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3D Raman mapping of complex fluid inclusions to shed light on fluid compositions in the deep lithosphere

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The micron-scale resolution of Raman spectroscopy revolutionized fluid and melt inclusion research in the past decades, which lead to a better understanding of fluid-related geological processes, widely known to be ubiquitously present in the Earth's deep lithosphere. Raman spectroscopy is a non-destructive and versatile vibrational spectroscopic technique that can be widely applied on mapping out the solid, liquid and vapor phase assemblages within fluid and/or melt inclusions, routinely used for 2D approach. In this study, our aim is to demonstrate the limitations, strength and applicability of 3D Raman mapping, with special attention to the determination of volume proportions of each constituent phases within the inclusions. We studied garnet-hosted multiphase fluid inclusions (MFI) in eclogites and granulites from the Cabo Ortegal Complex (NW-Spain) from where a robust petrographic and geochemical database, including 2D Raman maps is available[1]. These MFI represent deep subduction fluid and contain complex, optically non-distinguishable solid-fluid phases at room temperature.

As an independent control, 3D Raman models were compared to Focused Ion Beam - Scanning Electron Microscopy (FIB-SEM) submicron-scale slicing of three previously 3D Raman mapped inclusions, allowing us to optimize Raman spectroscopy data evaluation and provide volume proportion calculations. Using model analysis (e.g. classical least squares) with curated reference spectra, together with empirically determined Raman cross sections of the phases inside the inclusions can vastly improve the quality of Raman maps. Adjustment of the reference spectra should be done based on 2D map layers, to adjust the size of the inclusion to the optical image. In case of garnet-hosted MFI, the host garnet is always overestimated, thus an adjustment factor of 300-400% should be applied, meanwhile the Raman intensity signal of underrepresented high-density CO_2/CH_4 fluids need to be adjusted to 30% of the intensity of the host garnet. We found that a non-optimized evaluation procedure might result in even an order of magnitude over- or underestimation of phase volume properties. The results provide more precise, valuable input data for thermodynamic modelling and draw implications for fluid/rock interactions.

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References

[1] Spránitz et al., 2022; *J.Metamorph.Geol.,* 40:1291–1319.