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# In situ CO<sub>2</sub> mineralization in mantle-derived ultramafic basements: Insights from laboratory experiments and field studies (Oman ophiolite)

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## Abstract

Carbon trapping in ultramafic (UM) and basaltic basements is one of the options explored to mitigate industrial CO<sub>2</sub> emissions in the Earth's atmosphere. UM rocks and basalts comprise silicates rich in divalent cations (Mg, Ca, Fe) that are dissolved to form carbonates when in contact with CO<sub>2</sub>-rich fluids, thus trapping CO<sub>2</sub> over geological time scales. UM rocks have the highest concentrations in divalent cations and thus they have the highest potential for carbon trapping by CO<sub>2</sub>-mineralization. Nevertheless, because of their low permeability, UM basements have been overlooked for possible *in situ* CO<sub>2</sub> storage in favor of basaltic basements. Recent research shows that CO<sub>2</sub>-mineralization is active and efficient in UM basements, and that it is associated to potential benefits, such as the production of H<sub>2</sub>. However, the hydrodynamic, physical and chemical mechanisms driving CO<sub>2</sub>-mineralization whilst sustaining fluid flow are still poorly understood and numerous scientific and technological challenges remain before implementing industrial CO<sub>2</sub> geological storage in UM basements. Here we present an overview of our recent results on CO<sub>2</sub>-mineralization in UM rocks combining (i) laboratory experiments, and (ii) field studies of carbonated UM basements with a focus on the Semail ophiolite (Sultanate of Oman), in relation to the recently completed ICDP (International Continental Scientific Program) Oman Drilling Project.

*Keywords:* CO<sub>2</sub> mineralization; ultramafic rocks; peridotites; scientific drilling; laboratory experiments, borehole monitoring, pilot injection site

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## 1. Introduction

*In situ* CO<sub>2</sub> storage in mantle-derived ultramafic (UM) and basaltic basements is one of the options explored to mitigate excess CO<sub>2</sub> emissions produced by industrial activities, and limit the steady increase of greenhouse gases in the Earth's atmosphere [1]. UM rocks (peridotites, serpentinites) and basalts comprise abundant silicates rich in divalent cations (Mg, Ca, Fe) that are dissolved to form carbonates when in contact with CO<sub>2</sub>-rich fluids (CO<sub>2</sub>-mineralization), thus potentially trapping CO<sub>2</sub> over geological time scales [2-3]. Basalts compose most of the oceanic seafloor as well as oceanic islands (e.g., Iceland) and peridotites and serpentinites are accessible in several regions around the world at the limits of tectonic plates in both oceanic and continental environments (Fig. 1, [4]).

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Oceanic peridotites are exposed where mantle is tectonically exhumed along oceanic slow and ultra-slow spreading ridges (e.g., Mid-Atlantic Ridge; Southwest Indian Ridge; American-Antarctic Ridge; Gakkel Ridge) and along hyper-extended avolcanic continental passive margins (e.g., the Iberia and Newfoundland margins), in deeps along the fast spreading East Pacific Rise and in some fore-arcs (e.g., Izu Bonin Mariana, South Sandwich). Continental peridotite massifs outcrop along ancient to still active orogenic belts. They preserve relics of subcontinental mantle (orogenic massifs, e.g., Ronda, Beni Bousera, Pyrenees -including Lherz-) and oceanic lithosphere (ophiolites, e.g., Philippines, Bay of Island, California, Internal Ligurides in the Western Alps, Troodos). Some of these massifs, such as the Oman and New Caledonia ophiolites, are several km thick and outcrop over 10's of km<sup>2</sup>, and are exploited today for metals, such as copper, manganese, chromium and nickel.

