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In-situ Cl/Br measurements in scapolite and fluid inclusions by LA-ICP-MS: A powerful tool to constrain fluid sources

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Chlorine and bromine are highly conservative elements, and are therefore widely used to trace the origin of fluids in sedimentary and hydrothermal/magmatic systems (e.g. Hanor & McIntosh, 2007; Nahnybida et al., 2009). Halogens are important ligands for metal transport in hydrothermal solutions and thus their behavior in hydrothermal environments is crucial for comprehending ore-forming processes. Besides fluid inclusions, scapolite-group minerals hold great potential as a tracer of igneous, metamorphic, and hydrothermal processes, as no Cl/Br fractionation in scapolite has been observed and therefore halogen ratios in scapolite are thought to mirror the halogen ratios in coexisting melts and fluids (Pan & Dong, 2003). Hence, Cl/Br ratios in fluid inclusions and minerals can be utilized to trace the origin of fluids and fluid-rock interaction pathways. Due to their high ionization energies, bromine and chlorine are not routinely measured by LA-ICP-MS and suitable standards are rare. Little is known about the potential interferences and analytical limitations of in-situ chlorine and bromine analysis by LA-ICP-MS. Nevertheless, Seo et al. (2011) showed that quantification of Br and Cl in single synthetic and natural fluid inclusions is possible.

In this study, we have analyzed several scapolite grains of known bromine and chlorine concentrations by LA-ICP-MS and assess the capabilities and limitations of this method. The results show that Cl/Br ratios measured by LA-ICP-MS closely reproduce known values determined by microprobe (Cl), the Noble Gas Method (Br) and INAA (Br) (Kendrick, 2011; Lieftink et al., 1993) using laser ablation spot sizes from 24–120 μm. The well-characterized scapolite grains cover bromine concentrations from 50–883 ppm and chlorine concentrations from 3 to 4 wt.%. In order to further assess the method, we analyzed Cl/Br ratios in natural fluid inclusions hosted in sphalerite that were previously characterized by crush and leach ion chromatography. This is the first time that bulk crush and leach Cl/Br analyses can be compared with Cl/Br ratios within individual fluid inclusions. Our LA-ICP-MS measurements are in good agreement with bulk crush and leach analyses. For instance, molar Cl/Br ratios of single fluid inclusions (183$\pm$33) in sphalerite form East Tennessee match those obtained by crush and leach (206$\pm$8) Additionally, scapolite in samples from dykes of the Burstall granite, associated banded skarns and metasediments from the Mary Kathleen Fold Belt, Queensland, Australia are being studied.
Scapolite is highly luminescent and therefore, cathodoluminescence images resolve chemical zoning in scapolite group minerals that, in combination with in-situ Cl/Br analyses, is a powerful tool to better understand fluid sources and fluid-rock interaction within various geological environments.

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