



Mass transfer in the oceanic lithosphere: Serpentinization is not isochemical



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ABSTRACT

Whereas the serpentinization reaction leads to stark differences in the physical properties of mantle rocks at mid-ocean ridges, the chemical changes associated with this reaction are thought to be restricted to the addition of water and the generation of hydrogen (“isochemical” reaction). Here, I compile a geochemical dataset of serpentinized peridotites at mid-ocean ridges evidencing that a decrease by up to 11% of the MgO/SiO₂ ratio is associated with serpentinization. This MgO/SiO₂ decrease is consistent with the calculated distribution of Mg in the minerals since, during isochemical serpentinization, ~10% of the Mg should be contained in brucite, an Mg-hydroxide not commonly observed in serpentinized peridotites, which are typically composed of serpentine (Mg₃Si₂O₅(OH)₄) and magnetite (Fe₃O₄). This latter mineralogical assemblage and a decrease of the MgO/SiO₂ ratio were only reproduced in numerical models of peridotite reacting with fluids containing aqueous silica at fluid to rock (F/R) ratios greater than 20. At higher F/R ratios, talc (Mg₃Si₄O₁₀(OH)₂) was found to be stable, in agreement with observations in extremely altered samples found at mid-ocean ridges. The potential sources for aqueous silica in the fluid are the alteration of mafic units intruding mantle rocks at slow-spreading ridges. The mineralogical and chemical changes associated with SiO₂ gain during serpentinization at mid-ocean ridges will have consequences on abiotic hydrogen production, contribute to a volume increase of 50% and decrease water incorporation during serpentinization by more than 10% compared to “isochemical” serpentinization. These changes will also increase the depth at which fluids are released by dehydration reactions in subduction zones by more than 20 km.

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1. Introduction

Slow-spreading ridges exhume large tracts of mantle-derived ultramafic rocks (Cannat, 1993) and receive a moderate magma supply. Magmatic heat is released through the convection of aqueous fluids in the oceanic crust. When these fluids reach ultramafic rocks, alteration minerals such as serpentine (Mg₃Si₂O₅(OH)₄), brucite (Mg(OH)₂), talc (Mg₃Si₄O₁₀(OH)₂) and magnetite (Fe₃O₄) are formed, and show stark differences in physical and chemical properties compared to the primary minerals. The effect of serpentinization on these properties is already perceptible before the reaction progress even exceeds 10%, with an increase in the recorded magnetization by two orders of magnitude through the production of ferromagnetic magnetite (e.g. Malvoisin et al., 2012a), the formation of a reducing environment through the production of hy-

drogen (Charlou et al., 2002) and the decrease of the rock strength due to the precipitation of serpentine (Escartin et al., 2001). As the reaction progresses further, serpentinization impacts other properties such as rock density since the newly formed hydrated phases have lower densities ($\rho_{\text{serpentine}} = 2900 \text{ kg/m}^3$) compared to the primary minerals ($\rho_{\text{olivine}} = 3300 \text{ kg/m}^3$). This density change contributes to lower mantle seismic velocities (Miller and Christensen, 1997) and is associated with a positive increase in the solid volume by up to 50% in the case of a reaction where water is the only component to be exchanged (Coleman and Keith, 1971). This volume change has two opposite effects on the permeability which both increases through the generation of fluid pressures high enough to fracture the rock (e.g. Plümpner et al., 2012) and decreases when the porosity is filled with secondary phases (Godard et al., 2013). Constraining the chemical and mineralogical changes during serpentinization is thus crucial for understanding reaction-induced evolution of the properties of the oceanic lithosphere.

Fulfilling this latter objective is not straightforward because serpentinization occurs in an open system where the conditions of alteration evolve through time (Andreani et al., 2007). Although the

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effect of temperature on the distribution of iron between brucite, magnetite and serpentine during serpentinization has been quantified with increasing precision (Malvoisin et al., 2012b; Klein et al., 2013), the influence of reacting system composition on the mineralogical evolution during the reaction is still not constrained. In spite of the extensive geochemical datasets available for peridotites (Niu, 2004; Bodinier and Godard, 2003) and fluid compositions (Charlou et al., 2002) at mid-ocean ridges, the respective roles of the initial composition of the system and the mass transfer during the reaction on the final composition have yet to be determined. Mass transfer during serpentinization mainly involves the addition of water and fluid-mobile elements (e.g. B, Li, Cl, As, Sb, U, Th, Sr) susceptible to be released when entering subduction zones (see Deschamps et al., 2013 for a review). Based on the measurement of constant oxides/SiO₂ ratios with the increase in reaction progress (Coleman and Keith, 1971; Komor et al., 1985), the transfer of major elements during serpentinization is thought to be negligible in the general case and the reaction is thus called “isochemical” (Mével, 2003; Deschamps et al., 2013). However, some studies reported changes in the bulk composition of mantle-derived rocks at mid-ocean ridges. Snow and Dick (1995) and Niu (2004) measured a decrease of the MgO/SiO₂ ratio but they attributed this change in composition not to serpentinization itself but instead to low-temperature alteration on the seafloor. Some instances of large changes in the MgO/SiO₂ ratio of abyssal peridotites have been reported during talc formation. At the Atlantis Massif, these changes were interpreted as resulting from the serpentinization reaction itself with a synchronous formation of talc and serpentine (Boschi et al., 2008). At the 15°20′ fracture zone, they were attributed to a high temperature event with Si-rich fluids forming talc at the expense of serpentine (Paulick et al., 2006; Harvey et al., 2014).

