

Element detection in sulfate-rich fluid inclusions via Raman spectroscopy and LA-ICP-MS

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Rare earth elements (REEs) are critical metals that are essential for the global transition to green energy and are supplied mainly by carbonatite-related REE deposits. The ore-forming fluids of many world-class REE deposits (e.g., the Maoniuping, Dalucao deposits in China) are commonly sulfate (SO₄²⁻)-dominant [1-2]. Sulfate-rich fluids may transport the REE [3-4] that form many carbonatite-related rare earth element (REE) deposits and are responsible for over half the global REE resource. These fluids are often trapped as fluid inclusions, distributed throughout every stage of the mineralization. Accurate measurement of the elemental concentrations in these fluids inclusions is critical for understanding the genesis of carbonatite-related REE deposits and for guiding their exploration. Quantitative determination of the concentrations of REEs and other elements in such fluids inclusions is, however, very challenging. Here, we report the development of an analytical method that combines Raman spectroscopy and laser ablation-inductively coupled plasma-mass spectrometry (LA-ICP-MS) in determining the major and trace element concentrations of sulfate-rich fluid inclusions (Fig. 1). We show that using sulfur (measured by Raman spectroscopy) as an internal standard, our method enables determination of even the trace element concentrations to an accuracy of ±10%, and the limit of detection (LOD) for trace elements in synthetic fluid inclusions can indeed be quite low, potentially reaching levels as low as 0.4 µg/g. This provides an important new tool for understanding the mobilization and enrichment of REEs during REE ore deposit formation.

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[3] Migdisov & Williams-Jones (2008), *Geochimica et Cosmochimica Acta* 72, 5291-5303.

[4] Wan, Y., Wang, X., Chou, I.-M. & Li, X (2021), *Earth and Planetary Science Letters* 569, 117068.

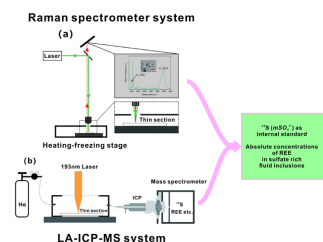


Fig.1 Schematic representation of the quantification method using Raman spectrometer and LA-ICP-MS.
 a The procedure for determining sulfate content in sulfate-bearing fluid inclusions via Raman spectrometer.
 b Following the determination of sulfate concentration using Raman spectroscopy, the same thin section was transferred to the LA-ICP-MS chamber for subsequent analysis.
 The results from both techniques were then integrated to calculate the absolute concentrations of REEs and other trace elements in the sulfate-rich fluid inclusions.