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New insights into the solubility of graphene oxide in water and alcohols

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Abstract

© The Owner Societies 2017. One of the main advantages of graphene oxide (GO) over its non-oxidized counterpart is its ability to form stable solutions in water and some organic solvents. At the same time, the nature of GO solutions is not completely understood; the existing data are scarce and controversial. Here, we demonstrate that the solubility of GO, and the stability of the as-formed solutions depend not just on the solute and solvent cohesion parameters, as commonly believed, but mostly on the chemical interactions at the GO/solvent interface. By the DFT and QTAIM calculations, we demonstrate that the solubility of GO is afforded by strong hydrogen bonding established between GO functional groups and solvent molecules. The main functional groups taking part in hydrogen bonding are tertiary alcohols; epoxides play only a minor role. The magnitude of the bond energy values is significantly higher than that for typical hydrogen bonding. The hydrogen bond energy between GO functional groups and solvent molecules decreases in the sequence: water 4 methanol 4 ethanol. We support our theoretical results by several experimental observations including solution calorimetry. The enthalpy of GO dissolution in water, methanol and ethanol is 0.1815 0.0010, 0.1550 0.0012 and 0.1040 0.0010 kJ g1, respectively, in full accordance with the calculated trend. Our findings provide an explanation for the well-known, but poorly understood solvent exchange phenomenon.

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