Goldschmidt 2022 Abstract
https://doi.org/10.46427/gold2022.11547

Chemically induced strain during solid state diffusion in minerals: An experimental study using pyrope – spessartine and pyrope – grossularite diffusion couples

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Reactions in solids, including the diffusive exchange of chemical elements, always involve a change of volume that results in a build-up of stress. The resulting strain depends on the rheological parameters and boundary conditions (e.g. confined / unconfined). The nature and extent of such chemically induced stress-strain relationships have been discussed in different contexts (e.g. tectonic overpressure, inclusion thermobarometry) using theoretical models but experimental observations under controlled laboratory conditions have not yet been available. We report here the results of two kinds of diffusion experiments using natural, gem quality garnet crystals where such effects could be studied: (i) pyrope-rich garnet crystals coupled with spessartine-rich crystals (diffusion-pyrope-spessartine, DPS experiments) and (ii) grossularite-rich with pyrope-rich crystals crystals (diffusion-grossularite-pyrope, DGP experiments). The experiments were carried out in a piston cylinder apparatus (e.g. 26 kbar, 1200 °C, 164 hours); experimental procedures were identical to those of Chakraborty and Ganguly, (CMP, 1992, 111: 74-86 and Ganguly et al., CMP, 1998, 131: 171-180). Concentration profiles measured in those earlier studies could be modeled using multicomponent diffusion theory. However, the experiments with the DPS and DGP couples yielded very irregular diffusion fronts. Chemical analysis at any point within the front correspond to stoichiometric garnet compositions. Structural analysis using electron backscatter diffraction (EBSD) confirmed that each point in the complex diffusion front could be indexed as crystalline garnet. However, the orientations of the crystals change with progressive diffusion. Specifically, as a larger cation enters the lattice of a garnet crystal containing a smaller cation, the build-up of strain results in the production of planes of lattice mismatch (subgrain boundaries or dislocation walls, these remain to be determined), disrupting a simple planar diffusion front. The effect increases in the order sps-alm / pyr-sps (non-existent) << pyr-sps (DPS) < pyr-gross (DGP) i.e. in the order of increasing difference in unit cell volumes between the two sides of the couple. These results suggest that the upper limit of strain / overpressure that can be sustained by a crystal lattice is constrained by the energy required to produce a new dislocation / grain boundary.