

Supplementary Material S1: Field emission electron probe microanalyzer (FE-EPMA) chemical characterization of starting olivine grains

Major element concentrations in olivine were determined at the University of Lausanne (Switzerland) using a JEOL JXA-8530F Hyper Probe field emission electron probe microanalyzer (FE-EPMA). The instrument was operated at an acceleration voltage of 15 kV and a probe current of 10 nA, resulting in a beam diameter of 3 μm . Chemical compositions for the olivine grains used in the experiments and for San Carlos olivine grains are provided in Tables A1 and A2, respectively.

Table A1. EPMA chemical compositions for the olivine grains used in the serpentinization experiments (number of grains = 3).

n = 15	MgO	SiO ₂	Al ₂ O ₃	MnO	Cr ₂ O ₃	CaO	TiO ₂	NiO	FeO	Mg#
Average	49.80	41.12	0.01	0.13	0.01	0.05	0.01	0.37	8.52	0.91
Minimum	49.42	40.98	0.00	0.12	0.00	0.04	0.00	0.32	7.59	0.91
Maximum	50.75	41.24	0.02	0.15	0.03	0.06	0.03	0.40	8.95	0.92
St. deva.	0.41	0.09	0.01	0.01	0.01	0.01	0.01	0.02	0.43	0.01

Table A2. EPMA chemical compositions of San Carlos olivine from Jarosevich et al., 1980 for comparison [1].

	MgO	SiO ₂	Al ₂ O ₃	MnO	Cr ₂ O ₃	CaO	TiO ₂	NiO	FeO	Mg#
Average	49.42	40.81	-	0.14	-	<0.05	-	-	9.55	0.90
Sigma ratios	1.00	0.81							0.9	

Sigma ratios are for 10 grains and equal to the observed sigma for 10 grains divided by the sigma predicted from counting statistics [1]

1. Jarosevich, E.; Nelen, J.; Norberg, J. A. Reference samples for electron microprobe analysis. *Geostandards Newsletter* **1980**, *4*, 43–47.

Supplementary Material S2: Olivine serpentinization in deionized water

The reaction experiment carried out in high-purity deionized water was used as a reference for comparison with the experiments run in NaOH and NaHCO₃ solutions. Even after 12 months of reaction in deionized water, olivine alteration remained very superficial, thus testifying to lower reaction rates than in alkaline solutions. The grains were partially covered with a 5-20 μm thick layer of reaction products (Fig. A1) consisting of lizardite with minor amounts of magnetite. The areas of the olivine free of reaction products revealed mammillated or etch pitted surfaces. Dissolution notches rarely exceeded 20 μm in depth and only a few cracks were observed. Compared to reactions in NaOH solution, layered veins filled with reaction products were few and narrow (Fig. A2).

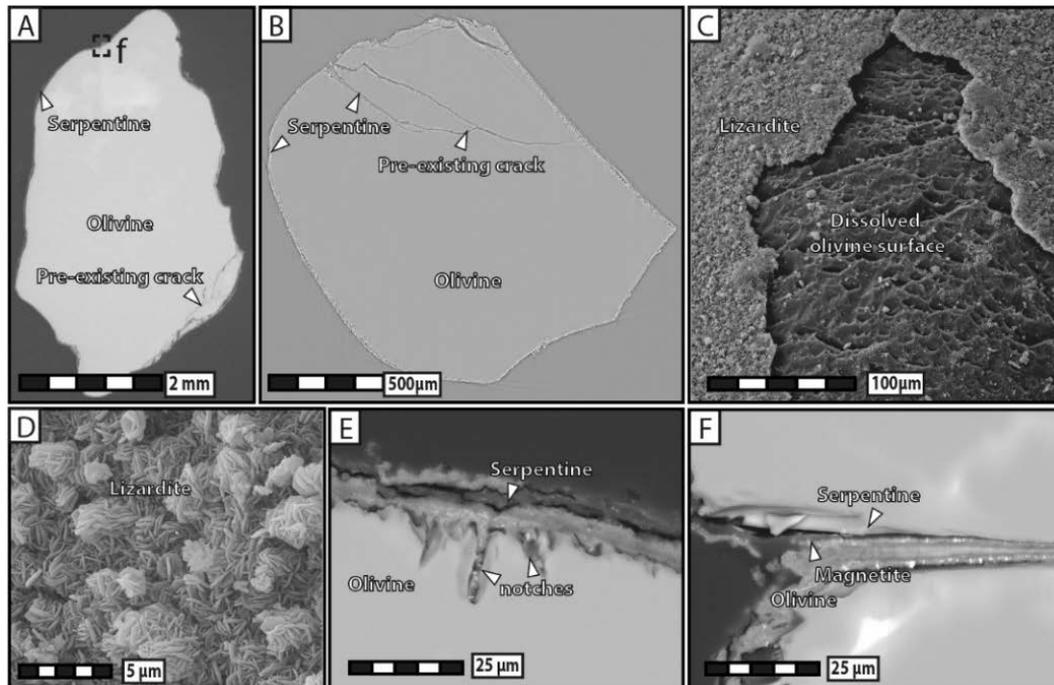


Figure A1. Olivine alteration after 12 months of reaction in high-purity deionized water. (A) BSE image of a starting olivine grain crosscut by a pre-existing crack. (B) Phase contrast X-ray synchrotron micro-tomography slice of an olivine grain filled with serpentine. (C) SE image of mammillated olivine surface covered by a thin lizardite cover. (D) SE image of well-developed lizardite crystals growing onto olivine. (E) BSE image through the alteration rim showing the thin serpentine cover layer and dissolution notches. (F) BSE image of a layered vein filled with serpentine and magnetite.

