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Geological reactive systems from the mantle to the abyssal sub-seafloor: Preface Godard Marguerite^{1,*}, Fumagalli Patrizia², Jamtveit Bjørn³, Ménez Bénédicte⁴ 1 Géosciences Montpellier, CNRS, Université de Montpellier, 34095 Montpellier, FRANCE ² Dipartimento di Scienze della Terra "Ardito Desio", Università degli Studi di Milano, ITALY ³ The Njord Centre, Department of Geosciences, University of Oslo, 0316 Oslo, NORWAY ⁴ Institut de Physique du Globe de Paris, Sorbonne Paris Cité, Université Paris Diderot, CNRS, 75005 Paris, FRANCE

Contact :

Godard Marguerite Géosciences Montpellier Université de Montpellier Campus Triolet cc60 Place Eugène Bataillon 34095 Montpellier FRANCE Email : Marguerite.Godard@umontpellier.fr

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

2 Godard Marguerite, Fumagalli Patrizia, Jamtveit Bjørn, Ménez Bénédicte

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4 The formation and alteration of the oceanic lithosphere represent one of the main processes for 5 energy and chemical exchanges between the deep Earth and its outer envelopes. However, the steep 6 thermal gradients characterizing these environments, especially at the main thermal and lithological 7 interfaces along mid-ocean accretion zones (Figure 1), mean that the physical and chemical 8 mechanisms controlling these exchanges remain poorly understood. Yet, these interfaces are the 9 main transitions for the physical and rheological properties of rocks, such as permeability and 10 viscosity, that control melt focussing and transport from the partially molten mantle to the surface, 11 as well as deformation mechanisms and the influx of seawater into the cooling oceanic lithosphere. 12 These processes also give rise to hydrothermal systems that produce economically valuable ore-13 deposits and play a major role in the global chemical budget. Some hydrothermal reactions produce 14 hydrogen and abiotic hydrocarbon, hence these extreme environments sustain life and they are 15 potentially implicated in its origin. Finally, these processes determine the architecture and 16 composition of the lithosphere plunging into the deep Earth along subduction zones, and contribute 17 to a broad range of mechanisms driving arc magmatism and localization of earthquakes in these 18 regions.

19 Traditionally the formation of oceanic lithosphere has been envisioned as a suite of mantle and 20 crustal magmatic processes, followed by high- to low-temperature hydrothermal processes, which in 21 turn supported the development of diverse ecosystems; in this scheme, each step was considered as 22 occurring independently and in different domains, whilst the interactions and feedbacks controlling 23 mass and energy transfers at their boundaries had been mostly overlooked. However, over the 24 recent years, the role of the physical and (bio-)chemical processes occurring at the interfaces 25 bounding the Earth's envelopes has been progressively recognized. These boundary layers 26 characterize the architecture of Earth's oceanic lithosphere and they are determined by two 27 competitive processes at mid-ocean ridges: conduction and advection of heat from depth, which is 28 dominantly controlled by mantle upwelling (spreading rate) and melt transport from the mantle to 29 the surface, and cooling by conduction and hydrothermal circulation. These boundary layers 30 correspond to isotherms marking transitions in rocks properties and therefore, in turn, they depend 31 on the composition of the lithosphere and are expected to vary greatly from fast-spreading layered 32 oceanic lithosphere (Penrose Model) to slow-spreading heterogeneous oceanic lithosphere (Figure 33 1). Understanding such complex highly reactive geological systems requires a shift of approach and 34 the development of new scientific tools to comprehend and integrate the role of transport of 35 magmatic melts and hydrothermal fluids through the oceanic lithosphere, and their feed-back on its 36 physical and (bio-)chemical properties. This volume brings together a series of articles addressing 37 these challenges with natural observations and laboratory experiments.

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39 *From mantle lithosphere to magmatic crust*

