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# Distinct metamorphic evolution of alternating silica-saturated and silica-deficient microdomains within garnet in ultrahightemperature granulites: an example from Sri Lanka

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#### ABSTRACT

Here we report the occurrence of garnet porphyroblasts that have overgrown alternating silica-saturated and silica deficient microdomains via different mineral reactions. The samples were collected from ultrahigh-temperature (UHT) metapelites in the Highland Complex, Sri Lanka. In some of the metapelites, garnet crystals have cores formed via a dehydration reaction, that had taken place at silica-saturated microdomains and mantle to rim areas formed via a dehydration reaction at silica- deficient microdomains. In contrast, some other garnets in the same rock cores had formed via a dehydration reaction which occurred at silica-deficient microdomains while mantle to rim areas formed via a dehydration reaction which at silica-saturated microdomains.

Based on the textural observations, we conclude that the studied garnets have grown across different effective bulk compositional microdomains during the prograde evolution. These microdomains could represent heterogeneous compositional layers (paleobedding/laminations) in the precursor sediments or differentiated crenulation cleavages that existed during prograde metamorphism. UHT metamorphism associated with strong ductile deformation, metamorphic differentiation and crystallization of locally produced melt may have obliterated the evidence for such microdomains in the matrix. The lack of significant compositional zoning in garnet probably due to self-diffusion during UHT metamorphism had left mineral inclusions as the sole evidence for earlier microdomains with contrasting chemistry.

Keywords – UHT, metapelites, garnet, microdomains, Highland Complex, Sri Lanka

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#### **1. Introduction**

Garnet is a common mineral formed in a wide range of pressure-temperature conditions from lower greenschist facies rocks to ultrahigh-temperature (UHT) granulites and ultrahigh-pressure (UHP) eclogites. Microstructural relationships, e.g., inclusion phases and patterns, and chemical and isotopic zoning in garnet provide useful records of metamorphic processes and rates (Ague et al., 2012, Ague and Carlson, 2013, Caddick and Kohn, 2013), especially in medium-grade metamorphism. In most high-grade granulite terrains, however, the prograde evidence is frequently obliterated due to strong ductile deformation-associated high to ultrahigh temperatures.

In the literature, garnet growth is commonly attributed to a single metamorphic reaction or to a series of successive reactions assuming constant bulk chemistry for the reaction domain. Complications that have been taken into account include melt loss that changes the chemistry of the reaction domain in a homogeneous manner (e.g., White et al., 2003). However, medium-grade garnet has commonly been observed to overgrow micro-layers with highly contrasting chemistries either reflecting original layering or differentiated crenulation cleavages. Classical examples are snowball garnets discussed by structural geologists (e.g, Schoneveld, 1977; Bell and Cuff, 1989; Kim and Bell, 2005). As a result, garnet grains with dimensions larger than the diffusion path lengths of major elements in the microtexture may be expected to show subdomains that have formed by different metamorphic reactions.

We report here the growth of large, single garnet grains in UHT metapelitic granulites from the Highland Complex, Sri Lanka, via different mineral reactions in silica-saturated and silica-deficient bulk compositions. In this context, we propose a model to explain the formation of such complex garnets and show that such garnet bearing mineral assemblages require multiple pseudosections to reconstruct their *P-T* history.

#### 2. Geological Setting and sample description

#### 2.1. Geological setting

Sri Lanka is a small but important crustal fragment in the East Gondwana. Based on Nd model age determinations (Milisenda et al., 1988, 1994) and lithological characteristics, the basement rocks of Sri Lanka have been subdivided into four lithotectonic units (e.g. Kröner et al., 1991; Cooray 1994) named as the Wanni Complex (WC), Kadugannawa Complex (KC), Highland Complex (HC) and Vijayan Complex (VC; Fig. 1a). All samples related to this study were collected near the WC-HC boundary (Fig. 1a, b).

