Supplementary Information of Phonon Softening Induced Phase Transition of $CeSiO_4$: A Density Functional Theory Study

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Because quantum mechanics algorithm is based on a local energy minimization method, it is possible to dive into the local minima scope as meta-stable structure instead of ground state, when the unit-cell structure relaxes under high-pressure. To avoid this problem, the strain was added linearly by two different means, one way was to compress the unit cell from low-pressure to high-pressure, and the other way was to expand the unit cell from high-pressure to low-pressure. A cross check of the relaxed structures revealed that the structures were identical from compression and expansion, which proved the high-pressure structures are in the global energetic minima. To further validate that the structure resides within a global saddle point, a series of random perturbations for each atom within the structure, collecting energy values from thirty such perturbation tests. During these tests, random perturbations were introduced to the atomic positions in stetindite, varying from -0.1 Å to 0.1 Å from their equilibrium distances under high-pressure. The result is shown in Fig. S1, it was observed that the atoms consistently relaxed back to their original positions in every instance.

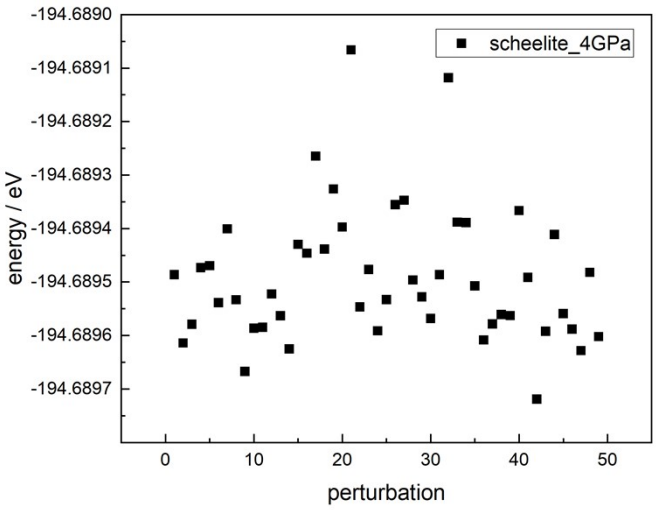

Figure S1 Random perturbations to the stetindite-Ce and scheelite-Ce systems. The x axis is the order label of each perturbation, and the y axis is the energy of relaxed structure after each perturbation. The energy difference is less than 0.001 eV, which indicates the lattice always relaxes to the same energetic minima each time, further proving the high pressure structure is in a global energetic minima point.

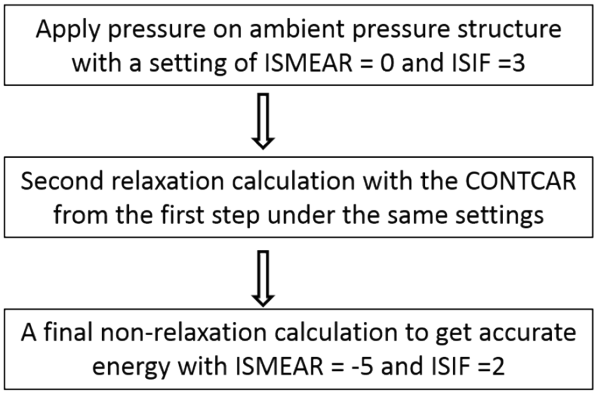

Figure S2 Three steps methodology of Pulay-stress addition to the lattice structure under high pressure.

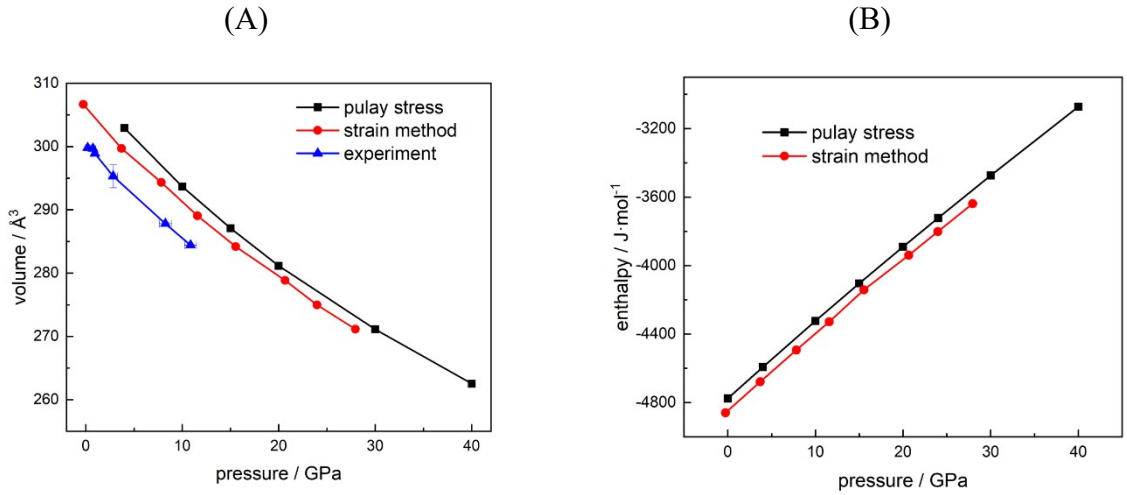

Figure S3 Comparison of the stress-strain and strain-stress methods in terms of the pressure-volume and pressure-enthalpy (A and B respectively).