Under what conditions does $(\text{SiO})_N$ 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 code1 with the B3LYP hybrid functional2 and a 6-31G basis set on the Si10O10 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.
