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The role of cyclic hydro-fracturing and Si-rich fluids on efficiency of serpentinite carbonation: the example of Malentrata magnesite deposit (Tuscany, Italy)

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Carbon dioxide storage by carbonation of magnesium silicate minerals such as serpentine and olivine is a possible way to reduce greenhouse gas emissions. This process is relatively unknown and consists in an exothermal reaction enhanced by temperatures lower than 200 ˚C where carbon dioxide is bind into the lattice of carbonate minerals forming stable and environmentally benign by-products such as magnesite. Fossil mineral carbonation systems, which occur as magnesite deposits hosted by intensively altered serpentinites, are well-exposed in Tuscany (Italy). Magnesite deposits are hosted by ultramafic rocks involved in a complex stack of units made up of ophiolitic and sedimentary slices (Ligurian units) dissected by NNW-SSE extensional structures bounding the Tuscan Neogene sedimentary basin. Steeply dipping fractures and extensional faults control the mineralized veins, whose Upper Pleistocene age has been constrained by U-Th dating of hydrothermal carbonates. These hydrothermal systems are particularly suitable for studying the processes of carbonation in order to implement the industrial techniques of *in situ* and *ex-situ* CO_2 sequestration. Here, we present the first investigations of the Malentrata magnesite vein deposit (Colline Pisane) and its altered serpentinitic host rocks.

Serpentinites have been mostly altered to a red-brown and green, friable rock characterized by an assemblage of opal, chalcedony, Al-Mg-Cr phyllosicates, with less Fe-rich magnesite and minor amounts of iron sulfides and oxides. Some darker lenses appear more cohesive as a result of the complete silicification and the lack of phyllosi-

cates. The observed mineral assemblages, together with the absence of talc, are indicative of a hydrothermal alteration triggered by a Si- and CO_2-rich fluid, under relatively high pH, and low temperature/pressure condition. The rocks have been fractured, brecciated and crosscut by at least three subsequent generation of veins: i) early irregular network of thin veins of brownish Fe-rich magnesite; ii) intermediate layered, fine-grained zoned magnesite veins with variable Fe content, and iii) thick whitish coarse-grained dolomite veins. The early stage of veining is intermittently associated with the silicification of the rock and repeated brecciation events, leading to the formation of a complex brecciated-veined texture. This intense and multiple hydro-fracturing enhanced the process, increased the porosity and created new conduits for the input and output fluids.

The main ore bodies consist of sub-vertical veins up to 4 m thick and hundred meters wide embedded in serpentinites. The veins are essentially made up of Fe-rich magnesite, dolomite, quartz, and minor amounts of iron sulfides and oxides. Field observations and petrographic studies indicate a complex sequence of crystallization of i) early microgranular magnesite, ii) comb-textured dolomite cementing the brecciated magnesite veins, and iii) late precipitation of quartz, chalcedony and opal in the cavities. The observed crystallization sequence suggests a progressive temporal change in composition of the hydrothermal fluid, at micro and mega-scale.

The example of Malentrata deposit and the altered serpentinitic host rocks highlight the role of cyclic hydro-fracturing and Si- and CO_2-rich fluids on efficiency of serpentinite carbonation and suggest a multi-step process of dissolution of serpentinite, substitution and carbonate precipitation. The concomitant presence of CO_2 and silica in the fluid enhanced the CO_2 mineral sequestration by triggering the serpentine destabilization and inhibiting the early and large precipitation of carbonates. The supersaturated solutions were able to escape the reactor zone being focused in the main structures where massive carbonate precipitation occurred.