Garnet as a monitor for melt–rock interaction: Textural, mineralogical, and compositional evidence of partial melting and melt-driven metasomatism

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Abstract
In this study, we focus on a partially melted garnet-bearing granulite from the Salvador–Esplanade Belt (Salvador da Bahia, Brazil), and examine the behaviour of major and trace elements during partial melting and melt-driven metasomatism. Phase equilibria modelling and U–Th–Pb geochronology show that the sample underwent partial melting during the heating segment of the decompression path from ~1.2 GPa and 675–700°C to ~0.8 GPa and 790°C at ~2.06 Ga. During the final stage of decompression, from 0.8 to ~0.5 GPa, physical segregation of melt resulted in the establishment of chemical potential gradients and mass transfer between the host granulite and the leucosome. Modelling shows that H₂O, CaO, K₂O, and Na₂O diffused from the melt into the host residue, whereas SiO₂ was transferred from the host granulite into the adjacent leucosome. Opposed senses of diffusional transfer resulted in the formation of a quartz-rich anhydrous leucosome and a quartz-depleted selvedge in the host granulite. Compositional maps show that garnet exhibit contrasting major and trace element distributions depending on their textural position. The largest garnet located in the quartz-depleted selvedge preserve their original Ca and trace element growth zoning. A transition from bell-shaped profiles for Y and heavy rare earth elements to bowl-shaped profiles for light rare earth elements is consistent with a typical Rayleigh fractionation model, fast intergranular element mobility, and rock-wide equilibrium during prograde partial melting. In contrast, smaller garnet away from the leucosome show prograde growth zoning modified by intragranular diffusion, as evidenced by a network of open channels and healed cracks that act as connecting pathways between the matrix and garnet core. This results in either subtle modification of both major and trace elements adjacent to the inner core inclusions, or a complete re-equilibration. Recognition that the original Ca growth zoning was later modified by intragranular diffusion implies that misleading thermobarometric results and tectonic interpretations would be obtained if the core composition was used to fingerprint the early garnet nucleation stage. This study demonstrates that at high temperature (>750°C) and in the presence of melt, REE are not less vulnerable to diffusive resetting than divalent cations like Ca²⁺.

KEYWORDS
chemical potential, diffusion metasomatism, garnet zoning, LA-ICP-MS, partial melting
Partial melting is a fundamental process for differentiating (Brown, 2010) and weakening the deep crust (Piazolo et al., 2020; Rosenberg & Handy, 2005), which in turn exerts a first-order control on deformation style, lateral redistribution of mass, crustal thickness, and surface topography (Beaumont et al., 2004; Hollister & Crawford, 1986; Jamieson et al., 2011; Rabin et al., 2015; Royden et al., 1997; Vanderhaeghe, 2012). Exhumed migmatites preserve, through their mineralogy and structure, a wealth of information that can be used to decipher the thermo-mechanical behaviour of orogenic continental crust. With this motivation, the petrological analysis of partially melted felsic rocks is now commonplace in metamorphic petrology, thanks to extended experimental work (e.g. Clemens, 1984; Grant, 1985; Le Breton, 1988; Thompson, 1982; Vielzeuf & Holloway, 1988; Vielzeuf & Montel, 1994) and thermodynamic phase relation modelling (Holland & Powell, 2001; Spear et al., 1999; Vielzeuf & Schmidt, 2001; White & Powell, 2010; White et al., 2001). Despite this progress, modelling phase relations in migmatites is not trivial because they are subject to redistribution and transfer of the highly mobile melt component from grain- to crustal scales (Etheridge et al., 2020; Sawyer, 2001). Therefore, phase relation modelling in migmatites requires the consideration of not only changes in environmental variables like pressure and temperature but also the variation of the equilibrium volume and composition of the system (i.e. open system processes).

Modelling the role of melt transfer (loss and gain) on phase relations has been extensively investigated (Alessio et al., 2017; Bartoli, 2017; Dumond et al., 2015; White et al., 2001; Yakymchuk & Brown, 2014). Among the many benefits of open system forward modelling is the application of a melt-reintegration approach that consists of recovering the bulk composition prior to any melt transfer. This method allows investigating the prograde evolution of melt-bearing systems that tend to be obliterated by continuous re-equilibration up to peak metamorphic conditions. Another complexity arising from the segregation of melt from the solid residue is that the system becomes physically and chemically differentiated into melt-rich domains (leucosome) and solid-rich domains (melanosome). The thermodynamic system is therefore composed of two sub-systems that will interact with changing pressure and temperature conditions, due to the establishment of chemical potential gradients of components at the interface between both sub-systems (White & Powell, 2010). The diffusion of components (H₂O, SiO₂, etc.) between the two sub-systems produces significant variations in composition, mineral reactions, and assemblages (Fitzsimons, 1996; White & Powell, 2010). Despite this conceptual framework being generally accepted, melt-rock interaction in segregated migmatites remains rarely identified via textural, geochemical, and/or mineralogical evidence, nor investigated via thermodynamic modelling (Kriegsman & Hensen, 1998; Lin & Sawyer, 2019; White & Powell, 2010).

In migmatites, garnet is very common as a peritectic phase and has the ability to preserve zoning of divalent cations (Fe, Mg, Ca, and Mn) that reflect changing $P–T$ conditions and/or bulk-rock chemistry, either due to garnet fractionation or mass transfer during melt segregation, transport, and interaction with the garnet-bearing residual rock (e.g. Caddick et al., 2010; Konrad-Schmolke et al., 2008). To retrieve the maximum information related to garnet chemical evolution, the characterization of major element zoning can be complemented with trace elements (Hyppolito et al., 2019; Raimondo et al., 2017). Trivalent cations, like rare earth elements (REEs), have lower diffusion coefficients than major divalent cations. They are therefore more resistant to diffusional modification and allow the preservation of prograde and high-$T$ processes (Carlson, 2012; Gaidies et al., 2020; George & Gaidies, 2017; Moore et al., 2013; Rubatto et al., 2020), including those involved during partial melting (Dumond et al., 2015).

The trace element characteristics of garnet can be linked to the evolution of accessory phases used for geochronology (e.g. monazite, zircon, and rutile) to better constrain tec-tonometamorphic histories (Dumond et al., 2015; Hermann & Rubatto, 2003; Mahan et al., 2006; Prent et al., 2019; Pyle & Spear, 2003a, 2003b; Tomkins & Pattison, 2007; Weller et al., 2020). They also provide insight into the mechanisms controlling the uptake (interface- vs diffusion-controlled growth) and mobility of trace elements during metamorphism and metasomatism (George et al., 2018; Konrad-Schmolke et al., 2008; Moore et al., 2013; Raimondo et al., 2017; Skora et al., 2006). With the recent development of in-situ laser ablation inductively coupled plasma mass spectrometry (LA-ICP-MS), it is now possible to rapidly and quantitatively map the 2D distribution of trace elements across major and accessory minerals of interest (George et al., 2018; Raimondo et al., 2017; Ubide et al., 2015). Yet, despite the known benefits of analysing trace elements in garnet for the purposes listed above, there remains a limited (but increasing) number of studies using this approach to understand the grain-scale controls on garnet crystallization (Gaidies et al., 2020; George et al., 2018; Rubatto et al., 2020).

In this contribution, we report the integrated petrological and geochemical characterization of a high-grade rock that preserves evidence of changes in $P–T$ conditions, partial melting, and melt-driven metasomatism. The selected sample, from the São Francisco craton in Brazil, is a layered composite rock that consists of (a) a garnet-bearing
quartz-saturated host granulite; (b) a quartzofeldspathic leucosome that may correspond to crystallized segregated melt or differentiated melt residue (Guernina & Sawyer, 2003); and (c) a quartz-poor garnet-bearing selvedge formed at the interface between the host granulate and leucosome (Sawyer, 2008). Petrological analysis and thermodynamic modelling (using $P$–$T$–$\mu$–$X$ phase diagrams, where $\mu = \text{chemical potential}$ and $X = \text{composition}$) shows that the distinct mineralogy of the selvedge is the result of mass transfer driven by chemical potential gradients established between the host granulate and the segregated melt during decompression. In addition, garnet show a consistent change in grain size, morphology, major and trace element composition, and zoning patterns across the migmatitic layering, as revealed by quantified LA-ICP-MS raster maps. The strong correlation between textural position and zoning pattern in garnet gives insight into the grain-scale petrogenetic processes involved in partially molten granulites. We distinguish the role of (a) intergranular mobility of elements and accessory phases during metamorphic garnet growth; and (b) post-growth intragranular diffusion of major and trace elements during melt-driven metasomatism.

2  |  GEOLOGICAL SETTING

2.1  |  São Francisco Craton and the Salvador–Esplanade Belt

The São Francisco Craton, located in the east-central part of Brazil, consists of an Archean to Palaeoproterozoic crystalline basement mostly overlain by Meso to Neoproterozoic sediments in its western part (Figure S1). The craton is bounded by Neoproterozoic marginal fold belts (Figure S1). The eastern part of the craton, where most of the metamorphic rocks are exposed, is subdivided into five major geological units: three Archean blocks (Gavião, Jequié, and Serrinha) and two Palaeoproterozoic belts: the Itabuna–Salvador–Curaçá Orogen (ISCO) and the Salvador–Esplanada Belt (SEB; Figure S1; Barbosa & Sabaté, 2004, see references therein). The southernmost part of the SEB, where this study has been conducted, trends broadly NE and consists mostly of mafic to felsic granulites and migmatites.

