Heterogeneous crystal nucleation on bubbles in silicate melt

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ABSTRACT

Experiments reported herein document heterogeneous crystal nucleation on bubbles in supercooled lithium disilicate melt. Crystalline lithium disilicate (Li₂Si₂O₅) nucleated and grew on small bubbles (~1 μm) with a one-to-one correspondence between the number of bubbles and crystals (ranging from <10⁴ to ~10⁶ bubbles/mm³). Crystals grew on large bubbles (>100 μm) only in samples fused in N₂, suggesting a chemical control on nucleating efficiency. Bubbles ~1 μm in diameter served as nucleation sites for polycrystalline lithium disilicate spherulites; bubbles smaller than ~1 μm served as nucleation sites for the more common ellipsoidal crystalline form. This difference in behavior might be due to the additional surface area available for crystal nucleation on the 1 μm bubbles.

Our findings suggest that superliquidus thermal history can influence crystal nucleation via bubble formation induced by supersaturation, and has implications for both natural samples and experimental studies. Heterogeneous crystal nucleation on bubbles may serve as an efficient nucleation mechanism in natural degassing magmas and may aid in the formation of fine-grained groundmasses common to many volcanic rocks. Furthermore, we have documented a new mechanism of spherulite formation in highly supercooled silicate melt, similar to conditions thought to exist during devitrification of natural glasses. The ability of crystals to nucleate on bubbles can be exploited in the production of commercial glass-ceramic materials.

INTRODUCTION

Nucleation kinetics play a central role in the development of crystalline texture in igneous and metamorphic rocks, and in commercial glasses and glass-ceramic materials. Nucleation can occur on pre-existing surfaces (heterogeneous: Berkebile and Dowty 1982; Holand et al. 1995) or in the absence of such surfaces (homogeneous: James 1974; Christian 1975). The effects of added impurities (Schlesinger and Lynch 1989; Narayan et al. 1996), water content (Gonzalez-Oliver et al. 1979; Davis et al. 1997), and thermal history (Lofgren 1983; Baker and Grove 1985; Mishima et al. 1996; Davis 1996) on both homogeneous and heterogeneous crystal nucleation in silicate melts have been investigated. In addition, the role of crystal surfaces on bubble nucleation has been analyzed (e.g., Wilcox and Kuo 1973; Hurwitz and Navon 1994). Here we report experimental results that document crystal nucleation on vapor bubbles in silicate melt. Crystal nucleation on bubbles has been observed only rarely. One notable example is Schmelzer et al. (1993), who reported crystallization on bubbles in NaPO₃ melt. To our knowledge, crystal nucleation on bubbles has not been reported for silicate melts, either natural or synthetic. We believe the possible consequences of crystal nucleation on bubbles to erupting natural magmas are significant enough to warrant further investigation.

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Previous studies of nucleation kinetics in lithium disilicate (Li₂Si₂O₅) melts have noted the presence of two populations of crystals in experimental charges: (1) the common ellipsoidal form of stoichiometric lithium disilicate; and (2) spherulites of the same material (e.g., James 1974; Davis 1996). The first morphology, the more common of the two in crystal nucleation experiments (e.g., Matusita and Tashiro 1973; James 1974; Davis et al. 1997), has been attributed to a homogeneous nucleation mechanism. During a systematic study of the influence of water on nucleation kinetics in silicate melt (Davis et al. 1997), we observed that a preliminary sample preparation method resulted in the (reproducible) development of a very high number density (~10⁴ crystals/mm³) of spherulites. This sample preparation method consisted of re-fusing glass cubes at a lower temperature than the initial fusion temperature of the starting material (1120 vs. 1350 °C). Upon closer inspection of the spherulites, it was discovered that every spherulite contained a ~1 μm bubble at its center. In contrast, when we re-fused our samples at a temperature equal to that of the initial fusion, no spherulites were observed in our quenched samples. The latter sample preparation method was used for the experiments reported in Davis et al. (1997) to ensure that only homogeneous crystal nucleation occurred. We consider in this report the nucleation of crystals on bubbles. As discussed below, the presence of crystals serves to “tag” very small bubbles that might otherwise have gone un-