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Thermodynamically consistent Phase Field modeling for freeze crystallization in a binary system

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Abstract

Freeze crystallization is a commonly employed separation process in Chemical Engineering, widely utilized in the food, pharmaceutical, and wastewater treatment industries. It is known for its ability to preserve delicate molecules and its energy efficiency. This process consists of solidifying water from an undercooled solution.

Among the liquid-solid (L-S) phase diagrams for binary mixtures (A-B), three basic groups are classified regarding the miscibility in the solid phase: (1) the complete miscibility in solid phase, which is called solid solution with a homogeneous mixture, such as the Cu-Ni mixture (Fig. 1a); (2) the partial miscibility of components A and B in solid phase, so that A-rich phase α and B-rich phase β exist corresponding partial dissolved solution (e.g. Pb-Sn, Fig. 1b); (3) the complete immiscibility with the solidus overlaps with the y-axis (Fig. 1c).

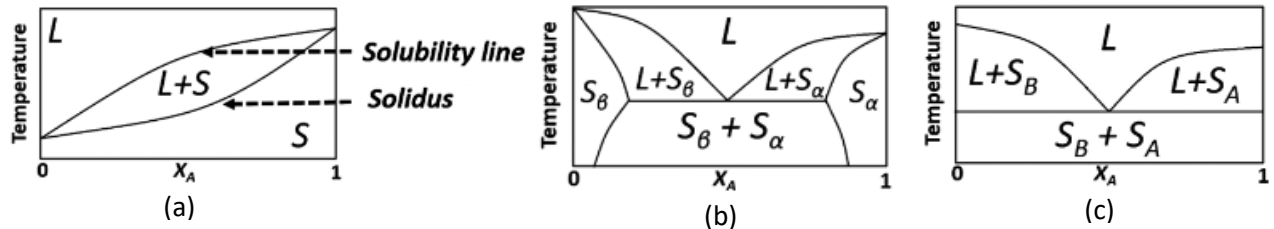


Figure 1: Basic classifications of phase diagram function of x_A concentration of component A : (a) complete miscible solid phase; (b) partial miscible solid phase; (c) immiscible solid phase (Toro-Vazquez, J. F. & Pérez-Martínez, J. D. - 2018).

So far the Phase Field Model (PFM) has mainly focused on the first two kinds of mixtures (complete miscibility and partial miscibility). It is yet the third kind of system (immiscible solid phase), where the solid remains pure during the phase change process, that is predominant in among water based solution. However, the lack of solid concentration profile raises the problem when introducing the equilibrium conditions.

A few studies focused on the solid immiscibility systems: Yuan et al. (2020) introduced a partition coefficient in liquid and solid phases without considering the thermodynamic aspect, the solute had integrated in solid phase; while Van der Sman (2016) addressed this issue using an interaction coefficient between solute and solvent dependent on the Phase-Field variable (ϕ). Importantly, their non-variational approach is not fully thermodynamically consistent, insofar as it deviates from developing the model from the same energy functional.

Our study investigates such immiscible solid systems using the KKS (Kim-Kim-Suzuki) model. We introduce a pseudo-component to reproduce the solute-eliminated effect, namely the pure ice formation. To ensure thermodynamic consistency, our model is developed from the same energy functional, and the thermodynamic properties are calculated and deduced using the CALPHAD method. Our 2D dynamic model focuses on the crystallization of ice from a synthetic wastewater solution (H₂O-NaCl mixture) considering the solute diffusion effect. The major result shows the pure solid phase and the formation of brine pocket due to the solute accumulation at interface and the development of side branches, which are largely dependent on ice growth velocity.