2.2  |  Farol da Barra: Metamorphism and age constraints

In the Salvador da Bahia city, high-grade metamorphic and magmatic rocks of the SEB are exposed in a horst that is bound on the western side by the Recôncavo sedimentary basin and on the eastern side by the Atlantic coastal margin. The studied area is located at the Barra lighthouse (Farol da Barra; 13°00′41″S, 38°31′57″W; Figure 1a). Four distinct lithologies are identified (de Souza et al., 2010; Souza-Oliveira et al., 2014; Figure 1a): (a) orthopyroxene-bearing granulites that constitute most of the basement of the city of Salvador da Bahia and are interpreted to be derived from late Archean tonalitic magmatic rocks; (b) garnet-bearing granulites and sapphirine-bearing Al–Mg granulites interpreted as meta-sediments; (c) granulitized ultramafic and mafic rocks that occur as centimetre- to metre-scale boudins within the paraderived granulites; and (d) monzonitic to syenitic intrusive dykes, oriented ~N10° (~Figure 1a), that are either deformed or undeformed with sharp intrusive contacts.

Metamorphic conditions have been estimated at 0.75–0.9 GPa and 840–900°C and 0.7–0.75 GPa and 525–550°C on mafic and felsic granulites from the Farol da Barra (Fujimori, 1988). The age of the granulitic metamorphism has been constrained by SHRIMP U–Pb dating at 2,089 ± 11 Ma on zircon overgrowths from the orthopyroxene-bearing granulites (Da Silva et al., 1997). On the same samples, zircon cores yielded an age of 2,561 ± 7 Ma, interpreted as the crystallization age of the protolith magmatic rocks. The later syenitic to monzonitic metasomatism has been dated by Pb–Pb evaporation on zircon at 2,064 ± 6 Ma (Souza et al., 2010).

2.3  |  Farol da barra: Structural relationships

The Barra lighthouse outcrop is characterized by a complex ductile finite strain pattern that results from the superposition of two main phases of deformation ($D_1$ and $D_2$; Figure 1b). The overall geometry consists of metre to decametre lenses, in which $D_1$ structures are best preserved, surrounded by $D_2$ ~ E–W-striking high strain zones (Figure 1b and Figure S2). The first deformation phase ($D_1$) is characterized by a gently dipping gneissic foliation ($S_1$) that strikes NE–SW (Figure 1b and Figure S2). This foliation is associated with isoclinal folds with axial planes parallel to $S_1$ (Figure 1b). The $S_1$ foliation bears a sub-horizontal $L_1$ stretching and mineral lineation plunging 0–20°, marked by the preferred orientation of feldspars, quartz ribbons, mafic minerals, and elongated ultramafic to mafic boudins. The second deformation phase ($D_2$) is characterized by heterogeneous deformation that progressively steepens and transposes the previous $D_1$ fabric along upright $D_2$ folds and steeply dipping high strain zones (>55°) that form an anastomosing network of conjugate shear zones trending approximately E–W (Figure 1b and Figure S2). An $L_2$ lineation is defined by stretched quartz and feldspars, and by
the preferred orientation of biotite with a moderate plunge of 10°–35° towards the NE.

Kinematic analysis in the D₂ low strain domains shows that the two sets of ductile conjugate shear zones consist of a dominant 108/72 SSW dextral shear zone and minor sinistral 021/79 ESE shear zones (Figure S2). This pattern of D₂ finite strain is consistent with NNW–SSE shortening and a bulk dextral sense of shear. The NW–SE cross-section from the Santa Maria Forte towards the Cristo hill (Figure 1b) shows that the studied area (the Barra lighthouse) corresponds to a major localized D₂ high strain zone within a preserved D₁ low strain domain characterized by a gently dipping foliation. However, the D₁ domains (Santa Maria fort, Hispaniola hospital, and Cristo Hill) consist mainly of orthopyroxene-bearing granulites, the Barra lighthouse D₂ high strain zone is characterized by an abundance of garnet-bearing granulite. This apparent relationship between deformation and lithology is beyond the scope of this contribution and will not be discussed further.
3 | TEXTURAL AND MINERALOGICAL CHARACTERISATION

3.1 | Outcrop and sample descriptions

The studied sample is located in an S2 high strain zone that surrounds a large decametric lens of low strain rocks where an earlier fabric (S1) is preserved (Figure 1). The S2 granulite-grade migmatitic foliation is steeply dipping (072/80 S) and contains a gently plunging mineral lineation (09→075) defined by the preferred orientation of biotite and sillimanite. The migmatitic foliation is marked by a compositional banding, with centimetre-wide quartzofeldspathic leucosomes in a garnet-bearing granulite (Figure 2a). Leucosomes are usually less than a 3 cm in width and are systematically surrounded by a centimetre-thick, dark, and coarse-grained quartz-poor selvedge developed at the expense of the surrounding garnet-bearing granulite (Figure 2b).

The studied sample 06SB06 encompasses the three lithologies described above: The host quartz-saturated granulite, a 1 cm wide quartz-poor selvedge and a thin leucosome (Figure 2b). The peak metamorphic assemblage of the quartz-saturated granulite consists of garnet, sillimanite, plagioclase, biotite, quartz, and K-feldspar (Figure 3a). Garnet is interpreted as a peritectic phase produced during biotite dehydration melting via the reaction:

\[
\text{biotite} + \text{sillimanite} + \text{plagioclase} + \text{quartz} + \text{K-feldspar} = \text{garnet} + \text{melt}
\]

Spinell is locally observed in the matrix, but is always surrounded by a thin corona of sillimanite and is never in contact with quartz. The preferred orientation of large sillimanite blades (up to 5 mm), K-feldspar and biotite (which occurs either as small flakes or large resorbed grains) defines the main S2 foliation observed at the outcrop scale. Garnet contains numerous quartz inclusions and polyphase aggregates of plagioclase+quartz, with minor biotite inclusions that all show a preferred orientation at high angle with respect to the S2 foliation (Figure 3a). This alignment is interpreted as a relict of the former shallow-dipping S1 foliation. Peak metamorphic garnet in contact with sillimanite is partially resorbed and surrounded by a moat of retrograde cordierite (Figure 3b), consistent with the classic reaction below that proceeds during decompression and/or cooling:

\[
\text{garnet} + \text{sillimanite} + \text{quartz} = \text{cordierite} + \text{melt}
\]

The selvedge in direct contact with the leucosome is composed of the same peak metamorphic assemblage, consisting of prismatic sillimanite, plagioclase, garnet, biotite, and K-feldspar, except it lacks quartz. Biotite is more abundant than in the quartz-saturated granulite. Garnet cores contain numerous quartz inclusions in association with plagioclase, suggesting that the onset of partial melting and crystallization of peritectic garnet occurred under quartz-saturated conditions. Garnet porphyroclasts are partially broken down into cordierite, and sillimanite is strongly resorbed and replaced by cordierite+spinel symplectites (Figure 3c) via the reaction:

\[
\text{sillimanite} + \text{garnet} + \text{melt} + \text{biotite} = \text{spinel} + \text{cordierite}
\]

The leucosome consists mainly of quartz and thin elongated K-feldspar ribbons. Plagioclase is very rare and occurs as late myrmekite intergrowths next to K-feldspar.

Mineral compositions are available in Data Set S1, and analytical conditions in Appendix S1. Biotite has \(X_{\text{Og}}(=\text{Mg}/\text{Mg}+\text{Fe})\) of 0.67–0.69 and 0.71–0.73 for the small flakes and large grains, respectively, with a near-constant Ti content of 0.25–0.30 pfu. Plagioclase in the matrix has a variable range in anorthite content from 0.43 to 0.52. Spinel is very homogeneous in composition with a \(X_{\text{Mg}}\) of 0.31. Garnet compositions are described in Section 4.
3.2 Evolution of garnet morphology and inclusions

One of the most conspicuous trends perpendicular to the migmatitic layering across the quartz-poor selvedge and the granulite is the evolution of garnet size, composition, and distribution of inclusions. Although no quantitative textural analysis has been conducted, it appears that garnet grain size decreases and the number of garnet grains increases away from the leucosome, with the biggest grains observed in the quartz-poor selvedge (Figures 4 and 5). Furthermore, the amount, shape, and composition of inclusions in garnet varies greatly and continuously across the migmatitic foliation. To describe this evolution, seven garnet (A–G) were selected (see locations in Figure 4).

