

Surface-specific measurements of olivine dissolution by phase-shift interferometry

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ABSTRACT

Natural olivine dissolution and replacement often occurs preferentially along specific crystallographic planes. Thus, olivine reactivity at specific surfaces was examined *in situ* using phase-shift interferometry, which has a detection limit $<10^{-5}$ nm/s, by dissolving two smoothed olivine crystal faces and a third sample corresponding to a surface that was generated by preferential dissolution along structural defects. The experiments were conducted at 22 °C and ambient pressure in 0.1 M NaCl solutions that were acidified to pHs between 1 and 4 using 0.1 M HCl. These experiments show that olivine dissolution can vary from one surface to another as well as in different areas of the same surface that have similar characteristics. The fastest vertical retreat occurred at the surfaces related to defects. However, only vertical advancement was observed at pH 1 on this surface consistent with the observation of isolated islands on the surface during atomic force microscopy investigations after the experiment. Raman analysis of the precipitated phase showed that it was not one of the thermodynamically stable phases expected from PHREEQC modeling. However, the correlation between the siloxane ring peak of amorphous silica with a similar peak in the precipitate spectrum, in conjunction with previous experimental and natural observations, indicates that the precipitate was a Si-enriched amorphous phase. Therefore, precipitation can facilitate the further dissolution of olivine on this surface as long as it does not completely armor the surface. Precipitate formation on surfaces associated with outcropping defects supports the natural observations of preferential dissolution and serpentinization along these defects implying that the fast dissolution of these surfaces will play a critical role during olivine replacement. In addition, comparison with flow-through experiments indicates that outflow fluid chemistry may provide an incomplete picture of processes occurring during olivine dissolution.

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