Development of the geobarometer using size and mineral species dependence of residual pressure of fluid inclusions

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Accurate measurements of the residual pressure of the fluid inclusions at given temperature has a potential to guess the depth provenance and *P–T–t* path of the rocks from the equilibrium temperature of the rocks and the equation of state of the fluid (Levresse et al. 2016; Oglialoro et al. 2017). Since this method does not depend on mineral species, it can also be applied to rocks which cannot use conventional geobarometer based on the mineral composition. Furthermore, since fluid density quickly responds to ambient temperature and pressure change, it is possible to trace a phenomenon on a short time scale. Although the existence of mineral species and size dependence of residual pressure (Yamamoto et al. 2008; Campione 2018) has been pointed out, but its geological significance is not well understood. Therefore, in this study, we measured the residual pressure of the fluid inclusions in the various mineral species by Raman spectroscopy, and discussed the possibilities of fluid density variations as the geothermometer, geobarometer, and geospeedmeter.

However, in order to assess re-equilibration processes of the fluid inclusions, it is necessary to obtain physicochemical information of fluid inclusions of various sizes including very small ones since the fluid retentivity depends on the sizes of the fluid inclusions (Bodnar et al., 1989; Doppler et al., 2013; Campione, 2018). Particularly with respect to the residual pressure, the most reliable size range of fluid inclusions for density calculation is close to the submicrometer level since the smaller the fluid inclusions are, the higher the pressure retentivity is (Bodnar et al., 1989; Wanamaker et al., 1990; Campione et al., 2015; Campione, 2018). Although microthermometry is available to determine the residual pressure of H_2O–CO_2 fluid inclusions (Bakker and Diamond, 2000), for fluid inclusion smaller than ~5 μm and/or those in which the CO_2 liquid and vapor homogenize to the vapor phase upon heating, it is difficult to estimate the internal pressure of the fluid accurately (Rosso and Bodnar, 1995; Kobayashi et al., 2012).

Alternatively, in order to measure the residual pressure of the smaller fluid inclusions, several previous studies have described the application of Raman spectroscopy for quantitative analysis of the residual pressure for various fluid composition. Several groups have improved CO_2 barometers (densimeters) and shown the pressure (density) dependence of the wave number difference (Δ) between the two large peaks of the Raman spectrum of CO_2 (e.g. Hagiwara et al. 2018). In case of H_2O–CO_2 mixture, relationship between Δ and pressure (density) can be altered since some H_2O will be dissolved in the CO_2 depending on fluid pressure even at ambient temperature. However, how the H_2O affects the CO_2 peak positions is still unknown. Practically, some studies have shown that other fluids in a gas mixture affect the Raman spectral features of Δ (Seitz et al. 1996; Lamadrid et al. 2018). Therefore, we experimentally confirmed that the Raman based CO_2 barometer can extend to high pressure H_2O–CO_2 system that may be encountered in natural systems, which is common in fluid inclusions in mantle xenoliths (Roedder, 1984; Frezzotti et al., 2012). By using this method, we measured the residual pressure of fluid inclusions in submicron to 10 μm in mantle xenoliths.

Raman spectra were acquired using excitation by a diode pumped solid-state laser (532 nm, Gem 532; Laser Quantum), and using a spectrometer with 75 cm focal length (Acton SP-2750; Princeton Instruments, Inc.) and a CCD camera (1650 ×200 pixels, 16 μm width, iVac; Andor Technology). The

laser power was ~10 mW at the sample. The respective sizes and depths of the fluid inclusions were measured using a 2000× objective (VH-ZST; Keyence Co.) with a digital microscope (VHX-5000; Keyence Co.). The fluid inclusion size was measured automatically by identifying the outline of a fluid inclusion using the brightness contrast.
The results show that the residual pressure of the fluid inclusion systematically changes depending on the diameter of the fluid inclusion, and the smaller the fluid inclusions are, the higher the pressure retentivity is. Furthermore, the absolute value of the residual pressure retained by the mineral species are also different. Based on these data, we discuss the cause of the variation of the residual pressure of the fluid inclusions from the viewpoint of the brittleness, elasticity and plasticity characteristics of the fluid inclusion-host mineral system.

Keywords: Fluid inclusion, Raman spectroscopy, Geobarometer, Mantle xenolith