

## THE COGNE MAGNETITE DEPOSIT (WESTERN ALPS, ITALY): A JURASSIC SEAFLOOR ULTRAMAFIC-HOSTED HYDROTHERMAL SYSTEM?

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### INTRODUCTION

The Cogne mining district (southern Valle d'Aosta region, Western Alps, Italy) exploited a magnetite-rich serpentinite unit from the Middle Ages to 1979. The Cogne deposit is the largest (18 Mt with an iron grade of 45-50%) in a series of apatite and sulphide-free serpentinite-hosted magnetite orebodies that crop out in ophiolitic units along the western Alpine collisional suture in Valle d'Aosta (Castello, 1981; Diella *et al.*, 1994; Rossetti *et al.*, 2009) and in its southern extension in Corsica (Farinole mine; Routhier, 1963). In southern Valle d'Aosta, most of these magnetite orebodies occur in the Mt. Avic serpentinite massif (located *ca.* 15 km ENE of the Cogne serpentinite) and have been interpreted as former metasomatized podiform chromitites, based on their high Cr concentration and the presence of chromite relicts (*e.g.*, Diella *et al.*, 1994).

The Cogne deposit differs from the above occurrences because its magnetite has a nearly pure endmember composition and contains only trace amounts of compatible elements such as Cr, Ti and V. This geochemical fingerprint, which is unusual for an ultramafic setting, as well as the relatively high tonnage of the deposit, make the Cogne deposit an interesting and still poorly studied example of ophiolite-hosted magnetite deposit. In this work, the geochemistry of the Cogne deposit has been investigated, presenting new petrographic and geochemical data as well as focusing on the textural relationships and the trace element composition of magnetite. In addition, the radiometric age of the magnetite orebody has been determined, for the first time, by U-Th-Pb dating of uraninite. The results evidence that the magnetite geochemistry and age support a seafloor oceanic hydrothermal setting for the Cogne deposit; following this assumption, the possible formation mechanisms, using constraints from geochemical modeling of seawater-rock reactions, have been explored. The demonstration of the oceanic origin of the Cogne deposit could eventually have implications for the interpretation of positive magnetic anomalies present at modern ultramafic-hosted hydrothermal sites on slow-spreading mid-ocean ridges (*e.g.*, Tivey & Dymant, 2010).

### GEOLOGY OF THE COGNE DEPOSIT

The Cogne serpentinite is a 2.5 km long sliver, with an average thickness of 100 m (Di Colbertaldo *et al.*, 1967), tectonically sandwiched between two different metasedimentary sequences. The basal contact of the serpentinite is a thrust fault (Elter, 1971), whereas the upper limit is marked by a thin (< 3 m) boudinaged rodingite. The Cogne serpentinite and the hanging wall metasediments form the core of a km-scale isoclinal fold that repeats the footwall sequence in its upper limb (Elter, 1971). The Cogne serpentinite and the hanging wall metasediments are considered to be part of the same greenschist- to blueschist-facies ophiolite-bearing unit (Aouilletta Unit; Polino *et al.*, 2014), for which, however, no P-T-time estimate exists. Two eclogite-facies ophiolitic units (Grivola-Urtier Unit and Zermatt-Saas Unit; Dal Piaz *et al.*, 2010), which represent the remnants of the Jurassic Piedmont-Liguria ocean (Alpine Tethys; Schmid *et al.*, 2004; Stampfli, 2000), envelop the Cogne serpentinite and the associated metasedimentary sequences. In the Zermatt-Saas Unit (southern Valle d'Aosta), the high-pressure (eclogitic) alpine metamorphic peak was reached in the Eocene (45-42 Ma; Dal Piaz *et al.*, 2001), contemporaneously with the closure of the ocean, and was followed by a greenschist-facies overprint during Late Eocene-Early Oligocene (Dal Piaz *et al.*, 2003). The Cogne magnetite mineralization is confined to the serpentinite body and it is exposed in three zones, referred to as Site 1 (Liconi and Colonna mines), Site 2 (western slope of Montzalet) and Site 3 (Larsinaz mine).

