

Metastable equilibrium in the C-H-O system: Graphite deposition in crustal fluids

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ABSTRACT

The presence of graphite in natural environments is linked to the redox and thermal conditions of C-H-O fluid/graphite equilibrium in hydrothermal veins and metasomatic contacts. A time-series experimental study was performed to investigate the graphite undersaturated C-H-O system at 600 °C and 1000 MPa, and with f_{O_2} ranging from highly reducing (10^{-23}) to highly oxidizing (10^4). A non-volatile intermediate carbon phase exhibiting the Raman spectral features of poorly ordered graphite was formed as the system evolves toward equilibrium as a function of run duration. The thermometric empirical expressions using the G and D bands in the spectra of graphite failed to accurately estimate the experimental temperature. Thus, the existing Raman geothermometers appear inadequate to address graphite formation under conditions of metastable equilibria and to account for kinetic effects such as, for example, the degree of crystallinity. The presence of poorly ordered graphitic carbon at all the redox conditions investigated suggests that the disordered structure of the mineral attains an extensive thermodynamic stability field, and that it may be more readily deposited than crystalline graphite. Metastable graphitic carbon could, therefore, function as a precursor and substrate for the formation of the well-ordered phase. Such metastable graphite may provide an intermediate state that facilitates subduction of carbonaceous material, while imposing constraints on the formation mechanisms and the $^{13}\text{C}/^{12}\text{C}$ isotopic systematics of deep seated carbonaceous fluids and minerals such as diamonds.

Keywords: Graphite, eclogitic diamonds, Raman spectroscopy, metastable equilibrium, C-O-H system, metamorphism, hydrothermal deposition, $^{13}\text{C}/^{12}\text{C}$ isotopic fractionation