

Studies of the Dissolution of Magnesite (104) in Acidic Aqueous Solutions: A new Approach Using Hydrothermal Scanning Probe Microscopy Under Controlled Hydrodynamic Conditions

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One of the key problems that is generally difficult to address in many studies pertaining to mineral dissolution and growth kinetics is the prediction of the near surface chemistry of the aqueous phase. With very sluggish surface kinetics it is usually assumed that mass transport near the surface is not important and thus, the kinetics of many oxide/water systems are not influenced by fluid transport. However, kinetic investigations by Scanning Probe Microscopy (SPM) necessarily require that the net flux at the surface is high enough for step motion to be observed over reasonable time scales. If the step kinetics are rapid enough to observe by SPM, then mass transport begins to be of concern with regard to its influence on the measured flux. Only recently has this problem been addressed quantitatively in SPM studies of heterogeneous kinetics by use of a precisely designed inlet jet (Coles et al., 1997). We will discuss this solution to the problem of controlled fluid transport in SPM fluid cells with special attention directed toward incorporation of the technology into the Hydrothermal SPM (Higgins et al., 1998). Initial results for studies of magnesite dissolution both

with and without the hydrodynamic inlet system will be discussed. We find in both experimental arrangements that the step velocity anisotropy defined as v_{st+}/v_{st-} is approximately 50 compared to 2-3 for that of calcite (Shiraki et al., 2000) at similar bulk pH conditions. The large anisotropy for magnesite may be used to infer relative kink detachment frequencies along the different steps. These inferences are further supported by the observed changes in step orientation with pH. Heterogeneous rate constants for proposed macroscopic mechanisms will be examined in light of the corresponding SPM images.

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