## MATERIALS AND METHODS

Sixty-eight rock samples from the Cogne mining area were studied by means of optical and scanning electron microscopy on thin polished sections. Mineral identification was aided by micro-Raman spectroscopy. The compositions of minerals (Na, Mg, Al, Si, K, Ca, Ti, Cr, Mn, and Fe) were determined by electron microprobe analysis (EPMA) and trace element concentrations in magnetite (Mg, Si, Ca, Ti, V, Cr, Mn, Fe, Co, Ni, Zn, Zr, and Mo) were measured by laser-ablation inductively-coupled plasma mass spectrometry (LA-ICP-MS).

The geochemistry of selected rock samples was investigated by means of X-ray fluorescence and inductively coupled plasma mass spectrometry and emission spectroscopy analyses. Thin sections of magnetite ore were scanned for uraninite by electron microscopy and uraninite grains were analyzed for Mg, Si, Ti, Ca, Cr, Fe, U, Th, and Pb using an EPMA. Both single spot analyses and traverses were performed, obtaining a large number of data points, which were necessarily filtered by excluding mixed or poor-quality analyses. Successive uraninite U-Th-Pb dating was performed following the indications of Montel *et al.* (1996) and Cocherie & Albarede (2001).

In an attempt to simulate the genesis of the Cogne deposit in a seafloor hydrothermal system, fluid-rock interactions were modeled with the EQ3/6 (Version 8.0a) software package (Wolery, 2013), using the database compiled by Klein *et al.* (2009), which contains thermodynamic properties of minerals and solutes in the 0-400 °C range at the fixed pressure of 500 bar. In the model, as a first step seawater is heated to 400 °C at 500 bar (considered to be typical P-T conditions of seafloor reaction zones in mid-ocean ridge settings), reacting with 1 g of harzburgite, thus simulating the descending fluid path with limited reactions with substrate rocks. Then, as a simulation of a reaction zone, the modified hot water reacts with either a harzburgite or a Fe-gabbro (the most common type of abyssal peridotite and the Fe-richest rock that can be found in W-Alpine ophiolites, respectively). The reaction path is terminated after the maximum value of dissolved Fe is reached. Finally, the fluid containing the maximum dissolved Fe is titrated with either fresh or serpentinized mantle rocks (dunite, harzburgite, troctolite, pegmatoid serpentinite), reproducing the reaction of the fluid with the final host rocks in the discharge zone. The titrations were performed at either 300 °C or 400 °C, in accordance with the temperature range estimated for Cogne hydrothermal mineral assemblages by Carbonin *et al.* (2014).

## RESULTS AND DISCUSSION

### *Petrographic features*

At Site 1, the magnetite ore forms a 60-m thick lens concordant with the foliation of the serpentinized harzburgites that envelop the mineralized rock volume. In the other sites the mineralized portions are much



Fig. 1 - Typical nodular magnetite ore from the Cogne deposit.

smaller and consist of disseminations and dismembered veins at Site 2 and of a thin lens (< 10 m) at Site 3. The ore shows different textures and mineral assemblages across the three sites. At site 1, the ore (up to 90 vol.% of magnetite) has a nodular texture, given by mm- to cm-sized magnetite crystals (actually polygonal aggregates of subgrains) in a serpentinite gangue, composed of dominant interlocking antigorite, minor Al-rich lizardite, forsterite, brucite, clinocllore and accessory carbonates and Ti-rich chondrodite. These rocks, have a strikingly texture similar to that of nodular chromitites (Fig. 1). At Site 2, the

disseminated ore occurs as magnetite rich bands (up to ~ 70 vol.%) in serpentinized harzburgite, which preserves orthopyroxene sites (bastites) and relict Mg-Al-rich chromite grains. A generation of pure diopside usually accompanies the magnetite mineralization. In other cases, the serpentinized harzburgite hosts dismembered cm-thick veins, composed of magnetite, antigorite, and chalcopyrite. At Site 3, the ore shows again nodular textures, but the gangue is composed only of interlocking antigorite or can be characteristically enriched in pure diopside and minor clinocllore. Diopsidites containing variable proportions of magnetite have been observed at both Site 1 and Site 3. The largest magnetite crystals are frequently rich in inclusions, represented by dominant clinocllore, minor brucite and calcite (Site 1), relict lizardite (Site 1 and 3), minor sulfides (sphalerite at Site 1, Cu-sulfides and pyrrhothite at Site 2), infrequent andradite (Site 2 and 3) and rare uraninite (Site 1 and 3).