40 Basch et al. (2018), Ferrando et al. (2018) and Borghini et al. (2018) investigate the nature of the 41 transition from mantle to gabbroic lower crust with the aim to characterize and identify the 42 magmatic processes controlling this transition. Basch et al. (2018) document a transition from mantle 43 peridotites to olivine-rich troctolites, the most primitive end-member of the lower gabbroic crust, 44 preserved in the Mt. Maggiore Ophiolite (Corsica, France). Ferrando et al. (2018) focus on the study 45 of olivine-rich troctolites interlayered within a drilled gabbroic section of heterogeneous oceanic 46 lithosphere (Integrated Ocean Drilling Program (IODP) Expeditions 304/305 Site U1309 Atlantis 47 Massif 30°N, Mid-Atlantic Ridge). Basch et al. (2018) and Ferrando et al. (2018) combine petro-48 structural and geochemical analyses and numerical modelling of major and trace element variations 49 to determine the respective roles of fractional crystallization and reactive porous melt flow during 50 the incipient stages of the formation of gabbroic oceanic crust. This approach provides criteria to 51 identify and quantify melt/rock reactions and to estimate the contribution of mantle rocks to the 52 formation of the lower crust. These studies give evidences for a likely progressive formation of the 53 lower oceanic crust at the expense of the shallow mantle, and shed light on the control that such 54 processes can exert on melt evolution in the oceanic crust. To better constrain the chemical and 55 physical parameters driving these processes, Borghini et al. (2018) investigate the origin of olivine-56 rich troctolites and studied melt-olivine reaction using specifically designed experiments. They 57 observe textural development comparable with disequilibrium features observed in natural olivine-58 rich troctolites (e.g., Basch et al. (2018), Ferrando et al. (2018)) and demonstrate the control of 59 starting melt composition and melt-olivine ratio on modal composition and mineral chemistry of 60 olivine-rich troctolites.

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62 High temperature hydrothermalism and cooling: Impact on architecture and composition of newly 63 *formed crust*

64 Koepke et al. (2018), Grant and Harlov (2018), Currin et al. (2018a, b) and Zihlmann et al. (2018) 65 investigate the role of fluids and of hydrothermalism on differentiation processes during cooling of 66 the gabbroic mafic lower crust and their impact on the architecture and composition of the newly 67 formed crust. Koepke et al. (2018) present the first phase-equilibria study of the late stages of MORB 68 differentiation and explore the role of water activity in these systems. On this basis, they propose a 69 two-step differentiation model for the formation of oxide gabbros in slow spread magmatic crust and 70 at the transition from lower to upper layered crust as well as for the formation of highly evolved 71 lavas at fast- and intermediate spreading mid-ocean ridges. This model emphasizes the prevalence of 72 oxygen fugacity on oxide differentiation suites including late formation of minerals such as apatite 73 and amphibole during cooling down to temperatures of 800-900°C. Grant and Harlov (2018) and 74 Currin et al. (2018a) use experimental approaches to study the behaviour of these systems in the 75 presence of NaCl-brines in order to understand fluid-rock interactions associated with high 76 temperature hydrothermalism (900° to 500°C) not only in oceanic gabbros but also in the deep 77 continental mafic crust. Grant and Harlov (2018) show a correlation between the reactivity of olivine-78 plagioclase assemblages, water activity and fluid NaCl concentrations. They identify the chemical and 79 temperature conditions the most favourable for the formation of Cl-rich amphiboles in these 80 systems. Currin et al. (2018a) further investigate the processes leading to the formation of 81 amphiboles, and in particular of Cl-rich amphiboles, at the pressure and temperature conditions 82 expected during hydrothermal reactions between seawater-derived fluids and the gabbroic oceanic 83 lower crust. They show that the composition of amphibole is highly variable in hydrothermal systems 84 and that their Cl content is affected not only by the composition of hydrothermal fluids but also by 85 the extent of fluid/rock interactions. Zihlmann et al. (2018) and Currin et al. (2018b) provide new 86 constraints on the role of deep hydrothermal systems at mid-oceanic ridges through detailed field, 87 petrologic and geochemical studies of lower gabbroic sections of layered oceanic crust preserved by 88 the Oman Ophiolite. Zihlmann et al. (2018) identify fault zones as the main flow paths for high 89 temperature hydrothermalism at mid-ocean ridges. They establish that such focussed flow zones 90 contribute to the fast cooling of the gabbroic lower oceanic crust and to the global hydrothermal 91 geochemical fluxes. Currin et al. (2018b) focus on the formation of Cl-rich amphiboles and show that 92 fluid-rock interactions leading to their formation occur in rock-dominated environment.

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94 *Serpentinization, sulphides, carbon and geo-resources*

95 The linkages between magmatic, hydrothermal and (bio-)geochemical processes becomes even 96 more prominent when investigating the mechanisms building the shallow oceanic lithosphere and 97 their impact on global geochemical cycles, deep sea natural resources and the development of life as 98 illustrated by Früh-Green et al. (2018). These authors present an overview of the results of the first 99 oceanic drilling expedition dedicated to investigate the interplay between magmatism, 100 serpentinization processes and microbial activity in the shallow subsurface (IODP Expedition 357, 101 Atlantis Massif). This expedition provided a unique sampling of the suite of altered and deformed 102 ultramafic and mafic rocks forming one of the major detachment fault zone along the Mid-Atlantic 103 Ridge as well as first biogeochemical and microbiological characterization of the shallow ultramafic 104 subseafloor.

