In-situ monitoring of mineral reactions using synchrotron X-ray diffraction

L.C. GÖTZE[1]*, R. MILKE[1], S. SCHORR[1,2], R. ABART[3], AND R. WIRTH[4]

[1]Free University of Berlin, Institute of Geological Sciences, Berlin, Germany (*correspondence: lutz.c.goetze@fu-berlin.de)

[2]Helmholtz Centre Berlin for Materials and Energy, Berlin, Germany

[3]University of Vienna, Department of Lithospheric Research, Vienna, Austria

[4]GFZ German Research Centre for Geosciences, Potsdam, Germany

We monitored mineral reactions of geologically relevant phases in-situ using energy-dispersive as well as angle-dispersive synchrotron X-ray diffraction in combination with an ex-situ assessment of the run products by focused ion beam (FIB) assisted transmission electron microscopy (TEM). We used the pulsed laser deposition technique (PLD) to deposit nanoscale reactant layers on top of single crystalline substrates. By using a thin film geometry and by applying low temperatures and short run durations, nanoscale reaction bands were synthesized. The in-situ experiments provide constraints for the temperature-dependent onset of a mineral reaction and the subsequent growth kinetics.

For one experimental setup initially amorphous MgO layers were deposited on top of (0001)-oriented corundum (α-Al_2O_3) substrates. In this setup spinel ($MgAl_2O_4$) growth was detected at temperatures ≥900°C. The spinel grows (111)-oriented into the substrate and the reactant layer. A corundum-grown spinel sublayer can be distinguished from a periclase-grown sublayer in TEM micrographs. The diffusion mechanism in this system – namely counterdiffusion of the cations through a rigid oxygen sublattice – can be deduced from the relative thickness proportions of these sublayers. Largely diffusion-controlled reaction kinetics were inferred from the evolution of the integrated intensity of the 111 spinel Bragg reflection at 900°C whereas at higher temperatures the interface reactions became increasingly rate-limiting [1]. A thermodynamic model [2] was applied to the data to extract the kinetic parameters characterizing spinel rim growth. Faster reaction kinetics were found using periclase substrates. Coherent interfaces and a negative reaction volume at the periclase/spinel phase boundary seem to promote the spinel-forming reaction in this setup.

[1] Götze *et al*. (2014) *PCM* **41**, 681-693. [2] Abart and Petrishcheva (2011) *AJS* **311**, 517-527.