Revision 2

Title:

Aluminum speeds up the hydrothermal alteration of olivine

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Abstract

The reactivity of ultramafic rocks under hydrothermal conditions controls chemical fluxes at the interface between the internal and external reservoirs of silicate planets. On Earth, hydration of ultramafic rocks is ubiquitous and operates from deep subduction zones to shallow lithospheric environments where it considerably affects the physical and chemical properties of rocks and can interact with the biosphere. This process also has key emerging societal implications, such as the production of hydrogen as a source of carbon-free energy. To date, the chemical model systems used to reproduce olivine hydrothermal alteration lead to the formation of serpentine with sluggish reaction rates. Here, we use in situ diamond anvil cell experiments and show that the presence of aluminum in hydrothermal fluids increases the rate of olivine serpentinization by one to two orders of magnitude. Aluminum increases the solubility of olivine and enhances the precipitation of aluminous serpentine. After two days, olivine crystals were fully transformed to aluminous serpentine under conditions typical of natural hydrothermal environments, i.e., 200°C and 300°C, 200 MPa. This result motivates a re-evaluation of the natural rates of olivine serpentinization and of olivine hydrolysis in general in a wide range of settings. This discovery also opens the potential of the serpentinization reaction for industrial scale production of hydrogen at economically feasible timescales and temperature.

Keywords: olivine, hydrothermal, serpentinization, aluminium
1) Introduction

The hydrothermal alteration of olivine is a widespread process that occurs in many geological settings wherever warm aqueous fluids react with ultramafic mantle rocks or olivine-rich magmatic rocks (e.g. wherlite, troctolites, gabbros). Most of the time, olivine is transformed into the hydrous sheet silicate serpentine, plus variable amounts of iron oxides and magnesium hydroxides as illustrated by the olivine serpentinization reaction:

\[
(Mg_{0.9}Fe_{0.1})_2SiO_4 + 1.37 H_2O \rightarrow 0.5 Mg_3Si_2O_5(OH)_4 + 0.3 Mg(OH)_2 + 0.067 Fe_3O_4 + 0.07 H_2 \quad \text{(Eq1)}
\]

olivine (Fo90) Mg-serpentine brucite magnetite

If the system chemistry differs from that locally imposed by olivine, the hydrated product minerals can differ. A silica input from either dissolving neighbouring minerals or the hydrothermal fluid would inhibit brucite formation, and even lead to talc precipitation for higher Si activity. Chlorite and tremolite may also form at olivine-plagioclase contact within olivine-rich magmatic rocks. Talc-chlorite-tremolite-serpentine is actually observed within oceanic detachment faults as a result of Si-Al-Ca metasomatism by fluid arising from distant mafic units (Boschi et al. 2006; Jons et al. 2010).

The hydrothermal alteration of olivine (e.g. Eq1) presents several fundamental and societal implications. It alters the physical, mechanical and chemical properties of ultramafic rocks and it is accompanied by a redox reaction producing hydrogen coupled to reduction of water and oxidation of olivine ferrous component. The ferric iron is integrated into iron oxides and/or into the hydrous phase. Indeed, chlorite and serpentine can both incorporate important amounts of ferric iron in their structure (e.g.
O’Hanley and Dyar 1993; Muñoz et al. 2006; Andreani et al. 2013). Hydrogen, resulting from serpentine and/or chlorite formation, has been identified within fluids venting at ultramafic-hosted hydrothermal sites along mid-ocean ridges (Charlou et al. 2002; Seyfried et al. 2011) as well as in experiments performed in large-volume reactor (Allen and Seyfried 2003; Seyfried et al. 2007; Marcaillou et al. 2011). Production of abiotic H₂ is also thermodynamically predicted using reaction path modelling (Klein et al. 2009). It provides a source of metabolic energy for deep chemoautotrophic ecosystems and could also act as an industrial-scale energy source. In the presence of CO₂, the availability of molecular hydrogen also controls the balance between CO₂-mineralisation and abiotic CO₂-reduction, including Fischer Tropsch-type reactions (McCollom and Seewald 2007; Proskurowski et al. 2008). Ultramafic-hosted hydrothermal systems have thus been proposed as a site for the emergence of the earliest ecosystems (Martin et al. 2008), as well as for CO₂ storage and remediation both at industrial and geological time scales (Kelemen and Matter 2008; Nakamura and Kato 2004; Andreani et al. 2009).

