Phase separation of hydrous Vesuvius melt: vesicle nucleation or spinodal decomposition?

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The phase separation of H_2O fluids from supersaturated hydrous silicate melts determines the starting point of explosive volcanic eruptions. The number of fluid vesicles per unit volume of silicate melt (*VND*) is a basic property that controls the efficiency of fluid-melt separation and as a result the acceleration of magma ascent. To simulate the phase separation of single phase hydrous silicate melts during ascent, continuous decompression experiments were performed at superliquidus temperatures with AD79 Vesuvius white pumice composition. This pumice buried Herculaneum and Pompeii and is representative of other catastrophic phonolitic and trachytic explosive eruptions like the violent 39 ka Campi Flegrei and the 1815 AD Tambora eruption.

Experiments were conducted in an internally heated pressure vessel equipped with a high-pressure valve that facilitates continuous decompression. 5.3 wt% H_2O were dissolved in the melts at 200 MPa and 1523 K for 96 h. Subsequently, the slightly H_2O undersaturated melts were isothermally decompressed at 1323 or 1373 K with rates of 0.024-1.7 MPa·s^{-1}. At final pressures (P_{final}) between 110 and 70 MPa, the samples were isobarically quenched with ~150 K·s^{-1} to room temperature. The *VND* of the vitreous samples were determined with optical microscopy and quantitative backscattered electron image analysis combined with computational 2D to 3D transformation.

Homogeneously dispersed vesicles are observed in the samples decompressed to a $P_{final} \leqq 100$ MPa which corresponds to a ΔP of ~95 MPa that is necessary to induce homogeneous phase separation. High log *VND*'s of ~5.2 (in mm^{-3}), corresponding to average vesicle distances of 10 μm, are observed irrespective of decompression rate within the investigated range. A nucleation theory based decompression rate meter (Toramaru, 2006), which is commonly used to estimate magma ascent velocity from *VND* of volcanic ejecta, fails to explain the experimentally observed decompression rate independent *VND*. Alternatively, decompression induced H_2O-silicate melt phase separation may be described by the theory of spinodal decomposition (Cahn, 1965) at the limit of thermodynamic stability. Critical behavior of hydrous silicate melt has already been evidenced experimentally by e.g. Shen and Keppler (1997) and is attributed to spinodal decomposition.

Irrespective of the phase separation mechanism, a decompression rate independent *VND* has profound consequences for the degassing dynamics of natural polyphase hydrous magma, that contain crystals and pre-existing fluid vesicles. During ascent, H_2O supersaturation is decreased in melt shells surrounding pre-existing vesicles by H_2O diffusion. Homogeneous phase separation occurs in interstitial melt volumes in which sufficient supersaturation is still preserved. If the decompression rate is fast enough to maintain such supersaturated volumes, the formation of vesicles with high log *VND* of 5.2 inevitably causes efficient melt degassing within seconds due to short H_2O diffusion distances on a 10 μm scale from the melt into fluid vesicles. This causes rapid density decrease accompanied by sudden increase of magma buoyancy required for explosive volcanism.

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