At both Site 1 and 3 atypical serpentinites showing a pegmatoid texture are associated with the magnetite ore and can be variably enriched in magnetite. The pegmatoid texture is given by cm-sized amoeboid domains made of coarse-grained antigorite that replaces yellow Al-rich lizardite, interlobate with regions composed of antigorite, Al-rich lizardite and very fine-grained magnetite, locally forming mesh textures. A corona of fine-grained antigorite lines the boundaries between the two domains. In magnetite-enriched rocks, the early fine-grained magnetite is overgrown by a new coarser-grained generation and lizardite is replaced by antigorite and clinocllore.

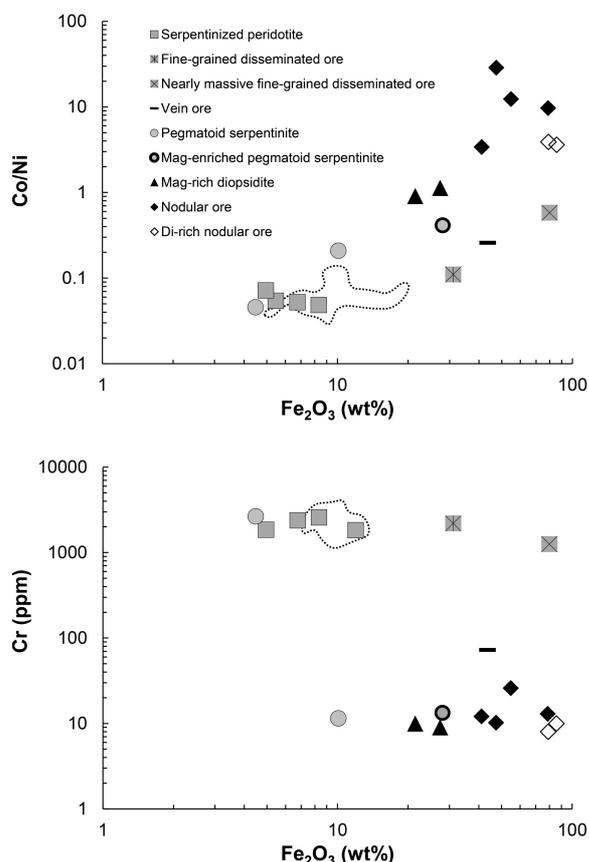


Fig. 2 - Bulk rock Cr and Co/Ni ratio plotted against  $\text{Fe}_2\text{O}_3$  content in Cogne (this work and Carbonin *et al.*, 2014). Contoured regions enclose data for abyssal peridotites after Niu (2004), Paulick *et al.* (2006) and Andreani *et al.* (2014). Redrawn and modified after Toffolo *et al.* (2017).

#### Bulk rock geochemistry

The magnetite poor ( $\text{Fe}_2\text{O}_3 < 8.5$  wt.%) serpentinized peridotites have major and trace element concentrations typical for abyssal peridotites (*e.g.*, Cr = 1900-2600 ppm and Co/Ni = 0.05-0.07; Niu, 2004; Fig. 2). The magnetite-poor pegmatoid serpentinites ( $\text{Fe}_2\text{O}_3 < 10$  wt.%) have very variable Cr content (10-2700 ppm) and Co/Ni ratio (0.05-0.2). The magnetite-enriched pegmatoid serpentinites and nodular ores from both Site 1 and 3 are characterized by low Cr (< 30 ppm) and high Co/Ni (0.4-30); the disseminated ore from Site 2 have high Cr (> 1200 ppm) and Co/Ni positively correlated with the amount of magnetite. The diopsidites have similar geochemical features to the nodular ores.

The geochemistry, the texture, and the relict mineralogy (presence of bastites and Mg-Al-rich chromite grains) of disseminated ore from Site 2 suggest that the host rocks are mantle harzburgites. However, the nodular ores, being geochemically and texturally akin to pegmatoid serpentinites, were emplaced in more atypical rocks. Similar textures, *i.e.*, interlobate domains separated by coronae structures, have been described for olivine-plagioclase assemblages in some troctolites from modern oceanic and ancient ophiolitic settings (*e.g.*, Blackman *et al.*, 2006; Renna & Tribuzio, 2011).

