



Incipient hydration of mantle lithosphere at ridges: A reactive-percolation experiment



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ABSTRACT

A reactive percolation experiment was conducted by injecting seawater into a permeable olivine aggregate at 190 °C and 19 MPa to explore the relationships between hydration reactions and hydrodynamic properties during the onset of serpentinization in the ultramafic lithosphere exposed at mid-ocean ridges. The experiment was stopped after 23 days when the sample became impermeable. The initial flow rate was 0.2 mL/h and then was decreased down to 0.06 mL/h after 8 days. Permeability decreased continuously throughout the experiment. The analyses of fluid chemistry time series and of the mineralogy and structure of the reacted sample showed olivine dissolution and precipitation of proto-serpentine, brucite and Fe-oxides. These reactions are controlled by coupled hydrodynamic and chemical processes interacting at different time and spatial scales. Their first characteristic is the production of silica rich outlet fluids in disequilibrium with the observed reacted mineral assemblages. These compositions are interpreted as resulting from a suite of coupled dissolution–precipitation reactions, controlled at the pore scale by surface kinetic processes, that rapidly reaches steady-state (constant fluid composition independent of fluid flux). Hence, the effective rate of serpentinization was controlled, to the first order, by the transport of reactants during the experiment. Mass balance calculations show that the rate of olivine conversion was fast (0.2–0.5 wt%/day), yet only ~8 wt% of the olivine sample reacted, because the permeability drop limited fluid circulation. Porosity varied little during the experiment compared to permeability changes: the decrease of permeability was explained by the structure of the newly formed serpentine favouring the clogging of fluid paths. The rate at which permeability decreased was the fastest at low flux conditions. This suggests that permeability changes were not controlled simply by the kinetics of the serpentinization reactions, but mainly reflected the development of low Peclet pore scale zones forming micro-environments that control the serpentinization reaction-paths. These results outline the need to take into account the coupling of hydrodynamic and chemical processes when modelling the effects of reactive transport within the hydrating oceanic lithosphere.

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1. Introduction

The hydration of the mantle-dominated oceanic lithosphere represents a major means of mass exchange between the mantle and the Earth's envelopes, first at ridges (German et al., 2004; Wilcock et al., 2004) then in subduction zones through recycling and dehydration processes (Deschamps et al., 2011; Li and Lee, 2006). The replacement of olivine by serpentine minerals is the main hydration reaction observed in ultramafic rocks. Serpentinization

leads to significant changes in the rheological (Escartin et al., 1997), physical (Dyment et al., 1997; Germanovich et al., 2012; Miller and Christensen, 1997; Toft et al., 1990) and geochemical properties (Paulick et al., 2006; Vils et al., 2008) of the mantle-dominated lithosphere exposed mostly by large oceanic detachment faults along the volcanic segments of slow and ultra-slow spreading ridges (e.g., Lagabriele et al., 1998). It gives rise to distinctive hydrothermal systems characterized by the emission of fluids enriched in H₂ and CH₄ (Charlou et al., 1998) and the production of complex carbon molecules (e.g., Holm and Charlou, 2001); it is also considered as a major process in sustaining a deep biosphere in ultramafic environments (Früh-Green et al., 2004). Oceanic serpentinization encompasses a protracted suite of fluid–rock reactions occurring at moderate to relatively low temperatures (500–100 °C) during mantle cooling and exhumation (see review in Mevel, 2003). Its main effect is the uptake of large amounts of water to form serpentine minerals

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(~13 wt% H₂O) and other hydrous minerals such as brucite and talc, and minor opaque minerals (magnetite, Ni-Fe sulphides and alloys). The onset and development of these hydrothermal processes requires, first, effective penetration of seawater at depth and then, continuous and efficient renewal of seawater and/or hydrothermal fluids at the surface of primary minerals during hydration reactions.

Geophysical measurements and models suggest that brittle deformation can occur down to 4–8 km along spreading ridges, and tectonically and thermally induced cracking are proposed as possible mechanisms allowing seawater penetration and serpentinization down to these depths (de Martin et al., 2004; Toomey et al., 1988). It is anticipated however that serpentinization will progressively modify the permeability of mantle peridotites by dissolution/precipitation reactions (changes in porosity and flow paths), the density of serpentine being much lower than that of the main constitutive mineral in mantle peridotites, olivine (~2.6 g/cm³ and ~3.3 g/cm³ respectively (Deer et al., 1996)). If the system is mass-conservative, serpentinization will increase the volume of the rock and effectively close porosity. To counteract this effect, crystal growth-induced cracking was proposed as a reaction-driven mechanism maintaining the permeability of serpentinizing peridotites (Iyer et al., 2008; Jamtveit et al., 2008; Kelemen and Hirth, 2012). If the system is volume-constant, serpentinization needs a sustained and efficient fluid flow so that chemical elements are leached out to conserve the rock volume and its permeability. The different mechanisms proposed to explain sustainable permeability during serpentinization are based on the hypothesis that permeability depends solely on porosity. However determining realistic porosity–permeability relationships remains challenging as permeability also strongly depends on local hydrodynamic and chemical conditions and on the initial heterogeneities in the structure and composition of the geological system (Andreani et al., 2009; Luquot and Gouze, 2009), in particular in low porosity rocks such as serpentinizing peridotites. Finally, serpentinization conditions can change with time as recently documented in the MARK area (Andreani et al., 2007). In this zone, serpentinization is a two-step process involving, first, constant-volume reactions with diffusion-dominated mass transfer, then constant-mass reactions with advection-dominated mass transfer. So, sustained fluid circulation and serpentinization within the oceanic lithosphere are controlled by a suite of different and interacting mechanical, hydrodynamic and chemical processes, but the actual coupling and/or feed-back effects between these different mechanisms are still poorly understood.

