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Syn-tectonic, low-temperature meteoric water-derived carbonation of the New Caledonia Peridotite Nappe

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ABSTRACT

Exceptional outcrops recently exposed in the Koniambo Massif allow the study of the serpentine sole of the Peridotite Nappe of New Caledonia. Many magnesite veins are observed, with characteristics indicating that they were emplaced during pervasive top-to-SW shear deformation. The oxygen isotope composition of magnesite is homogeneous ($27.4 ‰ < \delta^{18}O < 29.7 ‰$) while its carbon isotope composition varies widely ($-16.7 ‰ < \delta^{13}C < -8.5 ‰$). These new data document an origin of magnesite from low temperature meteoric fluids. Laterization on top of the Peridotite Nappe and carbonation along the sole appear to represent complementary records of meteoric water infiltration. Based on the syn-kinematic character of magnesite veins, we propose that syn-laterization tectonic activity has enhanced water infiltration, favoring the exportation of leached elements like Mg, which led to widespread carbonation along the serpentine sole. This calls for renewed examination of other magnesite-bearing ophiolites worldwide in order to establish whether active tectonics is commonly a major agent for carbonation.

INTRODUCTION

Carbonation of ultramafic rocks is the process by which CO_2-bearing fluids react with olivine and/or serpentine to form magnesite ($MgCO_3$) (e.g., Klein and Garrido, 2011). Based on stable isotope and structural evidence, Kelemen et al. (2011) recently showed that present day carbonation of the Oman ophiolite is due to downward infiltration of meteoric waters in the absence of significant tectonic activity. Other stable isotope studies have also established the meteoric origin of the fluids from which magnesite has formed in a number of ophiolite occurrences (Jedrysek and Halas, 1990; Fallick et al., 1991; Gartzos, 2004; Jurkovic et al., 2012). Some of these ophiolites include laterites and associated iron-nickel ore deposits capping the ultramafic rocks (e.g., Eliopoulos et al., 2012).

The main ophiolite of New Caledonia, referred to as the Peridotite Nappe, has also undergone intense laterization since its emergence. This has led to supergene nickel ore formation, a process which implies a well drained percolation system through the peridotites (Trescases, 1975). Recently exposed outcrops in the Koniambo Massif (Fig. 1A) show large surfaces of the serpentine sole that forms the base of the nappe (Figs. 1B and 2), providing unprecedented access to fresh samples. Numerous magnesite veins are observed along these outcrops, attesting for widespread carbonation.

Here we present oxygen and carbon isotope compositions of the magnesite veins and argue that they must originate from meteoric water. Furthermore, in contrast with the situation depicted in Oman (Kelemen et al., 2011), most veins appear to have formed syntectonically. This leads us to discuss possible genetic links between laterization, carbonation and tectonics.

GEOLOGICAL SETTING

New Caledonia lies 2000 km east of Australia, in the SW Pacific. About 40% of the island‘s surface consists of peridotite. Peridotites overlie rock units of the Norfolk Ridge microcontinent with a sub-horizontal contact materialized by a strongly deformed serpentine sole (Avias, 1967). This geometry results from the southwestward obduction of the Peridotite Nappe, initially rooted in the Loyalty Basin, sometimes between ~37 and 27 Ma (Cluzel et al., 2001, 2012; Paquette and Cluzel, 2007).

On top of the nappe, laterites have developed at the expense of the peridotites (Trescases, 1975). Several planation surfaces attest for distinct episodes of weathering during the Neogene and probably the Oligocene (Latham, 1986; Chevillotte et al., 2006; Sevin et al., 2012). This is consistent with biogeographic and phylogenetic studies indicating that New Caledonia was already emerged in the Late Oligocene (Grandcolas et al., 2008).

Magnesite is widespread in New Caledonia and occurs as veins within the serpentine sole of the Peridotite Nappe, and as nodular heaps in recent alluvial deposits and present-day soils.

Since Glasser (1904), the origin of the veins is supposed supergene, possibly linked to the laterization process.

OBSERVATIONS AND SAMPLING

The Koniambo Massif is one of the klippes of the Peridotite Nappe located along the West Coast (Fig. 1A). Recently, Koniambo Nickel SAS initiated a large industrial site for nickel production. As a result, new outcrops of exceptional quality and size have been created in the serpentine sole of this massif (Figs. 1B and 2).

In the serpentine sole, rocks are highly deformed, either finely schistose and/or intensely brecciated. A dense network of meter-thick shallow-dipping shear zones attests for pervasive non-coaxial deformation with a top-to-SW sense of shear. Magnesite occurs as veins, up to ~30 cm thick and irregularly distributed. Two main vein types are recognized (Figs. 3A, 3B and 3C, and Figs. A and B in Data Repository). Type 1 veins are located within or along the margins of the main shear zones. Open to tight asymmetric folding of some of these veins indicates that they formed during, or possibly before, shearing. Type 2 veins are steeper and occasionally crosscut by the shallow-dipping shear zones, demonstrating they do not represent younger structures. The obliquity of these veins with respect to the shear zones (Fig. 3D) and the local occurrence of magnesite as coarse fibers orthogonal to vein walls (Fig. 3C) are consistent with their interpretation as tension gashes opened during top-to-SW shearing.