The HC yields Nd model ages of 3400-2000 Ma (Milisenda et al., 1988, 1994) (Hö1zl et al., 1991, 1994; Kröner et al., 1994a; Malaviarachchi and Takasu, 2011a), evolved during the assembly of Gondwana Super Continent. The HC is composed of granulitic metaquartzites, metapelitic gneisses, marbles, calc-silicate granulites and charnockites (e.g. Cooray, 1994; Mathavan and Fernando, 2001). The detrital zircon ages are in the range of ca. 3200 - 1900 Ma and hence the deposition of sediments of the HC had taken place prior to ca.1900 Ma. Most of the granitoid plutons later transformed into orthogneisses, subsequently intruded into the sediments at 1800-1900 Ma and 670 Ma (Kröner et al., 1994a, Hö1zl et al., 1994; Santosh et al., 2014). High to ultra high temperature metamorphic ages of the rocks of the HC varies in the range of 610 - 530 Ma (Hö1zl et al., 1991, 1994; Kröner et al., 1994a; Malaviarachchi and Takasu, 2011a; Santosh et al. 2014; Dharmapriya et al. 2015b; Takamura et al., 2016). Syntectonic and post-tectonic pulses of mafic and charnockitic magmatism have also been reported in the HC (e.g. Santosh et al., 2014). Further, recent studies reported incorporation of Neoproterozoic detrital zircons in the HC metasediments (Dharmapriya et al., 2015b, 2016; Takamura et al., 2016; Osanai et al., 2016b).

In the HC, metamorphic pressures and temperatures (*P*-*T*) decrease from 8-9 kbar and 800-900  $^{0}$ C in the east and southeast to 4.5-6 kbar and 700-750  $^{0}$ C in the southwest (Faulhaber and Raith, 1991; Kriegsman, 1994, 1996; Raase and Schenk, 1994; Schumacher and Faulhaber, 1994; Kriegsman and Schumacher, 1999; Malaviarachchi and Takasu, 2011b; Dharmapriya et al., 2014a). UHT granulites have been reported only from few localities in the central HC (Osanai, 1989; Kriegsman and Schumacher, 1999; Bolder-Schrijver et al., 2000; Osanai et al., 2000, 2006, 2016a, b; Sajeev and Osanai, 2004a; Sajeev et al., 2007 and 2010, Malaviarachchi and Dharmapriya, 2015; Dharmapriya et al., 2015a, b, 2016) and rarely in the southwestern part (Sajeev and Osanai, 2004b) from pelitic, mafic and quartzo-feldspathic granulites. Estimated *P-T* conditions of UHT granulites are in the range of 900 - 1150  $^{0}$ C at *P* of 9 - 12.5 kbar (Kriegsman and Schumacher, 1999; Osanai et al., 2006; Sajeev and Osanai, 2004a, 2004b; Sajeev et al., 2007 and 2010, Dharmapriya et al., 2015a, b, 2016).

The WC, KC and VC have yielded Nd model ages 2000-1000 Ma, 1600-1000 Ma and 1800-1000 Ma (Milisenda et al., 1988, 1994) respectively and metamorphosed under granulite to amphibolite facies conditions (e.g. Kröner et al., 1991, 2013; Cooray, 1994; Kriegsman, 1991, 1993, 1994, 1996; Mathavan et al., 1999). The reader is referred to Santosh et al., 2014; Dhatmapriya et al., 2016 and He et al., 2015, 2016 for further details.

#### 2.2. Sample Description

Sapphirine, spinel, gedrite and sillimanite bearing garnet-orthopyroxene gneiss (Rock A) was collected from a quarry at Kotmale (Supplementary file 1a). Rock A occurs as thin layers (about 20-30cm in size) in the upper portion of the quarry of which the main lithology is garnet–orthopyroxene  $\pm$  clinopyroxene gneiss (metadioritic to charnockitic). The sample contains garnet-rich layers (thickness ~ 2 cm) parallel to the major foliation (Supplementary

file 1b,c), demarcated by stretched quartz grains. Garnet in these layers are rimmed by tiny biotite flakes (<0.2 - .0.4 mm).