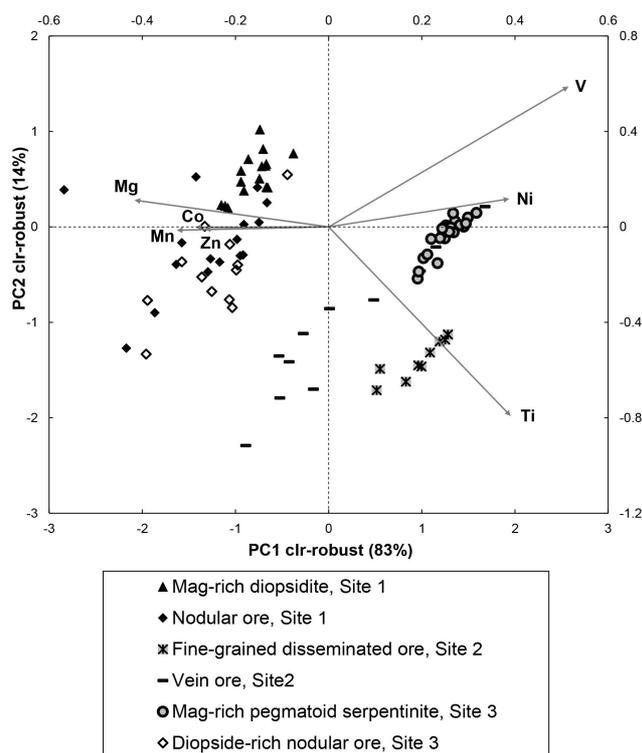


Fig. 3 - Robust-PCA of magnetite trace element composition obtained by LA-ICP-MS. Coordinates of datapoints (scores) are on left and lower horizontal axes. Coordinates of variables (loadings) are on right and upper horizontal axes. Redrawn after Toffolo *et al.* (2017).

### Geochemistry of Cogne magnetite

Cogne magnetite is characterized by high Mg and Mn (median values up to ~24,100 and ~5000 ppm, respectively), and low Cr, Ti and V (median values up to ~30, ~570 and ~60 ppm, respectively). The low Ti content (< 600 ppm) and the very high Ni/Cr ratio ( $\gg 1$ ), in particular, are considered to be typical of hydrothermal magnetite (Dare *et al.*, 2014). The statistical relationship between trace elements in magnetite was explored by a robust principal component analysis (PCA). The variability explained by the first two principal components is very high (97%), hence the PCA can adequately describe magnetite populations. As can be seen in Fig. 3, Mg, Mn, Co, and Zn are highly correlated, whereas Ni is anti-correlated and V and Ti behave more independently. The large cluster of data with relatively high Mg, Mn, Co, Zn and low Ni encompasses magnetite in nodular ores (and diopsidites) from both site 1 and 3. High Ni and V characterize magnetite in pegmatoid serpentinites while high Ni and Ti are distinctive of magnetite in disseminated ores. Magnetite in vein ore has intermediate compositions. These clear distinctions suggest different controls on magnetite composition:

the magnetites rich in more fluid-compatible elements may indicate fluid-controlled conditions, while the others, more enriched in weakly mobile elements such as Ti and V would suggest host-rock buffering. The first principal component (PC1) may thus be interpreted as reflecting magnetite formation under different water-to-rock ratios (W/R) from possibly similar parent fluids, whereas the PC2 further discriminates between the different host rocks (mantle serpentinites vs. pegmatoid serpentinites). In addition to this, the high Co/Ni ratios ( $> 1$ ) of magnetites in nodular ores would suggest a mafic metal source (*cf.* Melekestseva *et al.*, 2013), which is in contrast with the ultramafic nature of most of the Cogne host rocks. Hydrothermal fluids carrying a significant load of transition metals (high Fe, Mn, Cu, Zn  $\pm$  Co  $\pm$  Ni) are observed issuing from ultramafic substrates in high-T ( $> 350$  °C) hydrothermal systems associated with oceanic core complexes in slow-spreading mid-oceanic ridges, such as at Rainbow and Logatchev on the Mid-Atlantic Ridge (Douville *et al.*, 2002; Andreani *et al.*, 2014). In particular, the hydrothermal vent fluids at Rainbow are the richest in Co (Douville *et al.*, 2002), have the highest Co/Ni ratios ( $\sim 4$ ). These fluids produce sulfide deposits at the seafloor, due to mixing with seawater, but they are saturated in magnetite + chlorite + talc (Seyfried *et al.*, 2011). Moreover, positive magnetic anomalies centered on these ultramafic-hosted hydrothermal sites could be related to magnetite concentrations at depth (Tivey & Dyment, 2010). Thus, it seems not unlikely that Cogne mineralization represents an ancient analogue of a deep portion of such hydrothermal sites.

