INTRODUCTION

The importance of fluids in subduction systems is well established, as they control rheology, metamorphism, metasomatism, and volcanism (Bürgmann & Dresen, 2008; Gerya & Meilick, 2011; Peacock, 1990; Scambelluri & Philippot, 2001; Spandler & Pirard, 2013). In subduction zones, the volumes of fluids released from the slab (Van Keken et al., 2011) and the difference in fluid composition from dehydration at the source rock to solute-rich fluid in the mantle wedge, require that fluid–rock interaction takes place along the intervening pathways (Bebout, 2007; Brovarone et al., 2020; Dragovic et al., 2015; Hermann et al., 2006; Herms et al., 2012; Taetz et al., 2018). Despite the relevance of fluid–rock interaction for interpreting geological processes, several factors, such as complex solubilities and speciation, retrograde overprinting, limited exhumation and exposure, and other gaps in the rock record challenge our progress in this field.

A viable process of fluid interaction with high-pressure rocks is dissolution–precipitation (e.g. Ague &
Nicolescu, 2014; Angiboust et al., 2017; Ferrando et al., 2017; Giuntoli et al., 2018). As this mechanism can take place at low differential stresses, it is widespread in subduction zones (Wassmann & Stöckhert, 2013b; Wassmann et al., 2011). However, as precipitation does not necessarily occur at or near the site of dissolution, this fluid-driven process can facilitate mass loss at the reaction sites and mass transfer in subducting slabs while leaving barely perceptible traces. Tracing dissolution can be further complicated by retrograde overprinting, which is common in high-pressure terranes. Considering the long distances of up to several kilometres over which mass transport is hypothesized in subduction settings, finding an intact fluid pathway on this scale within dismembered high-pressure terranes is unlikely. Therefore, field-based studies will only probe small portions of fluid pathways, which should be treated as open systems (Plümper et al., 2017). Consequently, to understand the process of mass transfer through the slab, the single components of such large-scale systems must be studied individually.

The ultrahigh-pressure metamorphic (UHPM) Lago di Cignana unit (LCU) is a famous coesite- and diamond-bearing Alpine meta-ophiolite (Frezzotti et al., 2011; Reinecke, 1991) and a rare example of oceanic crust that reached 90–110 km depths (Groppo et al., 2009; Reinecke, 1998). Garnetite is common to various LCU lithologies, and although garnetites are typically sedimentary, metasomatic, melt- or fluid-related in origin (e.g. Angiboust et al., 2017; Hadjzobir et al., 2017; Spry et al., 2000; Vrijmoed et al., 2006), no origin mechanism has been established for the LCU garnetites. Infiltration of external fluids and extensive fluid–rock interaction within this unit have been revealed by detailed fluid-inclusion and geochemical studies, suggesting influx of locally solute-rich serpentinite-derived fluids (Frezzotti et al., 2011; Halama et al., 2020; Hawkins et al., 2009). Therefore, the LCU potentially includes remnants of a large-scale fluid pathway, which has not yet been identified and described in detail.

In this study, we examine the relationship between the LCU Mn-rich quartzites and the garnetite lenses that they host using a combination of petrological, mineralogical, and microstructural data. The observations elucidate how fluid pathways shaped the metasediments, how garnetite formed by a newly described mechanism involving pressure solution, how fluid–rock interaction evolved over time during metamorphism, and the stress regime during this process.

## 2 | GEOLOGICAL BACKGROUND

The Alpine high-\(P\) ophiolites of the Piemonte zone (Figure 1) are the dismembered remnants of the Mesozoic Tethyan Ocean that was subducted during Alpine plate convergence and orogeny (Dal Piaz et al., 1999 and references therein). The LCU is a coesite- and diamond-bearing UHPM ophiolitic sliver cropping out between two major metaophiolitic units: the underlying Zermatt-Saas Zone (ZSZ) recording eclogite-facies metamorphic conditions of 520 ± 20°C, 2.3 GPa (Angiboust et al., 2009), and the overlying Combin Zone (CZ) recording blueschist-facies conditions of 300–345°C, ~0.9 GPa (Reddy et al., 1999).

The relationship between the LCU and the ZSZ is still debated. One possibility is that the LCU is one of several minor bodies of diverse origin that crop out along the contact between the ZSZ and CZ (Gilio et al., 2019; Dal Piaz et al., 2001), which differ in their peak-metamorphic age, \(P–T\) evolution, and provenance (Theodul Glacier Unit, Etrol-Levaz Slice, Allalin Gabbro; Beltrando et al., 2010; Bucher & Grapes, 2009; Weber et al., 2015). The alternative is that the LCU is located within the upper levels of the ZSZ, and only deviates in terms of peak metamorphic pressure, while metamorphic ages and temperatures match (Groppo et al., 2009; Skora et al., 2015).

