Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Under what conditions does $(SiO)_N$ nucleation occur? A bottom-up kinetic modelling evaluation

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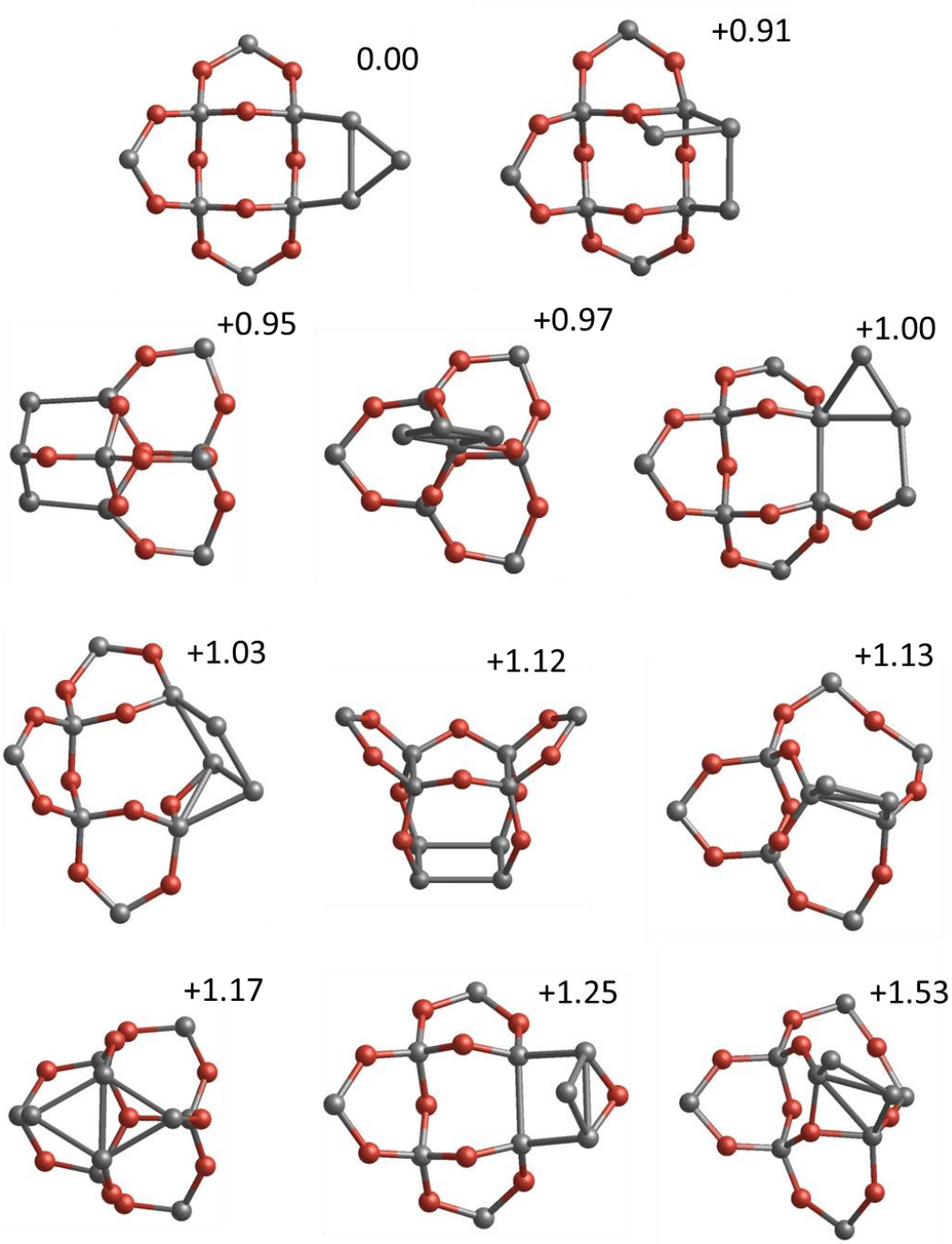
Supplementary Information 3

Simulated Annealing calculations using *ab initio* Langevin molecular dynamics:

We have performed *ab initio* Langevin molecular dynamics (LMD) based simulated annealing (SA) calculations using the Terachem code[1] with the B3LYP hybrid functional[2] and a 6-31G basis set on the $Si_{10}O_{10}$ system. The *ab initio* LMD-SA method was first reported as successfully applied to the global optimisation of small Si_n clusters.[3] Here we used three separate calculations, each from a different initial structure (the original $Si_{10}O_{10}$ structure as reported in the main text and two randomly generated connected structures) and each following the same protocol. Each LMD-SA run consisted of a series of contiguous LMD runs starting at 2000 K and each with a temperature subsequently reduced by 200 K until a temperature of 200 K was reached. We used a timestep of 2 fs and a Langevin damping time of 0.5 ps. For temperatures above 800K LMD runs of 5 ps were used and for the lower temperature runs we used 10 ps. Spherical boundary conditions were employed to avoid cluster evaporation and to encourage more compact cluster isomers. In addition to the final cluster structure, during each LMD-SA run we locally optimised the structures immediately after any isomerisation event was judged to occur. The lowest energy $Si_{10}O_{10}$ isomers found during the LMD-SA runs together with their total energy (eV) with respect to the reported putative global minimum are shown below.



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[2] P. J. Stephens, F. J. Devlin, C. F. Chabalowski and M. J. Frisch, *J. Phys. Chem.*, 1994, **98**, 11623-11627.

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