Both vein types have been sampled along two cross sections (BMS and VAV) located in the Vavouto peninsula, just above the basal contact of the Peridotite Nappe (Fig. 1B, Table DR1). Samples were also collected in other highly serpentinized zones, in the Koniambo Massif (sample CONV, Fig. 1B) and in the Kopeto Massif, ~50 km to the south-east (samples NEP and GAIACS). In addition, sample DECH was collected from a series of nodular heaps in a soil. The large size of the heaps (~15–30 cm) and their isolated character within an abundant and homogeneous clayey matrix suggest that they formed in situ in the soil (i.e., they are unlikely to represent reworked pebbles).

CARBON AND OXYGEN ISOTOPE DATA

Isotopic analyses were performed at the stable isotope laboratory of the University of Rennes 1. Samples were finely crushed in a boron carbide mortar and reacted with anhydrous phosphoric acid at 75 °C for 24 h. The liberated CO_2 was analyzed on a VG SIRA 10 triple collector mass spectrometer. The experimental fractionation coefficient between magnesite and CO_2 is $\alpha_{CO2\text{-}Magnesite} = 1.009976$ at 75 °C (Das Sharma et al., 2002). In the absence of a magnesite standard, in-lab calcite standard samples were analyzed together with the magnesite samples under identical conditions in order to control the general reliability of the protocol. The analytical uncertainty is estimated at 0.3‰ for O and 0.2‰ for C.

Results are presented in Figure 4 and Table DR1. All magnesite samples display comparable and high oxygen isotope values, irrespective of their structural position or sampling site ($27.4‰ < \delta^{18}O < 29.7‰$); the carbon isotope composition is highly variable and negative ($-16.7‰ < \delta^{13}C < -8.5‰$). Focusing on the samples from the Vavouto peninsula (BMS and VAV), the $\delta^{13}C$ and $\delta^{18}O$ values do not show any correlation.

METEORIC ORIGIN OF CARBONATION

The isotopic compositions of magnesite reflect the conditions at which fluid/rock interactions occurred. As the $\delta^{18}O$ values are homogeneous, the physical conditions of carbonation were constant, including water origin and temperature. In addition, the large spread of $\delta^{13}C$ values indicates at least two sources of carbon. Based on the arguments to follow, we suggest that a strong interaction with meteoric fluids is the most likely process that led to carbonation along the serpentine sole. Firstly, the largely negative δ^{13}C values rule out marine water as δ^{13}C values would be centered around 0‰. Secondly, in New Caledonia, the oxygen isotope composition of serpentine minerals is between 1.7 and 13.9‰ (Ulrich, 2010). If magnesite had precipitated in equilibrium with these serpentines at 200 °C (a minimum temperature for serpentine formation; e.g., O'Hanley and Wicks, 1995), it would display δ^{18}O values between 12.2 and 24.4‰ (Fig. 5). The range would be displaced toward even lower δ^{18}O values if higher temperatures were considered. Given the observed values are higher, we conclude that serpentinization and carbonation were not synchronous, even though serpentine and the magnesite veins share a common structural record of top-to-SW shearing. Therefore, at least part of the pervasive deformation observed along the sole post-dates serpentinization, a fact consistent with the mechanical softness of serpentine (e.g., Byerlee, 1978). We also considered the δ^{18}O value of magnesite expected if formed at near-surface temperatures from meteoric water (Fig. 5). To date, no isotopic data is available for meteoric precipitations in New Caledonia. A δ^{18}O range between −1 and −7‰ is used, which corresponds to values of meteoric precipitations on isolated islands at inter-tropical latitude and low elevation. We obtain a theoretical δ^{18}O range for the corresponding meteoric-derived magnesite that compares well with our data (Fig. 5).

Three independent observations confirm the meteoric origin of the fluids that led to carbonation. Firstly, the magnesite sampled in a soil (DECH) has isotopic compositions comparable to the magnesite veins from the serpentine sole. Secondly, the highest δ^{13}C values from the veins are close to −8‰, which coincides with the expected value for carbonates in which carbon is taken from the atmosphere (as illustrated by sample DECH). Additionally, the lowest δ^{13}C values, around −15‰, point to a biogenic contribution, either from surface soils, or from a deep-seated carbon source such as biogenic methane liberated from sediments buried below the Peridotite Nappe. Part of the low $\delta^{13}C$ signal may also reflect magnesite precipitation from high-pH fluids (e.g., Fourcade et al., 2007), which are commonly associated with carbonation of ultramafic rocks (Jurkovic et al., 2012). Thirdly, in Figure 4, we compare our data set with data from the literature on magnesite veins hosted by ultramafic rocks and for which a meteoric origin of the fluids has been proposed. Our data show a more restricted range in $\delta^{18}O$ values centered on the right side of the literature data cloud. This observation may reflect a difference in the initial $\delta^{18}O$ value of rain water due to distinct paleogeographic position.

