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Bubbles and element clusters in rock melts: A chicken and egg problem

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The nucleation and growth dynamics of gas bubbles and crystals play a vital function in determining the eruptive behaviour of a magma. Their rate and relative timing, among other factors, are controlled by the magma's ascent rate. Investigating the kinetics of decompression-induced degassing and crystallization processes can thus give us insight into the rheology of magmas. For example, the rapid decompression of magmas inhibits microlite crystallization and bubble nucleation during ascent leading to crystallization and degassing at shallow levels. This results in a drastic increase in viscosity and an over pressured system, which can lead to violent eruptions. Although many experiments and numerical simulations of magma decompression have been carried out, nascent and initial bubble nucleation remain poorly understood. It is widely accepted that there are two ways bubbles can nucleate within a melt: heterogeneous (on a pre-existing surface) and homogeneous nucleation (within the melt), where homogeneous nucleation requires a higher volatile supersaturation. It has since been tentatively suggested that homogeneous nucleation is simply a variety of heterogeneous nucleation where nucleation occurs on the surface of submicroscopic crystals. However, evidence of these crystals is equivocal. Thus, we have combined novel 2D and 3D structural and chemical microscopy techniques including scanning transmission electron microscopy (STEM), electron energy-loss spectroscopy (EELS) mapping, and atom probe tomography (APT) to investigate the presence of sub-nanometer scale chemical heterogeneities in the vicinity of gas bubbles within an experimental andesitic melt. The combined STEM and EELS data reveal a heterogeneous distribution of bubbles within the melt ranging between 20-100 nm in diameter, some of which have Fe and/or Ca element clusters at the bubble-melt interface. Element clusters enriched in Fe, Ca, and Na are also observed heterogeneously distributed within the melt. The reconstructed APT data reveals bubbles as low ionic density regions overlain by a Na-, Ca-, and K-rich cluster and heterogeneously distributed Fe clusters within the bulk of the melt. Based on these observations, our data demonstrate the existence of nano-scale chemical heterogeneities within the melt and at the bubble-melt interface of bubbles that were previously interpreted to be nucleated homogeneously within the melt, therefore contributing to the proposed hypothesis that homogeneous nucleation could in fact be a variety of heterogeneous nucleation. These results highlight the need to redefine homogeneous nucleation and revisit whether bubbles or crystals occur first within volcanic melts.