementary material for a movie of the flash and the burning SWNT (*11*).

Electron microscopy studies of the SWNT samples on microscope grids flashed at atmospheric pressures of air, He, Ar, and in vacuum (about 1×10^{-4} torr) reveal that the material undergoes surprisingly large structural reconstruction even in the absence of burning. In air, much of the remaining carbonaceous material (which cooled off without burning) is transformed into single layered structures with many conical tips reminiscent of nanohorns (*5*) (Fig. 1D). Under vacuum and Ar, substantial reconstruction results in partially graphitized filaments and disordered graphene (*6*). In contrast to Ar, He atmosphere flash experiments exhibit very few remaining nanotube structures and large amounts of nanohorn material. The reason may be the much lower thermal conductivity of Ar (0.0177 W m^{-1} K^{-1}) than of He (0.152 W m^{-1} K^{-1}).

Because carbon nanotubes oxidize at about 600° to 700°C (*7*, *8*), the samples must have reached such temperatures at the light power threshold necessary for ignition. But local temperatures must be much higher for the extensive reconstruction to occur. SWNTs are known to fuse into large tubes at temperatures between 1500° and 2000°C (*9*, *10*). The reconstruction into new morphologies requires bond breakage and rearrangements of several carbon atoms. Therefore, the effective transient local temperature within the tubes must be at least ~1500°C.

Heat confinement in nanostructures can thus lead to drastic structural reformations and, under oxidizing environments, induces ignition under conditions not expected for bulk materials. The heat pulse here is created by light absorption by the nanotubes from a proximal light flash. This opto-thermal behavior suggests applications in remote light-induced ignition and triggers.

References and Notes

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10. Upon fast annealing of SWNTs at ~1500°C in He, SWNTs underwent structural transformation into multiwalled tubes and coalesced, larger SWNTs.
11. Supplementary material is available on *Science* Online at www.sciencemag.org/cgi/content/full/296/5568/705/DC1.
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