Ab initio study on lattice thermal conductivity of minerals

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Thermal transport property of materials under pressure and temperature is of importance for understanding the dynamics of the solid Earth and the thermal history. However, both experimental and theoretical determinations of the thermal conductivity still remain technically challenging particularly at the deep mantle condition.

Recent progress in *ab initio* computational method based on the density functional theory (DFT) is making it possible to examine the transport phenomena including the lattice thermal conduction. The intrinsic bulk thermal conduction of insulator is essentially caused by lattice anharmonicity owing to phonon-phonon interaction. Determination of the anharmonic coupling constant is therefore the key to predicting the thermal transport property. Earlier theoretical works calculated the lattice thermal conductivity of MgO with *ab initio* molecular dynamics (MD) simulation or direct evaluation of the anharmonic force constants [1,2]. However, in these approaches, the simulation cell size could often be insufficient for accurate description of the long wavelength phonon scattering. As an alternative approach, the anharmonic coupling strength between phonon modes can be more efficiently and more accurately evaluated within the density functional perturbation theory (DFPT). In this approach, the higher-order force tensors are calculated based on the perturbative scheme taking care only of the primitive cell. We developed a technique for calculation of the phonon linewidh to obtain the phonon lifetime. Then the lattice thermal conductivity is evaluated combining with additional harmonic-level properties. In this presentation, we show the decay process of phonons and the lattice thermal conductivity of MgO and some typical minerals as a test for the applicability of our technique.

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Modelling of long-term diffusion-reaction in the Callovo-Oxfordian clay for radioactive waste confinement

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One of the French options for the final disposal of high-level radioactive waste is a deep geological repository into the Callovo-Oxfordian formation (COX, Haute-Marne, Meuse, France). In this concept, COX clay associated with compacted bentonite, or not, may constitute the barrier system. In such systems, the interactions between groundwater and clays, as well as between the corrosion products of steel overpacks and clays under the influence of temperature elevation, may modify the chemical and physical properties of the selected clay buffers. Clay material has a low permeability, and consequently molecular diffusion is the main mechanism of mass transport in a clay barrier. The system is modeled in reducing conditions using the KIRMAT code (KInetic Reactions and MAss Transport). The software has been developed [1] from the single-reaction path model KINDIS [2] generated from the purely thermodynamic code DISSOL [3, 4].

This study is focused on the possible feedback effects of geochemical reactions on the transport properties (porosity and diffusion) of COX clay. The results of modeling obtained after 10,000 years of simulated mass transport-reaction are compared to the previous studies on an engineer pure bentonite barrier [5, 6, 7, 8, 9]. The mineralogical modifications of clays in contact with the geological interacting fluid, and with Fe^{2+} and OH^- ions provided by the corrosion of the steel overpacks as well as the evolution of porosity and molecular diffusion will be discussed.

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