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Moho carbonation at an ocean-continent transition

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ABSTRACT

Carbonation of mantle rocks during mantle exhumation is reported in present-day oceanic settings, both at mid-ocean ridges and ocean-continent transitions (OCTs). However, the hydrothermal conditions of carbonation (i.e., fluid sources, thermal regimes) during mantle exhumation remain poorly constrained. We focus on an exceptionally well-preserved fossil OCT where mantle rocks have been exhumed and carbonated along a detachment fault from underneath the continent to the seafloor along a tectonic Moho. Stable isotope (oxygen and carbon) analyses on calcite indicate that carbonation resulted from the mixing between serpentinization-derived fluids at ~175 °C and seawater. Strontium isotope compositions suggest interactions between seawater and the continental crust prior to carbonation. This shows that carbonation along the tectonic

Moho occurs below the continental crust and prior to mantle exhumation at the seafloor during continental breakup.

INTRODUCTION

At magma-poor ocean-continent transitions (OCTs), mantle exhumation occurs along detachment faults (e.g., the Iberia margin; Boillot et al., 1987), in a comparable way to (ultra)slow spreading ridges (Tucholke et al., 1998; Cannat et al., 2006). Associated hydrothermal fluid circulation triggers serpentinization of mantle rocks, high-temperature (>350 °C) mineralization (Cu-Fe-Co-Zn-Ni; Coltat et al., 2019b), and carbonation of ultramafic rocks (Lemoine et al., 1987). While the tectonic processes responsible for mantle exhumation are well investigated at present-day OCTs (Peron-Pinvidic et al., 2013) and fossil OCTs (Epin et al., 2019), syn-exhumation carbonation of mantle rocks remains poorly constrained so far (e.g., Coltat et al., 2019a, 2020). This is mainly due to the fact that present-day OCTs are buried and accessible only by drilling, whereas fossil analogues are commonly overprinted by metamorphic events during late orogenesis disturbing the former hydrothermal systems.

We focus on the exceptionally well-preserved Tasna OCT section exposed in southeastern Switzerland. The continuous 6-km-long section shows mantle rocks exhumed along a detachment fault that defines a tectonic Moho in the south and the exhumed seafloor sealed by deepwater post-rift sediments in the north. Hydrothermal carbonates occur in mantle rocks all along the detachment. Hence, the Tasna section offers a unique opportunity to unravel, through coupled field work and oxygen, carbon, and strontium isotope geochemistry, the conditions of carbonation (i.e., structural setting, fluid sources, thermal regimes) during mantle exhumation.

GEOLOGICAL SETTING

The Tasna OCT belongs to the Penninic Tasna nappe exposed in the Engadine window near Scuol in southeastern Switzerland (Ribes et al., 2020, and references therein; Figs. 1A–1C). It has been interpreted to represent the most distal Briançonnais and/or European margin of the Alpine Tethys (Florineth and Froitzheim, 1994; Ribes et al., 2020). The Tasna outcrops preserve, across 6 km, an exhumed subcontinental mantle that underlies a wedge of thinned mid- to lower continental crust composed of pre-rift migmatites and Permian gabbros (Florineth and Froitzheim, 1994). Both the contact separating the crust and the mantle as well as the top of the crust represent detachment faults, namely the Lower and Upper Tasna detachments (LTD and UTD, respectively; Florineth and Froitzheim, 1994). The UTD was crosscut by the LTD (Figs. 1D and 1E), and both show a southeast-northwest–oriented stretching lineation in present-day coordinates (Manatschal et al., 2006). The detachments show well-developed damage zones formed by cataclasites whereas core zones are characterized by foliated cataclasites and gouges (Manatschal et al., 2006) and are sealed by Upper Jurassic to Lower Cretaceous deep-marine post-rift sediments (Ribes et al., 2020). Ar-Ar phlogopite dating and stratigraphic constraints suggest late Middle to early Late Jurassic exhumation (Manatschal et al., 2006; Ribes et al., 2020), predating the Alpine orogeny.