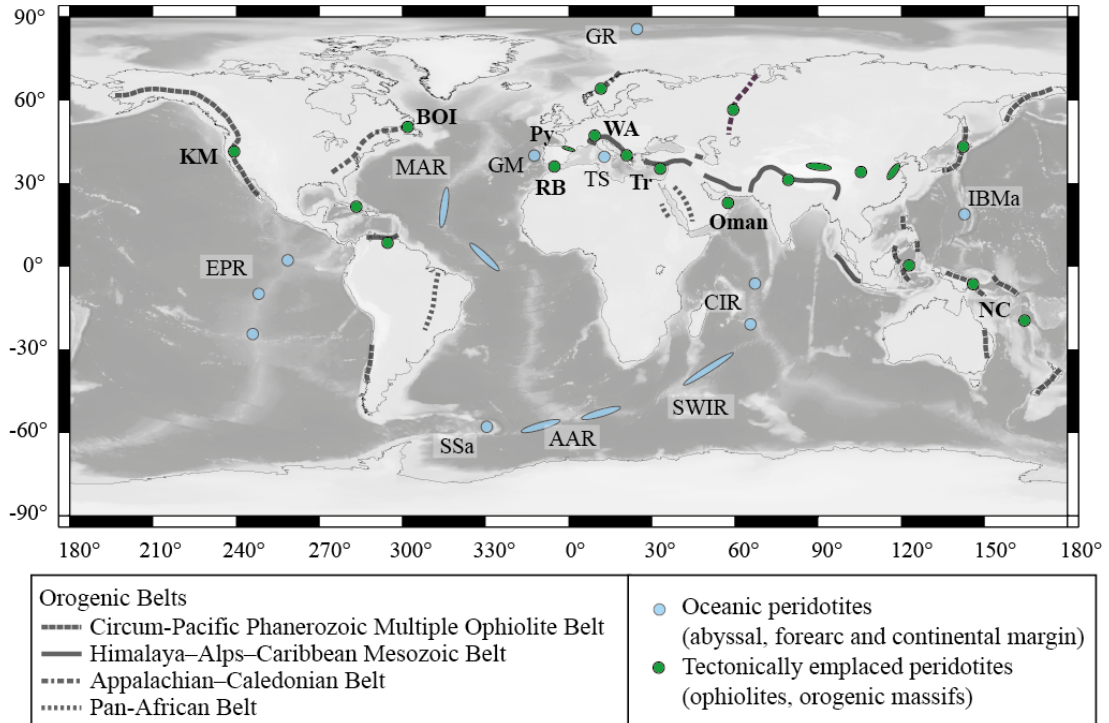


Fig. 1. Location of main peridotite occurrences mentioned in the text. (after [4]). Oceanic peridotites: *Fast-spreading ridge systems*: EPR—East Pacific Rise. *Slow-spreading ridge systems*: MAR—Mid-Atlantic Ridge; CIR—Central Indian Ridge. *Ultraslow-spreading ridge systems*: SWIR—Southwest Indian Ridge; American-Antarctic Ridge; Arctic Ocean (GR—Gakkel Ridge). *Supra-subduction settings*: SSa—South Sandwich Arc-Basin system; IBMa—Izu–Bonin–Mariana Forearc; TS—Tyrrhenian Sea. *Continental Margins*: GM—Galicia Margin and Gorrige Bank; Tectonically emplaced peridotites: Orogenic massifs (sub-continental mantle): Pyr—Pyrenees, southern France; RB: Betico-Rifean Neogene belt : Ronda, Betic Cordilleras, southern Spain & Beni Bousera, Rif Mountains, northern Morocco: Main ophiolitic belts and massifs: *Circum-Pacific Phanerozoic Multiple Ophiolite Belt* (450–0 Ma): KM—Klamath Mountains (Trinity and Josephine, western USA); NC—New Caledonia; *Himalaya–Alps–Caribbean Mesozoic Belt* (150–80 Ma): Oman (=Semail ophiolite), Sultanate of Oman and Unites Arab Emirates; Tr—Troodos, Cyprus; WA—Western Alps (Internal Ligurides, Lanzo). *Appalachian–Caledonian Belt* (450 Ma): BOI—Bay of Islands, Newfoundland. *Pan-African Belt* (750 Ma).

Field studies provide abundant evidences of natural CO<sub>2</sub> mineralization in basaltic and UM basements with the common occurrence of carbonates in basalts (e.g., [5]) and serpentinized peridotites (e.g., [6,7]) in both continental and oceanic environments. Enhancing this process could provide long-lasting and safe storage of CO<sub>2</sub> [1-3]. Peridotites and serpentinites have the highest potential for CO<sub>2</sub> mineralization, with divalent cation oxides typically representing more than 45% of their mass (e.g. [4]).

Enhanced *in situ* CO<sub>2</sub> mineralization requires efficient fluid flow to renew the reactants at the fluid-rock interface. The main issue for achieving efficient CO<sub>2</sub> mineralization in UM basements is the low porosity and permeability of

peridotites ( $10^{-21} - 10^{-17} \text{ m}^2$ ; e.g., [8-9]) that limits fluid flow. Furthermore, the  $\text{CO}_2$  mineralization reaction itself involves the precipitation of low density carbonates and hydrous phases at the expense of dense mantle minerals, therefore clogging flow paths [10]. For these reasons, UM basements had initially been overlooked for possible *in situ*  $\text{CO}_2$  storage in favor of highly permeable basaltic basements (e.g., CARBFIX project; [www.or.is/en/projects/carbfix](http://www.or.is/en/projects/carbfix) [3]). Recent research shows that  $\text{CO}_2$ -mineralization is active and efficient in the oceanic UM basements (e.g., [6, 11]) and in continental peridotite massifs (e.g., [7, 12]), and that it is associated to potential benefits, such as the production of  $\text{H}_2$ , therefore requiring further investigations of their potential for enhanced *in situ*  $\text{CO}_2$  mineralization.

We present an overview of our recent research realized as part of several research projects: ANR-CO2FIX (2008-2012, [www.anr.fr/Project-ANR-08-PCO2-0003](http://www.anr.fr/Project-ANR-08-PCO2-0003)) and LISZT (2018-2023, [www.anr.fr/Projet-ANR-18-CE01-0014](http://www.anr.fr/Projet-ANR-18-CE01-0014)), FP7 ITN ABYSS (2013-2018) and the ICDP Oman Drilling Project [13] (OmanDP 2017-2018; [www.icdp-online.org/projects/world/asia/oman/](http://www.icdp-online.org/projects/world/asia/oman/) and [www.omandrilling.ac.uk/](http://www.omandrilling.ac.uk/)). These studies aim to identify and measure the physical and chemical mechanisms driving  $\text{CO}_2$ -mineralization whilst sustaining fluid flow in UM basements. They involve a dual approach combining (i) field studies of carbonated UM basements with a focus on the Semail ophiolite (Sultanate of Oman) and (ii) laboratory experiments to quantify the efficiency and sustainability of  $\text{CO}_2$ -mineralization over the range of pressure, temperature, compositional and flow rate conditions characterizing natural environments and expected for industrial applications.

