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Accounting for Multi-Phase Carbon in Melt Inclusion Bubbles

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Constraining the carbon flux from volcanism is critical to understanding Earth's volatile budgets and the drivers of explosive eruptions. Our best estimates of pre-degassing CO_2 contents can be obtained from olivine-hosted melt inclusions (MI). Up to ~90% of the CO_2 contained in MI can be found in a vapor or "shrinkage" bubble, and thus, the total CO_2 concentration of a MI requires measurement of exsolved CO_2 in the bubble as well dissolved CO_2 in the glass. An additional challenge is that some MI include CO_2 sequestered as carbonates on the bubble walls or CO_2 found as co-existing liquid (L) and gas (G) at ambient temperature (T_{amb}=18-25 °C). Raman spectroscopy is a powerful tool to study MI, as it is possible to measure their density using a calibration of the P-V-T-X properties of CO_2 related to spectral features (e.g., Fermi diad separation). But for this technique to properly work, CO_2 must be a single phase (L, G, or supercritical). To solve the carbonate problem, we produced fast re-heating experiments on olivine-hosted MI from the Fall Stratified deposit of Etna to resorb carbonate crystals back to CO_2. We measured the bubble CO_2 and glass H_2O before and after heating with our calibrated Raman microscope. These short-duration heating experiments were sufficient to successfully resorb the carbonates as CO_2 and allow accurate density measurements (with change of up to ~0.2 g/mL) without losing H_2O. A set of these MI have also been fully re-homogenized by internally heated pressure vessel resulting in comparable CO_2 contents, validating the Raman results. The second challenge is related to many MI bubbles containing both L+G CO_2 at T_{amb} (densities of ~0.2-0.7g/mL which should not be measurable by Raman spectroscopy, but often reported). Therefore, we conducted a series of experiments at T_{amb} and 37°C (above the critical temperature T_{crit} of CO_2) with different instrument parameters (e.g., laser power) on natural L+G bearing bubbles in MI from Ross Island. Based on these results, we recommend laboratories to measure MI bubbles while heating above T_{crit} to homogenize coexisting phases, and to use low power to minimize the effect of further laser heating.