Serpentinized or metasomatized ultramafic rocks are common in the oceanic lithosphere particularly along slow and ultra-slow spreading ridges (Cannat, 1993), in subduction zones within and above the downgoing slab (Peacock 1990; Reynard et al. 2007), along active continental faults (Andreani et al. 2005; Solum et al. 2006) and within Archean supracrustal terranes (Blais and Auvray 1990; Arndt et al. 2008). In extraterrestrial environments, serpentines have been identified in meteorites (Tomeoka and Tanimura 2000), and on the surface of Mars together with chlorite (Ehlmann et al. 2010). The presence of serpentine has also been hypothesized in the core of icy satellites like Titan (Choukroun et al. 2010).
Despite the ubiquity of fully to partially altered olivine in nature, the thermochemical conditions and kinetics of this combined hydration and redox reaction are not yet well established. Serpentine as a result of olivine hydration is widespread. The stability field of serpentine in the MgO-SiO$_2$-H$_2$O (MSH) system extends up to a maximum temperature of 730°C at 2.1 GPa, i.e., the invariant point of the dehydration curve of antigorite. At lower pressure (< 1 GPa) the stability field of serpentine is enlarged toward high temperature as a function of Al content in the MgO-Al$_2$O$_3$-SiO$_2$-H$_2$O (MASH) system (O’Hanley 1996). Serpentinization is active at T < 150°C near ridge axes (Proskurowski et al. 2006), and is suspected at even lower temperature within ophiolites (Barnes and O’Neil 1978). Experimental studies have reproduced the serpentinization reaction of olivine, pyroxene or bulk peridotite at temperatures between 188 and 560°C, and pressures from 20 to 300 MPa (e.g. Allen and Seyfried 2003; Seyfried et al. 2007; Marcaillou et al. 2011, McCollo and Seewald 2007; Martin and Fyfe 1970; Wegner and Ernst 1983; Luquot et al. 2010). These experiments were performed in either pure water or an aqueous fluid closer to seawater composition. They have mostly focused on the chemical evolution of the fluid phase or on the rate of gas or organic compound production. Serpentinization rates were measured in four of these studies only and were found to be very low (Martin and Fyfe 1970; Wegner and Ernst 1983; Luquot et al. 2010; Marcaillou et al. 2011). None of the above-mentioned experiments has used an aqueous fluid with a seawater-modified chemistry, as expected in open natural settings, e.g. after it has interacted with magmatic units or sediments. Although aluminumaluminum is one of the common elements associated with the metasomatism of ultramafic rocks (e.g. Boschi et al. 2006; Jons et al. 2010), its role in the hydrolysis of olivine has only been investigated at low pH (2 < pH < 5) and low T conditions, T=65°C (Chen and Brantley 2000). This is probably due to the fact that aluminum is often considered as an immobile
element because of the very low solubility of aluminous minerals in pure water. Nevertheless, several studies of metamorphic rocks describing ubiquitous Al-bearing mineralization in veins have highlighted the mobility of Al within fluids (e.g. Kerrick 1990). Experimental works have also shown that the presence of Si or Na in solution leads to the formation of Si-Al or Na-Al complexes respectively, for pH >4 to 5 (Diakonov et al. 1996; Manning 2006). This complexation considerably enhances the solubility of alumino-silicates and the mobility of aluminum in metamorphic or metasomatic fluids despite current thoughts on the low solubility of corundum in pure water (Manning 2007). Moreover, abundant aluminum supply exceeding provision by pyroxenes in ultramafic rocks is expected in fluids released from the dehydration of metapelites in subduction zones as well as during the hydrothermal alteration of gabbros at mid-ocean ridges. Aluminum was also abundant in primitive environments of both the Earth and Mars, stored in either Al-rich minerals like plagioclase or Al-enriched ultramafic lavas like Munro-type or other high Al₂O₃/TiO₂ komatiites (Arndt et al. 2008).

In order to investigate the influence of Al on the hydrothermal alteration of olivine in those contexts, we have performed a series of experiments using a low-pressure diamond anvil cell (lp-DAC, Supporting Figure S1 and Oger et al. 2005) under P-T conditions representative of lower crust while following the reaction progress in situ by optical imaging and by confocal Raman spectroscopy.

