Abstract Submitted
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Density functional theory simulation of hydrogen-bonding vibrational densities of states at the quartz (101)-water interface and its relation to dissolution in aqueous solutions of ions[1] MARK DELLOSTRITTO, Department of Physics, The Pennsylvania State University, University Park, PA 16802, US, JAMES KUBICKI, Department of Geosciences and the Earth & Environmental Systems Institute, The Pennsylvania State University, University Park, PA 16802, US, JORGE SOFO, Department of Physics, The Pennsylvania State University, University Park, PA 16802, US — Physical processes at aqueous interfaces are strongly dependent on structure at the interface, and for water/metal-oxide interfaces, the structure of the local H-bond network dominates the behavior at the interface. In particular, for silica it has been hypothesized that the increase in dissolution rate due to dissolved salts is due to a reorganization of the H-bond network by ions near the surface. This would explain the order of magnitude increase in dissolution rate despite the fact that the activation energy of the dissolution reaction does not change with the addition of salts. We investigate two hypotheses of the dissolution of SiO_2 in ionic solutions using ab-initio molecular dynamics simulations. These hypotheses are 1) that the presence of ions induces orientations in interfacial H_2O molecules which are preferential for proton transfer to bridging oxygen (BO) atoms, and 2) the presence of ions induces stronger H-bonding between terminal hydroxyl (TH) groups and BO atoms, allowing proton transfer. It is found that although elements of these hypotheses are true, the model structures produced by density functional theory simulations do not support the former as valid mechanisms of dissolution.

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