American Mineralogist, Volume 97, pages 220–230, 2012

Experimental growth of diopside + merwinite reaction rims: The effect of water on microstructure development

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ABSTRACT

The growth rate and internal organization of bimineralic diopside ($CaMgSi_2O_6$)-merwinite ($Ca_3MgSi_2O_8$) reaction rims produced by a solid-state reaction between monticellite ($CaMgSiO_4$) and wollastonite ($CaSiO_3$) single crystals was determined at 900 °C, 1.2 GPa, and run durations from 5 to 65 h using conventional piston-cylinder equipment. Overall reaction rim thickness ranges from 3.8 to 20.9 μm and increases with the square root of time, indicating that rim growth is diffusion controlled. Symmetrical makeup of the internal microstructure implies that rims grow from the original interface toward both reactants at identical rates, indicating that diffusion of MgO across the rim controls overall growth, with an effective bulk diffusion coefficient of $D^{\mathrm{Di+Mw}}_{\mathrm{bulk,MgO}} = 10^{-16.3 \pm 0.2}$ m^2/s. At the initial stages, a "lamellar type" microstructure of alternating palisade-shaped diopside and merwinite grains elongated normal to the reaction front is generated, which gradually transforms into a "segregated multilayer type" microstructure with almost perfectly monomineralic Mw|Di|Mw layers oriented parallel to the reaction fronts at long run durations. This is due to changes in relative component mobilities. Whereas the "lamellar" microstructure develops when MgO is substantially more mobile than the other components, formation of the "segregated multilayer" microstructure requires additional mobility of at least one of the other components, CaO or SiO_2. We assume that a significant change in relative component mobilities is caused by continuous entrance of minute amounts of water from the piston-cylinder solid pressure medium through the capsule walls, as revealed by the presence of OH-defects in a reactant after the runs, and supported by water-containing powder experiments that only produce monomineralic Mw|Di|Mw layers. Traces of water have a major influence on relative component mobilities, internal rim organization, and the microstructural development of reaction zones.

Keywords: Reaction rims, reaction kinetics, solid-state diffusion, lamellar microstructure, IR spectroscopy, segregation, water