2) Methods

Temperature in the low-pressure diamond anvil cell (lp-DAC, Supporting Figure S1) was monitored with a thermocouple in contact with the gasket. Aluminum was introduced either as a gel of aluminum hydroxide formed by reaction of AlCl₃ with water and sodium hydroxide for experiments run at 200°C (pH set at 7, at 25°C), or as Al₂O₃ ruby
microspheres for experiments run at 300°C (pH measured at 25°C = 6.8). The most
appropriate source of aluminum has been used for the two different temperatures in
order to always ensure an excess of aluminum in the fluid and keep similar cations in the
initial fluid for all experiments. Indeed, the reactivity and dissolution kinetics of Al$_2$O$_3$ in
a 0.5 m NaCl aqueous fluid was tested separately in a temperature-step experiment,
which showed an almost instantaneous dissolution of Al$_2$O$_3$ microspheres at a
temperature above 280°C only. Therefore, another source of Al, minimizing chemical
changes, has thus been chosen for the 200°C experiments. Al$_2$O$_3$ ruby microspheres also
allowed pressure measurement and pressure cell calibration. Pressure was set and
maintained constant throughout experiments by having a tight control of the gas
membrane pressure of the Ip-DAC. The sample was sealed with a Ni-gasket lined with
gold to prevent reaction between the sample chamber and the gasket. Dimensions of the
reacting cell are 380 µm in diameter and 300 µm thickness (Figure S1).
The progress of reaction was monitored by optical microscopy and confocal Raman
microspectrometry using a Horiba Jobin-Yvon LabRAM HR800 Raman spectrometer at
the ENS (Lyon, France) using green laser at 514 cm$^{-1}$ with a beam size of ~3 µm.
X-Ray Diffraction (XRD) spectra have been acquired under an angle-dispersive mode
using a monochromatic radiation (33 keV, λ=0.3738 Å), focused into a spot of 8x8 µm in
size.

3) Experimental results

Experiments were run for 4.5 to 7.5 days with two olivine (Ol) grains reacted in saline
water (0.5 mol NaCl /kg) at 200°C and 300°C, and P=200 ±30 MPa. To ensure an excess
of Al in the fluid under the experimental conditions, aluminum was introduced either as
a gel of aluminum hydroxide for experiments at 200°C, or as Al₂O₃ ruby microspheres for experiments at 300°C. The importance of nucleation in the kinetics of olivine serpentinization has also been tested by adding lizardite (Mg-serpentine) nuclei in a subset of experimental runs. Table 1 summarizes the experimental conditions and main results of the experiments reported hereafter.

Aluminum-free experiments N°1 and N°2 do not show any visible reaction progress even after one-week duration, whereas reaction starts rapidly when Al is added to the system in experiments N°3 and N°4 at 300°C and in experiments N°5 and N°6 at 200°C. The first grains of reaction products appear on olivine within 2 hours (Table 1) in the absence of lizardite nuclei and the reaction goes to completion in 2 days. The progressive coating of initially transparent olivine and Al₂O₃ microspheres by brown crystals is illustrated in Figure 1. Figure 2 shows the corresponding evolution of the Raman spectra of olivine with time. It indicates the total disappearance of olivine after t=47h, that is confirmed by the XRD characterization of final reaction products (Figure 3C). The softness of the material noticed during its extraction from the lp-DAC also confirms the absence of any relict of olivine crystal in the core of the aggregate and the completion of the reaction. Figure 3A and 3B show the ghost olivine grain fully replaced by micrometric grains of a sheet silicate enriched in aluminum as evidenced by EDS-SEM analyses. X-Ray Diffraction shows that this silicate belongs to the serpentine group of minerals (Figure 3C). It was identified as the Al-rich serpentine amesite by Raman spectroscopy (Figure 2) in all the Al-bearing experiments void of lizardite nuclei (Exp. N°3a, 3b, 5, and 6). Serpentine is the only reaction product observed. Magnetite or brucite predicted by equation 1 for a pure MSH system have not been detected. The serpentine grains are enriched in iron and accommodate the iron released from olivine.
The very fast precipitation of Al-rich serpentine may inhibit magnetite nucleation here. However, this does not rule out the H₂ production since serpentines classically incorporate non negligible amount of ferric iron in their structure that contributes to H₂ production (e.g. O’Hanley and Dyar 1993; Marcaillou et al. 2011; Andreani et al. 2013). Unfortunately, H₂ could not be detected in situ since expected concentrations in the fluid are below detection limit by Raman spectroscopy. Indirect estimation of H₂ by quantifying the ferric iron in microscopic serpentine using synchrotron-based X-ray absorption was beyond the scope of this work. When lizardite nuclei are added to the starting materials at 300°C (Exp. N° 4), new serpentine grains formed after 1.5 days (Supporting Figure S2) with a chemical composition and Raman spectra close to that of the lizardite nuclei. The reaction, however, did not go to completion during the duration of the experiment. We documented a fast partial transformation of olivine into serpentine visible over 3 days and the reaction stopped or slowed down to a rate that precluded further observation. The latter experiment illustrates the importance of removing Al from the fluid or from the reactive interface in order to continue the reaction. Experiment N°3b includes a first temperature step at 250°C for 3 days followed by a second and final step of 300°C for 6 days. The latter experiment confirms that the Al₂O₃ microspheres do not dissolve at 250°C and that, as a consequence, the serpentinization reaction fails to start. Optical observations demonstrate that the onset of serpentinization is correlated with the dissolution of Al₂O₃ microspheres at 300°C and that the first grains of serpentine appear simultaneously on the surface of both olivine grains and Al₂O₃ microspheres (Figure 1B). This supports the critical role of dissolved Al to speed up both the dissolution kinetics of olivine and the nucleation and growth of Al-rich serpentine. It also demonstrates that olivine dissolution is the limiting parameter of
the serpentinization reaction and that olivine solubility is considerably enhanced by the presence of dissolved Al in the fluid.

