

Temperature dependence of surface tension between silicate liquid and H₂O

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According to the classical nucleation theory, surface tension of the liquid-bubble interface is one of the most critical parameters controlling the nucleation rate of bubbles in liquids. Even small changes in surface tension result in large difference in nucleation pressure. Although it is critical to quantify surface tension accurately to simulate bubble nucleation, we lack reliable data of and models on surface tension because of difficulty to measure the surface tension of volatile-bearing silicate liquids under pressure. Bagdassarov et al. (2000, *Amer. Mineral.*, 85:33-40) used the sessile drop method to measure surface tension for SiO₂-rich liquids (haplogranite, rhyolite) at 1 atm and under water pressure. They demonstrated that surface tension for anhydrous SiO₂-rich liquid is in the range of 0.3-0.4 N/m and that it decreases with increasing H₂O content, with increasing water pressure and with decreasing temperature. The third feature implies that temperature dependence of the surface tension is positive: e.g. $+6 \times 10^{-5}$ N/m/degree C for a haplogranite melt with 8.9 wt.% H₂O between 900 and 1200 degree C.

To examine the temperature dependence of surface tension in rhyolitic liquid containing about 7 wt.% H₂O, we compared the results of homogeneous bubble nucleation experiments performed at two different temperatures: 700 degree C (Hamada et al., 2010, *Bull. Volcanol.* DOI: 10.1007/s00445-010-0353-z) and 800 degree C (Mourtada-Bonnefoi and Laporte, 2004, *Earth Planet. Res. Lett.*, 218: 521-537). In both studies, bubble nucleation was triggered by decompressing the rhyolitic liquid well below its water saturation pressure at a rate $dP/dt = 1$ MPa/s. Bubble number densities in run products are comparable irrespective of temperature: $9 \times 10^{12} \text{m}^{-3}$ at 700 degree C and $6 \times 10^{12} \text{m}^{-3}$ at 800 degree C. On the other hand, bubble nucleation pressure decreases with decreasing temperature: 55 \pm 5 MPa at 700 degree C vs. 90 MPa at 800 degree C. These results suggest that surface tension increases with decreasing temperature and therefore that the temperature dependence of surface tension is negative. Using classical nucleation theory and assuming that surface tension remains constant during the decompression experiments (at given temperature), we obtained temperature dependence of surface tension = -4×10^{-5} N/m/degree C.

Temperature dependence of surface tension measured by Bagdassarov et al. (2000) is positive while that obtained from our study is negative. We attribute this contradiction to the fact that classical nucleation theory is not quantitatively accurate for the prediction of nucleation rates, especially their dependence with temperature: indeed one major limitation of the classical nucleation theory is that it treats small microscopic embryos as if they have bulk properties, such as macroscopic surface tension. The surface tensions computed from bubble nucleation experiments may not be directly comparable to macroscopic surface tensions because bubble nucleation is a microscopic process.

Keywords: bubble nucleation, surface tension, rhyolitic liquid, classical nucleation theory, sessile drop method