

Global Optimization of Si_xH_y and Si_xF_y Clusters at the *Ab Initio* Level

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Computational methods used in this research found F ligands cause dramatic change to the Si frameworks suggesting that different ligands could be used to tune the optoelectronic properties of the Si nanoclusters.

Background: Nanometer-sized Si clusters have promising optoelectronic properties which could be useful for various technological applications such as solar cells. Since the bare Si clusters are highly reactive, practical devices will most likely consist of Si clusters passivated by some ligand. H and F atoms are prototypical passivating ligands. In our research we

have been developing efficient strategies using non-empirical quantum chemistry calculations and genetic algorithms to globally search for the most stable Si_xH_y and Si_xF_y clusters. We obtained the diamond lattice-like global minima for the fully passivated $\text{Si}_{10}\text{H}_{16}$, $\text{Si}_{14}\text{H}_{20}$ and $\text{Si}_{18}\text{H}_{24}$ clusters and quite different structures for the incompletely passivated $\text{Si}_{10}\text{H}_{14}$, $\text{Si}_{14}\text{H}_{18}$, and $\text{Si}_{18}\text{H}_{22}$ clusters shown in Figure 1. We have also found the $\text{Si}_{10}\text{F}_{16}$ and $\text{Si}_{10}\text{F}_{14}$ global minima shown in Figure 2. Surprisingly, the F ligands cause dramatic change to the Si frameworks suggesting that different ligands could be used to tune the optoelectronic properties of the Si nanoclusters.

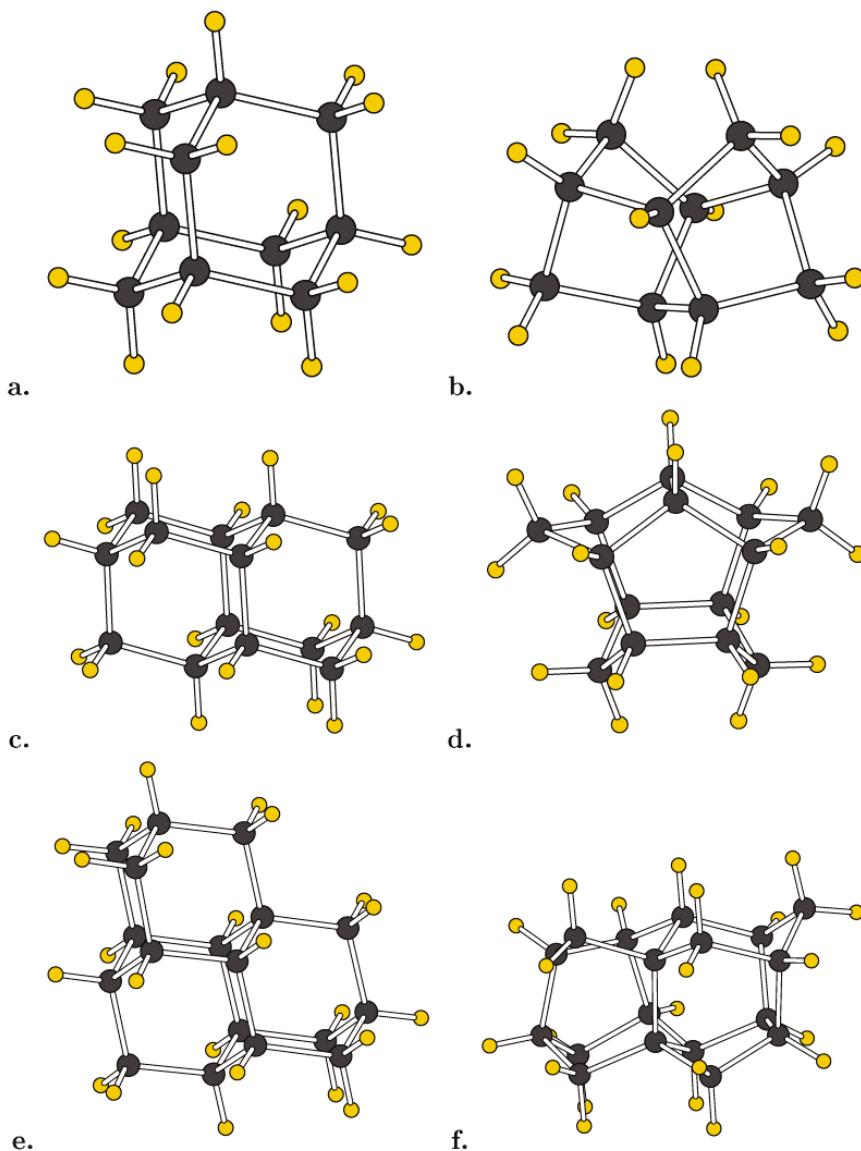


Figure 1. The MP2 global minima of various silicon hydride clusters: a. $\text{Si}_{10}\text{H}_{16}$, b. $\text{Si}_{10}\text{H}_{14}$, c. $\text{Si}_{14}\text{H}_{20}$, d. $\text{Si}_{14}\text{H}_{18}$, e. $\text{Si}_{18}\text{H}_{24}$, and f. $\text{Si}_{18}\text{H}_{22}$.

A recent publication describing the computational methods used in this work: Global Optimization of H-Passivated Si Clusters at the *Ab Initio* Level via the GAM1 Semiempirical Method, Y. Ge and J. D. Head, *J. Phys. Chem. B* 108, 6025-6034 (2004).

Research interests of this author include: Theoretical chemistry and electronic structure calculations of ground and excited state properties for large molecules, clusters, surfaces, and nanomaterials.

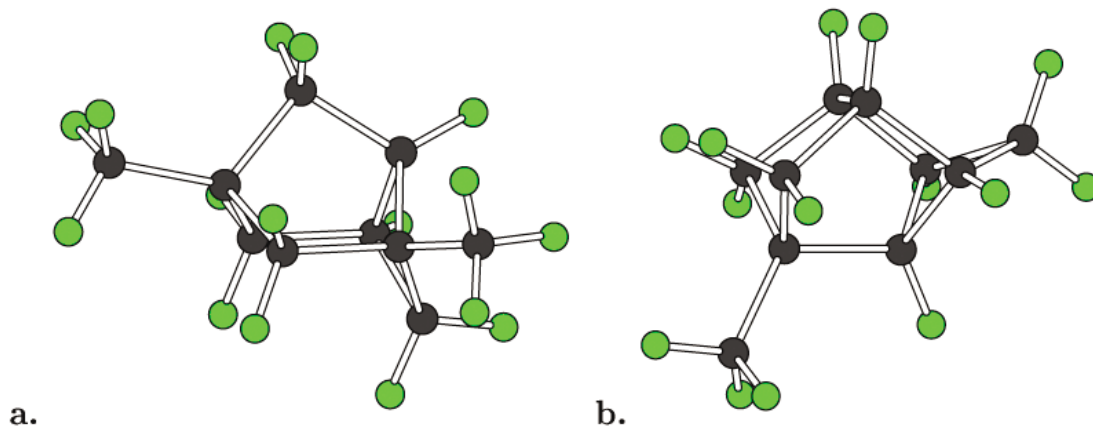


Figure 2. The B3LYP global minima of a. $\text{Si}_{10}\text{F}_{16}$ and b. $\text{Si}_{10}\text{F}_{14}$.

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