4) Mutually enhanced dissolution of olivine and alumina

Since Al is not a major component of olivine, it was not expected to directly control olivine’s solubility. At 300°C, 86 bars in aqueous solution, Salvi et al. (1998) nevertheless found that Al(OH)₃(H₂SiO₄)₋ is the dominant aluminous silica species at pH <2 and pH > 4.5. They also observed that the formation of Al-Si complexes increases the solubility of some aluminosilicates or oxide minerals such as kaolinite or boehmite, similar to that was observed at higher P-T conditions (Manning, 2007). Under the conditions of the present experiments, the best pH estimate is 6 at T = 300°C (CHESS code (Van der Lee et al. 2002)). Thus, dissolved Al in our experiments should be complexed to the Si released by dissolving olivine (Salvi et al. 1998). This may be responsible for the observed increase in olivine solubility in our Al-bearing experiments and the fast serpentine precipitation. Such a mutually enhanced dissolution of olivine and alumina leads to the precipitation of amesite under the present experimental conditions (Figure 3) but may also lead to the formation of other Al-rich hydrated silicate, like chlorite for instance, under slightly different chemical conditions. When lizardite is initially present in the system (Exp. N°4), aluminum still accelerates olivine dissolution. Serpentine growth starts on olivine as well as on available lizardite nuclei which leads to a more magnesian chemical composition of the newly formed serpentine. This slows down the reaction since the Al released by the alumina microspheres is less efficiently removed from the solution by serpentine precipitation. Thus, the equilibrium solubility of alumina under those conditions is more rapidly reached. This effect certainly does not apply to open
natural systems where local chemical gradients and fluid renewal at mineral interface are expected to refresh the fluid and thus remove excess Al from the reacting zone. The results of experiments N°3a, 3b, 5 and 6 are compared in Figure 4 to those of the Al-free experiment N°1 and to previously published serpentinization rates of olivine (Martin and Fyfe 1970; Wegner and Ernst 1983; Luquot et al. 2010) and of peridotite (Marcaillou et al. 2011). Assuming a linear conversion rate of olivine into serpentine, we obtain a mean serpentinisation rate of 49% per day in the present study. This is c.a. 7 to 50 times higher than the rates reported under very similar conditions (3.7 - 7.5% per day (Martin and Fyfe 1970); 1 - 3%/day (Wegner and Ernst 1983)). It is also 30 to 50 times higher than the rates of 1-2%/day reported for peridotite at lower pressure (P = 30 MPa) with a smaller grain size of 1 µm (Marcaillou et al. 2011). The latter work also shows that after 77% of serpentinization the conversion rate decreases down to 0.3%/day and approaches that reported at 200°C, 20 MPa during dynamic percolation experiments which was of 0.2-0.4%/day (Luquot et al. 2010); i.e. two orders of magnitude lower than those in the Al-bearing experiment reported here.

