



Nanodiamonds with defect centers formed using a doped carbon aerogel

The creation of defects in diamond by doping the lattice with foreign atoms can give rise to new properties that can be exploited in a wide range of applications. A team of researchers from the University of Washington, US Naval Research Laboratory, and Pacific Northwest National Laboratory has created various defect centers in nanodiamonds by exposing a doped carbon-based aerogel to high pressures and temperatures in a diamond anvil cell. Using their method, published recently in *Scientific Advances* (doi:10.1126/sciadv.aau6073), scientists may be able to create new, polyatomic defect centers in nanodiamonds using targeted molecular precursors. Because diamond is hard and inert, doped nanodiamonds could also be used for applications such as tracking the delivery of targeted drugs in the body.

Defects in nanodiamond are typically created by replacing carbon atoms in the diamond lattice with silicon or nitrogen atoms. One of the most important and well-studied defects is the negatively charged nitrogen vacancy (NV⁻) center. “This defect has a long electron spin coherence time at room temperature,

making it useful for a range of quantum sensing, quantum communication, and quantum computation applications,” says Peter Pauzauskie of the University of Washington, who led the research study.

In common with bulk diamond, nanodiamond fabrication requires simultaneous high pressures and temperatures where fabrication methods include chemical vapor deposition, high explosive detonation, or the milling of bulk diamond. Dopants are introduced through these processes or after synthesis using ion implantation. The synthesis conditions required for these methods, however, can often lead to significant damage of the nanodiamond crystal lattice because of the energies of the impinging ions, and offer little control over defect site formation.

“[This] most recent work incorporates silicon-vacancy defects and trapped argon into nanodiamonds without ion implantation and its associated lattice damage,” says Jagdish Narayan of North Carolina State University, who was not involved in the study.

To accomplish this, Pauzauskie’s team incorporated tetraethylorthosilicate (TEOS) molecules into a carbon-based aerogel. During processing, the doped aerogel was also exposed to nitrogen gas and nitrogen-based solvents. The team compressed the doped aerogel in an

argon-filled diamond anvil cell while being heated by a laser. After being subjected to pressure over 20 GPa and temperatures greater than 2000 K, the aerogel transformed into nanodiamonds with silicon, nitrogen, and argon lattice defects.

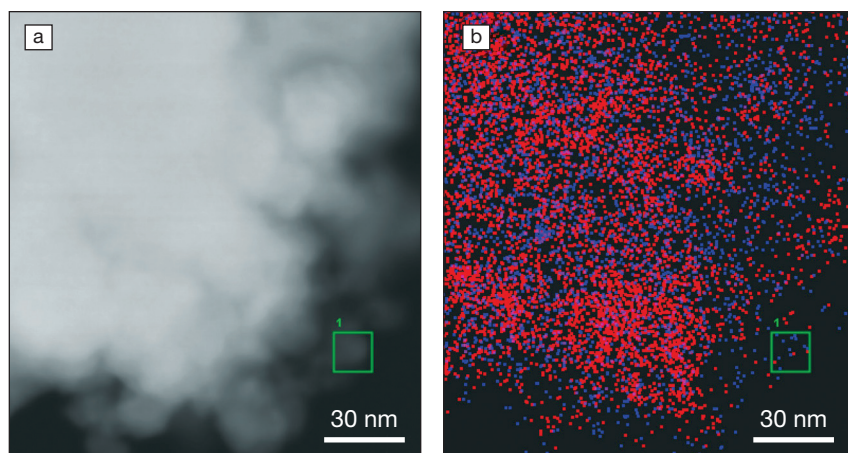
Many defects have specific spectroscopic signatures. Different spectroscopic methods were thus used to characterize the various defects in the nanodiamonds. Because of their higher atomic weight, silicon and argon defects could be observed using scanning transmission electron microscopy energy-dispersive x-ray spectroscopy (STEM-EDS). These revealed a homogenous distribution of silicon and argon defects. Photoluminescence revealed the presence of zero- and negatively charged nitrogen vacancies (NV⁰ and NV⁻, respectively) as well as negatively charged silicon vacancies (SiV⁻). The argon defect was not optically active.

This study thus presents a general method to produce, in a controlled manner, defect centers (also called color centers) in diamond, yielding designer defects.

“The approach we have developed has the potential to create or discover more complex, polyatomic point defects that cannot be made through conventional ion implantation and thermal annealing,” Pauzauskie says. This technique could allow new nanodiamond defect sites to be realized, if the atomic bonding and orientation are preserved from the molecular precursor to the final defect site. Understanding how various moieties, such as alkoxy groups, in the molecular precursor affect the behavior of the defect site will also be important, Narayan says.

Using this method, researchers could also investigate interstellar nanodiamond formation, potentially answering questions about their formation in systems outside of the solar system. According to Pauzauskie, “We are eager to learn more about how noble gases like xenon can be incorporated within nanodiamonds at extreme high pressure conditions to gain a better understanding of how noble gases are incorporated within extrasolar nanodiamonds.”

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(a) Wide-area high-angle annular dark-field scanning transmission electron microscope image and (b) corresponding scanning transmission electron microscopy energy-dispersive x-ray spectroscopy elemental mapping of argon (red) and silicon (blue) of the recovered tetraethylorthosilicate-doped carbon aerogel. The green squares correspond to the field of view in image (a). Credit: *Scientific Advances*.