The LCU consists of coesite-bearing eclogite, serpentinite (Gilio et al., 2019) and metasediments varying from calcscschist and marble to several types of manganiferous quartzites (Forster et al., 2004; Reinecke, 1998). The quartzite varies from schistose garnet–amphibole–phengite quartzites, to purple, hydothermally Mn-enriched quartzite similar to other Mn-rich metasediments from the ZSZ (Tumiani et al., 2010). Garnetite is present in several metasediments and eclogites as lenses and boudins. Certain LCU garnetites contain fluid inclusions with solutes that originated from the dissolution of carbonate and quartz (Frezzotti et al., 2011). Furthermore, Hawkins et al. (2009) observed an assemblage of daughter minerals in fluid inclusions in garnetite, consisting of jadeite, paragonite, phengite, chlorite, amphibole, titanite, and K-feldspar alongside Ca-sulphates, phosphates, and oxides. This evidence suggests
the dissolution of a polymineralic component of the system during garnet growth, beyond quartz and carbonates (Frezzotti et al., 2011; Hawkins et al., 2009). Boron concentrations and isotopic ratios in white mica within several lithologies in the LCU area indicate that a fluid derived from partially dehydrated serpentinites was the driver behind fluid–rock interaction in quartzite, but not in eclogite (Halama et al., 2020).

3 | METHODS

3.1 | Sample preparation

Samples were processed into sections 100 µm thick for petrographic analysis and electron microscopy. For the purpose of strain analysis on inclusions in garnet, sections 250 µm thick were prepared to maximize the number of inclusions without strain dissipation due to proximity to the surface of the section. Sections were polished with a 0.05 µm colloidal silica suspension in preparation for high-quality electron microscopy.

3.2 | Electron microscopy

Electron probe micro-analysis (EPMA) measurements of major elements were performed using a JEOL JXA 8200 Superprobe at the Department of Earth Sciences, University of Milan. An accelerating potential of 15 kV and a beam current of 15 nA were used for spot analyses with a 30 s counting time. Measurements for element distribution maps were performed with a combination of WDS and EDS, at 15 and 100 nA for high-detail maps. Element distribution maps were plotted using an inversion of the ‘roma’ scientific colour map (Crameri, 2018).

3.3 | Electron backscatter diffraction

Electron backscatter diffraction (EBSD) data were acquired on a Philips XL-30 field-emission gun scanning electron microscope equipped with an Oxford Instruments’ Nordlys-Nano EBSD detector and AZtec 3.3 acquisition software. An EBSD map of 1,638 × 1,228 pixels was acquired at a step size of 0.9 µm and an accelerating voltage of 30 kV. Data were processed using the MTEX v5.1.1 toolbox (Bachmann et al., 2010) for MATLAB®. Grain reconstruction was performed with a minimum misorientation value for grain boundaries of 2°. Although this value is low, no subgrain boundaries were misidentified as grain boundaries as subgrains are absent in the analysed garnet crystals. All grains below 5 pixels in size were considered to result from misindexing and were removed. Unindexed areas 1 pixel in size were filled in by interpolation. Grains in contact with the outer boundary of the map were removed from grain-size analysis. The EBSD map was colour coded by grain size using an inversion of the ‘roma’ scientific colour map (Crameri, 2018).

3.4 | Raman spectroscopy

Raman spectra of quartz inclusions in garnet were collected with a Horiba LabRam HR Evolution spectrometer (holographic gratings of 1,800 grooves/mm) equipped
with an Olympus BX41 confocal microscope at the controlled temperature of 20(1)°C. Raman spectra were excited using the 514.532 nm line of a solid state (YAG) laser. The laser power on the sample surface was approximately 1–2 mW. The spectrometer was calibrated to the silicon Raman peak at 520.5 cm⁻¹. The spectra were baseline corrected for the continuum luminescence background when necessary, temperature reduced to account for the Bose–Einstein occupation factor (Kuzmany, 2009) and normalized to the acquisition time. Peak positions, full widths at half maximum (FWHMs), and integrated intensities were determined from fits with pseudo-Voigt functions (pseudoVoigt = (1 − q) × Lorentz + q × Gauss, where q is the mixing coefficient).

3.5 | Elastic strain analysis

Elastic strains in quartz were determined for 17 quartz inclusions in garnet by using the relationship between strain and shifts in the position of selected Raman peaks (Angel et al., 2019; Murri et al., 2019). The shift, Δω, of each Raman peak is the difference between the peak positions in the inclusion, ωₚ, and an unstrained reference crystal, ω₀. To reduce shifts in peak position due to instrumental drift and/or minor changes to room temperature, the unstrained standard was measured multiple times per session. The ω₀ values were averaged and then subtracted from the ωₚ of the strained inclusions analysed in between two consecutive standards. Sets of Δω (modes 128, 206, 464 and, when available, 265) for each inclusion were converted to strain using the software stRAinMAN (Angel et al., 2019), applying the Grüneisen tensors for quartz (Murri et al., 2018) and relaxation of the strains (Angel et al., 2014a). Strain interference can occur due to complex inclusion geometries or inclusion proximity to the sample surface, outer edge of the garnet, or other inclusions. Therefore, we only analysed non-faceted, rounded, and sub-rounded inclusions, isolated by at least three times their radius (Campomenosi et al., 2018; Mazzucchelli et al., 2018; Zhang, 1998).

