MAR11-2010-002396

Abstract for an Invited Paper
for the MAR11 Meeting of
the American Physical Society

Theory of molecular hydrogen sorption for hydrogen storage[1]

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Molecular hydrogen (H_2) sorption has the advantage of fast kinetics and high reversibility. However, the binding strength is often too weak to be operative at near room temperatures. Research into such hydrogen sorption materials has branched into the study of pure van der Waals (vdW) physisorption and that of weak chemisorption (known to exist in the so-called Kubas complexes). In either case, however, theoretical tools to describe such weak interactions are underdeveloped with error bars that often exceed the strength of the interaction itself. We have used quantum-chemistry (QC) based approaches to benchmark the various available DFT methods for four classes of weak chemisorption systems [Sun et al., Phys. Rev. B **82**, 073401 (2010)]. These involve complexes containing Li, Ca, Sc, and Ti with increased strength of H_2 binding from predominantly vdW to mostly Kubas-like. The study reveals that most of the DFT functionals within the generalized gradient approximation underestimate the binding energy, oppose to overestimating it. The functionals that are easy to use yet yielding results reasonably close to those of accurate QC are the PBE and PW91. I will also discuss the effort of implementing vdW interaction into the currently available density functional methods [Sun, J. Chem. Phys. **129**, 154102 (2008)]. The rationale is that while the true vdW is an electron-electron correlation, a DFT plus classical dispersion approach may be too simple and unnecessary within the DFT. A local pseudopotential approach has been developed to account for the core part of the polarizability of the elements. Applications to a number of benchmark systems yield good agreement with QC calculations. The application of this method and the QC methods to vdW hydrogen binding will also be discussed.

[1]Work supported by DOE/BES and DOE/EERE Hydrogen Sorption Center of Excellence under RPI subcontracts No. J30546/J90336.