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Geological reactive systems from the mantle to the abyssal sub-seafloor: Preface

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The formation and alteration of the oceanic lithosphere represent one of the main processes for energy and chemical exchanges between the deep Earth and its outer envelopes. However, the steep thermal gradients characterizing these environments, especially at the main thermal and lithological interfaces along mid-ocean accretion zones (Figure 1), mean that the physical and chemical mechanisms controlling these exchanges remain poorly understood. Yet, these interfaces are the main transitions for the physical and rheological properties of rocks, such as permeability and viscosity, that control melt focussing and transport from the partially molten mantle to the surface, as well as deformation mechanisms and the influx of seawater into the cooling oceanic lithosphere. These processes also give rise to hydrothermal systems that produce economically valuable ore-deposits and play a major role in the global chemical budget. Some hydrothermal reactions produce hydrogen and abiotic hydrocarbon, hence these extreme environments sustain life and they are potentially implicated in its origin. Finally, these processes determine the architecture and composition of the lithosphere plunging into the deep Earth along subduction zones, and contribute to a broad range of mechanisms driving arc magmatism and localization of earthquakes in these regions.

Traditionally the formation of oceanic lithosphere has been envisioned as a suite of mantle and crustal magmatic processes, followed by high- to low-temperature hydrothermal processes, which in turn supported the development of diverse ecosystems; in this scheme, each step was considered as occurring independently and in different domains, whilst the interactions and feedbacks controlling mass and energy transfers at their boundaries had been mostly overlooked. However, over the recent years, the role of the physical and (bio-)chemical processes occurring at the interfaces bounding the Earth's envelopes has been progressively recognized. These boundary layers characterize the architecture of Earth's oceanic lithosphere and they are determined by two competitive processes at mid-ocean ridges: conduction and advection of heat from depth, which is dominantly controlled by mantle upwelling (spreading rate) and melt transport from the mantle to the surface, and cooling by conduction and hydrothermal circulation. These boundary layers correspond to isotherms marking transitions in rocks properties and therefore, in turn, they depend on the composition of the lithosphere and are expected to vary greatly from fast-spreading layered oceanic lithosphere (Penrose Model) to slow-spreading heterogeneous oceanic lithosphere (Figure 1). Understanding such complex highly reactive geological systems requires a shift of approach and the development of new scientific tools to comprehend and integrate the role of transport of magmatic melts and hydrothermal fluids through the oceanic lithosphere, and their feed-back on its

physical and (bio-)chemical properties. This volume brings together a series of articles addressing these challenges with natural observations and laboratory experiments.

From mantle lithosphere to magmatic crust

Basch et al. (2018), Ferrando et al. (2018) and Borghini et al. (2018) investigate the nature of the transition from mantle to gabbroic lower crust with the aim to characterize and identify the magmatic processes controlling this transition. Basch et al. (2018) document a transition from mantle peridotites to olivine-rich troctolites, the most primitive end-member of the lower gabbroic crust, preserved in the Mt. Maggiore Ophiolite (Corsica, France). Ferrando et al. (2018) focus on the study of olivine-rich troctolites interlayered within a drilled gabbroic section of heterogeneous oceanic lithosphere (Integrated Ocean Drilling Program (IODP) Expeditions 304/305 Site U1309 Atlantis Massif 30°N, Mid-Atlantic Ridge). Basch et al. (2018) and Ferrando et al. (2018) combine petro-structural and geochemical analyses and numerical modelling of major and trace element variations to determine the respective roles of fractional crystallization and reactive porous melt flow during the incipient stages of the formation of gabbroic oceanic crust. This approach provides criteria to identify and quantify melt/rock reactions and to estimate the contribution of mantle rocks to the formation of the lower crust. These studies give evidences for a likely progressive formation of the lower oceanic crust at the expense of the shallow mantle, and shed light on the control that such processes can exert on melt evolution in the oceanic crust. To better constrain the chemical and physical parameters driving these processes, Borghini et al. (2018) investigate the origin of olivine-rich troctolites and studied melt-olivine reaction using specifically designed experiments. They observe textural development comparable with disequilibrium features observed in natural olivine-rich troctolites (e.g., Basch et al. (2018), Ferrando et al. (2018)) and demonstrate the control of starting melt composition and melt-olivine ratio on modal composition and mineral chemistry of olivine-rich troctolites.

