Comparison of potential energy surface between carbon based and silicon based fullerene configurations

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Abstract

Carbon-based nanostructures such as fullerenes, nanotubes, nanosheets, and carbon based polymers are common building blocks. Similar structures do exist for Si as well, namely linear polysilanes, silicon nanosheets, and nanotubes. Therefore it does not come as a surprise that the discovery of such carbon based structures triggered the quest for analogous Si structures. While carbon atoms can readily adjust their valence states to participate in single, double and triple bonds, silicon strongly prefers sp^3 hybridization and single bonds. Therefore, although C_{20} is the lowest stable fullerene structure, quantum chemical calculations show the Si_{20} fullerene to be highly unstable The stability of a cage structure can be enhanced by modifications. Exohedral passivation of the silicon dangling bonds of the Si atoms in the Si_{20} dodecahedron by hydrogen attachment is one way to stabilize the cluster. Theoretical studies have identified the dodecahedron of $Si_{20}H_{20}$ as the lowest energy structure among many competing configurations several years ago. However even though, the carbon analogue, $C_{20}H_{20}$, has been synthesized more than two decades, this ground state structure has never been observed experimentally. The characteristics of an energy landscape are ultimately the key to understand the existence of a particular configuration. We employ a fingerprint distance (FP)- based exploration of the Potentail Energy Surface, [2, 1], to establish relationships between the minima and their connecting transition states (saddle points of the PES). We apply this in a comparative investigation of $Si_{20}H_{20}$ with two existing structures, namely the C_{60} and $C_{20}H_{20}$ fullerenes. The striking differences between the PES of these three systems are used to understand why the global minimum is experimentally observed only for C_{60} and $C_{20}H_{20}$. We show that the complex pathways between different metastable configurations and the ground state of the $Si_{20}H_{20}$ cluster explain the lack of observation of the global minima configuration in the experiment.

References

[1] Oren M. Becker and Martin Karplus. The topology of multidimensional potential energy surfaces: Theory and application to peptide structure and kinetics. *The Journal of Chemical Physics*, 106(4):1495–1517, 1997.

[2] Bastian Schaefer and Stefan Goedecker. Computationally efficient characterization of potential energy surfaces based on fingerprint distances. *The Journal of chemical physics*, 145(3):034101, 2016.