

On carbonatization fronts in serpentinite: Implications for *in situ* CO₂ storage

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In situ storage of CO₂ in ultramafic rocks is considered as a promising strategy to counteract global warming. In order to identify positive and negative feedback mechanisms for the reaction, examination of natural analogues is essential. Here we present observations from a massively carbonatized serpentinite complex in northern Norway. The investigated area is comprised of several ultramafic fragments of a dismembered ophiolite complex. Individual fragments are hydrothermally altered to varying degrees resulting in coexisting serpentinite, ophicalcinate, soapstone, and listvenite. Generally, reaction fronts between the soapstone and the serpentinite precursor are extremely sharp and can be traced for hundreds of meters. Locally, soapstone forms m-wide reaction halos around talc veins that cross-cut the serpentinite. The reaction fronts are defined by the complete replacement of the former antigorite by talc and magnesite. Where present, listvenite is spatially related to soapstone and cross-cut by abundant subparallel quartz-magnesite veins. The vein minerals exhibit a homogeneous O-isotopic composition and indicate a formation temperature of ~ 300 °C. The corresponding isotopic signature of the fluid ($\delta^{13}\text{C}_{\text{PDB}} = 2.2(5)\text{‰}$; $\delta^{18}\text{O}_{\text{SMOW}} = 7.8(1)\text{‰}$) suggests an interaction with crustal rocks and devolatilization of associated carbonates as a possible source for the CO₂. Assuming that during alteration temperature differences between directly adjacent soapstone and listvenite units are insignificant, thermodynamic phase stability modeling suggests that the conversion of serpentinite to listvenite requires a higher PCO₂ than that of conversion to soapstone.

In situ carbon storage schemes should aim to convert peridotite to listvenite as the complete inventory of divalent metal cations would be incorporated into the carbonates. However, our observations show that listvenite is distinctly less frequent than soapstone, implying that a high PCO₂ was not maintained during the natural alteration. Furthermore, alteration with a lower PCO₂ developed sharp fronts between the reacted and unreacted rocks, indicating that the carbonatization reaction ceased abruptly before completion.

Oxygen overshoot and recovery during the Early Paleoproterozoic

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During the Lomagundi Event, >2.22 to 2.06 Ga, marine carbonates recorded the largest and longest positive carbon isotope excursion, the earliest extensive marine sulfate evaporites were deposited, and the average Fe₂O₃/FeO ratio of shales increased dramatically. At the end of the Lomagundi Event, the first economic sedimentary phosphorites were deposited, and the carbon isotope values of marine carbonates returned to ~0‰ VPDB. Thereafter marine sulfate evaporites and phosphorites again became scarce, while the average Fe₂O₃/FeO ratio of shales decreased to intermediate values between those of the Archean and Lomagundi-age shales. We propose that the large isotopic and chemical excursions during the Lomagundi Event were caused by a positive feedback between the rise of atmospheric O₂ level, the weathering of sulfides in pre-2.3 Ga sedimentary rocks, and the flux of phosphate to the oceans. The rise in the terrestrial phosphate flux led to an increase in the burial rate of organic carbon and a major transfer of oxygen from the carbon to the sulfur cycle.

The end of the Lomagundi Event was probably caused by a decrease in the terrestrial phosphate flux related to the weathering of low-pyrite sediments that were deposited during the Lomagundi Event. The rate of deposition of organic matter and burial rate of sulfate evaporites decreased, the isotopic and chemical excesses of the Lomagundi Event were eliminated, and the ocean-atmosphere system entered the period frequently called the Boring Billion.