### Nomenclature

Mantle	Dense, ultramafic layer underlying the Earth crust; the upper mantle is composed of peridotites.
Ultramafic	Composed of >90% of Mg-Fe-rich minerals (mafic minerals) such as olivine, orthopyroxene, clinopyroxene and serpentine.
Olivine	Mineral solid solution between end-members forsterite ( $\text{Mg}_2\text{SiO}_4$ ) and fayalite ( $\text{Fe}_2\text{SiO}_4$ ); its composition in most peridotites sampled at the surface of the Earth is ( $\text{Mg}_{1.8}\text{Fe}_{0.2}\text{SiO}_4$ ).
Serpentine	Hydrous sheet silicate group with Mg-end member $\text{Mg}_3\text{Si}_2\text{O}_5(\text{OH})_4$
Peridotite	Ultramafic rock containing more than 40% olivine; <i>herzolite</i> (> 5% clinopyroxene), <i>harzburgite</i> and <i>dunite</i> (>90% olivine) are peridotites.
Serpentinite	Ultramafic rock composed mainly of serpentine resulting from the hydrothermal alteration ( <i>serpentinization</i> ) of peridotites.
Listvenite	Rock composed quartz and magnesite ( $\pm$ dolomite, more rarely calcite) and minor hydrous silicates and oxides. It is also spelled <i>listwaenites</i> .
Carbonation	Formation of carbonates by hydrothermal reaction of $\text{CO}_2$ with silicates; the carbonates produced are variously Fe-bearing magnesite ( $\text{MgCO}_3$ ), calcite ( $\text{CaCO}_3$ ) and/or dolomite ( $\text{MgCa}(\text{CO}_3)_2$ ); it is also called <i>carbonatization</i> , <i><math>\text{CO}_2</math> mineralization</i> and <i>carbon mineralization</i> .
Obduction	Tectonic emplacement of fragments of oceanic crust and upper mantle onto a continental margin; it occurs at zones of convergence between tectonic plates, typically in relation to <i>subduction</i> where one tectonic plate is thrust beneath the other.

## 2. $\text{CO}_2$ mineralization in peridotite basements

### 2.1. Insights from field studies : the Semail Ophiolite

The Semail Ophiolite (Sultanate of Oman) preserves a section of Tethyan oceanic lithosphere obducted onto the Arabian peninsula (Fig. 1). Its mantle section is variably serpentinized and it hosts several  $\text{H}_2$ -rich alkaline springs suggesting that serpentinization is still active today (e.g. [12, 14]) and carbonate-rich areas that can be used as analogues of  $\text{CO}_2$  geological storage [2].

Carbonate-rich areas are most commonly observed as outcrops of several 10s to 100s  $\text{m}^2$  along dry wadis. One of the most spectacular outcrops, set in the Wadi Dima area (south of Muscat), exposes partially altered to highly serpentinized harzburgites cross-cut by a high density (> 20 Vol%) carbonate vein network (Fig. 2a and b; [7]). The

vein network is composed of carbonates of variable Mg/Ca compositions, from dolomite to calcite, locally intermingled with serpentine. The majority of carbonate veins have serpentinite selvages (Fig. 2c). Carbonates were formed over several stages of veining and hydrothermal alteration. The onset of carbonation occurred as a fine diffuse network of dolomite and Mg-calcite penetrative into the serpentinized harzburgite, replacing olivine relics and, locally, co-precipitation with proto-serpentine (Fig. 2d-e). Analyses of carbon species and carbon isotopes indicate that minor fractions of reduced solid carbon species precipitate during this incipient stage of carbonation. This first step is followed by a sequence of veining during which carbonation occurred as a suite of dissolution/precipitation reactions at the expense of serpentine and early carbonates; locally, these processes replaced almost completely the harzburgite basement (Fig. 2b). Trace element analyses combined with oxygen and carbon isotope data indicate that carbonation reactions were triggered by interactions with CO<sub>2</sub>-bearing fluids of different compositions (salinity, cationic content, pH) and over temperatures ranging from hydrothermal (150-200°C) to ambient temperatures. Similar to serpentine veins [14], the field scale orientation of carbonate veins reflects the mantle fabric, suggesting that influx of CO<sub>2</sub>-bearing fluids occurred along preexisting serpentine veins and that these structural heterogeneities acted as fluid pathways throughout the different stages of carbonation and veining. Carbonation likely started as the oceanic mantle lithosphere was cooling during the onset of obduction (end of Cretaceous), and continued during the ophiolite uplift until today.