LINKS BETWEEN LATERIZATION, CARBONATION AND TECTONICS

The record of meteoric waters through carbonation along the serpentine sole implies that water circulated downward through the peridotite pile. An efficient drainage system has likely been provided by the dense network of fractures that characterizes the New Caledonia peridotites. This network is also recognized to have played a major role in peridotite weathering and the distribution of nickel ore (e.g., Leguéré, 1976). Hence, laterization on top of the Peridotite Nappe and carbonation along the serpentine sole may correspond to complementary records of meteoric water infiltration, as anticipated by Glasser (1904). In practice, laterization involves the leaching of large amounts of magnesium, a highly mobile ion that can be viewed as a tracer of fluid circulation from the surface down to the serpentine sole where it precipitated to form magnesite veins. Correlatively, nickel, which is less mobile, has accumulated at the base of the lateritic profile (e.g., Trescases, 1975). Throughout New Caledonia, the richest nickel ores are associated with a couple of planation surfaces associated with laterites up to 30 m thick (e.g., Chevillotte et al., 2006). They cap the Koniambo Massif at elevations between ~400 and ~800 m, in agreement with the island-scale mean elevation of 640 m reported by Chevillotte et al. (2006). The outcrops of the Vavouto peninsula, where most of our magnesite samples come from, lie near sea level, and so does the basal contact of the Peridotite Nappe around much of the Koniambo Massif. Therefore, downward infiltration of meteoric waters likely occurred across a vertical distance of at least ~600 m before magnesite formed along the serpentine sole. Greater vertical distances are also possible since older laterite-bearing planation surfaces are locally preserved at elevations up to ~1250 m in the nearby Kopeto Massif and in southern New Caledonia (Latham, 1986; Chevillotte et al., 2006).

Highly efficient drainage of meteoric water through the peridotite pile is attested for by the low temperature conditions at which magnesite formed along the serpentine sole. Figure 5 shows that a temperature of 25 °C fits well the data whereas 35 °C is hardly compatible. The minimum mean temperature at which laterites form is ~25 °C. Using 15 °C/km as a lower bound for the geothermal gradient in the Peridotite Nappe, surface water penetrating to depths of at least ~600 m would have reached temperatures of at least 34 °C if in thermal equilibrium with the host rocks. Our data are inconsistent with this hypothesis; instead, they imply rapid infiltration of meteoric water as far down as the serpentine sole. This is consistent with abundant morphological evidence documenting karst structures developed during laterization (Trescases, 1975; Latham, 1986).

Because active slip typically increases the permeability of faults, water drainage through the peridotites could have been strongly enhanced during active faulting. Indirect evidence for deformation-assisted fluid circulations across the peridotite pile is provided by the syn-kinematic character of the studied magnesite veins along the sole (Fig. 3, see also Data Repository) and the observation that at least some of the nickel mineralizations underlying laterites developed during brittle deformation (Cluzel and Vigier, 2008; our own observations in the Koniambo Massif).

Most magnesite veins of the Koniambo Massif have been emplaced during top-to-SW shearing deformation. Southwestward shearing recorded along the basal contact of the Peridotite Nappe may reflect obduction (e.g., Cluzel et al., 2001) or post-obduction reactivation of the contact as a SW-dipping extensional detachment (Lagabrielle and Chauvet, 2008). Deformation occurred sometimes between ~37 and 27 Ma if related to obduction, or later, but before ~20 Ma, if related to post-obduction NE-SW extension (Chardon and Chevillotte, 2006). The main laterites of New Caledonia were also formed before ~20 Ma (Chevillotte et al., 2006; Sevin et al., 2012). Hence, available time constraints make it possible that carbonation and laterization occurred at the same time, during tectonic activity.

As a result, considering the further requirement of fast downward circulation of surface waters for maintaining low temperature conditions during magnesite formation, we propose that syn-laterization tectonic activity enhanced water infiltration and played a major role in the exportation of leached elements like Mg, leading to widespread carbonation along the serpentine sole.

