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Transport phenomena in silicate melts: chemical diffusion and phase separation

In this talk, I will present two different studies of transport phenomena conducted at high temperature in silicate melts, in the joint Unit CNRS/Saint-Gobain SVI.

During her PhD, Corinne Claireaux studied the coupled diffusion of chemical species in the quaternary system $\text{Na}_2\text{O} - \text{CaO} - \text{SiO}_2 - \text{Al}_2\text{O}_3$. Diffusion experiments between melts of different initial compositions were realized, and chemical profiles were measured thanks to electron microprobe. The set of all chemical profiles is used to determine the complete diffusion matrix, relating the flux of one element to the gradients of all elements. From the diffusion matrix, eigenvectors and eigenvalues can be extracted. They are interpreted respectively as combinations of elements that rearrange cooperatively for diffusion to proceed, and as the probabilities of such exchanges. Eigenvalues of strikingly different magnitudes are found, confirming that fast and very slow diffusion processes operate simultaneously in the melts. Activation energies are measured for the different processes corresponding to the different exchanges.

Secondly, we studied the kinetics and the morphology of phase-separated domains during coarsening in barium borosilicate melts. During his PhD, David Bouttes used in-situ synchrotron microtomography to characterize the 3-D microstructure of phase-separated melts during isothermal heat treatments, in the temperature range 1000-1300°C, far above the glass transition.

Quantitative geometrical measurements and direct observations demonstrate that viscous coarsening is the dominant mechanism governing the evolution of the bicontinuous structure. This mechanism results in a linear growth of domain size with time, much faster than the $t^{1/3}$ growth associated to diffusive mechanisms, which have been observed so far in silicates. The activation energy for domain growth is consistent with the one of viscosity. Complementary experiments show that diffusive mechanisms are negligible compared to viscous coarsening at such high temperatures, in contrast to experiments of the literature performed closer to the glass transition. Furthermore, we observe a progressive fragmentation of one of the percolating phases, which we relate to the important viscosity contrast between phases.