During the Eocene–Oligocene, the Tasna OCT was emplaced onto the Lower Tasna unit (Ramosch and Roz-Champatsch zones; Figs. 1C–1E) along a north-vergent thrust. Thus, the Tasna OCT discussed here remained little affected by Alpine deformation and metamorphism and preserves its pre-Alpine, exhumation-related structures (Fig. 1E).

RESULTS

Structural relationships between carbonation and deformation were characterized by both field and microscopic analyses. We collected 32 samples of ophicalcites and carbonated rocks along the LTD (Fig. 1E) for C, O, and Sr isotope geochemistry (see details in the Supplemental Material[1]).

Field and Petrographic Data

At the Tasna OCT, spinel lherzolites display an advanced serpentinization, although ~~relics~~**[[relicts?]]** of primary minerals are locally preserved. Along the LTD, serpentinites display a cataclastic fabric, locally crosscut by green serpentine veins. Toward the top of the LTD, cataclasites are foliated and contain rounded serpentinite clasts. Carbonation in the LTD shows a gradient across 20 m, reaching a maximum in the core zone where it occurs both as discrete calcite + chlorite veins (Fig. 2A) and as pervasive replacement of serpentinites (foliated ophicalcites; Fig. 2B).

In the overlying thinned crust, migmatites and Permian gabbros display high-temperature mylonitic fabrics. At their base, the gabbros show carbonated, matrix-supported cataclasites with pervasively carbonated gabbro clasts (Fig. 2C; ~~App. 2~~ Fig. S1 in the Supplemental Material). The cataclasites are surrounded by chlorite-rich gouge zones with discrete calcite veins. Southeast-northwest striations on chlorite shear planes show the same orientations as kinematic indicators measured along the LTD. Foliated gabbros are locally crosscut by high-angle structures with respect to the LTD (Fig. S1). A greenschist-assemblage alteration (epidote + chlorite + albite + quartz) is commonly observed both in the gabbro groundmass and in high-angle structures, together with calcite veins (Fig. S1). Migmatites consist of quartz, feldspars, micas, garnet, and minor

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zircon, monazite, and opaque minerals. This assemblage is commonly altered into epidote + chlorite + quartz with graphite flakes locally associated with epidote (Fig. S1).

Stable (C and O) and Sr Isotope Compositions of Carbonates

The O, C, and Sr isotope compositions of carbonates are reported in Figures 3A and 3B and in ~~App. 3~~ Table S1 in the Supplemental Material. Along the LTD, the $\delta^{18}O$ values of carbonates range from 13.5‰ to 25.5‰ and the $\delta^{13}C$ values are from –4.75‰ to 1.49‰, being comparable to the isotopic signatures of Alpine Tethys ophicalcites (Fig. 3A). The $\delta^{18}O$ values of carbonates from the northern domain are between 16.8‰ and 25.5‰, higher than in the southern domain (13.5‰–16.6‰). The $\delta^{13}C$ values are between –0.69‰ and 1.36‰ in the northern domain, whereas they vary between –4.75‰ and 1.49‰ in the southern domain. Variations in both $\delta^{18}O$ and $\delta^{13}C$ values also exist within samples (Fig. 3C).

Along the LTD, the Sr isotope compositions range from 0.707043 to 0.710738 (Fig. 3B), with carbonates from the southern domain (with low $\delta^{18}O$ values) **[[generally?]]** being more radiogenic than in the northern domain. In addition, at a given site, the Sr isotope compositions of the carbonates are heterogenous (i.e., site Tas. 1; Fig. 3B).