POTENTIAL IMPLICATIONS FOR OTHER CARBONATED OPHIOLITES

Syn-tectonic carbonation along the serpentine sole of the New Caledonia ophiolite contrasts directly with the well documented case of post-tectonic subsurface carbonation of the Oman ophiolite (Kelemen et al., 2011). Studies documenting meteoric water-derived magnesite in other ophiolite occurrences lack a structural description that would allow the syn- versus post-tectonic character of carbonation to be evaluated (Jedrysek and Halas, 1990; Fallick et al., 1991; Gartzos, 2004; Jurkovic et al., 2012). Nevertheless, syn-laterization tectonically-driven carbonation of ultramafic rocks, as proposed here for New Caledonia, may concern other areas worldwide. For instance, the ophiolites of the Dinaric-Hellenic segment of the Alpine orogen include (i) large volumes of magnesite originated from meteoric water (e.g., Gartzos, 2004; Jurkovic et al., 2012), (ii) laterites capping the ultramafic rocks, with iron-nickel ore deposits (e.g., Eliopoulos et al., 2012) (iii) various time constraints showing that obduction occurred in the Late Jurassic, (iv) the unconformity of Late Jurassic sediments on at least some of the laterites (Robertson et al., 2012). These features strongly suggest that laterization occurred during obduction, which opens the possibility that carbonation occurred simultaneously, fostered by tectonic activity. This pleads for renewed examination of the Dinaric-Hellenic and other carbonated ophiolites worldwide in order to establish whether active tectonics is commonly a major agent for carbonation.

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FIGURE CAPTIONS

Figure 1. A) Simplified geological map of New Caledonia. B) Geological map of the southwestern margin of the Koniambo Massif, adapted from Maurizot et al. (2002). BMS, VAV and CONV indicate magnesite sampling sites. Laterites shown on this map belong to a planation surface that is younger than those having led to much thicker laterites at higher levels of the Koniambo Massif (at elevations between ~400 and ~800 m, see the text) (Latham, 1986; Chevillotte et al., 2006; Chardon and Chevillotte, 2006).

Figure 2. Field view along part of the ‘BMS’ cross-section (located in Fig. 1B), illustrating the exceptional size and freshness of the outcrops recently opened in the serpentine sole of the Koniambo Massif.

Figure 3. Field observations within the serpentine sole of the Koniambo Massif. A, B, C, field views illustrating the relations between deformation and the emplacement of magnesite veins. Numbers 1 and 2 refer to the two main vein types as described in the text. The two ellipses in B show sites where tight folding of a ‘type 1’ vein is well visible. In C, magnesite occurs as coarse fibers suborthogonal to the walls of this ‘type 2’ vein (sample BMS Gio 9). D, stereogram showing the orientation, along the ‘BMS’ cross-section, of major shear zones (shown as poles of planes) and of magnesite veins occurring in between the shear zones (‘type 2’ veins, shown as great circles).

Figure 4. $\delta^{13}C$ versus $\delta^{18}O$ diagram of magnesite veins hosted by ultramafic rocks in different regions worldwide. Black and gray symbols are from this study and the literature, respectively. **[[Spell out versus or place period after vs. abbreviation in figure (Figs. 4 and 5). Citations should be: Fallick et al., 1991; Jurkovic et al., 2012; Jedrysek and Halas, 1990.]]**

Figure 5. Histogram of oxygen isotopic compositions of magnesite. In black, data from this study. In gray, composition range of theoretical magnesite that would develop in equilibrium with serpentines, using the serpentine $\delta^{18}O$ data of Ulrich (2010). The dashed line shows the theoretical composition range of magnesite that would form at near-surface temperature from South-Pacific meteoric waters. The theoretical compositions are calculated using the serpentine-H_2O and magnesite-H_2O fractionation coefficients of Zheng (1993, 1999).

[1] GSA Data Repository item 2013xxx, xxxxxxxx, is available online at www.geosociety.org/pubs/ft2013.htm, or on request from editing@geosociety.org or Documents Secretary, GSA, P.O. Box 9140, Boulder, CO 80301, USA.

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SUPPLEMENTARY MATERIAL

Table DR1. O and C isotope compositions of magnesite samples from this study. The results are given in ‰ versus SMOW for $\delta^{18}O$ and ‰ versus PDB for $\delta^{13}C$.

Figure DR1. Additional field views in the serpentine sole of the Koniambo Massif. A, a low-dipping shear zone and associated ‘type 1’ magnesite veins. In this exemple, three subtypes of vein may be distinguished: (i) a planar vein running along, and parallel to, the roof of the shear zone, (ii) subplanar veins inside, and oblique to, the shear zone, representing Riedel- or C’-type shear planes, (iii) a folded composite vein inside, and broadly parallel to, the shear zone. Folding seems to result from both buckling and drag folding along the shear planes. B, a mylonite zone hosting ‘type 1’ magnesite veins. A first subtype consists of sheared (boudinaged?) veins paralleling the schistosity. A second subtype consists of thinner veins along Riedel- or C’-type shear planes.