The electronic properties of sulfur and SOx-based sp3 defects in single-walled carbon nanotubes from first-principles theory

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Semiconducting single-walled carbon nanotubes (SWCNT) containing sp3-defects are promising components in optoelectronic devices because of their bright tunable photoluminescence, long spin coherence, and demonstrated single-photon emission. Modifying the defect functional group leads to highly selective and tunable properties within the SWCNT. We present a first-principles computational study of the chiral (6,5) SWCNT, functionalized by SO_2 and its likely derivatives. Using density functional theory (DFT), we investigate the nature of binding of SO_2 and its four derivatives (S, SO, SO_3, and H_2SO_4) on the SWCNT including the role of hydrogen passivation of the defect site. Our calculations, in comparison with experiment, indicate that SO_2 preferably decomposes to its substituents. We show that the formation of a defect site for SOx-based defects is dependent on both the substituent and the location of binding on the SWCNT.

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