DISCUSSION

A Preserved Syn-Exhumation Hydrothermal Carbonation

At the Tasna OCT, several arguments can be made that carbonation was synchronous with Jurassic extension. First, undeformed Mesozoic post-rift marine sediments seal carbonated extensional structures (Manatschal et al., 2006), indicating that carbonation occurred during oceanic conditions. Secondly, southeast-northwest striations measured on chlorite shear planes surrounding matrix-supported cataclasites (Fig. 2C) are consistent with those measured along the LTD (Manatschal et al., 2006), suggesting that both chlorite gouge zones and cataclasites were formed during Jurassic detachment faulting. **[[Unless the following sentence is a continuation of the "Secondly" idea, please preface it with "Thirdly" for consistency]]** The presence of carbonate veins either found as clasts in matrix-supported cataclasites or crosscutting the latter indicates that carbonation occurred during the formation of the cataclasites and, by inference, during detachment faulting. Finally, high-angle structures characterized by a calcite-bearing greenschist alteration crosscut the Permian mylonitic gabbros and hence postdate this deformation. Such a greenschist alteration might be the consequence of oceanic hydrothermal alteration (Richardson et al., 1987**[[Richardson et al., 1987 is not in the reference list.]]**; Cann and Gillis, 2004**[[Cann and Gillis, 2004 is not in the reference list.**), previously recognized in mafic rocks along Jurassic detachment faults in Jurassic Tethyan OCTs (Coltat et al., 2019a, 2020). Also, this alteration could be inherited from oceanic conditions during the Jurassic rifting.

Even if carbonation along the LTD occurred during the Jurassic rifting, the preservation of the isotopic compositions of the carbonates might be questioned (see Bernoulli and Weissert [2020], the comment of Coltat et al. [2021], and the reply of Bernoulli and Weissert [2021]). Here, given the lack of Alpine compressional structures able to channelize fluids and the range of 12‰ observed for $\delta^{18}O$ values of carbonates along the LTD (Figs. 3A and 3B) as well as in a given sample (Fig. 3C), we infer that isotopic equilibration did not occur during Alpine metamorphism and that the isotopic compositions are indicative of Jurassic oceanic hydrothermal conditions.

Multiple Isotope Reservoirs for Carbonation

At the Tasna OCT, the C and Sr isotope compositions of carbonates are very heterogenous (Figs. 3A, 3B), suggesting different fluid sources and/or, for carbon, processes inducing isotopic fractionation. In the northern domain, the $\delta^{13}C$ values of carbonates are centered at 0‰–1‰, indicative of a marine origin (Bach et al., 2011). The low Sr isotope compositions measured in the northern domain (centered around ~0.7075), comparable to those of Jurassic seawater, also support a marine origin. In the southern domain, the Sr isotope compositions are higher (as high as 0.710738). Only the continental crust has $^{87}Sr/^{86}Sr$ values as radiogenic as those measured in the carbonates of the southern domain, suggesting interactions between seawater and the continental crust prior to carbonation. In the southern domain, some carbonates display $\delta^{13}C$ values as low as –4.75‰. Such negative $\delta^{13}C$ values have been reported in carbonates at the Lost City hydrothermal field (Mid-Atlantic Ridge; Früh-Green et al., 2003). There, they were attributed to the involvement of C-bearing species with a low $\delta^{13}C$ value (like methane) produced during Fischer-Tropsch–like reactions in serpentinization-derived fluids (SFs; Ludwig et al., 2006). Serpentinization is known to generate CH_4 and H_2 (Charlou et al., 2002), compounds that have been sampled in venting fluids at the Lost City hydrothermal field (Kelley et al., 2001). At the Tasna OCT, the circulation of such fluids during Jurassic mantle exhumation is expected to have occurred as well. Indeed, the graphite accompanying the greenschist facies alteration of migmatites (Fig. S1) is likely derived from the reduction of CO_2-bearing species reacting with H_2, a process proposed for other settings (Mastalerz et al., 1995). Hence, at the Tasna OCT, the involvement of SFs, which were affected by fluid–continental crust interactions prior to serpentinization, is deduced from the lowest $\delta^{13}C$ values and the highest $^{87}Sr/^{86}Sr$ values of carbonates, whereas seawater mostly controlled the carbon and strontium budgets during the formation of carbonates with $\delta^{13}C$ values close to 0‰–1‰ and low $^{87}Sr/^{86}Sr$ values.

