Phase decomposition in a three-component system under cooling conditions

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We address phase decomposition in a ternary feldspar by explicitly taking into account strong differences in diffusivities (mobilities) of the components. First, we adopt the standard Cahn-Hilliard model [1] to a multicomponent system with arbitrary diffusivities of components. To this end, the most general Onsager's approach is utilized. We assume substitutional interdiffusion and from this derive restrictions for the matrix of phenomenological coefficients. In a second step, a finite element routine is employed that yields numerical solutions of the generalized Cahn-Hilliard equation. Specifically the Comsol Multiphysics software is used. Third, we introduce and investigate several decomposition scenarios in which differences in diffusivities are important.

Our main result is that differences in diffusivities may produce systematic deviations from equilibrium element partitioning between the exsolved phases. Namely, the slowest component is essentially "frozen" initially, and phase decomposition takes place only for the two faster components. At this stage deviations from the equilibrium element partitioning are observed. To derive compositions of the new-born phases, one has to construct a histogram that contains information on all compositions observed throughout the system. The histogram shows two or three pronounced maxima revealing the compositions of the exsolved phases. During the separation process, the trajectories of the exsolved phases through the composition space deviate noticeable from the conodes corresponding to the compositions of the homogeneous precursor phase. The deviations from equilibrium element partitioning may be destroyed on a longer time scale by evolution of the third component. They may, however, also survive. In particular, this is a possibility under conditions of cooling. In the latter case the system remains unaccomplished at the observation point.

The model is applied to exsolution of ternary feldspar during cooling. A realistic free energy function for the uniform states is taken from the literature [2]. In ternary feldspar the orthoclase and albite components are substantially more mobile than the anorthite component. As a consequence, the exsolved phases do not evolve along the conodes of the ternary phase diagram but they evolve along lines of nearly constant anorthtite content. Only when the separation of the albite and orthoclase components is nearly completed, exchange of the anorthite component becomes effective and the system slowly returns to a correct conode. During cooling the system may, however, get stranded and non-equilibrium phase relations may be preserved. Such a behavior was indeed anticipated qualitatively and a model for compensation when applying two feldspar thermometry was suggested [3]. Our approach gives a quantitative account of this effect, where both microstructure and composition evolution are solved for simultaneously.

We believe that our results are of importance for a wide class of geological systems, as considerably different diffusivities are frequently encountered in mineral solid solutions. In such cases application of solvus thermometry may be difficult due to the potential preservation of non-equilibrium system states. The observed microstructures and phase compositions are rather coined by the cooling history than by peak temperatures and are thus potential geospeedometers.

References
[1] Cahn JW, Hilliard JE. (1958) J Chem Phys, 28:258–267; [2] Fuhrmann ML, Lindsley DH. (1988) Am Mineral, 73: 201–15; [3] Voll G, Evangelakakis C, Kroll H. (1994) Precambrian Research, 66:351–377.