In the quartz-poor selvedge, most garnet contain a few rounded inclusions of quartz in their core (Garnet A, Figure 5a). Away from the leucosome, but still in the selvedge, the largest garnet porphyroblast (Garnet B) contains in its core a polymineralic inclusion that consists of plagioclase and quartz surrounded by rounded isolated quartz inclusions (Figure 5b). Inclusion-rich garnet cores are surrounded by a large rim containing numerous and homogeneously distributed crystallized granitic melt inclusions that are less than 20 µm in diameter (Figures 5a,b and 6). The inclusions have been identified as crystallized melt based on the main microstructural criteria summarized in Cesare et al. (2015): (a) the development of a perfect negative crystal shape (Figure 6a); (b) a polycrystalline nature that consists of biotite, plagioclase, quartz, and apatite for the most common phases (nanogranitoids) (Figure 6b,c); and (c) the development of cracks (decrepitation) during decompression, filled with minerals similar to those in the melt inclusion (Figure 6). The presence of these crystallized melt inclusions attests to the growth of the garnet rim in the presence of melt.

In the quartz-saturated granulite, the abundance of polymineralic inclusions of plagioclase and quartz observed in garnet increases away from the leucosome (Figures 4 and 5c–g). The garnet core, defined as the domain containing plagioclase+quartz polymineralic inclusions surrounded by isolated quartz grains, can be as large as 1 mm and represent up to 50% of the garnet volume. Polymineralic inclusions have rounded (Figures 5c,d,f) to very irregular shapes with straight boundaries (Figure 5e). These facets are inferred to be parallel to crystallographic orientations of the garnet host and result from post-entrapment readjustment of the inclusion and host by dissolution–reprecipitation processes. Plagioclase in the polymineralic inclusions is chemically zoned, with a composition varying almost continuously from pure anorthite An$_{95}$Ab$_{05}$ to An$_{58}$Ab$_{41}$. The highest amounts of albite are measured in the vicinity of rounded quartz inclusions within plagioclase and the garnet host (Figure S3). All garnet have an inclusion-free rim without any melt inclusions, as for the garnet located in the quartz-poor selvedge (Figure 5). This difference in the ability for garnet rims to entrap melt inclusions may be related to a slowdown of radial growth rate as garnet continuously grows (Parisatto et al., 2018).

**FIGURE 3** (a) Photomicrograph of the metamorphic assemblage in the quartz-saturated granulite. (b) Cordierite development at the contact between garnet and sillimanite in the quartz-saturated granulite. (c) Spinel and cordierite symplectite developed at the expense of garnet and sillimanite in the quartz-poor selvedge.
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X-ray computed microtomography and sequential cuts through a single garnet crystal has been performed to highlight the volume, shape, and potential connectivity of the polyphase inclusions with the matrix. The selected garnet is located in the quartz-saturated granulite where the amount of inclusions is most abundant. The X-ray tomography reveals the very irregular shape of the polyphase inclusions, as well as the ‘spongy’ texture of the garnet core (Figure 7). Although all thin-section photomicrographs of garnet presented in Figure 5 suggest that polyphase inclusions are completely enclosed in the garnet, the 2D cuts rather suggest that they are connected to the matrix through narrow pathways (Figure 7d–f and Movie S1).

3.3 | Microstructures in the quartz-poor selvedge

Microstructures in the quartz-poor selvedge have been revealed using a cold cathode cathodoluminescence detector on an optical microscope. The matrix consists mainly of K-feldspar grains (in blue in the CL images) that form a fine-grained mosaic texture with straight boundaries and triple junctions at 120° (Figure 8a,b). K-feldspar is characterized by a core with fine-grained perthite textures surrounded by a K-feldspar rim with euhedral faces (Figure 8a). These boundaries are coated with thin plagioclase films or elongated grains (brown colour in Figure 8a,b) that can be connected to form a branching array. Terminations of the films and elongated grains are characterized by small dihedral angles (<20°). The connection of these films also leads to the formation of plagioclase pools along grain boundaries (Figure 8a,b) or at K-feldspar triple junctions. Where plagioclase films are abundant, K-feldspar grains show very irregular resorbed grain boundaries (Figure 8b) and plagioclase films are connected to myrmekite pools (Figure 8a,b). Finally, a large plagioclase (±quartz) moat surrounds sillimanite and garnet porphyroblasts (Figure 8b). All of these microstructures are indicative of the former presence of melt or crystallization from melt (see Section 7).

4 | GARNET COMPOSITION

To investigate the link between the textural and mineralogical features of melt–rock interaction and their compositional characteristics, major element data from multiple garnet grains were obtained by EMPA, and trace element data by LA-ICP-MS. X-ray maps and linear spot traverses were acquired to reveal the evolution of major element zoning patterns across the quartz-poor selvedge and granulite. Trace element mapping was performed to better inform the paragenetic context, particularly with regard to accessory minerals, and evaluate the record of geochemical mobility during garnet growth and dissolution. Major and trace element data from linear traverses are provided in Data Sets S1 and S2, respectively, and presented in Figures 9–13. Raw trace element maps from which the traverses were extracted are available as CSV matrices in Data sets S3–S5. LA-ICP-MS analytical methods are provided in Appendix S1.

4.1 | Major element zoning

Garnet are almandine–pyrope–grossular solid solutions with minor amounts of spessartine (<0.015; Data Set S1).
All garnet are zoned and their composition varies with respect to their location and distance from the leucosome. In the quartz-poor selvedge, a polymineralic inclusion-free garnet porphyroblast in direct contact with the leucosome (Garnet A; Figure 5a) is characterized by bell-shaped zoning in grossular, with a maximum content of 0.12 in the core that decreases continuously towards the rims down to 0.03 (Figure 9a). Almandine and pyrope content increases towards the rim from 0.56 to 0.61 and 0.30 to 0.35, respectively, which results in a constant $X_{Mg}$ of 0.34–0.35.

Garnet B has equivalent zoning, except that close to the polymineralic inclusions, grossular content decreases from a 700 µm wide and uniform plateau at 0.12 down to 0.08 (Figure 9b). This decrease in grossular content is mimicked by an increase in $X_{Mg}$ from 0.35 to 0.38. It is noteworthy that the depleted grossular core also corresponds to the domain containing smoothed and rounded inclusions of quartz (light grey area in Figure 9b). The outer rim of Garnet B is characterized by grossular and $X_{Mg}$ values of 0.03 and ~0.35 to 0.36 respectively (Figure 9b). The resorbed outer

**FIGURE 5** Photomicrographs of the seven analysed garnet. (a) Garnet A; (b) Garnet B; (c) Garnet C; (d) Garnet D; (e) Garnet E; (f) Garnet F; (g) Garnet G. The size of the scale bar shown is 50 µm in all cases, and dashed lines correspond to the location of major element profiles shown in Figure 9.
left rim, in contact with cordierite and biotite, shows a sharp decrease in $X_{\text{Mg}}$ down to 0.28 over a distance not greater than 100 µm (Figures 5b and 9b).

Garnet from the quartz-saturated granulite (Garnet C–G) show zoning profiles comparable in shape to Garnet B from the quartz-poor selvedge, except that the end-member modal abundances are different (compare Figure 9b and Figure 9c–g). Garnet cores in direct contact with the polymineralic inclusions are characterized by low grossular values (0.03–0.04) that are similar in composition to garnet rims in contact with the matrix. Grossular content then increases towards the rim to a maximum value that is systematically located at the transition between inclusion-bearing and inclusion-free domains (Figure 9c–g). The maximum grossular content varies from grain to grain, from 0.095 (Garnet E) down to 0.045 (Garnet F). Finally, the inclusion-free rims show a decrease in grossular content towards the matrix to a minimum value of 0.03, equivalent to the core compositions. $X_{\text{Mg}}$ ratio profiles for all samples are relatively flat, with values evolving from 0.36–0.37 (Garnet C) to 0.41–0.44 (Garnet G) with increasing distance from the leucosome. The smallest garnet grain analysed (Garnet F) does not follow this trend, showing a lower $X_{\text{Mg}}$ value of 0.36 and a near-constant grossular content of 0.03–0.04. Garnet E profile shows on its left side a sharp decrease in $X_{\text{Mg}}$ from 0.40 to 0.36, over a distance less than 100 µm, that is interpreted as late Fe–Mg exchange during partial garnet breakdown into cordierite, comparable to Garnet B.

4.2 | Trace element zoning

Similar to their major element characteristics, the analysed garnet show significant variations in trace element composition and zoning patterns depending on their location with respect to the leucosome. We will focus on Garnet A and B, located in the quartz-poor selvedge, and Garnet G, located in the quartz-present host granulite.

Garnet A is located directly adjacent to the leucosome and contains the least inclusions in its core except for a few rounded quartz grains. Figures 10a and 11 and Figure S4 illustrate the key features of the trace element zoning. Y, Zr, V, and heavy rare earth elements (HREEs), from Dy to Lu, show a symmetrical bell-shaped profile without sharp discontinuities except for an abrupt increase at the rim. In contrast, light rare earth elements (LREEs), from La to Gd, show a bowl-shaped zoning profile with a broad flat core that is mimicked by Li, P, and Sc. Notably, the radial positions of broad peaks along the zoning profile are correlated for all REEs and Y (Figure 11), and there is no difference in gradient with decreasing atomic number.

