

## Non-equilibrium thermodynamics: Diffusion controlled partial melting

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Melting is commonly assumed to take place in a rock assemblage that is in chemical equilibrium. The question we would like to answer is: does diffusion of major elements in mantle solid phases affect melt abundance and composition and if yes to what extent and at what conditions. The problem is addressed using our general thermodynamic/multiphase flow model described in some details in a separate communication (symposium S45) and applied in the context of ridges melting. To include the effect of diffusion in the model, transport equations for grain size and diffusion profiles are also solved and the problem is treated on two scales in space and time. The interesting feature is that the thermodynamic database for solids and melt (Ghiorso et al., G3, 2002) is still applied to the model through the Gibbs free energy minimization at every space and time step. When diffusion is taken into account, the thermodynamics model puts a constraint on the chemical transfer rate for every solid phase component but only at the solid-melt interface.

It is well known that during an equilibrium polybaric melting process, chemical elements are incorporated in the melt phase in different proportion as determined by their compatible/incompatible nature. Non equilibrium melting can alter the normal distribution of the chemical elements during melting process. We are expecting that if disequilibrium is maintained during mantle uprising, incompatible elements are retained in the solid phases for longer time compared to the equilibrium case. At the same time melt productivity should increase due to the preservation of a more fertile mantle at lower depths. This could have significant effects on Mg/Fe ratio and Na content in MORBs in particular for fast spreading ridges.

## *In-situ* diffusion at Mont Terri URL

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Clay-rich formations, such as the Opalinus Clay (Switzerland), are currently being considered as potential host rocks for the deep geological disposal of radioactive waste. Diffusion is the main transport mechanism for radionuclides in these impermeable rocks. Besides, sorption provides additional retardation for cationic species.

The objective of the DI-A in-situ diffusion experiment at the Mont Terri Underground Rock Laboratory (URL) was to confirm the expected diffusion-controlled transport and to compare the results of the experiment with those from small-scale (cm) through-diffusion experiments. The experimental setup at Mont Terri consisted of a borehole drilled in the rock, with a 1-meter-long injection interval at its bottom. Synthetic porewater containing an initial pulse of tracers (HTO, I<sup>-</sup>, <sup>22</sup>Na<sup>+</sup>, Cs<sup>+</sup>) was circulated through the borehole, and the evolution of tracer concentration was monitored. After about 10 months, a volume of rock around the injection borehole was excavated and tracer distribution profiles in the rock were measured. Reactive transport simulations allowed the fitting of (a) the temporal evolution of tracer concentration in the injection system and (b) the tracer profiles in the rock, which provided unique sets of effective diffusion coefficients (De) and accessible porosities (sorption parameters for sorbing tracers). The results for HTO, I<sup>-</sup> and <sup>22</sup>Na<sup>+</sup> were in excellent agreement with those from through-diffusion experiments, confirming the important effects of anionic exclusion (I<sup>-</sup>) and sorption (<sup>22</sup>Na<sup>+</sup>). There were no previous experimental values of De for Cs<sup>+</sup>, although batch sorption data were available. The results of DI-A indicated less sorption (by a factor of about 2) in the intact rock than in batch. Also, De for Cs<sup>+</sup> was about 5 times larger than for water (HTO). These results are now being confirmed by through-diffusion experiments.

A second experiment (DI-A2) is currently under way. This time, the tracers are HTO, I<sup>-</sup>, Br<sup>-</sup>, <sup>85</sup>Sr<sup>2+</sup>, <sup>60</sup>Co<sup>2+</sup>, Cs<sup>+</sup> and Eu<sup>3+</sup>. Additionally, scoping calculations are being performed for a new experiment (DR). The main objectives are to study in-situ diffusion for longer time scales (3-5 years) and to optimize the experimental concept for strongly sorbing cations and for measuring diffusion anisotropy in situ.