5) Implications

In natural open systems where a sustained H₂O flow allows hydration reactions, the rate of olivine alteration may be increased at any time depending on the inlet fluid flux and its Al content. Indeed, Al is always present in variable amount in serpentine minerals ranging from 1 to 27 wt. % of Al₂O₃ (O’Hanley 1996; Beard et al. 2009), and it is a major component of chlorite. Consequently our experimental results suggest that the hydrothermal alteration of olivine is probably much faster in natural settings where peridotites are intimately associated with Al-suppliers such as the common magmatic
bodies in slow-spreading oceanic crust (Cannat 1993). This is actually well illustrated at oceanic detachments where fault material results from peridotite-gabbro metasomatism and where active hydrothermal fields are observed (McCaig et al. 2007). The composition of fluids released at these hydrothermal vents is enriched in \( \text{H}_2, \text{CH}_4 \) and complex organic compounds and is mainly controlled by serpentinization at depth. The local abundance of Al issued from magmatic rocks certainly affects the fluxes of products released during serpentinization, and in consequence the development of the ecosystem associated with these hydrothermal fields (Schrenk et al. 2004). The effect of joined alteration of olivine and Al-bearing mineral, like plagioclase, on \( \text{H}_2 \) production in a natural system has indeed been described by Seyfried et al. (2011), based on the chemistry of the fluids that escape the Rainbow hydrothermal field (36°N, Mid-Atlantic Ridge).

The effect of Al on lower crust and upper mantle metamorphism and metasomatism is expected to be even stronger at high pressure in subduction zones where those reactions control the rheology and physical properties of the subducting plate and mantle wedge (Peacock and Hyndman 1999; Reynard et al. 2007). Indeed, the solubility of Al related to the stability of Na-Si-Al complexes in solution is greater at high pressure (Manning 2007), and may exceed the solubility of Mg or Fe in fluids equilibrated with mafic and felsic bulk compositions (Manning 2006).

Whatever the setting, the rate of metamorphic reaction is also a key parameter for determining the contribution of reaction-enhanced processes at the micrometer-scale, such as reaction-driven cracking, that can affect, in turn, macroscopic rock properties like permeability (Jamveit et al. 2008; Rudge et al. 2010). Reaction-driven cracking has
been proposed as an important positive feedback of the olivine hydration reaction that creates permeability for fluid supply and increases the reactive surface area (Kelemen and Hirth 2012). A well documented example is the case of fractured plagioclase grains between serpentinized or chloritized olivine crystals in troctolites or olivine gabbros, respectively. The present results suggest that the presence of Al in solution should, at least locally, increase the contribution of reaction-induced cracking during retrograde metamorphism of olivine-rich rocks.

The present results also provide a way to accelerate serpentinization reactions towards an economically feasible time-scale for industrial H₂ production and/or CO₂ remediation through the production of carbon compounds with a higher added value like CH₄ and hydrocarbons. In comparison with available technologies for H₂ production that mainly rely on fossil fuels (> 90% of H₂ production by steam reforming and < 1% via electrolysis), olivine serpentinization is a carbon-free process. Energy minimization is also obtained here by the exothermic character of the serpentinization reaction and by lowering the temperature of the reaction to 200°C, as long as Al is available in solution.

6) Acknowledgments

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References


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**Figure legends**

**Figure 1**: Optical images of the reaction progress in the Ip-DAC. Development with time (A to D) of aluminous serpentine at the expense of olivine and Al2O3 microspheres during experiment N°3a (Table1).

**Figure 2**: Monitoring reaction progress on the olivine grain with Raman spectroscopy. Example of reaction N°3a. Confocal Raman spectroscopy allows scanning the olivine grain in depth. The time steps are those illustrated on Figure 1. The final product of the reaction at t=47h is identified as amesite by comparison to the reference spectrum from RUFF database. Olivine is the only mineral detected at t=0 and 2h. Then
Raman modes characteristic of amesite (arrows) are observed with relict olivine at t=16h. Olivine is fully replaced by amesite at t=47h.

