

Exploring reaction pathways for efficient carbonatization and control of trace-metal mobility in ultramafic rocks

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The formation of listvenites (quartz-carbonate alteration assemblages) in ultramafic rocks has been studied as natural analogue for the development of effective industrial applications for CO₂ storage in the subsurface and *ex-situ* carbon mineralization methods. In Oman ophiolites, where the structural relationships and mineral associations have been studied with greater detail, it has been suggested that the carbonatization process occurred at the same time and after an initial stage of serpentinization, which led to complete obliteration of the pre-existing olivine-pyroxene assemblage. The conditions at which the processes of carbonatization and silicification took place are still debated but are reported to vary over a wide range of temperature values, reflecting a multi-stage process affected by percolation of CO₂-rich fluids at different times and structural levels.

With the goal of identifying the most favorable conditions to the carbonatization of ultramafic rocks and understanding the mobility of trace-metals involved in the overall process (Mn, Ni and Cr), we conducted an experimental study using a partially serpentinized harzburgite sample from the Oman ophiolitic complex. A series of experiments at T between 90 and 180 °C and pCO₂ of 15-40 bar was carried out using both batch and stirred-flow Ti-reactors. We studied the chemical evolution of the fluid and analyzed the sequence of reactions controlling the release of Fe and Mg and limiting the conversion of silicate minerals to carbonates. The highest extents of carbonation were achieved between 120 and 150 °C, with the appearance of abundant Fe-bearing magnesite and amorphous silica precipitation, whereas at 180 °C and under initially oxic conditions rapid Fe-oxidation reactions and formation of secondary silicates, such as talc, strongly limited the formation carbonate minerals reducing Mn and Ni aqueous concentrations. We also determined the dissolution rates of antigorite as a function of pH and pCO₂ up to 150 °C to better assess the role of serpentine minerals in the carbonatization process relative to olivine and pyroxenes. The collected data allow new quantification of the reaction sequence involved in ultramafic rocks carbonatization and should help to implement future strategies of CO₂ storage in the subsurface.