Abiotic organic synthesis in olivine-hosted fluid inclusions: The origin of abiotic CH_4 in (ultra-)mafic seafloor hydrothermal systems and ophiolitic gas seeps?

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Fischer-Tropsch type (FTT) processes are commonly invoked to explain abiotic CH_4 formation in serpentinization systems; however, established models fail to explain the occurrence of radiocarbon-free CH_4 in and seafloor hydrothermal systems and ophiolitic gas seeps, which suggests that CH_4 formation is decoupled from circulating fluids[1-3]. This study examines the formation of CH_4 in olivine-hosted secondary fluid inclusions from slow- to fast-spreading mid-ocean ridges, subduction zones, and ophiolites. Raman spectroscopy of inclusion contents reveals chrysotile, brucite, and magnetite as the dominant alteration mineral assemblage and CH_4 or H_2 (or both) as the dominant volatiles in most samples. The formation of CH_4 is envisioned as a 4-step process: 1) Cooling of igneous rocks to below the ductile-brittle transition temperature allows fracturing and creates pathways for hydrothermal and magmatic fluids. 2) At temperatures >~400°C olivine is stable in the presence of H_2O, which can cause healing of fractures and entrapment of fluids. 3) Cooling to lower greenschist facies conditions destabilizes olivine, which reacts with trapped H_2O to form daughter minerals and H_2. 4) During this process, aH_2O and f_{O2} decrease within the inclusion, creating conditions conducive to CH_4 formation. The olivine host appears to be sufficiently impermeable with respect to CH_4 and H_2 at low temperatures to store these volatiles until they are released by fracturing or dissolution of their host. The range of $\delta^{13}C$ of trapped CH_4 overlaps with the measured range of $\delta^{13}C_{CH_4}$ in submarine vent fluids and continental gas seeps in both mafic and ultramafic substrates[4]. The concept of CH_4 synthesis in fluid inclusions over geological timescales and subsequent mining of trapped CH_4 provides a plausible mechanism for the occurrence of radiocarbon-dead abiotic CH_4 in a wide range of geological settings, including mafic and ultramafic seafloor hydrothermal systems and ophiolitic gas seeps.
Refs.: 1) Abrajano T. A., et al., (1990) *Appl. Geochem.* 5, 625–630. 2) Etiope, G., et al. (2016) *Appl. Geochem.* 66: 101–13. 3) McDermott J. M., et al. (2015) *Proc. Natl. Acad. Sci.*, 7668-7672. 4) Grozeva et al. (submitted to *GCA*).