DFT studies of hydrogenated and defective carbon nanotubes

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The study of the effects of covalent functionalization on the mechanical properties of real-size carbon nanotubes (CNTs) by means of computer simulations requires the adoption of reliable models and of a robust and accurate methodology. On one hand the correct description of the changes induced by the addition of chemical bonds requires a quantum-mechanical approach; on the other hand the size of the systems (typically 1µm long) imposes the use of classical force fields. The latter however need validation against non-empirical methods.

We present the results of two projects: one aimed at identifying the evolution of the pattern of chemisorbed hydrogen on the outer surface of a CNT with increasing concentration [1]; the other at characterizing the stable reconstructions of the CNT after formation of vacancies. These studies include a comparison of results obtained with DFT using several exchange- and correlation-functionals and with the widely-used force-field AIREBO. The system under consideration is a zig-zag semiconducting (10,0) nanotube.

DFT calculations show that electron pairing and strain minimization lead hydrogen atoms to cluster. The underlying mechanism is explained as well On the contrary, AIREBO predicts a sparse distribution of the hydrogen atoms on the surface of the tube. We investigate the changes induced by hydrogen in the electronic structure and in the infrared spectrum of the nanotube [1].

Our DFT calculations predict that configurations with double vacancies are thermodynamically favored with respect to single vacancies, as found in previous studies [3]. A number of differences are however found with previous DFT calculations and especially with the results obtained with the AIREBO potential. The only agreement concerns the lowest energy state (5r8r5r) of a double vacancy.

References

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