Fragility and slow kinetics of melt homogenization in the As-Se binary

SRIRAM RAVINDREN, KAPILA GUNASEKERA, PUNIT BOOLC-HAND, University of Cincinnati — Two gram sized As$_x$Se$_{100-x}$ batches at various As content $x$ were synthesized using pure Se and As$_2$Se$_3$ as starting materials that were reacted at 700°C. Such melts typically took 3-12 days to homogenize, as monitored in punctuated, off-line FT-Raman line profiling experiments. We have now undertaken mDSC experiments as a function of modulation frequency to establish the compositional dependence of complex $C_p(x)$, and deduce the variation of fragility $m(x)$. We find the fragility to be rather low, $m < 20$, across the $22\% < x < 38\%$ range, and to rapidly increase at $x < 22\%$ to acquire a value of 43 near $x = 3\%$. We show that the slow melt homogenization is a direct consequence of the “strong” character of melts that serves as a bottleneck in melt-mixing at high temperatures. Once homogenized, physical properties of glasses, such as density, glass transition temperature $T_g(x)$, the Intermediate phase, and variation of enthalpy of relaxation at $T_g(x)$ differ significantly from their inhomogeneous counterparts.

$^1$This work is supported by NSF grant DMR 08-53957.