Garnet B is the largest analysed grain, with a small polyphase plagioclase+quartz inclusion in its core. It is located a few millimetres from Garnet A within the quartz-poor selvedge but further displaced from the leucosome. Trace element maps and profiles are shown in Figures 10b, 12, and Figure S4. Its zoning pattern shows the same symmetrical bell-shaped profile for Y, Zr, and the heaviest HREE (from Yb to Er), but with elevated concentrations compared to Garnet A (e.g. Y ~ 360 versus 270 ppm, Yb ~ 75 versus 45 ppm, and Lu ~ 15 versus 6 ppm). In contrast, rim compositions are strictly identical for all elements in both samples (compare Figures 11, 12, and Figure S4). Because Garnet A and B are located in the...
same petrographic setting, we suggest that the difference in absolute concentrations reflects a sectioning effect (i.e. Garnet A is not sectioned through its core) rather than a geological process. The outer rim of Garnet B is characterized by a sharp increase in Y, Zr, and HREEs, and LREEs and P have the same overall bowl-shaped zoning as Garnet A despite a more irregular profile that includes several narrow peaks. Concentrations in La, Ce, and P are significantly enriched next to the polyphase inclusion and the matrix on the left side of the profile (Figure 12). The major difference in comparison to Garnet A corresponds to a broad depletion in Dy, Ho, and Tb adjacent to the polyphase inclusion.

FIGURE 7  (a) Backscattered electron image of a representative garnet in the quartz-saturated granulite. (b) 3D reconstruction from computed X-ray microtomography of garnet. (c–f) sequential slices of the 3D reconstructed garnet (in red) showing the channels (white arrows) connecting the irregular inclusions (transparent) to the outside matrix.

FIGURE 8  (a) Cathodoluminescence images of K-feldspar texture (blue) with plagioclase films and pools (brown) along grain boundaries. Plagioclase films are connected to myrmekite pools (m). (b) Extensive replacement of K-feldspar by plagioclase and myrmekite.

FIGURE 9  Garnet compositional analyses (black square = molar amount of grossular and white square = $X_{Mg}$). The location of each line profile is shown in Figure 5. The dark and light grey stripes correspond to plagioclase+quartz inclusions and domains with minute inclusions of quartz respectively.
FIGURE 10  LA-ICP-MS trace element maps of (a) Garnet A; (b) Garnet B; (c) Garnet G. Concentration values are in ppm. The line profiles shown in Figures 11–13 are located in each case on the phosphorus (P) map.
located at the garnet core. This feature closely matches the grossular zoning profile (Figure 9b), and corresponds to the domain containing rounded inclusions of quartz.

Garnet G is derived from the quartz-present host granulite and is located furthest from the leucosome (Figures 4 and 5). It is the smallest analysed grain and contains a large irregular polyphase inclusion in its core. Trace element maps and profiles are shown in Figures 10c, 13, and Figure S4. Y and HREEs, from Lu to Tb, show a broad bell-shaped profile, but with maximum concentrations on average two to three times lower than the larger Garnet A and B (Figure 13). In contrast, rim compositions for all
elements are once again similar for all samples. La, Ce, and P show bowl-shaped profiles with a large flat core surrounded by an enriched rim. Sm (and to a lesser extent Gd) are no longer characterized by a typical bowl-shaped profile but rather mimic the grossular zoning, with a depleted core next to the polyphase inclusion (Figure 13). This similarity continues along the profile, with an identical location for the maximum Sm concentration and a core composition adjacent to the polyphase inclusion that is equivalent to the rim composition (2–3 ppm on average; Figure 13).

5 | PETROLOGICAL MODELLING

We use computed phase diagram sections to constrain (a) the pressure and temperature of garnet nucleation and growth, partial melting, and retrogression; and (b) the most likely
processes responsible for the formation of the quartz-poor selvedge. For this purpose, the model system \( \text{Na}_2\text{O}–\text{CaO}–\text{K}_2\text{O}–\text{FeO}–\text{MgO}–\text{Al}_2\text{O}_3–\text{SiO}_2–\text{H}_2\text{O} \) was chosen to calculate phase relationships using Perple_X 6.6.6 software (Connolly, 2005), with the thermodynamic database of Holland and Powell (1998), as revised in 2002. Although a bulk-rock TiO\(_2\) content of 0.6 wt% (Table 1) can have a significant effect on biotite modal abundance and the \( P–T \) location of equilibria, TiO\(_2\) was not considered in the modelling. Titanium content in biotite remains constant and ilmenite is the only Ti-bearing oxide present during partial melting, decompression, and subsequent melt-driven metasomatism. Therefore, adding the component TiO\(_2\) to the model should not provide critical extra information. Similarly, we did not consider Fe\(_2\)O\(_3\) because there is no Fe\(^{3+}\)-bearing phase such as hematite or magnetite, and the amount of Fe\(^{3+}\) in garnet based on structural formulae is negligible. Solution models and end-member phases considered in the modelling are listed in Table S1.

**5.1 | \( P–T \) conditions of garnet nucleation and growth**

Deciphering the \( P–T \) conditions of early garnet growth and prograde evolution relies essentially on the observation of garnet inclusions and the valid interpretation of garnet composition and zoning. Garnet cores are characterized by the occurrence of quartz and polyphase plagioclase+quartz inclusions, whereas the inner rim of the largest garnet grains shows abundant crystallized melt inclusions. We therefore suggest that the onset of garnet growth may have occurred under sub-solidus conditions.

Garnet core compositions are typically used to retrieve the approximate \( P–T \) conditions of garnet nucleation. However, we will show later that major and trace element zoning patterns (and more specifically garnet core compositions) have been modified, most likely as a result of post-growth re-equilibration during partial melting and retrogression. The biggest garnet located in the quartz-poor selvedge (Garnet A...
and B; Figures 4 and 5) show the least modified zoning profiles, with typical bell-shaped zoning in grossular (Figure 9a,b) and trace elements (Figures 11 and 12). Although these garnet are located in the quartz-poor selvedge, the presence of quartz inclusions within their cores suggests that the onset of garnet growth occurred under quartz-saturated conditions. We therefore suggest that the loss of SiO$_2$ and the formation of the quartz-poor to quartz-free layer is a post-garnet growth process. Hence, to retrieve the $P$–$T$ conditions of early garnet growth and constrain the prograde path, we have computed a $P$–$T$ phase diagram section with an XRF bulk composition corresponding to the quartz-saturated domain (Table 1 and Figure 14). The phase diagram section has been contoured for grossular content, $X_{Mg} (=\text{MgO}/\text{MgO}+\text{FeO})$ in garnet, and garnet modes (Figure 14).

The amount of H$_2$O is known to be a critical variable when modelling phase relations in melt-bearing systems (e.g. White et al., 2001). A classic strategy to estimate this parameter consists of adding enough H$_2$O to the system such that the solidus is H$_2$O saturated. However, if we follow this approach, high-grossular garnet core compositions cannot be reproduced. Therefore, the system was kept H$_2$O undersaturated, such that measured garnet core compositions (i.e. the highest grossular values) are predictable, and the amount of major rock-forming phases are consistent with observed mineral modes. The amount of H$_2$O was adjusted and fixed at 1.67 mol.% (0.025 mol. or 0.46 wt.%). With such low H$_2$O content, partial melting was restricted to temperatures higher than ~750°C (Figure 14).

The maximum grossular content of ~0.12, coupled with the corresponding $X_{Mg}$ values of 0.35–0.38, allow estimating the minimum $P$–$T$ conditions of garnet growth at ~1.2 GPa and 675°C (black circles with letters in Figure 14). This corresponds to sub-solidus conditions where garnet is in equilibrium with plagioclase, biotite, kyanite, and muscovite. Although these last two phases have never been observed as inclusions in garnet, the predicted plagioclase composition (An$_{76}$-Ab$_{23}$) is consistent with the high anorthite compositions measured in plagioclase inclusions. Plagioclase inclusions are therefore primary and trapped under sub-solidus conditions. Furthermore, melt inclusions are absent in the high grossular cores (Figure 5), whereas they are abundant in the low-grossular rims. This observation is consistent with
early garnet nucleation and growth under sub-solidus conditions, with peak metamorphism and garnet rim crystallization occurring in melt-present conditions.

In Figure 14, we report the composition of Garnet C–G, located in the quartz-saturated domain, with the highest grossular content and corresponding $X_{Mg}$ values (grey circles with letters in Figure 14). The maximum grossular content, preserved in the inner rim at the transition between the inclusion-bearing and inclusion-free domains, varies from 0.095 (Garnet E) to 0.04 (Garnet F) and $X_{Mg}$ from 0.36 (Garnet C) to 0.43 (Garnet G) (Figure 9). The inferred $P$–$T$ conditions vary from $\sim 1.3$ GPa and 720°C to 0.85 GPa and 780°C, corresponding approximately to the $P$–$T$ conditions of garnet nucleation and peak metamorphism/partial melting respectively (see Section 5.2 below). We suggest that these estimates could reflect the progressive and incomplete re-equilibration of garnet during a prograde (heating/decompression) path from $\sim 1.2$ GPa and 675°C down to 0.8 GPa and 790°C. During this $P$–$T$ evolution, minor garnet growth is expected (vol.% = 14–16; Figure 14).