**Figure 3 : SEM and XRD characterization of the serpentine grains at the end of reaction.** Example of Exp. N°3a. A) SEM image of the serpentine grains formed at the expense of olivine. B) Zoom in of serpentine grains with EDS-SEM analysis of their mean composition. C) XRD spectra of the serpentine grains with identification of the characteristic peaks of serpentine minerals. No other phases are detected.

**Figure 4 : Comparison of the present results with serpentinization rates available in the litterature.** Results of experiments N°1, 3a, 3b, 5, 6, realized with olivine grain size ~100-150 µm, are plotted in comparison with the available data on serpentinization rate of olivine or peridotite obtained under similar P-T conditions and grain size, i.e. 60-80 µm (Martin and Fyfe 1970) and 105-150 µm (Wegner and Ernst 1983). Available results obtained at lower P conditions (Luquot et al. 2010) and smaller grain sizes ~ 1 µm (Marcaillou et al. 2011) are also reported.
### Table 1: Experimental conditions and main results

<table>
<thead>
<tr>
<th>Solid reactants</th>
<th>Duration (hours)</th>
<th>First serpentine grain (hours from start)</th>
<th>Reaction completion at the end</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.5 mM NaCl in deionized H₂O, pH&lt;sub&gt;ini&lt;/sub&gt;(25°C) = 7 reacted at T=300°C, P=200 MPa</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>N°1: Ol</td>
<td>168</td>
<td>No reaction observed</td>
<td>No</td>
</tr>
<tr>
<td>N°2: Ol + Liz</td>
<td>182</td>
<td>No reaction observed</td>
<td>No</td>
</tr>
<tr>
<td>N°3</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>a: Ol + Al₂O₃</td>
<td>119</td>
<td>2 (± 0.25)</td>
<td>Yes, completion at 48h (± 8)</td>
</tr>
<tr>
<td>b*: Ol + Al₂O₃</td>
<td>144</td>
<td>2 (±0.25)</td>
<td>65h (± 8)</td>
</tr>
<tr>
<td>N°4: Ol + Al₂O₃ + Liz</td>
<td>112</td>
<td>38 (±3)</td>
<td>No, reaction stopped or slowed down at t = 70h (±5)</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Solid reactants</th>
<th>Duration (hours)</th>
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</tr>
</thead>
<tbody>
<tr>
<td>0.5 mM NaCl in deionized H₂O, pH&lt;sub&gt;ini&lt;/sub&gt;(25°C) = 7 reacted at T=200°C, P=200 MPa</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>N°5: Ol + Al(OH)₃ gel</td>
<td>70</td>
<td>1.5 (± 0.25)</td>
<td>Yes, completion at 45h (± 4)</td>
</tr>
<tr>
<td>N°6: Ol + Al(OH)₃ gel</td>
<td>50</td>
<td>1 (± 0.25)</td>
<td>Yes, completion at 37h (± 6)</td>
</tr>
</tbody>
</table>

*Experiment N°3b includes a first temperature step at 250°C for 3 days, that is not reported in the table, followed by a second and final step at 300°C for 6 days, reported in the table. No reaction was observed at 250°C since Al₂O₃ microspheres did not dissolve.

### Figures
Figure 3
Supporting Figure S1. Scheme of the low-pressure diamond anvil cell. (1) diamond anvil, (2) diamond window, (3) Ni sample gasket lined with gold (inner diameter = 380 µm; thickness = 300 µm), (4, 5) tungsten carbide seats. The experimental volume (coloured in blue) is filled with reactants as illustrated by the optical image experiment load N°3a at t=0. For mode details on the lp-DAC, see Oger et al. (2005).
Supporting Figure S2: Optical images of the reacting cell. Development with time of serpentine at the expense of olivine and on initial lizardite nuclei during experiment N°4 (Table 1).
Figure 1
Figure 2
Figure 3
This study, exp. N°1 (300°C, 200 MPa, Al-free,)
This study, exp. N°5&6 (200°C, 200 MPa, with Al)
This study, exp. N°3a&b (300°C, 200 MPa, with Al)

200°C, 300°C, 200-300 MPa (Martin and Fyfe, 1970)
270°C, 320°C, 200-300 MPa (Wegner and Ernst, 1983)
200°C, 20 MPa (Luquot et al., 2010)
300°C, 30 MPa (Marcaillou et al., 2011)