For comparison with the strain state in inclusions measured with Raman, numerically predicted strains were calculated based on expected entrainment conditions using the software EosFit7C (Angel et al., 2014b). The prograde P–T path for the LCU (Groppo et al., 2009; Reinecke, 1998) was divided into four segments of 0.5 GPa by 100°C (varying with pressure along the P–T path), from 1.0 to 3.0 GPa. Pressure was extended into the coesite field to accommodate any potential overstepping of the quartz–coesite transformation. Precise P–T values and resulting calculated strain states are available in the Supporting information. The host garnet mainly consists of an almandine+spessartine mixture, however, a complete set of elastic and thermodynamic properties for these two garnet end-members is unavailable in literature. Thus, we calculated expected strains for given entrapment P–T conditions using data for grossular. Differences between elastic properties of garnet end-members are small, and as a result, only minor variations (<5% at near-identical ratio of ε₁ + εₑ versus ε₃) in expected strains should occur when using properties of a different end-member (Zaffiro, 2020).

4 | SAMPLE DESCRIPTION

Garnetite layers and lenses are present in several of the LCU lithologies, including the Mn-rich schistose quartzite investigated here (Figure 3a–c) and eclogite (Figure 3d). However, this garnetite best preserves the relationship between the host rock and the garnetite. In contrast, the eclogite contains only sparse garnetite, and the structural relationship between eclogite and garnetite is unclear.

The Mn-rich schistose quartzite forms a 5 m thick layer, at the centre of which is a 30 cm thick zone where the majority of the garnetite is located. Here, the garnetite occurs as lenses, discontinuous layers, dismembered fold hinges, and limbs transposed along a phengite foliation (Figure 3a–c). This garnet quartzite can thus be subdivided into (a) garnet- and phengite-bearing schistose host rock and (b) garnetite (Figure 3a). Boudinage of the garnetite indicates a competence contrast between the garnetite and the quartzite host rock (Figure 3b,c). The garnetite lenses, particularly the thicker parts, are structurally complex and appear heterogeneous in terms of garnet content based on colour variations. Some quartz-rich veins and patches cross cut the lens and its outer margins.

4.1 | Host rock

The matrix of the host-rock quartzite exhibits a foliation defined by phengite, and contains garnet as euhedral grains <1 mm in size (Figure 4b). Smaller anhedral garnet crystals are concentrated into lenses and layers <1 cm across. The host rock also contains strongly zoned epidotes and aggregates of amphibole of a composition between magnesiohornblende, actinolite, and winchite, alongside phengite, biotite, and quartz.

Two distinct mineral assemblages are present as inclusions in garnet (Table 1). In the cores of this garnet, inclusions of quartz, apatite, magnesite, dolomite, rutile, and zircon are present. Rutile is often <1 µm in size, giving the garnet core an orange hue. In the garnet mantle, an assemblage of coesite, kyanite, clinozoisite, phengite, rutile, and zircon is present. The euhedral garnet rims contain occasional quartz inclusions.
4.2 | Garnetite

The garnetite is microstructurally diverse (Figure 4a), and can be subdivided into an inner and outer garnetite. This subdivision is based on garnet size and on quartz content, with the outer garnetite having more interstitial quartz and larger garnet (Figure 4c). Besides being smaller and having less to no interstitial quartz, garnet in the inner garnetite have a ‘welded’ appearance (Figure 4d). Large euhedral garnet are locally present on the outer margin of the inner garnetite (Figure 4e). The outer garnetite contains up to ~50% interstitial quartz by volume and occasional quartz veins cross cut the entire garnetite (Figure 4a). Rare accessory minerals, including phengite, apatite, biotite, and sulphides, are present in the garnetite. A variety of structures are present within the garnetite. The garnet in the outer garnetite are mostly euhedral and are often distributed in chains separated by quartz (Figure 4c). Garnet within this zone also exhibit a weak shape preferred orientation, as their long axis is subparallel to the host-rock foliation and perpendicular to the garnet chains.

In both garnetite zones the main inclusion assemblage is similar to that of host-rock garnet cores and consists of quartz, apatite, magnesite, dolomite, rutile, and zircon (Table 1). As in the host-rock garnet, rutile is usually sub-micron sized and occurs in clusters, which give the garnet an orange hue (Figure 4d). In the outer garnetite an inclusion-free rim is present around the inclusion-rich garnet core. The euhedral garnet overgrowth on the outer margin of the inner garnetite contains quartz inclusions (Figure 4f). No coesite has been observed in the garnetite.

5 | Garnet Structures and Composition

5.1 | Host rock

The backscattered electron (BSE) image of a representative host-rock garnet displays strong compositional zonation that is locally complicated by irregularities (Figure 5a). Radial veinlets that appear with the same brightness in BSE as the euhedral rims cross cut the darker garnet. The garnet cores do not exhibit any irregularity in zonation besides radial veinlets. In contrast, the growth zones surrounding the core (garnet mantle) are separated by irregular boundaries between compositional zones.

EPMA analyses and element distribution mapping of representative host-rock garnet reveal a strong compositional zonation (Figures 7a,d,g and 8a). Garnet compositions corresponding to coesite inclusions in host-rock garnet are marked as representing UHPM garnet growth (Figures 7a,d and 8). From these analyses, two trends can be distinguished. The
first compositional trend (labelled 1 in Figure 8a) forms the common prograde metamorphic zoning trend. Mn-rich garnet cores progress to compositions richer in Fe, whereas Mg initially increases in the intermediate mantle before decreasing so that Mg concentrations are similar between earliest and latest compositions. This metamorphic zonation is interrupted by irregular compositional boundaries located within the Mg-richer UHPM zone.