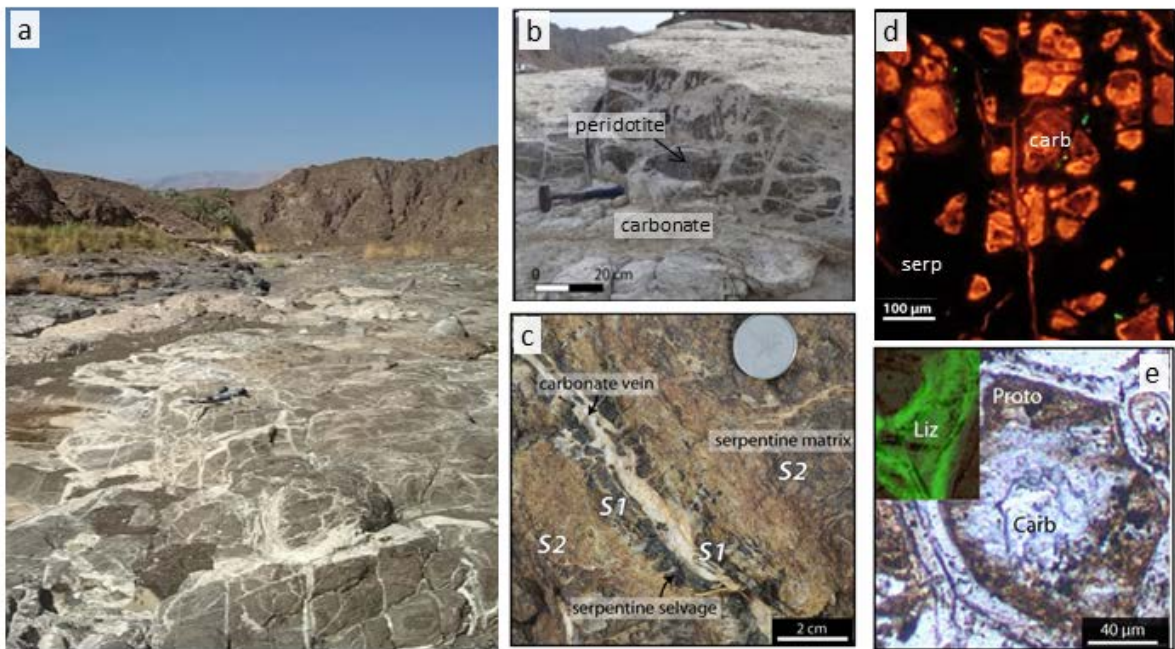


Fig. 2. Photographs of peridotite-hosted carbonate vein networks showing vein density, carbonate morphologies and relationships with serpentinized basement at different scale, in the Wadi Dima area (Oman ophiolite). (a, b) Photographs of Wadi Dima outcrops; (c) Close-up on a carbonate vein cross-cutting the serpentinized peridotite basement: serpentinitized selvages are observed (S1, S2) in which carbonation and secondary serpentinization occurs; (d) carbonate cores in S1 serpentinized matrix (cathodoluminescence); (e) Protoserpentine (proto) and carbonate (carb) replacing olivine in S2 serpentinized matrix – Liz : Lizardite background serpentine identified using Raman spectroscopy. (c, d, e from [7,15])

The Semail ophiolite exposes along its basal thrust a second type of carbonated peridotites, the listvenites (e.g., [16]). These UM-derived rocks are composed of quartz and magnesite ( $\pm$  dolomite) and minor hydrous silicates and oxides [17]. Listvenites outcrop in association to the metamorphic sole and/or to the serpentinite melange that characterizes the transition from the ophiolite to the underlying sedimentary lithologies. One of the largest exposures, set in Wadi Mansah (South of Muscat), was cored as part of OmanDP. Hole BT1B sampled a sequence



Ni. The collected data, including the results from Mg-isotope analyses of the fluid samples, will allow a precise determination of the reaction sequence involved in peridotite listvenitization and should help to develop more effective strategies of CO<sub>2</sub> storage in the subsurface.

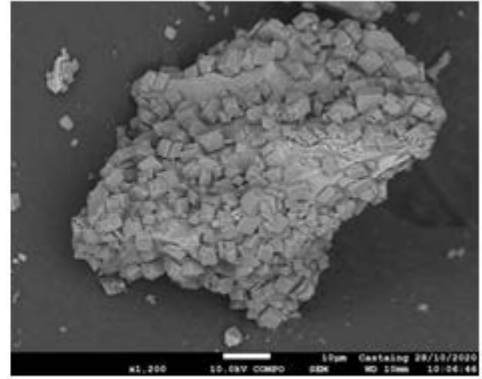
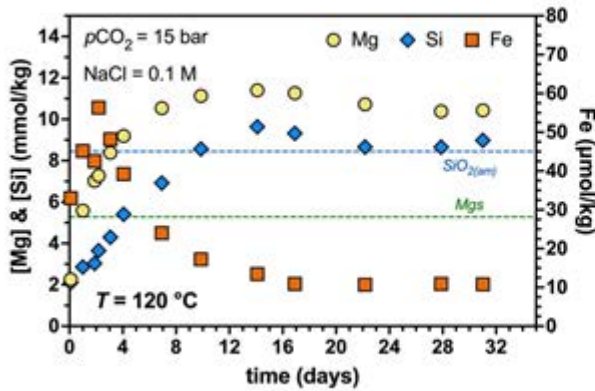


Fig. 3. Chemical evolution of the aqueous fluid (Mg, Si, Fe) during the interaction of Oman harzburgite with a NaCl solution at equilibrium with  $p\text{CO}_2 = 15$  bar and at  $T = 120$  °C (left). The rapid decrease of Fe concentration after 2 days is consistent with the precipitation of abundant ferroan magnesite observed in the solid product after the experiment (right). The dashed lines on the plot refer to equilibrium conditions relative to magnesite (Mgs) and amorphous silica ( $\text{SiO}_2(\text{am})$ ).

• *Reactive flow experiments*: This experimental work was conducted to quantify the efficiency and sustainability of CO<sub>2</sub>-mineralization over the range of pressure, temperature, fluid composition and flow rate conditions characterizing natural environments and expected for industrial applications. The kinetics of carbonate precipitation is strongly dependent on temperature but also on fluid chemistry, the precipitation for Mg-carbonates (magnesite) being sluggish compared to Ca-carbonates (e.g., calcite) [23-25]. Reactive-flow experiments revealed an additional control by the coupling between fluid flow, serpentinization and carbonate-forming reactions at the pore-scale that leads to an heterogeneous distribution of carbonates in peridotites percolated by CO<sub>2</sub>-bearing fluids (e.g., [26]): Mg-carbonates develop preferentially in low flow zones where fluid composition is controlled by silicate dissolution, whilst the composition of carbonates in the main fluid pathways are controlled by that of the inlet fluids [27-28]. These linkages result in complex reactive systems controlled by pore-scale changes in redox, fluid compositions (pH, solutes,...) and by heterogeneities in the structure and mineralogy of reacting peridotites. Recently, Peuble et al [29] showed that the development of these pore-scale redox domains was also strongly dependent on the  $p\text{CO}_2$  of the injected fluid with reduced solid carbon species developing at low  $p\text{CO}_2$  whilst solely carbonates precipitated when high  $p\text{CO}_2$  fluid is injected. These pore-scale processes could explain the occurrence of reduced carbon in the Wadi Dima peridotites. These early experimental studies were realized on peridotite analogues (sintered or compressed olivine core). As part of the ANR LISZT project, we investigated a more complex system and conducted a series of reactive percolation experiments designed to investigate carbonation reactions in a fractured peridotite basement, similar to the Semail ophiolite.

