

Modeling the effect of hydrogen bonding for understanding the dissolution of α -quartz surfaces in water

A.A. Skelton (1), D. J. Wesolowski (2) and P.T. Cummings (1,2)

(1) Dept. of Chemical and Biomolecular Engineering, Vanderbilt University, Nashville, Tennessee 37235-1604, USA

(2) Nanomaterials Theory Institute, Center for Nanophase Materials Sciences, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831-6494, USA

The interaction between water and two surfaces relating to different crystallographic faces of α -quartz have been atomistically modeled using two different force-fields, CLAYFF (1) and Lopes et al. (2). Comparisons with X-ray reflectivity experiments (3) have shown agreement between simulation and experiment for the $10\bar{1}0$ and $10\bar{1}\bar{1}$ α -quartz surfaces. The Lopes et al. force-field however has exhibited closer agreement with respect to axial densities, suggesting its greater accuracy in describing the quartz/water interface.

Although it has previously been reported that there are two possible terminations for the $10\bar{1}0$ α -quartz surface, only one of these surfaces shows agreement between experiment and simulation. It is postulated that the surface termination that is observed experimentally is the one more resistant to dissolution by water. In the less resistant surface, however, water-siloxane hydrogen bonds as well as hydroxyl-siloxane hydrogen bonds are formed. It is suggested that a concerted mechanism involving water, hydroxyls and siloxanes is responsible for the dissolution of the top surface layer thereby exposing the dominant termination.

Step edges for the $10\bar{1}0$ α -quartz (the dominant surface) have also been simulated in order to further illuminate the mechanism of dissolution. These simulations supply further evidence that the interplay between hydrogen bonds involving water, surface hydroxyls and siloxane oxygens is crucial in the dissolution of α -quartz.

1) R.T. Cygan et al, J. Phys. Chem. B 2004, 108, 1255

2) P.E.M. Lopes et al . J. Phys. Chem. B 2006, 110, 2782

M.L. Schlegel et al Geochimica et Cosmochimica Acta 2002, 66, 3037