A second trend in garnet compositions also depletes in Mn and enriches in Fe, yet without the Mg-richer excursion, essentially forming a series of intermediate compositions between the earliest prograde growth stage and the latest growth stage (Figure 8). Compositions related to this trend occur along the radial veinlets, in the cores of garnet, and as the outer rim (very similar BSE intensity in Figures 5a and 7a,d). The veinlets appear to radiate from the core of the garnet, where a ~50 to 100 µm zone of elevated Ti signal can be found, linked to the presence of sub-micron rutile inclusions (Figure 7g).

5.2 | Garnetite

Garnet in the outer garnetite exhibit several similarities with garnet in the host rock (Figure 5b–e). One irregular boundary between compositional zones is typically present (Figure 5c)
and, in addition, these garnet crystals display truncation of the metamorphic zones at garnet–garnet contacts (Figure 5b,c). The truncation involves portions of the garnet core and mantle, and is locally overgrown by another zone of garnet (Figure 5c). Radial veinlets are common to garnet in the outer garnetite (Figure 5b–e). Occasionally, garnet exhibit interlocking lobate structures between two cores (Figure 5d,e). The outer zone of the garnet is euhedral and free of veinlets. Particularly in the garnet-richer zones of the outer garnetite, garnet–garnet contacts are often sealed with a late growth of garnet (Figure 5b,e).

Garnet in the inner garnetite do not resemble the other garnet types, as they do not exhibit a distinct metamorphic zonation and lack radial veinlets (Figure 5f). In addition, these garnet exhibit irregular grain shapes, illustrated by interlocking lobes of dark grey garnet (Figure 5f). Interlocking features are more common in garnetite with lower quartz content. Most garnet–garnet grain boundaries are sealed with a late growth of garnet (e.g. Figure 5e), which also gives the inner garnetite a single-crystal appearance in thin section and hand specimen. The euhedral garnet at the outer margin of

**FIGURE 5** Backscattered electron (BSE) images with contrast adjusted so all minerals besides garnet are black or white and zones within garnet are highlighted. (a) Host-rock garnet exhibiting irregular boundaries between compositional zones, four of which are marked with white arrows. Brighter thin features often perpendicular to the zoning are radial veinlets. (b) Garnet in the outer garnetite exhibit truncated of zones at the grain contacts, as indicated by white arrows. Garnet–garnet contacts are sealed with late-stage garnet. Note the similar BSE contrast for the outermost garnet rim, the garnet inside garnet–garnet contacts and within the radial veinlets. The large, white inclusion is rutile. (c) Garnet in the outer garnetite in part of a garnet chain, with white markers indicating truncated zones with late-stage zones grown over the truncation. Black marker indicates an irregular boundary between garnet zones of different compositions. (d) Small aggregate of garnet in the outer garnetite exhibiting truncated zones, indicated by markers, alongside interlocking garnet structures. Selected grain boundaries have been marked by dashed white lines to emphasize the interlocking structure. (e) Outer garnetite with a low amount of interstitial quartz. Markers indicate a late growth of garnet along grain boundaries with a bright shade in BSE, similar in composition to radial veinlets throughout the garnet and related alteration focused in the cores. Black dashed box highlights an interlocking structure between two neighbouring garnet. (f) Mass of garnet in the inner garnetite. Several interlocking lobate structures have been marked.
the inner garnetite locally contain inclusions of garnet from the inner garnetite (Figure S1f).

Grain boundaries in the inner garnetite can be difficult to distinguish based on compositional zones alone. However, individual grains can be clearly separated based on crystal orientations measured by EBSD. Figure 6a presents an EBSD map of a portion of inner garnetite. Although the majority of the garnetite has a relatively consistent distribution of grain size, some bands of a consistently finer grain size, a few hundred micrometres thick (blue domain in Figure 6a), are present within the inner garnetite. In BSE images, these zones exhibit an increase in complexity of grain shape and zonation relative to the adjacent garnetite (Figure 6a,b), and abundant rutile and zircon grains, with strong BSE signal and often <1 µm in size. Figure 6c presents an extreme case of rutile and zircon enrichment along these fine-grained zones.

Despite differences in microstructures, the same two trends (1: metamorphic zoning, and 2: recrystallization) as are present in the compositions of host-rock garnet, are recognizable in the garnetite garnet (Figure 8). The compositions along this trend are somewhat enriched in Mn in the outer and inner garnetite relative to those corresponding to UHPM and late growth in the host rock. In the outer garnetite, the garnet zone with compositions matching the UHPM garnet is significantly thinner than in host-rock garnet, despite a similar size of garnet cores (Figure 7a,b,d,e,g,h). Garnet growth over the truncated zones (Figure 7b,e; see line labelled ‘Truncation boundary’ in Figure 8b) corresponds to compositions of coesite domains in host-rock garnet. Garnet cores in the garnetite, often displaying erratic zonation (Figure 7b,c,e,f), adhere to the recrystallization trend also found in the host rock (Figure 8). In the inner garnetite, garnet of the same composition as the coesite zone in host-rock garnet is barely present, only occurring as minor component in interlocking lobes and in the euhedral overgrowth at the outer margin. This overgrowth does not correspond to the euhedral late-stage growth that is observed in the outer garnetite and host rock, based on their different compositions.