Conditions of Fluid-Rock Interactions Leading to Carbonation

In the southern domain, carbonates display $\delta^{18}O$ values centered around ~15‰ (Fig. 3A). The limited range of the $\delta^{18}O$ values indicates precipitation from a unique fluid at constant temperatures. On the other hand, the $\delta^{18}O$ values of the Tasna serpentinites are centered around 8.4‰ (Engström et al., 2007). The ~6.6‰ difference is the qualitative sign of isotopic equilibrium between calcite and serpentine under hydrothermal conditions. The oxygen isotope fractionation factor between calcite and serpentine (Zheng, 1993) indicates a temperature of ~175 °C for equilibration, i.e., for both carbonation in the southern domain and serpentinization at depth.

Because seawater contains a higher concentration of dissolved carbon species than SFs (Tivey, 2007), even minor contribution of seawater may drastically change the $\delta^{13}C$ values of carbonates whereas the $\delta^{18}O$ values remain unaffected. Thus, in the southern domain, the C and O isotope compositions are better explained by a carbonation mechanism involving the mixing of small amounts of seawater with SFs.

The increase of the $\delta^{18}O$ values of the carbonates toward the northern domain is due to progressive involvement of larger amounts of cold seawater during carbonation. The progressive mixing of SFs with seawater may be modeled by simple isotopic mass-balance calculation. Whereas the carbonates with the lowest $\delta^{18}O$ values form from almost pure SFs at 175 °C mixing with minute amounts of seawater, the highest $\delta^{18}O$ values measured (25.5‰) can form under a SF/seawater ratio of ~1:2 at a temperature of

60 °C. The global temperature range (60–175 °C) encompasses the temperatures estimated in ophicalcites formed during mantle exhumation at oceanic core complexes (Kelley et al., 2001; Bach et al., 2011; Schroeder et al., 2015). In this context, the variable O, C, and Sr isotope values measured in a given sample from Tasna (Fig. 3C; Table S1) would be the record of superimposed carbonation events during mantle exhumation, the carbonates with low $\delta^{18}O$ values being formed at depth prior to those with high $\delta^{18}O$ values near the seafloor.

A Hydrothermal Model for the Moho Carbonation at Tasna

In Figure 4, we propose a hydrothermal model for syn-exhumation carbonation of the tectonic Moho at the Tasna OCT. During Jurassic rifting, seawater circulated downwards along normal faults cutting through the continental crust, increasing the Sr isotope composition of the modified seawater (Fig. 4A). At depth, serpentinization reactions occurred by alteration of the subcontinental mantle. During their way back to the seafloor along the detachment plane, the SFs (former seawater) were focused along the uppermost permeable damage zone of the detachment and progressively mixed with ambient or downgoing seawater, inducing carbonation (Fig. 4A). High fluid fluxes occurred in high-permeability fault zones of the detachment, leading to the formation of foliated ophicalcites, whereas less-permeable cataclasites were mostly affected by fractures sealed by calcite (Fig. 4B).

Carbonate precipitation from the mixing of SFs with seawater is predicted by numerical modeling (Palandri and Reed, 2004) and is proposed to explain carbonation at the Lost City hydrothermal field (Ludwig et al., 2006). Early carbonation in deep zones occurred from the mixing of low-$\delta^{13}C$, ^{87}Sr-rich, carbon-bearing SFs with minute amounts of seawater. During mantle exhumation, the proportion of seawater progressively increased in the hydrothermal ~~mixed~~ fluid (i.e., decrease of the SF/seawater ratio), explaining the increase of $\delta^{18}O$ and decrease of $^{87}Sr/^{86}Sr$ values at constant $\delta^{13}C$ values. The increasing amount of seawater induced a decrease of the formation temperatures of carbonates toward the northern domain.

CONCLUSIONS

Carbonation of the tectonic Moho interface is observed at the Tasna ocean-continent transition in the Swiss Alps. The exceptional preservation of the structural and isotopic systems allows highlighting the following major points:

(1) Carbonation occurred during syn-serpentinization mantle exhumation beneath the continent wedge and lasted after mantle exhumation at the seafloor.

