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Calcium Isotopes Record Rapid Mineral Precipitation Rates in Skarn Systems

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Skarn systems are formed through interactions between silicate melts or magmatic fluids and carbonate country rocks (e.g., limestones, marble, etc.). During exoskarn formation, magmatic fluids drive metasomatic dissolution-precipitation reactions that generate permeability, promoting fluid flow and host rock replacement by calc-silicate minerals. Although the overall lifetimes of skarn systems can be constrained through radiometric dating methods, fewer constraints exist on mineralization rates and on the duration of skarn-forming fluid pulses [1]. Unlike radiogenic isotope systems, Ca isotope ($\delta^{44}Ca$) fractionations are sensitive to the kinetics of crystal growth [2]. Faster precipitation rates lead to greater mineral-fluid isotopic disequilibrium, whereas slow precipitation rates favor isotopic equilibrium, governed mainly by temperature and mineral composition.

We present Ca isotope data for mineral separates, including garnet (grossular-andradite), epidote, johannsenite ($CaMnSi_2O_6$), diopside, and hornblende from three skarn deposits in Spain (Cala), Bulgaria (Petrovitsa), and Canada (Bancroft), and compare them with new PBE-based *ab-initio* predictions for Ca isotope equilibrium between calc-silicates and carbonates. Epidote has the lowest $\delta^{44}Ca$ values, down to ~0.5‰ lower than host carbonates, consistent with formation near isotopic equilibrium at temperatures of 300-400°C. Grossular, andradite, hornblende, and diopside also agree with our equilibrium predictions, with higher $\delta^{44}Ca$ than host carbonates (up to ~0.6‰).

On the other hand, banded layers of acicular johannsenite crystals from Petrovitsa appear to preserve disequilibrium signatures, with $\delta^{44}Ca$ up to ~0.4‰ lower than predicted at relevant temperatures. Taking into account current best estimates for input parameters (*i.e.* α_f, α_{eq}, α_p, temperature- and pH-dependent R_b), these effects require precipitation rates of ~10^{-6} to ~10^{-5} mol $m^{-2}s^{-1}$ that correspond to growth timescales of ~10^0 to ~10^1 years for 5-cm thick (prograde) johannsenite layers. These timescales are shorter than estimated for retrograde mineralization in skarns (~10^3 years, from Al diffusion in quartz) [3], but consistent with geophysical observations of magmatic-hydrothermal unrest [4]. Calcium isotope data and models will be discussed alongside various timescale estimates, many of which increasingly point towards rapid mineralization in ore-deposit systems.

[1] Chiaradia et al. (2013) *Econ. Geol.*; [2] DePaolo et al. (2011) *GCA*; [3] Li, Y. et al. (2022) *Geology*; [4] Petrosino et al. (2023) *Earth Planets Space*.