Here I use a statistical approach to determine the respective role on abyssal peridotite composition of both the system's initial composition and the change in composition due to fluid/rock interactions at mid-ocean ridges. Further, I thermodynamically model fluid/mantle rock interactions to determine the mineralogical and chemical processes controlling compositional changes and their impact on the physical and chemical properties of the mantle rocks exhumed at mid-ocean ridges.

2. Methodology

2.1. Determining changes in composition during serpentinization with geochemical data compilation

I assembled a dataset consisting of bulk rock major element analyses of 1075 serpentinized harzburgites, dunites and lherzolites, 551 of which were abyssal peridotites compiled from 18 sources and 524 of which were ophiolitic peridotites compiled from 34 sources (see Supplementary Table S1 for details and Bodinier and Godard (2003) for a referencing of the ophiolites).

I targeted the variations in MgO and SiO₂, the two most abundant components in abyssal peridotites, in order to estimate their deviation in composition from the “isochemical” model. These variations depend on the protolith composition and mass transfer during processes such as serpentinization. The influence of the protolith composition was modeled differently depending on its nature. Unaltered dunites are mainly composed of olivine with a constant MgO/SiO₂ ratio of 1.2 (Niu et al., 1997) which was considered representative of the protolith composition. More complex changes in protolith composition from fertile lherzolites to refractory harzburgites were modeled by a progressive magmatic depletion of a primitive mantle following Jagoutz et al. (1979) and Hart and Zindler (1986). This model predicts that the protoliths of serpentinized lherzolites and harzburgites should lie on

a straight line, the so-called “terrestrial array”, in MgO/SiO₂ vs. Al₂O₃/SiO₂ diagrams. As this array was determined with analyses of fresh continental peridotites, it must be tested against data of unaltered peridotites formed at mid-ocean ridges to be used here. From a geochemical perspective, serpentinization is a process during which water is incorporated into mineral phases. To estimate the weight fraction of this bound water in the rock, the loss on ignition (LOI) is measured by heating a powdered sample in a furnace to temperatures of ~1000 °C. Samples with low LOI and thus with low degrees of serpentinization were selected in the database for determining the deviation of unaltered abyssal peridotites from the “terrestrial array”. Other volatile components such as CO₂ can participate in an increase of the LOI (e.g. Schwarzenbach et al., 2013). Carbonate-bearing samples were therefore not included in the compilation. Abyssal peridotites are generally extensively altered as shown by the fact that only 3% of the selected samples have LOI below 4 wt.%. As this is not enough for accurately estimating the deviation from the terrestrial array, ophiolites samples were considered, since 25% of these samples have LOI < 4 wt.%. Moreover, irrespective of the mechanism of serpentinization of the two types of serpentinized peridotites, at decreasing levels of hydration these samples should project back to a common origin.

2.2. Thermodynamic modeling

Fluid/rock interactions were modeled at the scale of geochemical data acquisition, i.e. at the bulk rock scale rather than at the grain scale. Serpentinization relies on two kinds of mineralogical reactions: dissolution reactions releasing aqueous species in the fluid and precipitation reactions allowing the incorporation of new mineral species in the rock. The nature and the extent of these reactions depend both on their kinetics and on thermodynamic equilibrium. Malvoisin et al. (2012b) demonstrate experimentally using powders that the kinetics of serpentinization are controlled by the dissolution of olivine rather than by the precipitation of secondary phases. Therefore, the bulk composition of the reacting part of the rock was assumed here to depend on the kinetics of dissolution of the primary minerals. The mineralogical composition of the reacted peridotite was then determined with thermodynamic modeling tools by neglecting the kinetics of precipitation and by assuming a constant temperature and an initial composition of the reacting system only dependent on the reaction progress and on the initial composition of the peridotite (no spatial variability). These assumptions are shown to be valid in the Supplementary materials P1.

Seismic velocity profiles indicate that serpentinization mainly occurs within the first three kilometers of the oceanic lithosphere (Canales et al., 2000). In these domains, lithostatic pressure variations do not exceed 100 MPa whereas temperature can vary by more than 400 °C. Therefore, the variations of thermodynamic properties due to pressure variations are negligible compared to those associated with temperature variations. All numerical simulations were thus conducted at a constant pressure of 50 MPa. The four parameters considered here as having a potential impact on the composition of serpentinized peridotites at mid-ocean ridges are: temperature, bulk composition of the reacting protolith, composition of the fluid, and relative quantity of protolith and fluid during the alteration (fluid to rock ratio). Two different numerical models were used to understand compositional change during serpentinization.

2.2.1. “Isochemical” serpentinization model

A first model was built to determine the influence of the starting composition of the reacting protolith on the nature and the composition of the secondary minerals during “isochemical” serpentinization. This model was aimed at providing a basis for com-

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