(2) Carbonation resulted from the mixing of serpentinization-derived fluids (temperature of ~175 °C) with cold seawater.

(3) The contribution of unmodified seawater increased consistently with progressive mantle exhumation toward the seafloor.

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FIGURE CAPTIONS

Figure 1. (A) Simplified geological map of the Alpine system in Western Europe (from Schaltegger et al., 2002). (B) Geological map of the Engadine window in the southeastern Alps, Switzerland. (C) North-south–striking cross section along the Tasna nappe (modified after Ribes et al., 2020). See B for location.**[[Briefly explain the diagram at the lower right corner]]** OCT—ocean-continent transition. (D,E) Southwest-northeast–oriented field view of the Tasna section (from Ribes et al., 2020) (D) and corresponding line drawing after field analysis (E). Samples from sites Tas. 4 and Tas. 8 were collected along blocks slipped down from the cliff. Their corresponding location is projected. **[[In the figure, panel A, replace “&” with “and”; replace “units/ophiolites” with, e.g., “units (green) and ophiolites (black)”. In panel B, put “N” and “E” at the end of latitude/longitude labels instead of at the beginning; in the symbol explanation, insert a space on either side of the plus symbol (+). In panel C, label the vertical axis with a description—e.g., “Elevation (m)”; make instances of “unit” lowercase; spell out instances of “P.”. In panel E, make instances of “unit” lowercase; spell out instances of “P.”; insert a space after each instance of “Tas.” (before the numeral) to**

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match text and other figures; in the symbol explanation, insert a space on either side of the plus symbol; change "Samples" to "Sample"]].

Figure 2. Field pictures from the Tasna section along the the Lower Tasna detachment.**[[Provide location information (latitude/longitude) for each photo]]** (A) Calcite (Cc) + chlorite (Chl) veins cutting through serpentinites from the northern domain (site Tas. 4). (B) Foliated ophicalcites at contact with overlying gabbro along tectonic Moho interface from the southern domain (site Tas. 2). (C) Matrix-supported cataclasite with clasts of gabbro at the top of the Lower Tasna detachment in the southern domain (site Tas. 3).

Figure 3. (A,B) $\delta^{13}C$ (Vienna Peedee belemnite, VPDB) (A) and ^{87}/Sr/^{86}Sr versus $\delta^{18}O$ (Vienna standard mean ocean water, VSMOW) (B) isotopic diagrams of carbonates from carbonated rocks from the Tasna section plotted together with ophicalcites in the Alpine realm**[[Clarify how these are represented in the figure]]** (Weissert and Bernoulli, 1984; Schwarzenbach et al., 2013; Lafay et al., 2017; Coltat et al., 2019a, 2019b). Samples from Engström et al. (2007) were collected at the vicinity of sites Tas. 3 and Tas. 4.**[[Define "SW" (seawater?)]]** (C) Sawed sections of ophicalcites showing variety of carbonate textures and corresponding oxygen and carbon isotope values.

[[In the figure, panel A, make "eastern" lowercase; capitalize and correct the spelling of "Apennines"]]

Figure 4. Schematic tectonic-hydrothermal model for carbonation at thenMoho interface at the Tasna ocean-continent transition. (A) Mantle exhumation was accompanied by upward circulation of serpentinization-derived fluids along the detachment (geometry adapted from Ribes et al., 2020). **[[Explain the color gradient labeled “T°C”]]** Sr— **[[strontium?]]**; LTD—Lower Tasna detachment; UTD—Upper Tasna detachment. (B) Serpentinization-derived fluids progressively mixed with seawater, leading to carbonate precipitation. See text for details.

[[In the figure, panel A, insert a space between “175” and “°C”; change “T°C” to just “*T*” (with italics) and define in the caption as “temperature”? (see query above)]]

[1]Supplemental Material. **[[Please provide a brief caption here]].** Please visit https://doi.org/10.1130/XXXXX.1 to access the supplemental material, and contact editing@geosociety.org with any questions.