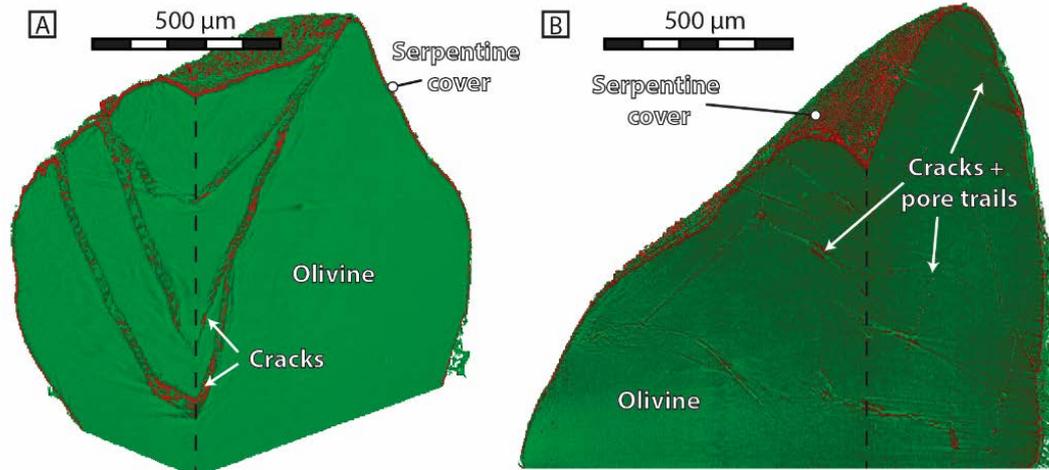


Figure A2. Coloured phase-contrast X-ray synchrotron microtomography slices of olivine grains (A) after 12 months of reaction in deionized water, and (B) after 6 months of reaction in NaOH (run # 2). The higher reaction rate in the latter leads to a widespread development of the fracturation and porosity networks, while only a few pre-existing cracks can be observed in the former.

Supplementary Material S3: Identification of serpentine polymorphs using Raman spectroscopy

In-situ Raman spectroscopy was the method of choice to differentiate the serpentine polymorphs chrysotile and lizardite (Fig. A3), as they are characterized by distinct Raman peaks on the OH stretching range, with a single band at 3697 cm^{-1} , and two bands at about 3684 cm^{-1} and 3704 cm^{-1} , respectively. The technique is based on the interaction of monochromatic light with the vibrational motion of atoms in the crystal lattice. This interaction results in an energy shift in the reflected signal, which can be quantified and used as an identification tool. Raman spectra of the reaction products were collected using a Horiba Jobin-Yvon LabRam HR800 apparatus generating an argon laser beam, 532 nm in wavelength. The instrument was equipped with an Olympus™ BX30 open microscope used for fine focusing of the laser beam on the sample surface (probe size $< 1\text{ }\mu\text{m}$ in diameter). The reflected Raman signal was sampled for 60 s in two cycles, and analysed using a 600 lines/mm grating.

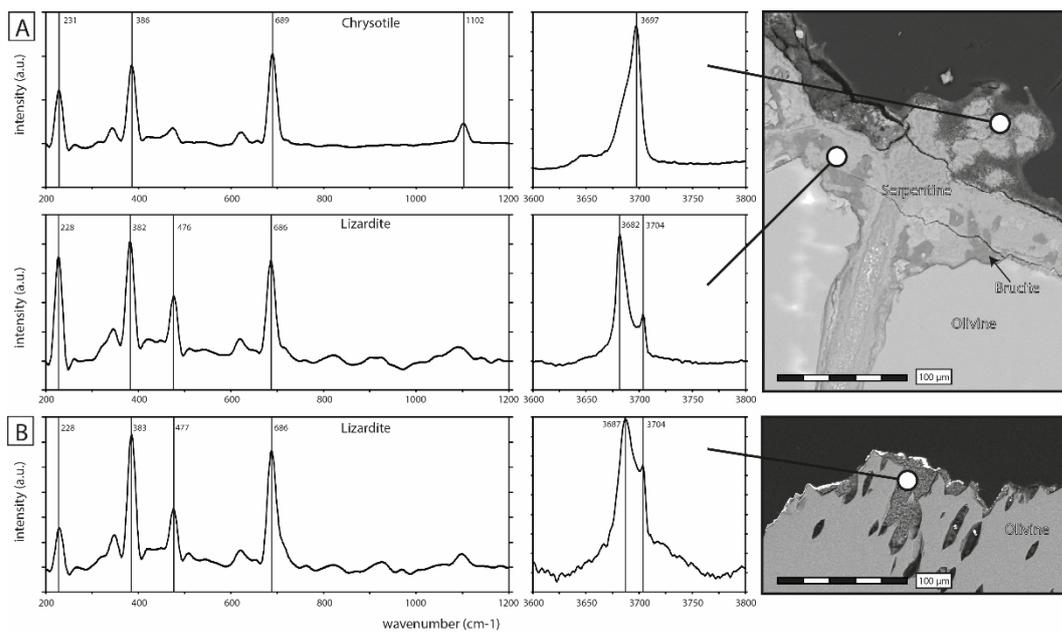


Figure A3. Raman spectra of serpentine polymorphs obtained (A) after 12 months of reaction in NaOH (Run # 3), and (B) after 6 months of reaction in NaHCO_3 (Run # 5). Locations of the analyses are reported on BSE images.