### 5.2 | $P$–$T$ conditions of peak metamorphism, partial melting, and retrogression

The computed $P$–$T$ phase diagram in Figure 14 shows the classic phase relations for partially melted aluminous pelites (e.g. White et al., 2001), with the divariant muscovite-dehydration melting equilibrium at high pressure ($1.2 > P > 0.9$ GPa) and the biotite-dehydration melting equilibrium that produces garnet and cordierite at lower pressure ($0.8 > P > 0.6$ GPa). Between these two narrow divariant fields lies a trivariant assemblage with garnet, biotite, sillimanite, K-feldspar, quartz, and melt in equilibrium, between 770 and 820°C (Figure 14). This calculated assemblage is consistent with the observed assemblage in the quartz-saturated granulite. Thermobarometry performed using garnet rim compositions or garnet core compositions in contact with the polyphase plagioclase+quartz inclusions (~3% grossular and $X_{Mg} = 0.26$–0.44) gives consistent results of $0.8 \pm 0.05$ GPa and 790 ± 20°C (white circles with letters in Figure 14), with the predicted $X_{Mg}$ of biotite (0.70) in good agreement with the measured biotite composition. We interpret these estimates to reflect partial melting during the development of the steeply dipping $S_2$ granulite-grade migmatitic foliation defined by biotite and sillimanite. As discussed in Section 7.2, however, the garnet trace element distribution indicates that core compositions may have been affected by post-growth re-equilibration.

Decompression in the quartz-saturated domain is characterized by the local development of cordierite at the expense of garnet and sillimanite (Figure 4b), through Reaction 1. This local and partial retrogression is also evidenced by the sharp decrease in $X_{Mg}$ from 0.40 to 0.36, at a constant
grossular content of ~0.30 (outer rim of Garnet E; Figure 9e). Figure 14 shows that the development of cordierite and the decrease in $X_{Mg}$, at an almost constant grossular content, would require further isothermal decompression at or below 0.7 GPa in the garnet+cordierite+sillimanite+K-feldspar+plagioclase+quartz+melt stability field.

5.3 | Formation of the quartz-poor to quartz-free selvedge via melt-driven metasomatism

In migmatites, the formation of quartz-undersaturated assemblages may either be inherited from the protolith or driven by partial melting processes. In the latter case, quartz-free assemblages have been interpreted as characteristic of advanced quartz-consuming reactions during partial melting coupled with local melt extraction (Fitzsimons, 1996; Owen, 1991). Alternative models consist of producing the quartz-free selvedge by small-scale silica metasomatism between the host rock and a quartz vein (Penniston-Dorland & Ferry, 2008), or a silica-rich leucosome formed after melt segregation and transport (Dunkley et al., 1999). In the latter case, selvedge formation is a post-anatetic process rather than being directly related to partial melting of the host rock (Lin & Sawyer, 2019). Following the approach of White and Powell (2010), we model and show that element exchange (diffusion metasomatism) between the segregated melt (leucosome) and the host granulite is likely to be responsible for the formation of the quartz-poor to quartz-free selvedge during post-peak decompression.

Our conceptual model is similar to Lin and Sawyer (2019) and requires a first stage of partial melting and melt segregation to produce two subsystems, corresponding to the quartz-saturated domain (~residue) and the melt domain. At the $P-T$ conditions of partial melting and melt segregation (i.e. 0.8 GPa and 790°C), chemical potentials are the same in the solid phases and the melt of both domains, if thermodynamic equilibrium is maintained. The second stage corresponds to decompression into the cordierite-bearing stability field below 0.7 GPa. As pressure decreases, chemical potentials evolve independently in both subsystems. Therefore, the diffusion of components is driven by the chemical potential gradient established at the interface between the quartz-saturated residue and the leucosome.
In order to determine the direction of diffusion, we have computed the chemical potentials of H\textsubscript{2}O, SiO\textsubscript{2}, CaO, and K\textsubscript{2}O for the quartz-saturated and melt domains along an isothermal decompression path from 0.8 to 0.5 GPa at 790°C (Figure 15). Values for the melt domain were determined using the chemical composition of melt produced at 0.8 GPa and 790°C (Table 1). Chemical potentials of the selected components were computed separately for each sub-system (segregated melt and residue) by assuming that during decompression these two sub-systems are closed to mass transfer and chemically isolated from each other (Figure 15). This ideal model is in a strict sense impossible, because the two sub-systems are physically in contact and therefore chemical interactions are expected at some scale, and chemical interactions
potential will continuously evolve with ongoing decompression due to diffusive mass transfer (White & Powell, 2010). However, our approach can be used as a first-order constraint to determine the direction of transport of the four selected species if chemical interaction at the interface is permitted. During decompression, the chemical potentials of H$_2$O, CaO, K$_2$O, and Na$_2$O (Na$_2$O not shown in Figure 15) become higher in the melt segregated domain (leucosome) than in the residue. In contrast, the chemical potential of SiO$_2$ is lower in the melt than in the residue. This suggests that species such as H$_2$O, CaO, K$_2$O, and Na$_2$O will diffuse from the melt into the host residue whereas SiO$_2$ is transferred from the host residue into the adjacent melt domain.

To model the effect of metasomatism and pressure on phase relations, a P–X phase diagram section was computed at 790°C (Figure 16). The initial amounts of CaO and Na$_2$O are very low (<1.3 and 0.6 wt% respectively) and do not significantly influence the phase relations. Therefore, variations in Na$_2$O and CaO content are not considered further. The bulk composition for X = 0 corresponds the quartz-saturated host rock composition prior to any mass transfer. The bulk composition for X = 1 is obtained assuming a gain
FIGURE 16  $P$–$X$ phase diagram section computed at 790°C with $X = 0$ corresponding to the quartz-saturated domain and $X = 1$ corresponding to a 55% loss of SiO$_2$ and 200% gain of H$_2$O. See Table 1 for compositions.

FIGURE 17  $P$–$T$ phase diagram section computed for a modelled quartz-undersaturated selvedge composition produced after a 47% loss of SiO$_2$ and 140% gain of H$_2$O. See Table 1 for compositions.
in H₂O of 200% (H₂O = 0.075 mol) and a SiO₂ loss of 55% (SiO₂ = 0.342 mol); these values are arbitrary. The bulk compositions used are provided in Table 1.

Figure 16 shows the location of the two main low-variance (V = 2) decompression equilibria that have been observed in the quartz-saturated domain and quartz-absent selvedge: Grt+SiO₂±Bt=Crd+L (Reaction 2) and Grt+SiO₂±Bt=Crd+Sp+L (Reaction 3) at 0.72 and 0.55 GPa respectively. These divariant equilibria together with their related trivariant assemblages are used to locate the quartz-out, cordierite-in and spinel-in boundaries (Figure 16). The formation of the selvedge assemblage (Crd+SiO₂+L±Grt/Bt) from the quartz-saturated host rock requires a bulk composition with X ranging from ~0.65 to 0.8 for a pressure lower than 0.7 GPa. The formation of the quartz-absent spinel-bearing symplectite (Figure 3c) would require further decompression down to 0.55 GPa. Taking a value of X = 0.7, mass transfer required to produce the selvedge from the host quartz-saturated granulite corresponds to a relative H₂O gain of ~140% coupled with a relative SiO₂ loss of 47%. The P–X phase diagram is also contoured for garnet mode (Figure 16). As expected, decompression is associated with garnet breakdown, but at pressures above the cordierite-in boundary (P > 0.72 GPa), mass transfer (SiO₂ loss coupled with H₂O gain) induces a slight increase in garnet mode, from 12 to 17 vol.%. This mineralogical evolution is consistent with the observation of a higher modal abundance of garnet in the metasomatic selvedge with respect to the host granulite.

Based on the estimation of mass transfer required to produce the quartz-depleted to undersaturated selvedge, a P–T phase diagram section has been computed with an estimated selvedge composition (Figure 17 and Table 1). This composition corresponds to X = 0.70 in Figure 16. Figure 17 shows the location in P–T space of the two main low-variance retrogression reactions 2 and 3 and the position of the quartz-out, cordierite-in and spinel-in lines. Once again, the selvedge formation is consistent with decompression from ~0.8 GPa down to 0.5 GPa at temperatures greater than 750°C.

6 | U–Th–Pb monazite and zircon geochronology

Monazite and zircon geochronology were acquired to establish a temporal framework for melt–rock interaction and the relationship of garnet growth and dissolution to the accessory mineral evolution. Chemical zoning in monazite and zircon were first characterized via X-ray mapping and cathodoluminescence respectively. In situ U–Th–Pb monazite and zircon ages were then acquired by LA-ICP-MS. Sample preparation and operational procedures for all techniques are described in Hurai et al. (2010) and Paquette et al. (2014) and presented in Appendix S1. U–Th–Pb geochronological data are provided in Data Set S6.