The first set of experiments consisted in injecting CO<sub>2</sub> rich saline solutions (0.5 M NaCl) with  $p\text{CO}_2$  values of 20 MPa and 2 MPa into artificially fractured partially serpentinized harzburgite cores (Oman ophiolite) at constant flow rates of 6 mL/h, except for one experiment realized at 2.4 mL/h. The experiments were conducted at constant temperature (170°C) and pressure (25 MPa) and lasted 6 to 11 days [30].

All experiments displayed outlet fluids characterized by high pH and high Si, Mg, Fe and Ca concentrations compared to inlet fluids hence revealing the dissolution of silicate minerals. At the end of experiments, reaction zones with strong evidences of dissolution (e.g., voids formation, etch-pits and saw-toothed textures) were observed along fractures in all samples (Fig. 4). Two reaction zones were systematically observed: the first, adjacent to the fracture (darker zone), denotes a highly porous area characterized by extensive dissolution and the second, more diffuse, further from the fracture denotes a less reacted slightly porous reacted zone (lighter grey areas). Both

reacted zones show the alteration of olivine and pyroxenes into hematite (a ferric iron oxide), and precipitation of Si-rich phase (i.e., amorphous silica). Away from the fracture, the reacted zone displays also abundant carbonates indicating changes of the fluid reactivity as it migrates into the peridotite matrix.

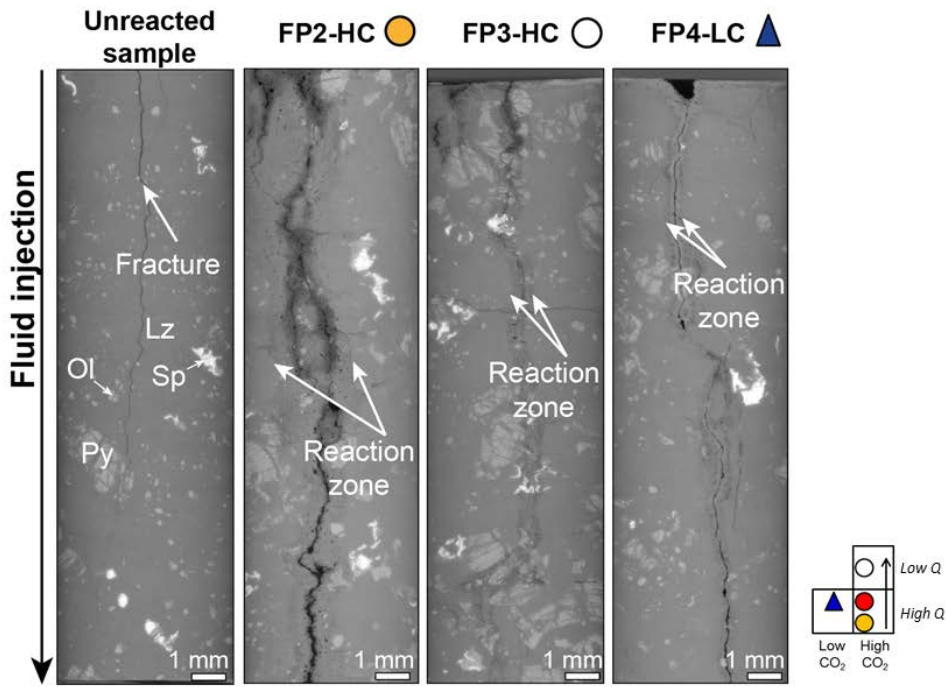


Fig. 4. X-ray microtomography images (XMT) acquired with a resolution of 2.2 microns on fractured peridotite cores before (unreacted sample) and after reactive percolation experiments at  $p\text{CO}_2=20$  MPa (FP2-HC; FP3-HC) and  $p\text{CO}_2=2$  MPa (FP4-LC). The peridotite cores sample a serpentinized peridotite from the Oman ophiolite; their initial mineralogy is lizardite (Lz), olivine (Ol), pyroxene (Px) and minor spinel (Sp). (After [30])

Detailed investigations of the mineralogy and mass balance calculations based on the chemistry of the outlet fluid time series indicate differences in reaction paths and reaction rates depending on  $p\text{CO}_2$  and solute transport conditions. At low  $p\text{CO}_2$ , olivine and pyroxenes are slightly altered to (Ca)-magnesite, hematite and (Ni-Fe) oxides and, at high  $p\text{CO}_2$ , olivine, pyroxenes and lizardite are strongly altered to (Fe)-magnesite, hematite and amorphous silica. At constant  $p\text{CO}_2$  values, carbonation is more efficient at high injection rates. This investigation provides a preliminary basis for the evaluation of UM fractured reservoirs as potential ways for safety  $\text{CO}_2$ -storage.