A flow-through experiment was conducted during which artificial seawater was injected into a sintered San Carlos olivine sample to investigate the relationships between hydrodynamic and chemical processes. The experiment was designed to simulate a simplified avolcanic ridge system in which serpentinization is initiated during exhumation and cooling of mantle-dominated lithosphere (here modelled by olivine). The objectives of this study were (i) to determine the mechanisms controlling the fluid chemistry, the secondary mineralogy and the porosity–permeability relationship in a simplified rock system (mono-mineral, no cracking) during serpentinization, and (ii) to better understand the role and time-scale of these hydrodynamic, mineralogical and chemical processes and their control on the reactivity of the percolated peridotites during the early stages of serpentinization at ridges.

2. Experimental and analytical methods

2.1. Starting material

The experiment was carried out on a cylindrical core (9 mm diameter (*D*) and 18 mm length (*L*)) of sintered olivine Fo90. The sample was prepared as follows: 2–3 mm olivine grains were

hand-picked from a crushed sample of a coarse-grain dunite from San Carlos (Arizona, USA (Andreani et al., 2009)); grains were then crushed in an agate mortar and sieved; 150–300 μm grains were selected for sintering, after being cleaned three times in MilliQ water in an ultrasonic bath; sintering was realized in a Patterson press (Géosciences Montpellier) during 6 h at 1175 °C and 150 MPa. The initial mass of the sample was 3.379 g and its porosity ϕ , determined by the triple weight method (Dullien, 1992), was 11.7%.

2.2. Experimental set-up

The experiment was carried out using the ICARE-Lab flow-through platform. The experimental device used in this study allows injecting fluids at controlled flow rates into low permeability samples at controlled temperature and confining pressure, while monitoring continuously permeability changes (Luquot et al., 2012; Appendix). A pressurized micro-sampling tool enables periodic sampling of the outlet fluids (max volume 3 mL). The experiment was performed with total pressure $P=19$ MPa and temperature $T=190$ °C. Artificial seawater (ASW; Table 1) was injected at a flow rate Q of 55.6×10^{-12} m³/s (0.2 ± 0.005 mL/h) for $t < 195$ h and then 16.7×10^{-12} m³/s (0.06 ± 0.005 mL/h) to the end of the experiment (550 h). These flow rates induce a low Peclet number ($Pe \sim 0.1$ at $t=0$), representative of diffusion-dominated transport and typical of natural systems. Pe is defined as vl/d , where l is the characteristic length of the pores ($\sim 50 \times 10^{-6}$ m), d the diffusion coefficient (5×10^{-9} m²/s at 190 °C (Philibert, 2005)) and v the mean fluid velocity ($\sim 7.5 \times 10^{-6}$ m s⁻¹) defined as $Q/\phi S$, with Q the flow rate (m³/s) and S the cross-sectional area of the sample (m²).

2.3. Permeability measurements

The permeability k of the sample is calculated, assuming flow is laminar, using Darcy's law:

$$k = -\mu LQ / S\Delta P$$

where k is the permeability (m²), ΔP the pressure difference upstream and downstream of the sample (Pa), L the length of the sample (m) in the flow direction, and μ the dynamic viscosity of the water (0.182×10^{-3} Pa·s at 190 °C and 19 MPa for a salinity of 1 mol/kg (Kestin et al., 1981)). The changes in ΔP are measured continuously during the experiment.

2.4. Fluid chemistry

Artificial seawater (ASW) was prepared using analytical-grade Alfa-Aesar chloride, nitrate and bicarbonate salts and deionized water. Its pH, measured at room temperature, is 8.21. Fluids were analysed at the CIRAD (Montpellier, France), using a Varian Vista Pro CCD Simultaneous ICP-OES for Si, Al, Mg, Fe, Ca, Na, Ni, and Cr and by colorimetry using an Alliance Instruments Continuous Flow Analyzer for Cl. The detection limit is 0.01 mg/L for all the analysed elements and the analytical reproducibility is within 5% at concentrations < 0.5 mg/L and 3% at higher concentrations. The minimum volume of water required for these chemical analyses was ~1.5 mL. It determined the minimum time interval between each fluid sampling (~8 h at $Q=0.2$ mL/h and 1 day at $Q=0.06$ mL/h).

2.5. Mineralogical and chemical characterization of the sample after experiments

After the experiment, the sample was longitudinally sectioned and two thin sections prepared for optical and electron microscopy

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