6.1 | Textural features and dates

Monazite is abundant in both quartz-saturated and quartz-poor domains but is totally absent in the leucosome. The analysed monazites (Figure 18) range in size from 50 to 250 μm and occur in the matrix, at grain boundaries and included in sillimanite and plagioclase (M1-2-13), in the vicinity of garnet, and within the decompressive cordierite moat (M3-6-4-8-9-10-11-12). Monazite grains M3, M6, and M12, located in cordierite moats, have an unusual worm-like shape that mimics the embayed garnet rim (Figure 18c). Only one grain (M7) was found included in a garnet rim.

All monazite grains are chemically homogeneous and Y-poor (Figure 18b,c), except for two grains (M9 and M10) that show subtle zoning with high Th domains. Grains located in the cordierite moat show discrete and discontinuous microscopic Y-rich rims (Figure 18c), whereas those included in other phases are devoid of such overgrowths (Figure 18b). These observations suggest that the crystallization of the low Y grains might precede or be coeval with the peak assemblage of garnet (rim), sillimanite, biotite, plagioclase, K-feldspar-quartz. The crystallization of discrete Y-rich monazite overgrowths in the vicinity of garnet porphyroblasts partially retrogressed into cordierite has been previously interpreted as an indicator of accessory phase growth during garnet breakdown (Mahan et al., 2006). We suggest that Y overgrowths are also contemporaneous with cordierite crystallization and selvedge formation.

Nine homogeneous Y-poor grains were analysed for a total of 37 analyses (Data Set S6). The Y-rich overgrowths were not analysed due to their small size, in order to avoid mixing during ablation. In a 206Pb/238U versus 208Pb/232Th diagram, all analyses are concordant or sub-concordant (Figure 19a). Excluding six analyses (see Data Set S6) gives a concordia age of 2,061 ± 6 Ma with an MSWD (C + E) of 1.3. The weighted mean of 208Pb/232Th dates gives an age of 2,069 ± 8 Ma with an MSWD of 1.6 (Figure 19b). The 208Pb/232Th dates show no systematic variation with textural location (see Figure 18a and Data Set S6), and more specifically, monazite included in garnet (M7) is not statistically different from matrix grains (Figure 19). The age of 2.06 Ga is interpreted as the timing of partial melting at 0.8 ± 0.05 GPa and 790 ± 20°C, before decompression responsible for partial garnet breakdown, crystallization of small Y-rich overgrowths, and selvedge formation.

Zircons are rare and do not exceed 50 μm in diameter. They are translucent with a rounded shape typical of zircons recrystallized under granulite facies conditions. Fourteen analyses were performed on 14 grains (Data Set S6). They do not show significant zoning in cathodoluminescence. A first group of
seven analyses are concordant and yield a concordia age of 2,073 ± 6 Ma with an MSWD (C + E) of 0.84 (Figure 19c). A second group of four analyses are discordant, and when plotted together with the previous seven analyses, define a discordia with an upper intercept of 2,073 ± 6 Ma (MSWD = 1.3). The final group consists of four analyses that do not match the previous two identified clusters (white ellipses in Figure 19), with 207Pb/206Pb dates that range from 2.07 to 2.33 Ga and one concordant analysis at 2,192 ± 15 Ma. The 2.07 Ga date is similar to that obtained for monazite and is interpreted as the timing of partial melting. The remaining three analyses are interpreted as inherited ages.

7 | DISCUSSION

Petrological and microstructural analysis carried out across the interface between the leucosome and the host granulite shows a systematic evolution in mineral paragenesis, microstructures, garnet grain size, and major/trace element composition with distance from the leucosome. The studied sample is therefore used to discuss small-scale melt–rock interaction processes. In this section, we will first summarize the petrological model of migmatite formation, and more specifically the quartz-depleted selvedge at the interface between the segregated melt (leucosome) and the partially melted host granulite. Based on this petrological evolution, we will discuss the significance of major and trace element zonation observed in garnet.

7.1 | Petrological model of migmatite and quartz-depleted selvedge formation through diffusion metasomatism

The phase relation modelling performed in this study suggests that the onset of garnet crystallization occurred under sub-solidus conditions at high pressure (P ~ 1.2 GPa) and temperatures of 675–700°C. These P–T conditions, estimated using the maximum grossular content of garnet cores and anorthite content of plagioclase inclusions, are consistent with a steady state geotherm considering average crustal thermal parameters (thermal conductivity k = 3 W.m⁻¹ K⁻¹, heat production A = 0.75 µW/m³, mantle heat flow Q = 30 Mw⁻², and density ρ = 2.7 kg/m³) at a depth of 45 km (1.2 GPa). We suggest that the gently dipping gneissic foliation (S₁) that trends NE–SW (Figure 2b and Figure S1) may have formed at these conditions and correspond to a crustal thickening event. This interpretation is also supported by the observation of quartz and plagioclase inclusion trails defining an S₁ foliation in the high-grossular garnet cores (Figure 3a).
Partial melting occurred at 0.8 GPa and 790°C at c. 2.07–2.06 Ga. The former presence of melt at peak metamorphic conditions can reliably be inferred from the occurrence of peritectic garnet, produced by biotite-dehydration melting (Reaction 1; Figures 4 and 5) and the presence of crystallized melt inclusions within the garnet (Figure 6). Interstitial elongated films along grain boundaries and pools at triple junctions (Figure 8) are alternative criteria diagnostic of the former presence of melt and melt movement at the grain scale (Holness & Sawyer, 2008; Sawyer, 2001; Vernon, 2011). The interstitial plagioclase may represent the plagioclase component crystallized from the melt (Hasalová et al., 2008; Stuart et al., 2018). The resorbed K-feldspar grains and the crystallization of plagioclase and myrmekite (Figure 8) are interpreted as the result of K-feldspar dissolution during the interaction between the host rock and a migrating melt (Hasalová et al., 2008). We suggest that the connection between the leucosome and the host granulate via the grain boundary melt array favoured efficient small-scale mass transfer in and out of the host granulate to produce the quartz-depleted to undersaturated selvedge.

Thermodynamic modelling shows that decompression from 0.8 to 0.5 GPa at 790°C coupled with melt segregation are responsible for the establishment of chemical potential gradients between the leucosome and the host granulite. Chemical potential gradients drive diffusive metasomatism of species between the two reservoirs and the formation of the quartz-undersaturated selvedge at the interface. The modelling suggests that during isothermal decompression from 0.8 to ~0.5 GPa at 790°C, species like H₂O, CaO, and Na₂O will be transferred from the leucosome to the host rock, whereas SiO₂ has an opposite diffusion direction from the host rock to the leucosome. Phase diagrams in Figures 16 and 17 show that mass transfer coupled with decompression resulted in the partial transformation of the quartz-saturated peak metamorphic assemblages (Grt, Bt, Sil, Qz+L) into a quartz-free assemblage with cordierite and spinel (see location of Qz-out, Crd-in, and Spl in Figure 16). Formation of the quartz-depleted selvedge via melt-driven metasomatism
(i.e. SiO$_2$ loss) is well supported by the rare occurrence of quartz relics in the matrix and the observation of quartz mostly as inclusions in garnet cores.

Several other petrological and microstructural observations provide support for the modelled mass transfer direction and the formation of the selvedge:

1. The quartz-depleted selvedge, where local mass transfer is inferred as the dominant process, is characterized by larger garnet grain sizes. This is consistent with modelled garnet modes in Figure 16, which show an isobaric and isothermal increase in garnet abundance at 0.8 GPa due solely to mass transfer (SiO$_2$ gain and H$_2$O loss).

2. Diffusion of H$_2$O from the melt into the host granulite will favour the crystallization of anhydrous products in the melt, producing almost an anhydrous leucosome, whereas the adjacent host granulite is hydrated, as demonstrated by the slight enrichment of the selvedge in hydrous phases like biotite and cordierite. This observation is consistent with results from White and Powell (2010), who predicted that the diffusion of H$_2$O into the melt will be responsible for the formation of anhydrous leucosomes during isobaric cooling under granulite facies conditions.

3. Diffusion of other mobile species like CaO, Na$_2$O, and K$_2$O from the leucosome into the selvedge is indirectly supported by the mineralogical composition of the leucosome, which consists mostly of quartz with very minor K-feldspar and almost no plagioclase. It is well known that leucosomes in migmatites do not have the expected minimum granitic composition (Ashworth & McLellan, 1985) due to post-anatectic magmatic differentiation processes. However, following Olsen and Grant (1991) who performed mass-balance (iscon) analysis on leucosomes, we suggest that in our samples the quartz-rich and K-feldspar and plagioclase-poor leucosomes most likely formed by loss of Na$_2$O and CaO by diffusion.

4. Diffusion of K$_2$O into the host rock is likely to be responsible for the formation of the distinct inclusion-free outer rim of K-feldspar located in the selvedge (Figure 8).

5. The introduction of an Na$_2$O/CaO-rich fluid or melt into the selvedge is consistent with the observation of plagioclase films and pools along K-feldspar grain boundaries (Figure 8). The formation of myrmekite in the selvedge is also consistent with fluid- or melt-mediated Na–Ca replacement of K-feldspar by plagioclase (Yuguchi & Nishiyama, 2008).