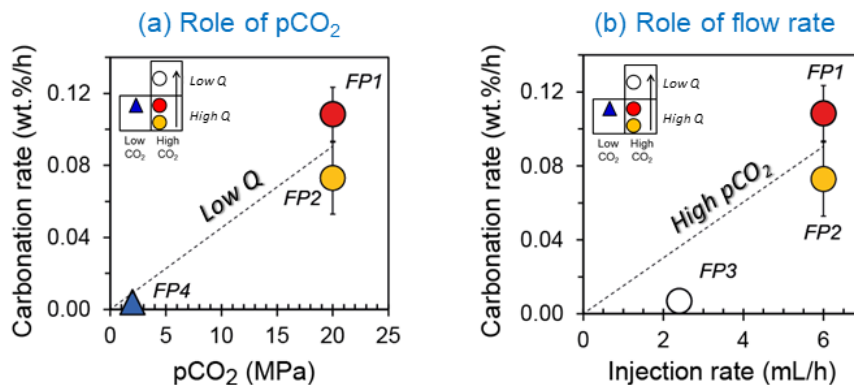


Fig. 5. Carbonation rates (wt.%/h) vs (a)  $p\text{CO}_2$  (MPa) and (b) injection rate  $Q$  (mL/h). (After [30])



### 3. Discussion and conclusions

Combining field studies with laboratory experiments provides insights on the strongly coupled processes that control natural CO<sub>2</sub>-mineralization in UM basements.

The petrostructural, mineralogical, and geochemical (elemental chemistry, C and O isotopes) study of the Semail ophiolite shows that it underwent widespread serpentinization induced by the ingress of seawater-derived fluids in Wadi Dima and by slab-derived fluids in Wadi Mansah. Carbonation reactions are closely associated with serpentinization, but show different reaction pathways. Carbonation reactions produce (i) Mg-carbonates (and quartz) and act as an efficient de-hydration process (de-serpentinization) in Wadi Mansah and (ii) Ca-Mg-carbonates co-precipitating with serpentinites and (possibly abiotic) formation of minor fractions of reduced-carbon species in Wadi Dima. The two reaction pathways are also distinguished by the remobilization of critical metals (Mn, Cr, Li, Pb,...) in Wadi Mansah and by evidences of H<sub>2</sub>-rich alkaline fluids in Wadi Dima. Carbonation reactions occur over the same range of temperature at both sites. This suggests that the composition of incoming fluids (salinity, pH, solute concentrations ...) and, in particular, their CO<sub>2</sub> concentration (high in Wadi Mansah compared to Wadi Dima), is critical in the development of these two different reaction pathways. Such variations in the fluid composition are expected at an industrial CO<sub>2</sub>-injection site, with the fluid CO<sub>2</sub>-contents decreasing away from the injection borehole, therefore studies of different carbonation analogues can contribute to provide guidance to predict the impact of CO<sub>2</sub>-injection in UM basements. However, because of the complex tectonic history of ophiolites, studies of carbonated UM basements provide little information on the kinetic and efficiency of carbonation reactions. These processes and specifically the role of the complex interactions between the chemical changes, rock microstructure evolution and hydrodynamic properties are better quantified by experimental studies where conditions are controlled and mass transfers monitored. Our results suggest that the (evolving) CO<sub>2</sub> content of percolating fluids is critical in the development of variable serpentinization and carbonation reaction paths by promoting (localized) dissolution/precipitation of hydrous phases and carbon species (Mg- or Ca-carbonates, reduced carbon) that, in turn, control the porous network (porosity, tortuosity) and thus the sustainability of the fluid flow (permeability...), the elemental spreading of the reactants over long distances and the local mixing that controls the reactions [26-31].

This research is a first step in the evaluation of the feasibility of industrial *in situ* CO<sub>2</sub> mineralization and of its possible environmental and economic impacts. Numerous scientific and technological challenges remain before implementing industrial CO<sub>2</sub> geological storage in UM basements; one of these challenges is improving knowledge on the linkages between hydrodynamic and (bio-)geochemical processes, at the scale of a borehole, in a region where serpentinization and carbonation are active.

A “Multi-Borehole Observatory” (MBO) was developed as part of OmanDP in an area of active weathering in the Semail ophiolite [32]. MBO is the first and unique site where the geochemistry, mechanics, and hydrology of peridotite alteration can be investigated *in situ*. It consists in 6 boreholes (including 3 cored holes), set mostly in a 500m x 500 m area, thus allowing for the first time a continuous acquisition of flow and (bio-)geochemical time series on UM hydrothermal alteration. Comparing the petro-structure and composition of drilled cores to downhole borehole physical and hydrodynamic measurements will provide a unique dataset to identify the reactive transport mechanisms and characterize the links between the basement structure, fluid flow, chemical fluxes, effective reaction rates and the deep biosphere during CO<sub>2</sub> mineralization in natural environments. Investigations are still in progress. The results of these studies will be critical for developing pilot test sites for CO<sub>2</sub> storage in UM basements.

