Fluid composition and carbon isotope evolution in the Borrowdale graphite deposit (United Kingdom)

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Graphite was discovered at the Borrowdale volcanic-hosted deposit (Cumbria, UK) by mid 16th century. The graphite deposit occupies about a 400 m length of a conjugate set of normal faults and is hosted by andesitic rocks of the upper Ordovician Borrowdale Volcanic Group, and by a probably coeval hypabyssal dioritic intrusion. The volcanic rocks are underlain by the low-grade metapelites of the Skiddaw Group. Graphite in the Borrowdale deposit occurs as: i) nodular masses (up to 1 m in diameter) in pipe-like bodies along fault intersections (1 x 3 m in cross-section and up to more than 100 m in length), ii) fault-veins in the volcanic rocks, usually associated with chlorite, and iii) as replacements (disseminations) within the volcanic host rocks. Graphite morphologies include flakes (>90 vol%), cryptocrystalline (colloform) aggregates, and spherulites. The textural sequence of graphite morphologies (spherulites \rightarrow cryptocrystalline \rightarrow flakes) suggests precipitation from fluids with progressively lower supersaturation in carbon.

Fluid inclusions were studied in quartz fragments associated with graphite in the pipes. Microthermometric and Raman data allowed the recognition of four types of inclusions. The composition trend of these fluid inclusion assemblages shows an overall fluid evolution characterized by: 1) depletion in volatiles, i.e. the carbonic species are transferred to the solid state as graphite, and 2) progressive decrease in the $XCO_2/(XCO_2+XCH_4)$ ratio. Bulk carbon isotope ratios of graphite nodules and microscale SIMS study of the different graphite morphologies show light $\delta^{13}C$ values (-34.7 to -23.4 ‰) compatible with the assimilation of biogenically-derived carbon from the Skiddaw metapelites. Within the main mineralized breccia pipe-like bodies, cryptocrystalline graphite is lighter than flaky graphite, which is consistent with the progressive loss of CO_2 in the mineralizing fluids. Late graphite-chlorite veins contain isotopically heavier spherulitic graphite than flaky graphite. This agrees with CH_4 -enriched fluids at this stage of the mineralizing process.