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Triple-Halogen (Cl, Br, I) Systematics of Scapolite from the Tietangdong Fe-Au Skarn Deposit, North China Craton

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Halogens, especially chlorine (Cl), bromine (Br), and iodine (I), are important complexing agents for various metals and have been used as indicators for the sources and evolution of hydrothermal systems. However, such a use has been limited due to the challenge in performing precise analysis of halogens of fluid inclusions trapped in minerals, notably for Br and I. Experimental studies have shown that the partition coefficients of halogen ratios (Br/Cl and I/Cl) between scapolite and the ore-forming fluids are close to 1. As such, the halogen ratios in scapolite-group minerals approximate to values of the fluids from which the minerals precipitated. Here, we present results of fs-LA-HR-ICP MS (femtosecond laser ablation high-resolution inductively coupled plasma mass spectrometry) halogen analysis of scapolite from the Tietangdong Fe-Au skarn deposit in the North China Craton to better understand triple-halogen behavior during ore-forming processes.

Scapolite at Tietangdong occurs as coarse-grained crystals (0.2–1.5 cm) closely associated with garnet, diopside, and magnetite. Both scapolite and diopside contain vapor-rich and daughter crystal-bearing fluid inclusions that form petrographically defined fluid inclusion assemblages, which indicate fluid unmixing during the skarn formation. The scapolite has high and relatively homogeneous halogen concentrations, with 1.84–3.41 wt % Cl, 388.8–806.2 ppm Br, and 8.4–24.4 ppm I, corresponding to high Br/Cl ratios of (6.1–14.7) × 10^{-3}, and I/Cl ratios of (90.8–302.0) × 10^{-6}. Besides, the scapolite displays a positive correlation between Br/Cl and I/Cl molar ratios (Fig. 1).

The halogen ratios are an order of magnitude higher than the values typical of magmatic hydrothermal fluids ([0.65–2.47] × 10^{-3}; for Br/Cl and [9.34–121.00] × 10^{-6} for I/Cl). This contrast cannot be explained by any mixing trends because no end-member fluids with higher halogen ratios exist (Fig. 1). We suggest that the high Br/Cl and I/Cl ratios in the Tietangdong scapolite most likely reflect the preferential partition of Br and I into brine during fluid phase separation. This view is partly supported by the halogen compositions of daughter mineral-bearing fluid inclusions in quartz of the Tsushima magmatic-hydrothermal system in Japan, which have Br/Cl ratios ranging from 0.49 × 10^{-3} to 5.5 × 10^{-3} (Kurosawa et al., 2016). Recent experimental study has also revealed preferential partition of Br into brine phases formed by fluid unmixing so that the brine has high Br/Cl ratios (Liebscher et al., 2006). Their experiments also showed that the partition coefficient of Br between the brine and vapor phases is pressure- and temperature-dependent and positively correlated with the extent of fluid unmixing. Using a Rayleigh trace element fractionation model, we found that the brine would have Br/Cl and I/Cl ratios of 18 × 10^{-3} and 500 × 10^{-6}, respectively, if the fluid unmixing occurred at temperature lower than 450ºC and pressure of ~300 bar (Fig. 1), conditions similar to those under which the scapolite at Tietangdong formed.

Fig. 1. Molar I/Cl vs. Br/Cl ratios of scapolite samples and the modeling results of halogen partition during fluid unmixing using a Rayleigh fractionation model. Various halogen reservoirs are also shown for references.