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«Modeling phase separation in the melt»

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The glass is a multi-components system which appears as a single liquid phase at high temperature. However, when it is lowered below a critical temperature, several observations show that the melted glass can separate into two liquid phases. Their density ratio is quite low but their viscosity ratio is around 100. Both liquid phases are separated by an interface and their bulk properties are characterized by the equilibrium compositions of each specie. In this talk we present the whole methodology carried out for simulating the phase separation of such systems. It is based on several complementary approaches: 1) a Calphad modeling for thermodynamics of simplified glasses, 2) a mathematical modeling based on the phase-field theory and 3) an efficient computational code (`LBM_Sac1ay`) implementing the models for the simulations.

The phase-field theory is a popular approach for modeling phenomena which involve interface dynamics and thermodynamic phase diagrams. A simple free energy functional composed of one double-well term plus one gradient energy term can simulate the phase separation. The well-known Cahn-Hilliard (C-H) equation, which is derived from that functional, mixes the thermodynamics and the interface properties. As an advantage, the interface position as well as the composition can be simultaneously recovered. As a drawback, the fit of the thermodynamics necessarily modifies the interface properties. A first illustration of phase separation is given on a melted glass composed of two components $\text{SiO}_2\text{-MoO}_3$ (binary mixture). The simulations illustrate the nucleation/growth process and the phase separation of a gaussian distribution.

That simplest free energy functional is next adapted for the ternary phase diagram of $\text{SiO}_2\text{-Na}_2\text{O-MoO}_3$ and the partial derivative equations are derived. In order to decouple the thermodynamic properties and the interface parameters, the C-H equation is advantageously replaced by an Allen-Cahn equation coupled with two diffusion equations. The first one tracks the interface and involves the surface tension and the mobility. The two other contain the diffusion coefficients of species and their equilibrium compositions in each bulk phase. The simulation of the Oswald ripening is presented as an illustration of that two-phase/three-component model. Finally several simulations will present the fluid flow effect with viscosity contrast when that model is coupled with the Navier-Stokes equations.

Several perspectives of this work are currently in progress. The first one consists to simulate the model expanded with a fourth component (Ba_2O) and coupled with the temperature equation. Next, comparisons will be done with the experiments performed at CEA/ISEC.