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Mantle oxidation initiated by slab-derived carbonate fluids: insights from CH_4±graphite inclusions in orogenic peridotites

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Carbonate fluids derived from deep-subduction slab are important agents in crust-mantle interaction and can alter the oxidation state of overlying mantle wedge. Here we compile direct evidence of how the evolution of carbonate fluids in subduction channels facilitates element transfer, initiates mantle oxidation and results in abiotic CH_4 formation.

In the mantle-type orogenic peridotites from the Maowu mafic-ultramafic body in the Dabie orogen, China, we observed two centimeter-scale metasomatic veins developed in harzburgite, with lithological composition transitioning from magnesite clinopyroxenite to magnesite orthopyroxenite. Petrographic structures, as well as high Ca/Al (56-144) and $(La/Yb)_N$ (56-144) ratios of clinopyroxene, indicate carbonate metasomatism. The metasomatic products consist of magnesite, Ti-clinohumite, garnet, clinopyroxene and orthopyroxene. The proportion of magnesite in clinopyroxenite is higher than that in orthopyroxenite, which suggests that the magnesite clinopyroxenite vein formed earlier.

Evolution of carbonate fluids is represented by the primary CH_4±graphite inclusions enclosed in magnesites with negative crystal shapes. Carbon isotope of the magnesites with relatively few inclusions is -2.89±0.69‰ (n=7), indicating an isotopically homogeneous carbonate fluid.

V and Cr abundance decrease from core to rim in clinopyroxene, whereas there is a core-to-rim increase in V and Cr in the adjacent garnets. U-Pb dating on zircons associated with the occurrence of the CH_4±graphite inclusions yielded ages of 226-220 Ma, a period of peak metamorphism or early exhumation stage of the deeply subducted continental crust. Given limited changes in temperature and pressure during that period, changes in fO_2 during metasomatism process is probably the dominant driver of the variation in partitioning of V and Cr between clinopyroxene and garnet. Other compelling evidences for change in redox state of the mantle include magnetite exsolutions in the olivines and Ti-clinohumites as well as core-to-rim variation in Fe content in magnesites. Thereby, CH_4±graphite inclusions are likely to form through redox reactions between the evolving carbonate fluids and mantle minerals with multivalent elements. This study also proposes a new formation mechanism of abiotic methane in subduction zones.