Sapphirine, kyanite and spinel bearing garnet-sillimanite-orthopyroxene gneiss (Rock B) was collected from a quarry from Gampola (Supplementary file 1d) south of Kandy. This foliated rock is composed of well-defined compositional domains with gradual variation from one domain to the other without a sharp contact, probably indicating variations of bulk composition within the same rock (Dharmapriya et al., 2015b). The Rock B (Supplementary file 1e) comprises porphyroblastic garnet (0.5- 2.5cm in diameter) surrounded by symplectites of orthopyroxene-cordierite and/or coronas of orthopyroxene-sillimanite (Supplementary file 1e). Leucosomes with subhedral to anhedral quartz and feldspar and tiny flakes of biotites may have formed during the retrograde evolution.

Sapphirine and spinel bearing garnet-sillimanite gneiss (Rock C) occurs as relatively large fragments (25 - 50 cm size) within the upper portion of the same quarry (Supplementary file 1a) in which the Rock B was collected. The samples contain garnets in various sizes (< 0.25 to > 3 cm in diameter, Supplementary file 1f,g), locally rimmed by symplectites of orthopyroxene+cordierite (Supplementary file 1g). Ribbon quartz (up to 6 cm in diameter) defines the main foliation. Medium- to fine-grained sillimanite needles are also present within ribbon quartz and show the same orientation. Local clusters of tiny biotite flakes have probably formed during a retrograde stage (Supplementary file 1f).

Corundum, kyanite and staurolite bearing garnet-sillimanite-graphite gneiss (Rock D, typical khondalite) was collected from a road exposure further south of Gampola (Supplementary file 1h) and, contains various sizes of porphyroblastic garnet (0.25 - 3 cm diameter, Supplementary file 1i), locally showing euhedral crystal shapes.

#### **3.** Petrography and textural evolution of garnet porphyroblasts

We have subdivided the porphyroblastic garnet grains into (1) core, being the most central part of the grain; (2) rim, i.e. the outermost part of the garnet grain, generally a few 100 microns thick; and (3) mantle, being the area between (1) and (2). The absolute size of each domain is variable and depends on the garnet grain size. Mineral Abbreviations after Kretz, (1983).

#### 3.1. In Rock A

The rock contains medium-grained subhedral to anhedral garnet porphyroblasts, (diameter about 0.5-1 cm). The distribution of inclusion phases in garnet is heterogeneous. Sapphirine, spinel, gedrite and sillimanite inclusions occur only within garnet grains that are concentrated in garnet rich layers (Grt<sub>1</sub>). Some of the garnets in the garnet rich layer (Fig. 2a contains rounded and prismatic sapphirine (25 - 80  $\mu$ m), anhedral spinel (20- 60  $\mu$ m), tiny, both needle shaped and anhedral sillimanite (< 20 – 150  $\mu$ m) and gedrite (20 - 100  $\mu$ m) and tiny rutile and apatite inclusions toward the core area (Fig. 2b, c, d, e, f). Tiny sapphirine grains are present as isolated inclusions (Fig. 2c,e,f,g), coexisting with spinel (Fig. 2f). Spinel also forms isolated inclusions (Fig. 2f), coexisting with sapphirine (Fig. 2f) or sillimanite (Fig. 2g). Sapphirine, sillimanite and spinel are always present very close to each other whereas gedrite inclusions are present rarely close to the clusters of sapphirine, spinel and sillimanite (Fig. 2g).