High temperature hydrothermalism and cooling: Impact on architecture and composition of newly formed crust

Koepke et al. (2018), Grant and Harlov (2018), Currin et al. (2018a, b) and Zihlmann et al. (2018) investigate the role of fluids and of hydrothermalism on differentiation processes during cooling of the gabbroic mafic lower crust and their impact on the architecture and composition of the newly formed crust. Koepke et al. (2018) present the first phase-equilibria study of the late stages of MORB differentiation and explore the role of water activity in these systems. On this basis, they propose a two-step differentiation model for the formation of oxide gabbros in slow spread magmatic crust and at the transition from lower to upper layered crust as well as for the formation of highly evolved

lavas at fast- and intermediate spreading mid-ocean ridges. This model emphasizes the prevalence of oxygen fugacity on oxide differentiation suites including late formation of minerals such as apatite and amphibole during cooling down to temperatures of 800-900°C. Grant and Harlov (2018) and Currin et al. (2018a) use experimental approaches to study the behaviour of these systems in the presence of NaCl-brines in order to understand fluid-rock interactions associated with high temperature hydrothermalism (900° to 500°C) not only in oceanic gabbros but also in the deep continental mafic crust. Grant and Harlov (2018) show a correlation between the reactivity of olivine-plagioclase assemblages, water activity and fluid NaCl concentrations. They identify the chemical and temperature conditions the most favourable for the formation of Cl-rich amphiboles in these systems. Currin et al. (2018a) further investigate the processes leading to the formation of amphiboles, and in particular of Cl-rich amphiboles, at the pressure and temperature conditions expected during hydrothermal reactions between seawater-derived fluids and the gabbroic oceanic lower crust. They show that the composition of amphibole is highly variable in hydrothermal systems and that their Cl content is affected not only by the composition of hydrothermal fluids but also by the extent of fluid/rock interactions. Zihlmann et al. (2018) and Currin et al. (2018b) provide new constraints on the role of deep hydrothermal systems at mid-oceanic ridges through detailed field, petrologic and geochemical studies of lower gabbroic sections of layered oceanic crust preserved by the Oman Ophiolite. Zihlmann et al. (2018) identify fault zones as the main flow paths for high temperature hydrothermalism at mid-ocean ridges. They establish that such focussed flow zones contribute to the fast cooling of the gabbroic lower oceanic crust and to the global hydrothermal geochemical fluxes. Currin et al. (2018b) focus on the formation of Cl-rich amphiboles and show that fluid-rock interactions leading to their formation occur in rock-dominated environment.

Serpentinization, sulphides, carbon and geo-resources

The linkages between magmatic, hydrothermal and (bio-)geochemical processes becomes even more prominent when investigating the mechanisms building the shallow oceanic lithosphere and their impact on global geochemical cycles, deep sea natural resources and the development of life as illustrated by Früh-Green et al. (2018). These authors present an overview of the results of the first oceanic drilling expedition dedicated to investigate the interplay between magmatism, serpentinization processes and microbial activity in the shallow subsurface (IODP Expedition 357, Atlantis Massif). This expedition provided a unique sampling of the suite of altered and deformed ultramafic and mafic rocks forming one of the major detachment fault zone along the Mid-Atlantic Ridge as well as first biogeochemical and microbiological characterization of the shallow ultramafic subseafloor.

Rouméjon et al. (2018), Escario et al. (2018) and Pastore et al. (2018) investigate the thermodynamic and hydrodynamic conditions and the localization of serpentinization reactions, the dominant hydration process in the shallow mantle lithosphere. Rouméjon et al. (2018) document the development of serpentine minerals along detachment faults at slow- and ultraslow-spreading ridges using samples drilled at Atlantis Massif (IODP Expedition 357) and dredged along the easternmost Southwest Indian Ridge (SWIR, 62–65°E). The studied serpentine minerals have similar textures in these environments, yet they have variable compositions when associated to gabbros, which suggest cross-contamination by hydrothermal fluids. They are also isotopically heterogeneous for oxygen isotopes down to scales of ~100 µm, and these variations depend on water/rock ratio. Pastore et al. (2018) developed an innovative scanning magnetic microscopy technique allowing mapping, at the millimeter to micrometer scale in serpentinized peridotites, the distribution of magnetite, one of the mineral products of serpentinization reactions. This technique sheds new light on the strong heterogeneity and variability in the direction of the magnetization with respect to the pristine sample in serpentinized samples. Escario et al. (2018) investigate the effects of solute transport on reaction paths during incipient serpentinization of olivine cores using a reactive-percolation experimental approach. They show that, for the same initial fluid and mineral compositions, serpentinization reaction paths vary depending on local flow distribution (at the microscale) and that kinetics- and transport-controlled reaction paths can coexist at the sample scale. These mechanisms favour the development of mineralogical and compositional heterogeneities. These results suggest a contribution of flow rate on the development of the different serpentinization reactions paths observed in the basement and in fault zones in serpentinized oceanic mantle lithosphere, where they are commonly ascribed to changes in the composition of hydrothermal fluids resulting from interactions with different lithologies along flow paths (e.g., Rouméjon et al, this volume).