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## References

- [1] Oelkers, E. H., S. R. Gislason, and J. Matter, Mineral Carbonation of CO<sub>2</sub>, *Elements*, 4(5), 333-337, 2008, doi:10.2113/gselements.4.5.333.
- [2] Kelemen, P.B., Aines, R., Bennett, E., Benson, S.M., Carter, E., Coggon, J.A., de Obeso, J.C., Evans, O., Gadikota, G., Dipple, G.M., Godard, M., Harris, M., Higgins, J.A., Johnson, K.T.M., Kourim, F., Lafay, R., Lambert, S., Manning, C.E., Matter, J.M., Michibayashi, K., Morishita, T., Noël, J., Okazaki, K., Renforth, P., Robinson, B., Savage, H., Skarbek, R., Spiegelman, M.W., Takazawa, E., Teagle, D., Urai, J.L., Wilcox, J.. In situ carbon mineralization in ultramafic rocks: Natural processes and possible engineered methods. *Energy Procedia*, 2018, 146: 92-102 doi: 10.1016/j.egypro.2018.07.013
- [3] Gislason, S.R., Sigurdardóttir, H., Aradóttir, E.S., Oelkers, E.H., A brief history of CarbFix: Challenges and victories of the project's pilot phase. *Energy Procedia*, 2018, 146: 103-114, doi:10.1016/j.egypro.2018.07.014
- [4] Bodinier, J.-L., Godard, M., Orogenic, ophiolitic, and abyssal peridotites, 2014. In: Carlson, R.W. (Ed.), *Treatise on Geochemistry*, vol. 2: Mantle and Core. *Treatise on Geochemistry* 2<sup>nd</sup> Edition. Elsevier Science Ltd., pp. 1-73.
- [5] Alt, J.C., Teagle, D.A.H. The uptake of carbon during alteration of ocean crust. *Geochim. Cosmochim. Acta*, 1999. 63(10): 1527-1535.
- [6] Früh-Green, G.L., Kelley, D.S., Bernasconi, S.M., Karson, J.A., Ludwig, K.A., Butterfield, D.A., Boschi, C., Proskurowski, G.. 30,000 years of hydrothermal activity at Lost City hydrothermal field. *Science*, 2003, 301: 495-498.
- [7] Noel, J., Godard, M., Oliot, E., Martinez, I., Williams, M., Boudier, F., Rodriguez, O., Chaduteau, C., Escario, S., Gouze, P., Evidence of polygenetic carbon trapping in the Oman Ophiolite: petro-structural, geochemical, and carbon and oxygen isotope study of the Wadi Dima harzburgite-hosted carbonates (Wadi Tayin massif, Sultanate of Oman). *Lithos*, 2018. 323: 218-237, doi:10.1016/j.lithos.2018.08.020.
- [8] Hatakeyama, K., Katayama, I., Hirauchi, K.-i., Michibayashi, K.. Mantle hydration along outer-rise faults inferred from serpentinite permeability. *Scientific Reports*, 2017, 7(1): 13870.
- [9] Katayama, I., Abe, N., Hatakeyama, K., Akamatsu, Y., Okazaki, K., Ulven, O.I., Hong, G., Zhu, W., Cordonnier, B., Michibayashi, K., Godard, M., Kelemen, P., The Oman Drilling Project Phase 2 Science Party. Permeability profiles across the crust-mantle sections in the Oman Drilling Project inferred from dry and wet resistivity data. *J. Geophys. Res: Solid Earth*, 2020. 125, e2019JB018698. <https://doi.org/10.1029/2019JB018698>
- [10] Xu, T., Apps, J.A., Pruess, K.. Numerical simulation of CO<sub>2</sub> disposal by mineral trapping in deep aquifers. *Appl. Geochem.*, 2004, 19: 917-936.
- [11] Früh-Green, G.L., Orcutt, B.N., Rouméjon, S., Lilley, M.D., Morono, Y., Cotterill, C., Green, S., Escartin, J., John, B.E., McCaig, A.M., Cannat, M., Ménez, B., Schwarzenbach, E.M., Williams, M.J., Morgan, S., Lang, S.Q., Schrenk, M.O., Brazelton, W.J., Akizawa, N., Boschi, C., Dunkel, K.G., Quéméneur, M., Whattam, S.A., Mayhew, L., Harris, M., Bayrakci, G., Behrmann, J.-H., Herrero-Bervera, E., Hesse, K., Liu, H.-Q., Ratnayake, A.S., Twing, K., Weis, D., Zhao, R., Bilinker, L. Magmatism, serpentinization and life: Insights through drilling the Atlantis Massif (IODP Expedition 357). *Lithos*, 2018: doi:10.1016/j.lithos.2018.09.012.
- [12] Chavagnac, V., Ceuleneer, G., Monnin, C., Lansac, B., Hoareau, G., Boulart, C.. Mineralogical assemblages forming at hyperalkaline warm springs hosted on ultramafic rocks: A case study of Oman and Ligurian ophiolites. *Geochem. Geophys. Geosyst.*, 2013, 14: 2474-2495, doi:10.1002/ggge.20146.
- [13] Kelemen, P.B., Matter, J.M., Teagle, D.A.H., Coggon, J.A., and the Oman Drilling Project Science Team, *Proceedings of the Oman Drilling Project, Report, College Station, TX (International Ocean Discovery Program)*, 2020, doi:10.14379/OmanDP.proc.2020.
- [14] Dewandel, B., Lachassagne, P., Boudier, F., Al-Hattali, S., Ladouche, B., Pinault, J.-L., Al-Suleimani, Z.. A conceptual hydrogeological model of ophiolite hard-rock aquifers in Oman based on a multiscale and a multidisciplinary approach. *Hydrogeology Journal* 2005, 13: 708-726.
- [15] Noel, J., *Etude pétro-structurale et géochimique des processus de serpentinisation et carbonatation des péridotites d'Oman*. PhD thesis 2018, University of Montpellier, 342p, HAL-02053492.
- [16] Nasir, S., Al Sayigh, A.R., Al Harthi, A., Al-Khribash, S., Al-Jaaidi, O., Musllam, A., Al-Mishwat, A., Al-Bu'saidi, S. Mineralogical and geochemical characterization of listwaenite from the Semail Ophiolite, Oman. *Geochemistry*, 2007, 67(3): 213-228.
- [17] Halls, C., Zhao, R.. Listvenite and related rocks: perspectives on terminology and mineralogy with reference to an occurrence at Cregganbaun, Co. Mayo, Republic of Ireland. *Mineralium Deposita*, 1995, 30(3): 303-313.
- [18] Kelemen, P.B., Carlos de Obeso, J., Leong, J.A., Godard, M., Okazaki, K., Kotowski, A.J., Manning, C.E., Ellison, E.T., Menzel, M.D., Urai, J.L., Hirsh, G., Rioux, M., Stockli, D.F., Lafay, R., Beinlich, A.M., Coggon, J.A., Warsi, N.H., Matter, J.M., Teagle, D.A.H., Harris, M., Michibayashi, K., Takazawa, E., Al Sulaimani, Z., the Oman Drilling Project Science Team. Listvenite Formation During Mass Transfer into the Leading Edge of the Mantle Wedge: Initial Results from Oman Drilling Project Hole BT1B. *Journal of Geophysical Research: Solid Earth*, 2022, 127(2): e2021JB022352, doi:10.1029/2021JB022352.