Compositions matching the second trend in the outer garnetite occur mainly in recrystallized garnet cores, alongside Ti enrichment attributed to rutile inclusions (Figure 7g–i). The interlocking lobes lack these rutile inclusions, similar to garnet of corresponding compositions in the outer garnetite and host rock. Garnet–garnet grain boundaries are also sealed with garnet of a Fe-rich composition along either trend. This trend of garnet compositions is also observed along radial veinlets and as an outer garnet rim. That the recrystallization of garnet cores resulted in a similar composition as garnet within veinlets is most visible in Figure 7b, where veinlets and the recrystallized core appear strongly related. Recrystallization processes partially overprinted and obscured original zonation of garnet cores in the garnetite.

6 | STRAIN STATE OF QUARTZ INCLUSIONS IN GARNET

Elastic strain analysis via Raman spectroscopy allowed for the determination of the strain state and associated 2σ variance–covariance ellipsoid for quartz inclusions located in the euhedral garnet overgrowth at the outside margin of the inner garnetite (Figure 9a, corresponding to Figure 4e,f; upside-down triangles in Figure 8c). These inclusions were measured to study the relationship between stress and fluid-driven mechanisms. Only the quartz inclusions in this specific garnet population were used, to avoid potential interference from radial veinlets or recrystallization as is observed in other garnet populations.

The method of elastic geothermobarometry was developed for the primary aim of obtaining entrapment \( P-T \) conditions. However, we can also numerically predict what values of \( \varepsilon_1 + \varepsilon_2 \) and \( \varepsilon_3 \) are expected for hydrostatic entrapment conditions and envisage deviations from these calculated strains.

**Figure 6** Zones of reduced grain size within the garnetite. (a) EBSD grain boundary map with grains coloured based on surface area, overlain on the band-contrast image. Grains on the edge of the map were removed. (b) BSE image of a similar zone of reduced grain size. (c) Zoom-in of area marked in (b), in which areas of high Ti content as measured with EPMA element mapping are highlighted in red.
**FIGURE 7** EPMA element distribution maps for Mn (a–c), Mg (d–f) and Ti (g–i) of garnet from three different zones. The host-rock garnet maps (a, d, g) correspond to Figure 5a. Quartz and coesite inclusions as identified by Raman spectroscopy, and the transect along which garnet composition was measured (Figure 8a), are marked in (a) and (d). Based on the inclusions, the garnet has been subdivided into prograde, UHPM (peak) and retrograde zones. Irregular boundaries marking a contrast in composition are indicated with arrows. The outer garnetite zone displayed here (b, e, h) corresponds to Figure 5c. In (b) and (e), markers indicate truncated zones with late stage zones grown over the truncation. The area of inner garnetite (c, f, i) corresponds to Figure 5f. Grain boundaries as obtained by EBSD, alongside areas that were not indexed, are indicated in black. Ti map (i) appears more enriched than (g) and (h), as a result of a beam current of 100 nA instead of the 15 nA used for the other maps. Scale bars represent 100 µm
as factors external to the host-inclusion system, either during formation or afterwards (Figure 9b; Alvaro et al., 2020; Bonazzi et al., 2019; see Tables S8–S11 for full data). The expected strain states based on LCU \( P-T \) data from literature (Figure 2b; Groppo et al., 2009; Reinecke, 1998) have been provided for comparison.

Typical strain values obtained from the quartz inclusions are between \(-0.024\) and \(-0.012\) for \( \varepsilon_1 + \varepsilon_2 \), and between \(-0.0054\) and \(0.0014\) for \( \varepsilon_3 \). Within error, most inclusions cluster along or near both the hydrostatic stress line and the expected strains for the LCU. Two inclusions (i14 and i20) have no apparent strain, probably having been affected by...
fractures in the host garnet. The strain states of the majority of measured quartz inclusions cluster around the line corresponding to 0.75 GPa inclusion pressure (Figure 9a,b), in accordance with entrapment conditions between 1.5 and 2.0 GPa for the prograde temperatures of the LCU.

7 | DISCUSSION

The garnet microstructures and compositions and the mineral inclusions in garnet described here provide key information on the subduction-zone evolution of the Lago di Cignana UHP garnet-quartzite and garnetite. Importantly, our data demonstrate that garnetite formed through a process of mineral compaction and pressure solution. Moreover, the inclusion populations in garnet provide information on the possible evolution of matrix minerals through time and during garnet growth, and Raman spectroscopy of quartz inclusions in garnet helps understanding the stress regime operating during fluid-mediated garnet deformation and dissolution in garnetite.

7.1 | Evidence for fluid–rock interaction

7.1.1 | Host rock

In the UHPM garnet quartzite presented here, host–rock garnet display zoning patterns that are interrupted by irregular boundaries of contrasting composition (Figures 5a,c and 7a,d). These boundaries are spatially related to resorption of Mn (Figure 7a), suggesting that periods of garnet growth were halted by stages of garnet dissolution driven by disequilibrium between garnet and an infiltrating fluid (Figures 5a–c and 7a,d). The cyclicity of dissolution and growth suggests that fluids pulsed over an extended period during metamorphism.

In host-rock garnet, ubiquitous radial veinlets of a late-stage garnet composition cross cut the majority of the zonation. These veinlets indicate a mechanism of selective garnet replacement along fractures formed during retrograde metamorphism (Angiboust et al., 2012; Broadwell et al., 2019; Giuntoli et al., 2018; Rubatto et al., 2020; Whitney, 1996). Currently known mechanisms for garnet fracturing during metamorphism, either seismic fracturing (Angiboust et al., 2012; Giuntoli et al., 2018), or hydrofracturing due to increased pore fluid pressure (Rimša et al., 2007), normally do not generate radial fractures. The radial nature of the observed fractures could derive from stressed mineral inclusions in the garnet cores (Van der Molen & Van Roermund, 1986).

