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Exploring new frontiers in fluids processes in subduction zones

Molecular modeling approaches to element speciation in aqueous fluids (invited)

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Aqueous fluids are key agents in subduction zone processes. They do not only change the physical properties of rocks but they are also able to dissolve and transport mineral components. They are released or absorbed during mineral reactions. A starting point for understanding the role of fluids in fluid-rock interactions is to investigate their molecular structure, especially the formation of various solute species, at relevant thermodynamic conditions. Joint experimental and numerical modeling efforts are required to reveal the full complexity of fluid speciation up to high temperatures and pressures. Here, we discuss different approaches for predicting thermodynamic properties of aqueous fluids from first principles simulations. By combining ab initio molecular dynamics simulations with advanced sampling methods such as thermodynamic integration or metadynamics we derive, e.g., species stability constants or acidity constants. The methodology is illustrated by examples of our recent studies of beryllium and yttrium speciation in fluorine- and chlorine-rich fluids. Finally, we discuss the reliability of present thermodynamic models of these systems and how first-principles simulations can help to improve them considerably, especially at high temperatures and pressures.