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

MATHIEU ROSKOSZ,1,* MICHAEL J. TOPLIS,1,† AND PASCAL RICHET2

1Centre de Recherches Pétrographiques et Géochimiques, CNRS UPR 2300, 15 rue Notre-Dame des Pauvres, BP 20, 54501 Vandoeuvre-lès-Nancy Cedex France
2Physique des Minéraux et des Magmas, UMR CNRS 7047, Institut de Physique du Globe, 4 place Jussieu, 75252 Paris Cedex 05, France

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-Al2O3-SiO2. The observed phases are yoshiokaita, 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.

INTRODUCTION

Crystal nucleation and growth in silicate melts are fundamentally important processes, controlling mineral textures, crystal size distribution, and chemical zoning in igneous rocks (e.g., Lofgren et al. 1974; Cashman and Marsh 1988; Albarède and Bottinga 1972). The kinetics of crystal growth are driven by the Gibbs free energy difference between the relevant crystalline and liquid phases. The rates are zero at the liquidus and increase at lower temperature until the rate at which atoms may be brought to the crystal-liquid interface becomes limited by rapidly decreasing element diffusivities. This is why crystal growth rates generally tend to zero near the glass transition after having passed through a maximum somewhere between the liquidus and glass-transition temperatures (e.g., Turnbull and Cohen 1960).

In the geological literature, the kinetics of nucleation and crystal growth have mainly been investigated near the liquidus, for instance in the ternary system Anorthite-Albite-Diopside (e.g., Kirkpatrick 1974; Klein and Uhlmann 1974; Kirkpatrick et al. 1976, 1979; Tsuchiyama 1983; Muncill and Lasaga 1987). Basaltic melts have also been studied (e.g., Donaldson 1975; Nabelek et al. 1978), but with an emphasis that was put on chemical gradients and textural relations rather than on crystal growth rates. The subsolidus temperature range has received less attention in the Earth sciences (e.g., Kirkpatrick 1974; Klein and Uhlmann 1974) although it is important in volcanic contexts where glasses may be heated (Burkhard 2001, 2002), or for rocks with complex thermal histories such as stony meteorites or the lunar regolith (Yanin and Bish 1990). On the other hand, because subsolidus conditions are critical for production of ceramics, studies at high degrees of supercooling have been performed on simple, congruently crystallizing compositions such as lithium disilicate, cordierite, and 3spodumene (e.g., Scherer and Uhlmann 1975; Zanotto and Galhardi 1988; Watanabe and Giess 1994, Barbieri et al. 1997; Diaz-Mora et al. 1998; Fokin and Zanotto 1999; Burgner and Weinberg 2001).

A conclusion drawn from these studies is that the standard model of crystal growth (Turnbull and Cohen 1960) is not universally applicable. Several alternatives have been proposed, such as the empirical approach based on crystal morphologies (e.g., Jackson and Hunt 1966; Christensen et al. 1973) and the theoretical approach using cluster dynamics (Gránásy and James 1994, Barbieri et al. 1997; Diaz-Mora et al. 1998; Fokin and Zanotto 1999; Burgner and Weinberg 2001).

In this work, we present experimental determinations of the kinetics of crystal growth from a series of CaO-Al2O3-SiO2 liquids. This system was chosen because its phase relations and