Molecular vs atomic encapsulation of hydrogen in metal cluster-cage assemblies

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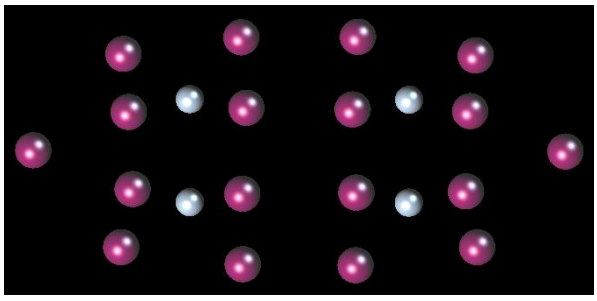
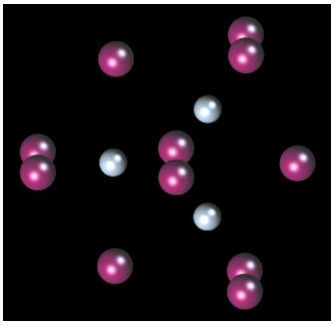
Presented are results of ab initio calculations for main-group light-metal cluster cages predicted to be able to accommodate hydrogen endohedrally, with a diatom or atom per cage for different isomers [1-3]. Such systems can also preserve their integrity when assembled into larger aggregates.

The H_2 molecules are found to dissociate due to electron-transfer from the surrounding cages which, however, confine the produced pairs of hydrogen anions electrostatically suspended inside, resulting in metastable systems. The system stabilities are characterized in terms of dissociation into hydrogen and empty cage/assembly, and of energy barriers for hydrogen exit.

Alterations of the cage structures and properties upon insertion of hydrogen are also discussed. Peculiar features concern simultaneous exit of two H atoms, relative stability of aggregates built of different isomers of monomers, specific geometry-stability relationships.

With each such hydrogen-filled cage representing a "nanobubble", the assemblies extrapolated to bulk material can be viewed as filled metal-nanofoams, with hydrogen storage capacity estimated as up to 10 weight-%. Furthermore, the metastability suggests low-temperature storage conditions and a relatively easy hydrogen release, unlike many alternatives schemes needing excessively high temperatures.





[1] Naumkin, F. N., Wales, D. J., *Chem. Phys. Lett.*, 545:44, 2012.
[2] Naumkin, F. N., Wales, D. J., *Int. J. Quantum Chem.*, 112:3068, 2012.
[3] Naumkin, F. N., Wales, D. J., *J. Phys. Chem. A*, 115:12105, 2011.