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Hydrothermal CO_2 Mineralization in Cabo Ortegal Dunite (NW Spain): From Silicate Thermal Activation to Magnesite Formation

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Dunite from the Cabo Ortegal Complex (NW Spain) was selected as a natural, Mg-rich feedstock to evaluate its potential for CO_2 capture by hydrothermal mineral carbonation. Pristine dunite is dominated by silicate phases, principally forsterite and lizardite, with additional minor components such as brucite. The material was subjected to a thermal pre-treatment at 650 °C to (i) activate the silicate assemblage by increasing its reactivity and (ii) minimize possible carbonate contribution prior to the CO_2–fluid experiments. Hydrothermal reactions were performed under controlled conditions at 175 °C, for reaction times ranging from 1 to 90 days, in a closed system (i.e., a hydrothermal reactor) with CO_2 injected at 10 bar. The study focused on the formation of carbonated mineral products, primarily magnesite ($MgCO_3$) as a stable mineral sink for CO_2, and the associated alteration of the silicate phases, which governs the supply of Mg and potentially controls the reaction progress. Scanning electron microscopy observations provided textural and morphological evidence for dissolution–precipitation processes, allowing the identification of carbonate growth rhombohedral habits and spatial relationships between newly formed phases and silicate substrates. FTIR spectroscopy was used to analyse features of carbonate groups and changes in silicate bonding environments. Moreover, X-ray diffraction allowed the identification of these crystalline phases under the different reaction conditions. To quantify overall carbonation efficiency, thermogravimetric analysis was used to determine total carbonate contents in reacted materials. Across the investigated conditions, carbonate yields were on the order of ~5-10 wt%, indicating measurable CO_2 uptake and highlighting the sensitivity of mineral trapping to reaction time and silicate alteration extent. These results demonstrate that Cabo Ortegal dunite constitutes a promising precursor material for mineral CO_2 storage and provide practical constraints for scaling mineral carbonation pathways, supporting the development of verifiable CCS strategies.