In addition, considerable amount of apatite and rutile (Fig. 2e) together with minor amount of monazite and zircon grains are present at the garnet cores. Fine-grained quartz is present mostly in mantle to rim of the garnet (Fig. 2b, c, d). Isolated and medium-grained biotites, quartz and sillimanite are also commonly distributed in the mantle (Fig. 2b, c, d),

where biotite and sillimanite shows a folded foliation (Fig. 2b,d). The rim of the garnet is surrounded by biotite (Fig. 2h).

The core area of other garnets of the rock (Fig. 2i) contains gedrite in addition to sapphirine, spinel and sillimanite while quartz and biotite are present in the mantle area (Fig. 2i, j, k). Gedrite coexists with spinel or sapphirine (Fig. 2l) while some spinel grains occur as isolated inclusions (Fig. 2j, k). Rare sillimanite is also present close to isolated spinel (Fig. 2k). In the mantle area, biotite and quartz are present as isolated grains (Fig. 2i,j).

The inclusion phases within the core areas of the above porphyroblastic garnets could indicate the FMASH reaction:

$$Spl + Ged + Sil => Grt + Spr + V(1)$$

while, the mantle to rim areas of the same garnets testify to further growth of garnet via the reaction:

$$Bt + Sil + Qtz \Rightarrow Grt + Kfs + V/Melt (2)$$

In contrast, a sapphirine, spinel and sillimanite bearing cluster is found towards the rim of one side in some other garnets (Fig. 2m) in garnet-rich layer (Fig.2n). Biotite is present in both core and rim of the garnet grain(Fig. 2m, o). Although isolated quartz is also present in the mantle and towards the rim (Fig. 2o) in the same garnet, there is no quartz inclusions present in the vicinity of sapphirine, spinel and sillimanite bearing cluster. Inclusion phases of this porphyroblastic garnet indicated that the core formed via reaction (2) and the rim via reaction (1).

Garnet grains, which are further away from the garnet-rich layer, are mostly inclusion free and contain plagioclase, quartz and biotite as minor inclusion phases (Supplementary file 2a).

The matrix contains, ribbon quartz (up to 5cm), anhedral to subhedral orthopyroxene (0.5-1.5 cm), subhedral to euhedral plagioclase (<0.25 - 0.5cm) and anhedral K-feldspar

(<0.1- 0.5cm) as major mineral phases (Supplementary file 2b). The stretched quartz grains demarcate the major foliation. Ilmenite and acicular fine biotite flakes (~0.5cm in length) are present as minor mineral while monazite and zircon occur as accessory phases. Biotite occurs mainly as an overprinting product of garnet and orthopyroxene.

#### 3.2. Rock B

The Rock B contain needle shaped sillimanite, anhedral quartz, plagioclase, K-feldspar, biotite and as major constituent minerals with Fe-Ti oxides (exsolved hematiteilmenite  $\pm$  rutile) as a minor phases with sapphirine, kyanite, monazite and zircon as accessory (Fig. 3a, b, c, d). Subhedral to anhedral shaped garnets (Grt<sub>2</sub>, diameter 0.40 – 1.5cm) contains coexisting kyanite+sapphirine at the core area (Fig. 3a, b, c). Supplementary file 3a shows the Raman spectrum of kyanite. Medium grained kyanite shows approximately prismatic shape (Fig. 3a,b). Isolated quartz is present in the mantle (Fig.3a, b) area and isolated sillimanite occurs towards the rim area of the same garnet grain (Fig. 3a,d). Biotite is present from mantle to rim (Fig. 3a, b). The coexisting sapphirine+kyanite in the core area could have formed via the prograde dehydration reaction in the FMASH system:

Mg-rich 
$$St => Spr + Ky + Grt + V(3)$$

while the isolated sillimanite needles and biotite flakes toward the rim area and presence of quartz towards the outer core area could indicate that a significant portion of  $\text{Grt}_2$  has grown via reaction (2).

