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Experiments on Pb diffusion in titanite

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Titanite ($CaTiSiO_5$) is a versatile mineral for petrochronology as it allows to connect the physico-chemical conditions of its formation with age determinations. U-Pb geochronology is most often applied to determine absolute ages of titanite formation. As there is only one experimental work published on Pb diffusion coefficients in titanite (Cherniak 1993) and the data therein differ by about four orders of magnitude from natural constraints there has been a debate for three decades about the closure temperature of Pb in titanite.

We conducted Pb diffusion experiments in natural titanites at temperatures between 850 and 1100°C and at atmospheric pressure and controlled $f(O_2)$ for durations between 16 and 184 hours. As Pb source we used thin films deposited on chemically polished titanite surfaces as well as synthetic perovskite powders. The thin-films and the powder source were produced from $Ca_{0.9}Pb_{0.1}TiO_3$. Diffusion profiles were measured with Rutherford backscattering spectrometry (RBS) as well as by time-of-flight secondary ion mass spectrometry (ToF-SIMS).

The measured diffusion profiles are between 150 and 600 nm long. Several samples show heterogeneities as nm-scale impurities or surface roughness up to 50 nm, which causes analytical artefacts for the depth profiling techniques used. Beyond that zone characteristic diffusion profiles were obtained, which can be approximately fitted with an error function or a numerical model.

Our preliminary results show that all calculated diffusion coefficients indicate Pb diffusion in titanite to be at least one, in some cases up to three orders of magnitude slower than in the experiments of Cherniak 1993. Furthermore, most of the diffusion profiles suggest the existence of two different diffusion mechanisms with diffusion coefficients differing by about one order of magnitude (cf. Beyer et al., 2019).

The existence of different diffusion mechanisms with significantly different diffusion coefficients would explain the discrepancy between experimentally determined diffusion parameters and natural constraints. However, more experiments are currently conducted that will shed further light on the reason for this decades-long discussion.

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