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Thermodynamic model for growth of reaction rims with lamellar microstructure

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Abstract

A thermodynamic model for the growth of a reaction rim with lamellar internal microstructure is derived. Chemical mass transfer across the rim and the chemical segregation within the reaction fronts, at which the lamellar microstructure is produced, are considered as the only dissipative processes involved in rim growth. Depending on their relative rates, either one of these processes may be rate limiting. Rim growth is parabolic when mass transfer across the growing rim is rate limiting, and it is linear when the material redistribution within either one or both of the reaction fronts is rate limiting. The transition between these two extreme scenarios is continuous. The controlling factors are the characteristic length scale of the lamellar microstructure and the relative rates of chemical mass transfer across the rim and within the reaction fronts. Reaction rims with both lamellar and layered internal microstructures have been produced experimentally in the ternary system CaO-MgO-SiO_2. Based on the thermodynamic extremum principle, parameter domains can be discerned, where reaction rims preferably develop lamellar microstructures or, alternatively, a sequence of monophase layers. For a given set of kinetic parameters, formation of the layered microstructural type is more likely during the initial growth stages, and the lamellar type is preferred at later growth stages.

Keywords: Reaction rims, reaction kinetics, thermodynamics, lamellar, segregation, solid-state diffusion, ternary system, microstructure