### Age of the deposit

Cogne nodular ores have significant uranium contents (up to  $\sim 3$  ppm), which can be related to the presence of uraninite grains. Uraninite crystals are invariably contained in magnetite and, in turn, they can

include magnetite and clinocllore: this is a good evidence that uraninite formed contemporaneously with the other two minerals. Thus, by dating uraninite the age of the ore can be obtained. The U-Th-Pb weighted average age calculated for a group of three small ( $< 10 \mu\text{m}$ ) uraninite grains from Site 1 is  $161.8 \pm 3.5 \text{ Ma}$  (mean square of weighted deviates, MSWD, = 0.51;  $n = 5$ ). The plateau age obtained for an aggregate of zoned grains from Site 1 is  $150.8 \pm 2.0 \text{ Ma}$  (MSWD = 1.03;  $n = 16$ ) and the age calculated for a big ( $\sim 40 \mu\text{m}$ ), unzoned uraninite crystal from Site 3, which is intergrown with magnetite and includes clinocllore, is  $151.9 \pm 1.4 \text{ Ma}$  (MSWD = 0.91;  $n = 29$ ). The two age determinations that yield the lowest uncertainties and best MSWD values (*i.e.*, close to unity) are within errors of each other and are considered to be the most reliable. However, considering the limited age scatter, it is reasonable to combine all the data into a single age determination, which yields a value of  $152.8 \pm 1.3 \text{ Ma}$  (MSWD = 1.3;  $n = 50$ ). This radiometric age places the ore-forming event in proximity of the Kimmeridgian-Tithonian boundary ( $152.1 \pm 0.9 \text{ Ma}$ ) and overlaps with that of the spreading stage of the Piedmont-Liguria ocean ( $\sim 170\text{-}145 \text{ Ma}$ ; Cordey *et al.*, 2012; Manatschal & Müntener, 2009).

#### *Insights from thermodynamic modeling*

The Cogne magnetite ore was emplaced in mantle serpentinites and, probably, in troctolites, during the spreading-stage of the Piedmont-Liguria ocean. The Cogne magnetite has a hydrothermal composition affected by water-to-rock ratios and has an apparent mafic signature. Based on this evidence and considering the general lack of sulfides, Cogne can be interpreted as a deep segment of a seafloor hydrothermal system. Thus, a geochemical model of a hydrothermal cell was created in the attempt of reproducing the observed mineral assemblages. The fluids produced by interaction of modified seawater with harzburgite and Fe-gabbro, at  $400 \text{ }^\circ\text{C}$  and 500 bar provide two potential endmember compositions for fluids circulating in and reacting with the original oceanic substrate rocks. The geochemical calculations show that the dissolved Fe in the harzburgite-reacted fluid reaches a maximum value of  $\sim 11 \text{ mmol/kg}$  (604 ppm) at W/R  $\sim 7$ ; the Fe-gabbro-reacted fluid, being more acidic, is Fe-richer, with a Fe concentration that reaches  $\sim 26 \text{ mmol/kg}$  (1439 ppm) at W/R  $\sim 80$ . Other major differences between the two fluid types at their Fe peak concern the concentrations of Mg and Si, which are about one order of magnitude lower and two orders of magnitude higher, respectively, in the Fe-gabbro-reacted fluid ( $\text{Mg}_{\text{harz.-reacted}} = 15.6 \text{ mmol/kg}_{\text{solution}}$ ,  $\text{Mg}_{\text{Fe-gab.-reacted}} = 3.1 \text{ mmol/kg}_{\text{solution}}$ ,  $\text{Si}_{\text{harz.-reacted}} = 0.3 \text{ mmol/kg}_{\text{solution}}$ ,  $\text{Si}_{\text{Fe-gab.-reacted}} = 17.5 \text{ mmol/kg}_{\text{solution}}$ ). The titration of the two fluids containing the maximum dissolved Fe with either fresh or serpentinitized mantle rocks, at  $300 \text{ }^\circ\text{C}$  or  $400 \text{ }^\circ\text{C}$ , produces mineral assemblages that are qualitatively consistent with those observed (antigorite + magnetite + clinocllore  $\pm$  brucite, at  $300 \text{ }^\circ\text{C}$ ,  $\pm$  forsterite, at  $400 \text{ }^\circ\text{C}$ ). However, the modal amount of magnetite is invariably too low to produce an iron ore.