- [19] Godard, M., Carter, E.J., Decrausaz, T., Lafay, R., Bennett, E., Kourim, F., de Obeso, J.-C., Michibayashi, K., Harris, M., Coggon, J., Teagle, D., Kelemen, P., the Oman Drilling Project Phase 1 Science Party. Geochemical Profiles Across the Listvenite-Metamorphic Transition in the Basal Megathrust of the Semail Ophiolite: Results from Drilling at Oman DP Hole BT1B. *Journal of Geophysical Research: Solid Earth*, 2021, 126(12): e2021JB022733, doi: 10.1029/2021JB022733.
- [20] de Obeso, J.C., Kelemen, P.B., Leong, J.M., Menzel, M.D., Manning, C.E., Godard, M., Cai, Y., Bolge, L., Oman Drilling Project Phase 1 Science Party, 2022. Deep Sourced Fluids for Peridotite Carbonation in the Shallow Mantle Wedge of a Fossil Subduction Zone: Sr and C Isotope Profiles of OmanDP Hole BT1B. *Journal of Geophysical Research: Solid Earth*, 127(1): e2021JB022704, doi:10.1029/2021JB022704.
- [21] Beinlich, A., Plümper, O., Boter, E., Müller, I.A., Kourim, F., Ziegler, M., Harigane, Y., Lafay, R., Kelemen, P.B., the Oman Drilling Project Science Team, 2020. Ultramafic Rock Carbonation: Constraints From Listvenite Core BT1B, Oman Drilling Project. *Journal of Geophysical Research: Solid Earth*, 125(6): e2019JB019060.
- [22] Falk, E.S., Kelemen, P.B., 2015. Geochemistry and petrology of listvenite in the Samail ophiolite, Sultanate of Oman: Complete carbonation of peridotite during ophiolite emplacement. *Geochimica et Cosmochimica Acta*, 160: 70-90.
- [23] Saldi, G.D., Jordan, G., Schott, J., Oelkers, E.H.. Magnesite growth rates as a function of temperature and saturation state. *Geochimica et Cosmochimica Acta*, 2009, 73(19): 5646-5657.
- [24] Saldi, G.D., Schott, J., Pokrovsky, O.S., Gautier, Q., Oelkers, E.H.. An experimental study of magnesite precipitation rates at neutral to alkaline conditions and 100–200°C as a function of pH, aqueous solution composition and chemical affinity. *Geochimica et Cosmochimica Acta*, 2012, 83: 93-109.
- [25] Bénézech, P., Saldi, G.D., Dandurand, J.-L., Schott, J. Experimental determination of the solubility product of magnesite at 50 to 200°C. *Chemical Geology*, 2011, 286(1): 21-31.
- [26] Andreani, M., Luquot, L., Gouze, P., Godard, M., Hoisé, E., Gibert, B., 2009. Experimental study of carbon sequestration reactions controlled by the percolation of CO<sub>2</sub>-rich brine through peridotites. *Environmental Science and Technology*, 43(4): 1226-1231, doi:10.1021/es8018429.
- [27] Peuble, S., Godard, M., Luquot, L., Andreani, M., Martinez, I., Gouze, P. CO<sub>2</sub> geological storage in olivine rich basaltic aquifers: New insights from reactive-percolation experiments. *Applied Geochemistry*, 2015, 52: 174-190, doi:10.1016/j.apgeochem.2014.11.024.
- [28] Peuble, S., Andreani, M., Godard, M., Gouze, P., Barou, F., Van De Moortele, B., Mainprice, D., Reynard, B. Carbonate mineralization in percolated olivine aggregates: Linking effects of crystallographic orientation and fluid flow. *American Mineralogist*, 2015, 100(2-3): 474-482, doi: 10.2138/am-2015-4913.
- [29] Peuble, S., Godard, M., Gouze, P., Leprovost, R., Martinez, I., Shilobreeva, S. Control of CO<sub>2</sub> on flow and reaction paths in olivine-dominated basements: An experimental study. *Geochimica et Cosmochimica Acta*, 2019, 252: 16-38, doi:10.1016/j.gca.2019.02.007.
- [30] Escario, S., Coupling between hydrodynamic properties and reactions during serpentinization : Impact on H<sub>2</sub> / CO<sub>2</sub> budgets, PhD thesis 2018, University of Montpellier, 346p. HAL 01928017.
- [31] Escario, S., Godard, M., Gouze, P., Leprovost, R. Experimental study of the effects of solute transport on reaction paths during incipient serpentinization. *Lithos*, 2018, 323: 191-207, doi:10.1016/j.lithos.2018.09.020.
- [32] Kelemen, P.B., Leong, J.A., Carlos de Obeso, J., Matter, J.M., Ellison, E.T., Templeton, A., Nothaft, D.B., Eslami, A., Evans, K., Godard, M., Malvoisin, B., Coggon, J.A., Warsi, N.H., Pézard, P., Choe, S., Teagle, D.A.H., Michibayashi, K., Takazawa, E., Al Sulaimani, Z., the Oman Drilling Project Science Team. Initial results from the Oman Drilling Project Multi-Borehole Observatory: Petrogenesis and ongoing alteration of mantle peridotite in the weathering horizon. *Journal of Geophysical Research: Solid Earth*, 2021: e2021JB022729, doi: 10.1029/2021JB022729.