The possibility that the migmatitic layering (and more specifically the quartz-depleted selvedges around leucosomes) is inherited from the sedimentary protolith is rather unlikely. In addition to the observations and modelling discussed above, field observations from the Farol da Barra outcrop show that quartz-depleted selvedges are present only at the margins of leucosomes. These features must therefore have formed after melt segregation and hence after partial melting.

Melt segregation and the development of the selvedge defines a steeply dipping S$_1$ migmatitic layering that transposes the previous S$_1$ fabric (Figure 3 and Figure S1). Kinematic analysis of the D$_2$ high strain zone is consistent with NNW–SSE shortening and a bulk dextral sense of shear. We conclude that melt transfer and metasomatism may have been enhanced by D2 transpressive deformation.

### 7.2 Major and trace element behaviour during partial melting and retrograde melt–rock interaction

The studied sample preserves mineralogical and textural evidence of prograde partial melting and melt segregation, near-isothermal decompression and melt-driven metasomatism. Compared to other minerals, garnet has the tremendous advantage to preserve records of these various processes via its chemical zoning in major and trace elements, because of the slow rates of intracrystalline diffusion (e.g. Carlson, 2006). However, in order to interpret accurately these zoning patterns in terms of large-scale and long-term geological/geodynamic processes, we have to understand the small-scale processes that control the behaviour of major and trace elements during garnet nucleation, growth, breakdown, and re-equilibration (George & Gaidies, 2017 and references therein). Although the zoning features we document vary from garnet to garnet, they show systematic variations and similarities that can be correlated with the $P$–$T$–$X$ evolution and petrographic setting. Via a qualitative analysis of zoning profiles, and with reference to previous work by others cited below, we suggest these features reflect the superposition of at least three grain-scale processes that control the intra- and intergranular mobility of chemical components (major and trace elements).

#### 7.2.1 Evidence of fast intergranular mobility of elements, including REE, and rock-wide equilibrium

The $P$–$T$ evolution of the studied sample occurred at high temperature (>650°C) and in the presence of melt, from peak-temperature conditions (0.8 GPa; 790°C) to the conditions of selvedge formation (~0.5 GPa; 800°C). Therefore, the high mobility of major elements and to a lesser extent trace elements is expected. We report below evidence of fast intergranular mobility of elements, including REE, and rock-wide equilibrium.

Rock-wide equilibrium and by extension fast intergranular transport of major and trace elements during peak
metamorphic conditions and subsequent melt-driven metasomatism is best demonstrated using garnet rim compositions. Although the analysed garnet are located in different petrographic settings (within the quartz-depleted selvage or in the host granulite displaced from the leucosome) and vary markedly in grain size and morphology, their outermost rims share consistent major element (grossular content = 3%; Figure 9) and trace element compositions (Y and REEs; Figures 11–13). This consistency in major element composition of the rims permits an accurate estimation of peak metamorphic conditions at 0.8 GPa and 790°C (Figure 14).

The length scale of equilibration during garnet growth, which is related to the degree of mobility of major and trace elements through the intergranular matrix, can be evaluated using the REE zoning pattern (Moore et al., 2013; Skora et al., 2006). In both studies, these authors have shown a dependency of REE distribution on atomic number in large and early nucleating garnet. Such garnet are characterized by pronounced central peaks for Y and HREEs (Lu to Er), central peaks and annular maxima for MREEs (Ho, Dy), and well-defined annular maxima for LREEs (Tb to Nd). This distribution is driven by diffusion-limited uptake that evolves during garnet growth. With increasing temperature, intergranular diffusivities increase progressively as garnet grows, but with more rapid diffusion for HREEs compared to LREEs (Moore et al., 2013; Skora et al., 2006). However, this dependency of REE distribution with atomic number is less valid at high temperature. Numerical modelling of garnet zoning along a temperature–time path performed by Moore et al. (2013) demonstrates that a uniform bell-shaped distribution for all REEs is consistent with garnet nucleation and growth at high temperature, whereas at lower temperature a sharp central peak with an annular maximum would be expected (see Figure 19 and figure 20b in Moore et al., 2013). From these previous studies, we can conclude that a uniform distribution for all REEs is the result of rapid intergranular diffusion that strongly minimizes the development of concentration gradients in the matrix surrounding the growing garnet.

All studied garnet are characterized by a bell-shaped distribution for Y and REEs, ranging from Lu to Dy (±Tb). The radial positions of peaks are identical for all elements. This distribution is consistent with a progressive depletion of the matrix in Y and REEs (Lu to Dy) due to Rayleigh fractionation. The uniform bell-shaped distribution for REEs, ranging from Lu to Dy, is consistent with rapid intergranular diffusivities. The inferred P–T path in Figure 14 suggests that garnet growth occurred with mineral assemblages evolving from Bt–Kfs–Ms–Grt–Ky–Pl–Qz to Bt–L–Gt–Kfs–Sil–Pl–Qz between 675 and ~800°C. In contrast to Moore et al. (2013) and Otamendi et al. (2002), we conclude that in our specific case, the changing mineral assemblage had no significant effect on the REE distribution.

All garnet grains are characterized by (a) the absence of progressive outwards migration of REE peak locations with decreasing atomic number; and (b) the lack of a central peak for LREEs between La and Gd. Instead, they show a large LREE-depleted core of constant composition surrounded by an enriched rim. This transition from bell- to bowl-shaped profiles with decreasing atomic number is similar to an example reported by Raimondo et al. (2017) from a metasomatic shear zone in central Australia. Both observations (a) and (b) are once again indicative of fast intergranular mobility and rock-wide equilibrium during garnet growth, without significant differences in diffusivity between HREEs and LREEs. However, we argue below that the bowl-shaped profile could also be related to (or at least accentuated by) post-growth modification.

Following Raimondo et al. (2017) and Konrad-Schmolke, Zack, et al. (2008), we suggest that the REE zoning reflects a typical Rayleigh fractionation model. As garnet nucleates and grows at high temperature (T > 750°C), the matrix is progressively depleted in HREEs on a large scale due to the presence of melt along grain boundaries that favour fast element mobility and rock-wide equilibrium. Simultaneously, the matrix is passively enriched in LREE on a rock-wide scale due to the low garnet/matrix partition coefficients for LREEs (Moore et al., 2013). The rapid intergranular mobility for all REEs with respect to garnet growth, without any dependence on atomic number, precludes the establishment of diffusion gradients in the matrix, resulting in smooth core-to-rim zoning and a transition from bell-shaped HREE to bowl-shaped LREE profiles.

Small garnet located in the quartz-saturated host granulite (e.g., Garnet G) are characterized by a significantly HREE-depleted composition compared to large garnet located in the quartz-depleted selvage adjacent to the leucosome (Garnet A and B), whereas their LREE content remains similar. This discrepancy in REE concentrations could be related to the later nucleation of garnet located in the quartz-present host granulite. Assuming a rapid intergranular mobility and rock-wide equilibrium for REEs, the early nucleation and enhanced growth of garnet in proximity to the leucosome may have induced a bulk-rock HREE depletion at the scale of the distance between Garnet A and Garnet G (~7 cm). In contrast, the small garnet/matrix LREE partitioning coefficient had a negligible effect on the bulk-rock composition, resulting in an approximately constant LREE concentration for garnet regardless of its nucleation and growth history.

### 7.2.2 Post-growth intragranular diffusion of major and trace elements

Calcium maps and profiles from Garnet B–G reveal that their prograde growth zoning is modified near the inner core inclusions. This post-growth modification resulted in chemical
re-equilibration of the grossular content to varying degrees, depending on the location of the garnet with respect to the quartz-depleted selvedge and the host granulite. In the quartz-saturated host granulite, the smallest garnet (which also contain the biggest polyphase inclusions) show a complete (Garnet F) or partial re-equilibration (Garnet C, D, E, and G) of the garnet core in contact with the inclusions, with grossular content decreasing to 3%, corresponding to the rim composition. This suggests that post-growth modification occurred near peak P–T conditions (0.8 GPa; 790°C). Located in the quartz-depleted selvedge, the largest analysed grain (Garnet B) also shows post-growth modification of bell-shaped HREE zoning, but chemical re-equilibration at peak-metamorphic conditions was only partial, with a minimum grossular content of 8% in the garnet core.

The modification of garnet cores in contact with polyphase inclusions and their strong similarity with matrix compositions requires a pervasive connectivity to permit such rock-wide re-equilibration to proceed. Both X-ray tomography (Figure 7) and LA-ICP-MS mapping (e.g. linear features highlighted in Li map; Figure 10) reveal the presence of a network of open channels and re-healed cracks that may have acted as potential connecting pathways. These features confirm that the garnet core was not isolated or armoured from the matrix, but was rather freely accessible to the extent that fluid access along these discrete open fractures or larger conduits (Figure 7) may favour cation transport and garnet re-equilibration.