In host-rock garnet, the mineral inclusion assemblages are: (a) quartz, rutile, dolomite, magnesite, apatite, zircon in the core, (b) coesite, kyanite, clinozoisite, phengite, zircon in the mantle, (c) quartz in the rim (Table 1). The distribution of quartz and coesite in this garnet (Figure 7a,d) indicates a primary origin of such inclusions, despite the fractures in the host garnet which could have re-equilibrated the inclusions (Griffiths et al., 2014; Whitney, 1996). The inclusions thus correspond to preserved remnants of the evolving rock matrix at different stages of garnet growth. Absence in the matrix of key inclusion minerals present in the host-rock garnet (e.g. carbonates) suggests that such minerals might have been involved in the same mineral dissolution process recorded by the garnet. Overall, the quartzite hosting the garnetite layers retains evidence of dissolution that affected the garnet and some matrix minerals.

7.1.2 | Garnetite

The majority of garnet–garnet contacts in the outer garnetite exhibit discontinuous zonation (Figure 5b–e) interrupted by an irregular pattern of garnet grain boundaries where garnet indent and flatten each other. These microstructures are the result of pressure solution, a fluid-mediated deformation mechanism that operates through the increased solubility at stressed grain contacts, resulting in localized dissolution and potential reprecipitation at lower-stress areas. This mechanism has previously been recognized for garnet (Smit et al., 2011; Wassmann & Stöckhert, 2013a), and can be distinguished from undeformed polycrystalline garnet aggregates as only deformation by pressure solution will lead to discontinuous zonation across grain boundaries (Smit et al., 2011). Figures 5c and 7b,e display garnet that has overgrown truncated garnet–garnet contacts, indicating that pressure solution was not a retrograde feature as described by Wassmann and Stöckhert (2013a) but instead took place during prograde to peak metamorphism.

Delicate garnet-chain microstructures are preserved in quartz-rich domains of the outer garnetite (Figure 4c) and coincide with garnet flattening and truncation of compositional zones by pressure solution. The garnet-chain microstructure resembles stacked garnet as described and interpreted by Massey et al. (2011) as the result of flattening in a direction parallel to the chains coupled with lateral stretching, accommodated by pressure solution and reprecipitation of interstitial quartz. In the case of the LCU outer garnetite, the host-rock foliation is near perpendicular to the garnet chains, supporting the notion that the shortening direction is parallel to the chains. Moreover, the origin and preservation of this microstructure relies on little to no simple shear. Truncated zonation from pressure solution is also observed in parts of garnetite that are not rich in quartz (Figure 5e), suggesting that flattening strain in garnetite did not rely on the local remobilization of quartz alone, as was suggested...
for this microstructure by Massey et al. (2011). The garnet stacking provides no constraints on strain perpendicular to the stacking and instead can also indicate uniaxial strain with no related stretching, which relies on the reduction of volume by local dissolution coupled with removal of mass from the system.

Most garnet–garnet contacts in the inner garnetite and a few in the outer garnetite have developed highly interlocking lobate structures by recrystallization through grain boundary migration (GBM; Figure 5d–f). This recrystallization mechanism is a process where grain boundaries sweep over part of an old grain, recrystallizing it as part of the neighbouring grain. As a result, the grain morphology is altered, yielding a higher degree of interlocking than is known from pressure solution (see microstructures of chemically induced GBM in calcite; Hay & Evans, 1987; Smit et al., 2011). The GBM-recrystallized garnet differs from the precursor garnet as the result of a change in garnet composition during recrystallization (Figures 5f and 7c,f,i). For the composition to change, an input and output of solutes is required, particularly as this mechanism should destroy and regrow equal amounts of garnet. GBM took place along all grain boundaries where garnet cores were in contact, and therefore a saturated wetting fluid was present throughout the garnetite. GBM in the outer garnetite relied on pressure solution to expose two garnet cores and bring them into contact; core–mantle contact yielded no GBM (Figure 5b).

The bands with finer grain size that cross cut the inner garnetite (Figure 6) differ from surrounding garnetite in grain size and in enrichment of rutile and zircon. Both differences can be explained by a higher degree of garnet dissolution within the fine-grained bands, leading to a grain-size reduction and the liberation of less soluble inclusions (zircon, rutile). Alternatively, release of Zr from the garnet could drive zircon precipitation. An alternative mechanism for the accumulation of rutile is an influx of a Ti-bearing fluid (Scambelluri & Philippot, 2001) leading to rutile precipitation. Either way, rearrangement of pre-existing rutile grains into an evolving garnet microstructure, or rutile precipitation in trails within this garnet, are both fluid-mediated processes.

