Kinetics of Nucleation and Crystal Growth in Glass Forming Melts in Microgravity

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Abstract

Numerous experiments conducted on melts in microgravity have concluded that homogenization and glass forming tendency of a melt are improved compared to when the same melt is processed at 1-g on earth. Glasses prepared in micro-g are more resistant to crystallization, more chemically homogeneous, and contain more uniform and improved short range structural order (SRO) than identical glasses prepared on earth. When doped with small amounts of platinum, the platinum particles in glasses prepared in microgravity were more uniformly distributed compared to glasses of identical chemical compositions prepared on earth. Similar results, viz., an improved microstructure for metals and alloys prepared by melt solidification in microgravity have also been reported.

The results cited above for melts solidified in microgravity are in contradiction to the general notion that chemical homogenization in a melt is controlled by gravity-driven convection. Since gravity-driven convection is ideally absent in microgravity, a solid prepared by cooling a melt in microgravity has the potential to be less chemically homogeneous and has less structural uniformity than the same melt processed under identical conditions at 1-g. However, the observed experimental results are just the opposite. Attempts to explain these seemingly contradictory results are limited and warrant needful investigation.

The mechanisms involving solidification of high temperature melts are essentially grouped under the generic terms nucleation and crystal growth. Glass formation requires that a melt be cooled no slower than a critical rate to avoid detectable crystallization. Whether a melt of a desired composition can be quenched to glass or crystallizes during cooling, is controlled by the kinetics of the structural transformations for nucleation and crystal growth. The puzzling differences observed for melts solidified in low gravity and on earth indicates that the transformation mechanisms in melts cooled in low gravity may be different from those that occur in melts cooled at 1-g. Since the transformation of a melt to either glass or crystal depends primarily on the combined roles of nucleation and crystal growth, a satisfactory explanation for the observed differences between the low-g and 1-g results lies in the proper understanding of the kinetics of nucleation and crystal growth in high temperature melts. The mass transport or diffusion processes associated with forming nuclei or crystal growth can be affected by gravity-driven convection in fluid melts at 1-g, but these processes may remain undisturbed in low-g where the gravity-driven convection is ideally absent. Studies of crystal nucleation and growth in glass melts in low-g may provide information that is difficult to obtain from similar studies on earth.

The objective of our NASA program (NASA Contract NAG8-1465) is to investigate this critical issue, namely, how the kinetics of nucleation and crystal growth in melts in low-g differ from those in identical melts at 1-g. The general plan is to obtain the following data for a lithium disilicate ($Li_2O.2SiO_2$ or LS_2) glass melted on earth (1-g) and in space (micro-g):

(1) nucleation and crystal growth rates as a function of temperature;

(2) activation energy for the overall crystallization process;

(3) primary crystallization mechanism, surface or volume;

(4) effective diffusion coefficient;

(5) chemical homogeneity, short and intermediate range order, and microstructure (for crystalline samples); and,

(6) selected properties such as the density, molar volume, refractive index, and transmission in the UV, VIS, and IR.

A lithium-disilicate composition has been chosen for this study since it is a well studied system both experimentally and theoretically. Furthermore, the thermodynamic and kinetic parameters for nucleation and crystal growth at 1-g are available for this LS_2 glass.

The proposed experiments include melting an LS_2 composition in low-g (flight experiments) and at 1-g (ground-based experiments) at temperatures between 1150 and 1500 °C for times between 1 and 4 h and then cooling the melts at predetermined rates. The quenched melts that are obtained as glasses will be heat treated at various temperatures between 420 and 700 °C for times between 30 min and 12 h where temperature isothermality will be a prime consideration. A part of this heat treatment scheme is designed for homogeneously nucleating the glasses and the other part is for growing these nuclei to crystals of detectable size. Some of the melts will be doped with small amounts of platinum (≈ 0.001 wt%) to investigate and compare the heterogeneous nucleation and crystal growth in this LS_2 melt in low-g and at 1-g.

It is currently planned to melt the glasses in platinum containers in low-g, but it would be advantageous if a containerless facility such as the AMPLE (Aerodynamic Materials Processing in Low-g Environment) developed by ORBITEC or the AAL (Aero-acoustic Levitator) developed by Containerless, Inc., is available for use. Containerless melts should be free of container-induced impurities and unplanned heterogeneous nucleation/crystallization at the melt-container interface.

This research is expected to show how the kinetics of nucleation and crystal growth in high temperature melts depend upon gravity and will greatly enhance our present understanding of the fundamental mechanisms of nucleation and crystal growth in deeply undercooled melts. The knowledge from this research is expected to improve our glass processing technology on earth and could lead to new high performance glasses such as laser glasses with much higher output power per unit volume, lower loss optical glass fibers, glasses with a high coefficient of nonlinear refractive index suitable for fast all-optical switching devices, or glass-ceramics with improved microstructures and properties.