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Experimental constrain of hydrogen production during early serpentinization stages

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The hydrothermal alteration of ultramafic rocks along axial valleys at low spread oceanic ridges lead to the production of hydrogen, as well as hydrocarbons and organic molecules in presence of CO2. The natural production of these compounds is of fundamental interest for the understanding of the formation of prebiotic molecules, and has a potential economical interest for the production of natural hydrogen.
In order to improve our knowledge of the processes involved in the hydrogen production, different sets of experiments have been conducted using either powder or centimetric fragments of natural peridotites as starting material. Powder samples were altered at 300 bar and different temperatures (50, 150, 250, 300, 350 and 450°C) for different durations from 0 to 170 days.. Rock fragments were altered at 300°C/300 bar, between 0 and 84 days. The characterization of the alteration products was first carried out using SEM, EMPA, XRF and XRD. Then, XANES investigations were used to study the evolution of the oxidation state and structural environment of iron. For each alteration time, the relative amount of phases was determined from linear combinations of the XANES spectra. On the other hand, high resolution μ-XANES maps were collected at the iron K-edge for the characterization of the centrimetric fragments. These maps give information on the speciation of iron from the core to the border of the alteration zones. The results allow us to constrain the kinetics of alteration in natural context.
Kinetics of serpentinization is more efficient between 250 and 450°C and is maximized around 300°C. At this temperature, two processes of iron oxidation and hydrogen production were identified from the experimental results. In the experiments shorter than two months, serpentine shows a significant incorporation of ferric iron (up to 60%) and the amount of newly formed iron oxide and hydroxide is less than XXX%. In contrast, iron oxides and hydroxides are produced in significant quantities above XXX months. It follows that the production of hydrogen is not directly related to the amount of iron oxide and hydroxide in the early serpentinization stages, but is related to the bulk Fe3+ content of the sample, included either in oxides or in phyllosilicates. Our results allow us to define a model of “Fe3+ in serpentine vs hydrogen production” as a function of time, which can be used to estimate the amount of hydrogen produced in natural rocks. The evolutions of the Fe3+/Fetotal ratio for both powders and rock fragments allow us to discuss in terms of kinetics of fluid-rock interactions and associated hydrogen production.