Staurolite is a common mineral in metasedimentary rocks subjected to Barrovian-type metamorphism (Yardley, 1977), but is also observed in Buchan-type metamorphism, e.g., in the Hercynian domains of the central Pyrenees (Kriegsman et al., 1989). The reason for the absence of staurolite in Rock B could be the complete consumption of staurolite during prograde metamorphism. Theses authors have identified Mg- rich staurolite ( $X_{Mg} \sim 0.50$ ) in a

locality approximately 2 km west of the sampling locality of Rock B and close to the Victoria dam at the UHT locality 7 in Fig. 1a), in metapelitic granulites. Hiroi et al. (1994) have also reported coexisting local sapphirine+kyanite enclosed by garnet in Highland Complex paragranulites. The authors have also interpreted this assemblage by the operation of reaction (3). Tsunogae and Van Reenen (2006) reported on Mg-rich staurolite ( $X_{Mg} = 0.47-0.52$ ) from the Beitbridge area in the Central Zone, Limpopo Belt of South Africa. The authors regarded the magnesium staurolite as a relict phase and inferred that dehydration reaction (3) took place during prograde metamorphism. Tsunogae and Van Reenen, (2011) also reported similar examples from the Limpopo Belt.

Most of the garnet grains in the rock are rimmed by orthopyroxene+sillimanite corona (Supplementary file 2c) and/or orthpyroxene+cordierite symplectites (Fig.3a, Supplementary file 2c,d). At the rim area of some garnet contains a box structure of orthopyroxene-sillimanite (Fig. 3a).

The matrix of the rock B is composed of ribbon quartz (up to 3cm in length), sillimanite (up to 4 cm in length), subhedral to anhedral K-feldspar (< 0.2 - 0.5 cm) and plagioclase (0.2 - 0.75 cm) as major mineral phases. Anhedral Fe-Ti oxide (mainly exsolved titanohematite-ilmenite±rutile grains, Supplementary file 2e) and disseminated biotite flakes (up to 0.25cm) present as minor constituents while zircon and monazite occur as accessory phases. Fine to medium grained anhedral sapphirine ( $\sim$ 200 µm), which are surrounded by two feldspars in some micro domains may represent melt ribs (Supplementary file 2f).

#### 3.3. Rock C

This rock contains subhedral to anhedral, medium to coarse-grained garnet porphyroblasts (Grt<sub>3</sub>, diameter about 0.5-2 cm). The garnet grains contain sillimanite, biotite,

quartz and rutile as major inclusion phases while plagioclase, K-feldspar and ilmenite as minor phases. Sapphirine, spinel, apatite and zircon are present as accessory phases.

Grains of Grt<sub>3</sub> contain quartz, sillimanite, K-feldspar, biotite, and rutile with minor apatite inclusion phases from core to rim (Fig. 4a,b). In the same grain, clusters of inclusion phases are present at the mantle area in two opposite sides (Fig. 4a,b,c,d). These clusters mainly contain sillimanite and rutile as tiny inclusions with no quartz. In some clusters (Fig. 4c,d), tiny sapphirine (~20  $\mu$ m in length) coexist with sillimanite (Fig. 4e). Close to those inclusions, tiny spinel (~10  $\mu$ m) and sillimanite co-exist (Fig. 4f). Isolated quartz grains are present surrounding the mineral cluster where the sapphirine and spinel are present (Fig. 4c, d).

Quartz, biotite and sillimanite inclusions from core to rim area indicate that the Grt<sub>3</sub> has mainly grown via reaction (2) in a silica-saturated environment. However, the mantle area of one side of this garnet lacks quartz inclusions and contains the assemblages sillimanite+spinel and spinel+sapphirine. Hence, this particular garnet domain has grown in a quartz-deficient environment, either by reaction (1) or by an alternative reaction,

Mg rich  $Bt + Sil => Grt + Spl \pm Spr + V/Melt (4)$ 

Orthopyroxene-cordierite symplectite is present at the rim of the garnet (Fig. 4a,b). However, some of the  $Grt_3$  lack symplectitic textures (Supplementary file 1g). