

Pyrophyllite dehydroxylation process by First Principles calculations

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ABSTRACT

The crystal structure of triclinic pyrophyllite and its dehydroxylate derivative was studied with quantum mechanical calculations. The standard Kohn-Sham self-consistent density functional theory (DFT) was used through a linear-scaling DFT method with periodical boundary conditions in the generalized gradient approximation (GGA) with numerical atomic orbitals as the basis set. The calculations reproduce the lattice parameters found experimentally in pyrophyllite and its dehydroxylate derivative. The geometrical disposition of the OH bond in the crystal lattice and the hydrogen bonds and other electrostatic interactions of this group were analyzed. The frequencies of different vibration modes were calculated and compared with experimental data; the results show a good agreement. The dehydroxylation process, including different intermediates of this reaction, was investigated theoretically. The energetic differences are according to the thermodynamics of the experimental process. The semidehydroxylate derivative is identified, for the first time, as an important intermediate in this process, clarifying previous questions concerning the mechanism reported from the experimental data.