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Once carbonated, does it carbonate better? The complex origin of peridotite-hosted carbonates and its implications for carbon storage

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This study investigates carbonates hosted by partially serpentinized peridotites from the Braszowice-Brzeźnica Massif, representing the Devonian oceanic lithosphere formed in the slow-spreading regime. The complex story of this massif includes prograde metamorphism, reaching its climax within the amphibolite facies, emplacement of syn- and post-Variscan magmas, as well as tropical weathering event(s) in the Cenozoic. Theoretically, carbonate mineralization could have been triggered by each of these processes. However, previous studies predominantly suggest a weathering origin and low-temperature alteration of ultramafic minerals. To reassess these findings, LA-ICP-MS, and isotopic studies were conducted.

Carbonate mineralization appears as extensive veins and vein-like structures. The majority of veins exhibit a high modal abundance of cryptocrystalline magnesite accompanied by chalcedony or quartz. Field investigations revealed that carbonate veins containing magnesite-dolomite and calcite-dolomite are less common. Based on both bulk and single-spot chemical composition, some discrepancies have been observed between carbonates. For example, varied chemical compositions of trace elements have been pinpointed among veins sampled at different depths. At least two groups of carbonate veins can be distinguished based on their $^{87}Sr/^{86}Sr$ ratios. Veins sampled from the pit floor, exhibiting Mgs or Mgs±Dol or Cal±Dol paragenesis, consistently display 0.7064 - 0.7065 values. Carbonate veins located in shallow depths in Braszowice show $^{87}Sr/^{86}Sr$ values ranging from 0.7070 to 0.7113 suggesting two sources of Sr during formation of the carbonates. However, the rough correlation between $^{87}Sr/^{86}Sr$ and Rb/Sr may also indicate that the majority of carbonates formed shortly after ultramafic rocks formation. The $\delta^{13}C_{(VPDB)}$ values for carbonates range from -13.8‰ to -10.6‰, while $\delta^{18}O_{(SMOW)}$ values vary from 22.0‰ to 29.6‰.

Additionally, peridotite was subjected to carbonation experiments for 48 hours at 185°C under a PCO_2 of 150 bar. The extent of carbonation was significant, with magnesite comprising ~47 wt.% of the post-experimental material. In contrast, two other rock samples from nearby ultramafic massifs, which underwent carbonation under the same conditions contained only 13 wt.% and 14 wt.% of magnesite, respectively. We hypothesize that the presence of primary carbonates may enhance carbonation efficiency, primarily by facilitating the nucleation of secondary carbonates.

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