In quartz-poor areas of the garnetite, garnet–garnet contacts are sealed by a final stage of garnet growth. This garnet is of similar composition to garnet in the radial veinlets and outermost garnet rim in the outer garnetite and host rock (Figure 5b), corresponding to the Fe-rich part of the composition range (Figure 8; note lower Mg and Mn of this garnet in Figure 7). Since no interstitial minerals were present to drive a garnet-forming reaction, either within fractures or along grain boundaries, the garnet must have precipitated from a fluid. An alternative mechanism relies on the selective replacement of garnet similar to the formation of radial veinlets by a fluid along fractures. The textures presented here thus indicate that the garnetite garnet was wetted by fluids during GBM and during late-stage garnet growth and/or replacement in fractures, grain boundaries, and as tiny rims.

In the inner and outer garnetite, carbonates only occur as inclusions in the garnet core and are absent as intergranular phases, pointing to dissolution of intergranular carbonate. A lack of interstitial quartz in the inner garnetite, where quartz inclusions are common, suggests that intergranular quartz was also removed. This mineral dissolution supports the finding of (a) bicarbonate and carbonate ions and silica monomers in fluid inclusions and of (b) associated microdiamonds in other, nearby, garnetite lenses in the LCU (Frezzotti et al., 2011).

### 7.1.3 | Reconstruction

Garnet microstructures can be linked to the well-established $P–T$ path of the LCU (Figure 2b) by comparing garnet composition linked to those microstructures to the presence of primary quartz and coesite inclusions in host-rock garnet of the same composition. Although garnet morphology varies between garnetite and host rock, the studied garnet exhibit a near-identical compositional evolution (Figure 8), the only difference being a slight Mn enrichment in the garnetite. This similarity, together with the similar size (Figure 7g–i) and reactivity of inner garnet cores in all garnet types, suggests a common initial garnet-forming reaction at a similar rate of nucleation versus growth. Based on this linked garnet growth, the composition and inclusions of quartz versus coesite in the host-rock garnet are used as a tool for relative timing and an estimate for UHPM conditions of garnet growth throughout the system.

The temporal link between the rock domains established through garnet composition is used to reconstruct their evolution in terms of garnet growth, dissolution, deformation, and recrystallization, and related fluid flow patterns (Figure 10). After the initial growth of garnet cores at approximately 1.0 GPa and 400°C (Reinecke, 1998), the first stage of compaction by fluid–rock interaction took place by dissolving and transporting interstitial material (stage 1, Figure 10b). The above processes concentrated garnet in what is now the inner garnetite, essentially forming a garnet stylolite. The euhedral garnet growing at the outer margins of the inner garnetite formed after this compaction, and the lower range of $P–T$ conditions obtained from quartz inclusions in this garnet (~1.5 to 2.0 GPa) indicate that compaction took place at conditions of 1.0–1.5 GPa and 400–450°C. No fluid–rock interaction during this phase is recorded in the host rock.

The second stage (stage 2, Figure 10b) was marked by the dissolution of matrix minerals adjacent to the inner garnetite and related arrangement of garnet in the garnet chain microstructure, while host-rock garnet continued regular
metamorphic growth. While the outer garnetite was compacting and partially dissolving into an infiltrated fluid, the inner garnetite underwent GBM and growth of euhedral crystals at the outer margin. This phase took place at conditions between stage 1 and UHPM conditions, thus 1.5–2.6 GPa and 450–550°C.

During UHP metamorphism (stage 3, Figure 10b), the host rock and to a lesser degree the outer garnetite were affected by fluid pulses that induced cycles of dissolution and growth in garnet. Only local fluid pathways still allowed fluid flow in the inner garnetite. By examining the youngest preserved garnet before pressure solution and the garnet that grew over truncated zonation (Figures 5c and 7b,e), a window for pressure solution in the outer garnetite is constrained as prograde to the first part of UHPM (Figure 8b,d), coincident with formation of the mantle of host-rock garnet.

In the final stage (stage 4, Figure 10b) during exhumation, garnet in the outer garnetite and host rock were fractured radially. These fractures provided a fast pathway for fluids, leading to selective replacement of garnet in cores, along
fractures, and along grain boundaries. This event was coeval with a final phase of garnet growth, sealing all garnet–garnet grain boundaries and forming a euhedral outer rim on garnet throughout the system.

The above evidence suggests that fluid flow and mineral dissolution during prograde metamorphism was localized in and directly adjacent to the inner garnetite, whereas most of the fluid pulses during UHPM were focused on the host rock, with lesser impact on the outer garnetite. This progression suggests a shift over time of the fluid pathway, not only migrating from the inner garnetite towards the host rock but also widening in the process.

7.2 Evaluation of stress conditions from quartz inclusions

The studied quartz inclusions are inside euhedral garnet crystals located at the outer margin of the inner garnetite. This euhedral garnet grew from circulating fluids, after formation of the inner garnetite and coeval with GBM, and is considered representative of the stress conditions during garnet growth at approximate $P$–$T$ conditions of 1.5–2.0 GPa and 450–490°C.

Factors external to the host-inclusion system, such as differential stress during entrapment, can change the strain components from the expected strain state. As a result, inclusions that are entrapped under hydrostatic conditions should correspond to calculated strains for the $P$–$T$ conditions of entrapment, while a significant differential stress during entrapment will lead to a deviation from the predicted strains, along a line of constant inclusion pressure (Figure 9b; Bonazzi et al., 2019). This method is not calibrated as quantitative, so currently no magnitude of differential stress during entrapment can be obtained.