#### *Hypotheses on the genesis of the Cogne ore*

The geochemical model indicates that the fluids are not sufficiently Fe-rich to account for the formation of the Cogne deposit. Thus, other processes and factors must be invoked. One possibility could be the presence of a Fe-richer substrate, but this can be excluded because such rocks are not observed in western alpine ophiolites. Another process, which could potentially increase the Fe concentration in the fluid, is the incorporation of a magmatic gaseous component that could promote acidification and thus increase Fe solubility. However, assuming a gas composition similar to that of gases emitted from mafic lavas (see Sawyer *et al.*, 2008), it can be calculated that a relatively high condensed gas/fluid mass ratio of 1:10 would increase the Fe concentrations only by a factor of  $\sim 2.3$  and this is not sufficient to allow a significant increase in the final amount of precipitated magnetite. A process that was not taken into account in the geochemical modeling but is often observed in hydrothermal fluids venting from modern seafloors is phase separation. Phase separation can produce high-chlorinity brines enriched in weakly volatile Fe (and transition metals) and, at the same time, promotes  $\text{H}_2\text{S}$  partitioning into the vapor phase, causing sulfide undersaturation in the brine and thus delaying sulfide precipitation. Therefore, phase separation occurred in the hydrothermal fluid could actually be responsible for the production of the large amount of magnetite and the scarcity of sulfides observed in the Cogne deposit.

Alternative hypotheses that might account for the genesis of the Cogne ore include *i*) the metasomatism of chromitite bodies (Diella *et al.*, 1994; Rossetti *et al.*, 2009) and *ii*) alpine metamorphism of pre-existing seafloor Fe-sulfides or Fe-oxyhydroxides. In the first case, the temperatures predicted for hydrothermal alteration (300-400 °C) and for alpine metamorphism (< 400 °C, based on the coexistence of antigorite and lizardite; Schwartz *et al.*, 2013) were too low to cause any significant Cr mobilization (temperatures in excess of 500-550 °C are required, according to Arai & Akizawa, 2014): the immobility of Cr during magnetite mineralization is testified by the mantle peridotites containing disseminated magnetite from Site 2, which have similar bulk-rock Cr content as their magnetite-poor counterparts (Fig. 2). In the second case, the radiometric ages presented here demonstrate that the magnetite-forming event was coeval with the spreading of the Piedmont-Liguria ocean and thus predates Alpine metamorphism.

## CONCLUSIONS

The Cogne magnetite deposit was formed at ~ 150 Ma by hydrothermal processes during an advanced stage of the opening of the Piedmont-Liguria ocean. Geological, geochemical and mineralogical similarities with some modern ultramafic-hosted VMS deposits on mid-ocean ridges suggest that the exposed mineralized section at Cogne may represent the deep segment of a seafloor, high-temperature (~ 300-400 °C) hydrothermal system, which was possibly associated with shallower, now eroded, sulfide-rich bodies. As suggested by thermodynamic modeling, simple seawater-rock interactions cannot produce a magnetite ore, thus, fractionation processes such as phase separation were probably critical to generate sufficiently Fe-rich hydrothermal fluids capable to precipitate large amounts of magnetite in various types of mantle host rocks. The possible occurrence of similar ultramafic-hosted magnetite deposits in present-day oceanic settings could contribute to explain the presence of significant magnetic anomalies centered on active and inactive ultramafic-hosted hydrothermal fields.

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