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Experimental determination of crystal growth rates in highly supercooled aluminosilicate liquids: Implications for rate-controlling processes

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ABSTRACT

The kinetics of crystal growth have been investigated between 100 and 200 K above the glass-transition temperature for four liquids in the ternary system CaO-Al_2O_3-SiO_2. The observed phases are yoshiokaite, gehlenite, larnite, anorthite, and wollastonite. They crystallize either congruently or incongruently through heterogeneous nucleation on gas-liquid interfaces. In all cases, the growth rate at high degrees of supercooling is independent of time, while its temperature dependence is Arrhenian. When the crystal has the same Al/Si ratio as the parent liquid, the growth rate of congruent and incongruent crystallization shows a 1:1 correlation with the viscosity of the melt. We do not interpret this observation to imply that growth rate is controlled by viscosity but, rather, that both phenomena are controlled by the rate of rearrangement of bonds between oxygen and network-forming cations. When Al/Si varies across the crystal-melt interface, the effects of interdiffusion must also be considered and the temperature dependence of growth rate is no longer correlated directly with that of viscosity, the former being smaller than the latter. Our observations show that, at constant viscosity, the fastest growth rates occur when the Gibbs free energy difference between liquid and crystal is greatest, rather than when crystalline and liquid phases are structurally most similar. A simplified equation that describes crystal growth rates from highly supercooled liquids is proposed.