With the exception of two outliers, the calculated strain states of all measured quartz inclusions fall within uncertainty of the line that indicates the strain ratio expected for quartz under hydrostatic stress, and the expected strain states for hydrostatic entrapment along the prograde $P$–$T$ path for the LCU. Depending on the peak metamorphic temperature of the LCU (590–605°C, Groppo et al., 2009; 630°C, Reinecke, 1998), little to no plastic deformation occurred in garnet, and therefore the state of strain and inclusion pressure of the quartz is reliable (Voegelé et al., 1998; Zhong et al., 2020).

The inferred hydrostatic entrapment conditions for quartz in garnet fit the fluid-driven deformation process envisaged, with a fluid wetting the garnet crystals. Load-bearing and consequently stressed grain contacts resulted in local pressure solution, while other parts of the garnet were coated by a fluid, where quartz subsequently recorded (near-) hydrostatic stress during entrapment.

7.3 Implications

Reconstructing the evolution of the host-rock–garnetite system in the LCU quartzite has yielded several gaps in the rock record, most notably when comparing the inclusion mineral assemblages to the current matrix mineral assemblages. The extensive evidence for garnet dissolution presented here is not coupled with concomitant reprecipitation, which implies that the whole process took place under open system conditions, with overall loss of material. The fluids responsible for mineral dissolution were externally derived and likely made available by dehydration of nearby serpentinite (Halama et al., 2020). Because garnet was concentrated into garnetite by dissolution of more soluble interstitial minerals along a fluid pathway, the garnetite can be considered a stylolite. The genesis of garnetite is generally linked to precipitation from a fluid or melt, or sedimentary processes (e.g. Angiboust et al., 2017; Hadjzobir et al., 2017; Stry et al., 2000; Vrijmoed et al., 2006); therefore, the process described here provides a new mechanism for garnetite formation.

The fluid pathway described in this study involves mass removal via dissolution and transport of quartz, garnet, and carbonates, due to circulation of large amounts of fluids (Vernon et al., 1987). When considered as a whole, the host-rock–garnetite system acted as a fluid pathway throughout metamorphism. The observed fluid-mediated deformation in combination with near-hydrostatic to hydrostatic stresses inferred for garnet growth based on the elastic strain of quartz inclusions in garnet, showcases the capacity of fluids to result in significant compaction at low differential stress.

The mechanisms that operated in the LCU quartzite and garnetite provide new insights into the possible evolution of aqueous-carbonic fluids released by dehydration of oceanic slabs. By dissolving minerals located in specific rock domains through the pressure-solution process documented here, fluids may become enriched in rock-forming components, driving enrichment of silicates in water- and carbon-rich solutions when moving across different lithologies within the subducting plates.

8 CONCLUSIONS

The combination of microstructures, mineral inclusions, and garnet compositions provide a record of metamorphism and fluid–rock interaction for the UHPM garnetite-bearing quartzite of Lago di Cignana. This fluid–rock interaction record showcases the longevity of this system as a major fluid pathway throughout subduction. The externally derived fluids locally led to a significant compaction and mass removal by pressure solution of minerals such as quartz, carbonates, and garnet. Due to its relatively low
solubility, garnet was concentrated into garnetite stylolites, constituting a new potential mechanism for the formation of garnetite. Quartz inclusions entrapped in euhedral garnet overgrowths on garnetite yield strains that indicate near-hydrostatic stress conditions during garnet growth. Considering pressure solution as the main mechanism of mass removal, this fluid–rock interaction also operated as a means of exerting control on deformation during metamorphism.

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Simplified Raman spectra of inclusions and standards, given in cm\(^{-1}\). Fitted peak positions and uncertainties for Raman spectroscopy of inclusions in comparison to an unstrained standard crystal.

Additional supporting information may be found online in the Supporting Information section.

**Table S1:** EPMA measurements in wt% of transect 1L1, and corresponding calculated garnet composition in atoms per formula unit.

**Table S2:** EPMA measurements in wt% of transect 2L1, and corresponding calculated garnet composition in atoms per formula unit.

**Table S3:** EPMA measurements in wt% of transect 3L1, and corresponding calculated garnet composition in atoms per formula unit.

**Table S4:** EPMA measurements in wt% of transect 3L2 and point measurement 3–3, and corresponding calculated garnet composition in atoms per formula unit.

**Table S5:** EPMA measurements in wt% of transect 4L1, and corresponding calculated garnet composition in atoms per formula unit.

**Table S6:** EPMA measurements in wt% of transect 5L1, and corresponding calculated garnet composition in atoms per formula unit.

**Table S7:** EPMA measurements in wt% of transect 6L1, and corresponding calculated garnet composition in atoms per formula unit.

**Table S8:** Fitted peak positions and uncertainties for Raman spectra of inclusions and standards, given in cm\(^{-1}\).

**Table S9:** Shifts in peak positions in Raman spectra of quartz inclusions in comparison to an unstrained standard crystal.

**Table S10:** Calculated strains, estimated standard deviations (esd) and errors for quartz inclusions.
Table S11: $P$–$T$ conditions representing prograde metamorphism of Lago di Cignana, and corresponding strain estimations for quartz inclusions.

Figure S1: BSE images including the locations of all EPMA measurements, of (a, b) representative host-rock garnet, (c) outer garnetite, (d, e) inner garnetite where (e) is a close-up of the marked area in (d), and (f) euhedral overgrowth on the outer margin of the inner garnetite.