105 Rouméjon et al. (2018), Escario et al. (2018) and Pastore et al. (2018) investigate the 106 thermodynamic and hydrodynamic conditions and the localization of serpentinization reactions, the 107 dominant hydration process in the shallow mantle lithosphere. Rouméjon et al. (2018) document the 108 development of serpentine minerals along detachment faults at slow- and ultraslow-spreading ridges 109 using samples drilled at Atlantis Massif (IODP Expedition 357) and dredged along the easternmost 110 Southwest Indian Ridge (SWIR, 62-65°E). The studied serpentine minerals have similar textures in 111 these environments, yet they have variable compositions when associated to gabbros, which suggest 112 cross-contamination by hydrothermal fluids. They are also isotopically heterogeneous for oxygen 113 isotopes down to scales of ~100 µm, and these variations depend on water/rock ratio. Pastore et al. 114 (2018) developed an innovative scanning magnetic microscopy technique allowing mapping, at the 115 millimeter to micrometer scale in serpentinized peridotites, the distribution of magnetite, one of the 116 mineral products of serpentinization reactions. This technique sheds new light on the strong 117 heterogeneity and variability in the direction of the magnetization with respect to the pristine 118 sample in serpentinized samples. Escario et al. (2018) investigate the effects of solute transport on 119 reaction paths during incipient serpentinization of olivine cores using a reactive-percolation 120 experimental approach. They show that, for the same initial fluid and mineral compositions, 121 serpentinization reaction paths vary depending on local flow distribution (at the microscale) and that 122 kinetics- and transport-controlled reaction paths can coexist at the sample scale. These mechanisms 123 favour the development of mineralogical and compositional heterogeneities. These results suggest a 124 contribution of flow rate on the development of the different serpentinization reactions paths 125 observed in the basement and in fault zones in serpentinized oceanic mantle lithosphere, where they 126 are commonly ascribed to changes in the composition of hydrothermal fluids resulting from 127 interactions with different lithologies along flow paths (e.g., Rouméjon et al, this volume).

128 Los et al. (2018) studied experimentally the formation of sulphides in relation to low temperature 129 hydrothermalism at mid-ocean ridges and more particularly the role of the composition of basement 130 rocks on sulfidization efficiency. Basalt, troctolite, dunite and serpentinite were reacted with H₂S-rich 131 hydrothermal fluids. Sulphides could be observed only in the basalt experiment indicating that 132 olivine-rich seafloor lithologies are not favourable to sulphidation reactions. These results challenge 133 the commonly proposed anhydrite formation models and open new research paths.

134 Noel et al. (2018) and Menzel et al. (2018) studied in natural samples the formation of carbonates 135 at the expense of ferromagnesian minerals, a process commonly associated to the alteration of 136 mantle peridotite. They carried out a detailed petro-structural, geochemical and carbon and oxygen 137 isotope study of variously altered peridotites from the Oman and the Newfoundland ophiolites. Noel 138 et al. (2018) document successive episodes of carbonate-forming reactions from the Oman Ophiolite, 139 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 CO_2 -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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167 Figure Caption

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169 Figure 1: Boundary layers marking the transition of mantle asthenosphere to the shallow sub-170 seafloor at mid-ocean ridges for heterogeneous (left-hand side) or layered (right-hand side) oceanic 171 lithosphere. They correspond mainly to isotherms : (1) Asthenosphere-lithosphere boundary layer: (i) 172 mechanical boundary (*LithM*): transition between a visco-plastic asthenospheric mantle and a rigid 173 lithosphere (ca. 1000 °C; note that it defends also on the deformation and composition of the 174 mantle); (ii) chemical/petrological boundary (*LithT*): marks the crystallization of clinopyroxene and / 175 or plagioclase (ca 1180° C), this process locally blocks magma flow and induces a change in 176 permeability (and viscosity?) of the partially molten mantle; (2) Moho: seismic interface interpreted 177 as marking (i) a change in lithology in layered lithosphere, from mantle peridotite to magmatic crust 178 (max 1180°C at mid-ocean ridges), and (ii) the transition from serpentinite to peridotite in 179 heterogeneous mantle lithosphere (350-500 °C); (3) Lithosphere / hydrosphere interface i.e. limit of 180 penetration of hydrothermal fluids in the lithosphere (Hy) : (i) In a layered lithosphere, this limit is 181 generally assumed to be the brittle-ductile transition in the oceanic crust (ca. 700°C-750 °C); 182 however, recent works suggest that very high temperature (up to 975°C, *HyHT*) hydrothermalism 183 could develop in lower gabbros; (ii) the dominant mechanisms driving the hydration of the 184 heterogeneous lithosphere remain debated (permeability and role of large faults vs. diffuse 185 penetration); assuming that the lithosphere / hydrosphere limit corresponds to the Moho implies 186 that hydrothermal fluids can interact with the lithosphere down to several kilometre deep.

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