Furthermore, we suggest that REEs are also susceptible to the post-growth re-equilibration process. In contact with the polyphase inclusions in Garnet B, the bell-shaped HREE profile is modified with a decrease in Ho, Dy, and Tb that correlates with the Ca zoning (Figure 12). The remaining HREEs are apparently unaffected by this modification, whereas the modification of LREEs such as Sm, Gd, La, and Ce is more challenging to interpret. This is largely because the bowl-shaped profile for Sm and Gd, as described above, could also be enhanced by a depletion in LREEs next to the inner core inclusions. However, Grain G does show a pronounced depletion in LREEs at the garnet core, resulting in zoning that closely mimics the Ca distribution (compare Figures 9g and 13). Unlike Grain B, the absence of significant depletion in Ho, Dy, and Tb, coupled with its lower HREE content and well-preserved bell-shaped HREE profile, may support the interpretation that Grain G represents a late nucleating garnet in the host granulite at peak metamorphic conditions. Post-growth modification proceeded at a lower temperature where only LREE remained mobile, in the presence of F, Cl, and/or P complexes (Ague, 2017; Migdisov et al., 2009; Williams-Jones et al., 2012). Alternatively, it may suggest that selective REE mobility is coupled to Ca substitution via retrograde garnet–plagioclase exchange that favours LREE over HREE according to kinetic controls and element compatibility.

### 7.2.3 | Role of changes in accessory phases and major rock-forming minerals

In the studied sample, the LREE+P whole-rock budget is largely controlled by the stability of monazite. As such, an approximately constant LREE concentration for garnet whatever its nucleation and growth history, coupled with a smooth bowl-shaped LREE+P profile prior to post-growth modification (i.e. Garnet A; Figure 13), suggests that monazite did not grow on the prograde path. Bulk-rock fractionation of this kind is incompatible with garnet–monazite partitioning relationships if their growth was coeval (Warren et al., 2019). Rather, the apparent lack of monazite competition for LREE+P throughout the garnet growth history necessitates that monazite first appeared at peak conditions, consistent with the U–Pb age interpretation documented in Section 6.1 and further explained below.

Superimposed on the bowl-shaped LREE profile, the rims of Garnet B and G show a sharp coincident enrichment in Ce, La, and P where garnet is in contact with the matrix assemblage consisting of plagioclase and biotite. The same sharp enrichment is observed next to polyphase plagioclase and quartz inclusions located in garnet cores. There are two possibilities for explaining these features. First, we suggest that they may reflect modification of garnet zoning during post-peak breakdown of monazite. This interpretation is consistent with the pervasive occurrence of thin Y-rich overgrowths on monazite located in the matrix (Figure 18c), which may represent dissolution–precipitation events during the retrograde evolution. Second, plagioclase breakdown may also contribute to the LREE enrichment since garnet rims are locally enriched in large ion lithophile elements such as Ba and Sr, further suggesting that post-peak garnet–plagioclase exchange is responsible for the modified grossular zoning and that monazite ages reflect the timing of partial melting prior to breakdown on the retrograde path.

The outermost rims of garnet are characterized by a sharp increase in Y and HREEs along grain boundaries (Figures 10–13). This local enrichment is interpreted to be the result of partial garnet breakdown during decompression via Reactions 2 and 3, causing back-diffusion of elements that strongly partition into garnet with respect to the matrix. Notably, the absence of sharp HREE and Y enrichment in core domains, despite an apparently pervasive matrix connectivity during post-growth re-equilibration and simultaneous monazite replacement, confirms that garnet resorption must have post-dated these events.

Similar to the HREEs, Zr zoning is radial in all garnet except for sharp spikes adjacent to grain boundaries (Figure S4). There are no satellite peaks or annular maxima indicative of zircon breakdown, nor are there any pronounced depletions indicative of sudden zircon crystallization. This suggests that zircon grew on the prograde P–T path, with the bulk-rock
Zr budget progressively depleted as zircon and garnet grew under rock-wide equilibrium. The distribution of U–Pb dates supports this interpretation, with concordant zircon analyses at 2.07 Ga being similar to the 2.06 Ga monazite age inferred to represent peak metamorphism and partial melting. Rimward spikes in Zr are coincident with the HREE and Y enrichments noted above and slope away from the grain boundary, thus most likely due to garnet resorption rather than zircon breakdown.

8 | CONCLUSIONS

The petrological and geochemical evidence for melt-driven metasomatism documented in this study highlights the complexity of processes involved during partial melting, at scales ranging from a few microns to centimetres. We demonstrate that partially melted rocks are open systems in which changes in P–T conditions, deformation and mass transfer are preserved by mineral assemblages and their major/trace element distributions revealed by X-ray microprobe and LA-ICP-MS mapping respectively. One of the major challenges is to be able to distinguish the mineralogical records of metasomatism from those related to changes in P–T conditions.

A key outcome of this study is that melt segregation and extraction, coupled with changes in P–T conditions (due to decompression in this example), induce local disequilibrium between the segregated melt and the residual host rock. The establishment of chemical potential gradients at the interface of these two sub-systems drives diffusive mass transfer. Our thermodynamic modelling shows that while H₂O, Na₂O, CaO, and K₂O are transferred from the melt to the host rock, SiO₂ diffuses in the opposite direction. This combined mass transfer results in the formation of a quartz-rich and almost anhydrous leucosome and a quartz-depleted selvedge at the expense of the host granulate. Depending on the initial bulk-rock composition, P–T conditions of melt extraction and shape of the P–T path, the diffusive behaviour of elements in or out of the melt domain and residue will vary. We suggest that the variety of mineral compositions encountered in selvedges surrounding leucosomes in migmatites relates directly to these environmental variables and the induced melt-driven metasomatism.

Garnet shows complex major and trace element distributions that systematically vary with petrographic setting. We suggest that preserved chemical zoning reflects the superposition of changes in P–T conditions and melt-driven metasomatism. Major and trace element growth zoning was first acquired during prograde to peak metamorphism from 675 to 790°C. Garnet grew mostly as a peritectic phase via biotite-dehydration melting reactions. Calcium, Y, and HREEs (Lu to Dy), are characterized by a bell-shaped distribution, and LREEs (La to Gd) show a bowl-shaped pattern.

The chemical zoning is consistent with a typical Rayleigh fractionation model, with fast intergranular element mobility of all elements, including REEs, and rock-wide equilibrium. The fast mobility is enhanced by the presence of melt along grain boundaries.

Post-peak melt-driven metasomatism is responsible for modification of the prograde garnet growth zoning. Both Ca and REEs are susceptible to this process, resulting in partial or complete re-equilibration through diffusion at the P–T–X conditions of metasomatism (isothermal decompression from 0.8 to 0.5 GPa at 790°C). Garnet cores in contact with polyphase inclusions are affected by post-growth modification due to the presence of a network of open channels and cracks that act as connecting pathways between the matrix and garnet core. Once again, at these high-T conditions and in the presence of melt (and fluid), divalent cations (Ca²⁺) and REEs are characterized by fast inter- and intra-granular element mobility and rock-wide equilibrium. We conclude that in this peculiar case (high temperature and melt/fluid-bearing open system), REEs are not less vulnerable to diffusive resetting than divalent cations.

Besides our interpretations regarding the formation of the quartz-depleted selvedge and the mobility of major and trace elements at high-T conditions, this contribution shows that an inaccurate interpretation of chemical zoning in garnet could result in misleading thermobarometric results. Indeed, if the modified garnet core compositions (i.e. low Ca contents) were used as the chemical fingerprint of the early garnet nucleation stage, an erroneous low-P estimate would have resulted.

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**SUPPORTING INFORMATION**

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

**Figure S1.** Simplified geological map of the São Francisco Craton modified after Barbosa et al. (2003).

**Figure S2.** (a) Simplified structural map of the southwestern part of the city of Salvador da Bahia. (b) Simplified structural cross-section. (c) Equal-area Schmidt lower hemisphere projection of poles to S1 and S2 foliations (dashed lines = conjugated shear zones associated the S2 mylonitic foliation).

**Figure S3.** (a) Backscattered electron image of a polyphase plagioclase+quartz inclusion in garnet. (b) Anorthite versus albite content in moles of plagioclase. Grey and black diamonds are plagioclase compositions in contact with quartz and garnet respectively. (c) Line profile showing the evolution of plagioclase composition in contact with quartz and garnet. See location of the profile in (a).
Figure S4. LA-ICP-MS Zr line profiles of Garnet A, B, and G.

Table S1. Solid solution models used in Figures 14–17.

Data Set S1. Electron microprobe analyses of biotite, plagioclase, spinel, and garnet. Garnet compositions include the seven profiles collected from Garnet A to G in Figure 9 (see Figure 5 for line profile locations).

Data Set S2. LA-ICP-MS trace element data of line profiles collected from Garnet A, B, and G (see Figures 11–13).

Data Set S3. Raw LA-ICP-MS trace element data of Garnet A (see maps in Figure 10a).

Data Set S4. Raw LA-ICP-MS trace element data of Garnet B (see maps in Figure 10b).

Data Set S5. Raw LA-ICP-MS trace element data of Garnet G (see maps in Figure 10c).

Data Set S6. LA-ICP-MS isotopic U-Th–Pb data of monazite and zircon.

Movie S1. 3D reconstruction from computed X-ray microtomography of garnet (see Figure 7).

Appendix S1. Analytical methods.

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