Los et al. (2018) studied experimentally the formation of sulphides in relation to low temperature hydrothermalism at mid-ocean ridges and more particularly the role of the composition of basement rocks on sulfidization efficiency. Basalt, troctolite, dunite and serpentinite were reacted with H₂S-rich hydrothermal fluids. Sulphides could be observed only in the basalt experiment indicating that olivine-rich seafloor lithologies are not favourable to sulphidation reactions. These results challenge the commonly proposed anhydrite formation models and open new research paths.

Noel et al. (2018) and Menzel et al. (2018) studied in natural samples the formation of carbonates at the expense of ferromagnesian minerals, a process commonly associated to the alteration of mantle peridotite. They carried out a detailed petro-structural, geochemical and carbon and oxygen isotope study of variously altered peridotites from the Oman and the Newfoundland ophiolites. Noel et al. (2018) document successive episodes of carbonate-forming reactions from the Oman Ophiolite, first associated to serpentinization close to a mid-ocean ridge, then to the different stages of the

140 emplacement of the ophiolite. They demonstrate structural and chemical linkages between
141 serpentinization and carbonate-forming reactions during the cooling of the oceanic mantle
142 lithosphere in the presence of CO₂-bearing fluids, and evidence the control of inherited mantle fabric
143 on the field scale orientation of late carbonate veins. Menzel et al. (2018) document a complete
144 sequence of carbonate-forming and redox reactions preserved at the interface in the Advocate
145 Ophiolite Complex (Newfoundland, Canada). They show that this reaction sequence was triggered by
146 an influx of CO₂-rich fluids and that the devolatilization of neighbouring meta-sediments during
147 subduction is the most likely source of these fluids. They suggest that carbonate-bearing peridotites
148 can act as a carbon flux pathway beyond sub-arc depths.

149 Carbon occurs also as abiotic organic compounds as well as organic metabolic byproducts or
150 remnants of microbial ecosystems in the shallow mantle lithosphere. Ménez et al. (2018) show that
151 organic carbon can influence secondary mineral formation as well as the speciation and mobility of
152 transition metals during low temperature serpentinization reactions (< 200 °C). This mechanism has
153 possible implications for understanding ore formation during late active serpentinization of ophiolitic
154 massifs and/or for subsurface carbon dioxide storage in ultramafic rocks.

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164 lithosphere.

Figure Caption

Figure 1: Boundary layers marking the transition of mantle asthenosphere to the shallow sub-seafloor at mid-ocean ridges for heterogeneous (left-hand side) or layered (right-hand side) oceanic lithosphere. They correspond mainly to isotherms : (1) Asthenosphere-lithosphere boundary layer: (i) mechanical boundary (**LithM**): transition between a visco-plastic asthenospheric mantle and a rigid lithosphere (ca. 1000 °C; note that it depends also on the deformation and composition of the mantle); (ii) chemical/petrological boundary (**LithT**): marks the crystallization of clinopyroxene and / or plagioclase (ca 1180°C), this process locally blocks magma flow and induces a change in permeability (and viscosity?) of the partially molten mantle; (2) **Moho**: seismic interface interpreted as marking (i) a change in lithology in layered lithosphere, from mantle peridotite to magmatic crust (max 1180°C at mid-ocean ridges), and (ii) the transition from serpentinite to peridotite in heterogeneous mantle lithosphere (350-500 °C); (3) Lithosphere / hydrosphere interface i.e. limit of penetration of hydrothermal fluids in the lithosphere (**Hy**): (i) In a layered lithosphere, this limit is generally assumed to be the brittle-ductile transition in the oceanic crust (ca. 700°C-750 °C); however, recent works suggest that very high temperature (up to 975°C, **HyHT**) hydrothermalism could develop in lower gabbros; (ii) the dominant mechanisms driving the hydration of the heterogeneous lithosphere remain debated (permeability and role of large faults vs. diffuse penetration); assuming that the lithosphere / hydrosphere limit corresponds to the Moho implies that hydrothermal fluids can interact with the lithosphere down to several kilometre deep.

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