



## **Free energy minimization calculation: an efficient method to investigate serpentine minerals at variable pressures and temperatures**

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Atomistic simulation is a powerful tool to investigate the physical properties of minerals. Using the Gulp code (J. Gale, Molecular Simulation, 2003), energy minimization has been performed on lizardite, a hydrous magnesian 1:1 layer-silicate which belongs to the serpentine family. This mineral is of large geological interest, as this is the main serpentine variety occurring in hydrated oceanic peridotites. Its own stability field is still not clearly established (Evans, 2004), mainly because of the lack of reliable thermodynamic data concerning the serpentine minerals in general. Modeling lizardite during a structural relaxation including variations of pressure and temperature should provide insights on its behaviour along a geotherm as well as on a better understanding of its spectroscopic behavior.

Evolution of the total energy as a function of the lizardite unit-cell volume along with constant volume relaxations of all atomic degrees of freedom, converges to an energy that corresponds to the unit-cell dimensions and symmetry consistent with that of the experimental structure. These calculations also provide a cohesion energy between the layers of 0.32 eV (ie. 13 kJ per interlayer O-H...O bond). This is consistent with the accepted value for hydrogen bonding and also not so far from the energy obtained by *ab initio* calculations on the same mineral (Balan *et al.*, 2002 ; 19 kJ per bond). Finally, bulk and Young moduli are deduced from the elastic matrix analysis and the fit of the energy potential well using the Murnaghan equation.

The effect of pressure and temperature was also investigated. Remaining in serpen-

tine stability conditions, we evidenced that structural variations are mainly pressure-induced, temperature only playing a minor role which vanishes with increasing pressure. Pressure is mainly accommodated by a decrease of *c*-lattice parameter up to 10 GPa. These results are consistent with experimental data from Mellini & Zanazzi (1987), who investigated the structural modifications of lizardite up to 1.2 GPa. Lizardite density strongly increases with pressure. The bond length and angle analysis are compared to the results obtained in a high pressure Raman spectroscopic study (Auzende *et al.*, 2004) and are in good agreement with the observed shifts of the Raman modes.

This study also shows that the chosen set of interatomic potentials provides a reasonable description of lizardite. The extension of such work to antigorite, the higher pressure, modulated variety of serpentine, is now in progress.

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