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Thermodynamic model for microstructure evolution during reaction rim growth

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The formation of different microstructural types of reaction rims in a closed system is investigated theoretically. In particular multilayer types that are comprised of a sequence of monomineralic layers and cellular (symplectite) types are addressed. A thermodynamic model for cellular reaction rim growth in a ternary model system is derived. It is found that either the chemical mass transfer across the rim or the material re-distribution that must occur within a reaction front, at which a cellular microstructure is produced, may be rate limiting. Based on the thermodynamic extremum principle, parameter domains can be discerned, where reaction rims preferably produce cellular or multilayer microstructural types. The controlling factors are the characteristic length scale (wavelength) of mineral intergrowth in the cellular microstructure and the relative efficiencies of chemical mass transfer across the rim and within the reaction fronts of cellular layers. For a given set of kinetic parameters formation of the multilayer type is preferred during the initial growth stages, and the cellular type is preferred at later growth stages. If component mobilities remain constant throughout reaction rim growth, a transition from the multilayer to the cellular type is expected to occur as the rim thickness increases. The reverse transition is unlikely.

Both transitions have been observed in experiments done in the CaO - MgO- SiO_2 system. The transition from multilayer to cellular type was observed in experiments, where monticellite and wollastonite reacted to akermanite and diopside and some low-SiO_2 phase at 1200^{o}C, 0.5 GPa and durations between 5 min and 60 h at very dry conditions. The reverse transition was observed in experiments, where monticellite and wollastonite reacted to diopside and merwinite at 900^{o}C , 1.2 GPa and durations between 5 h and 65 h. In the latter case the reverse transition clearly indicates a change in the relative mobilities of MgO and CaO during the experiment, where CaO became more mobile relative to MgO due to the successive diffusion of water into the capsule. The microstructural evolution during solid state transformations is very sensitive to the kinetic parameters underlying the respective reaction. Irreversible thermodynamics provides a tool to infer kinetic parameters from observed microstructures.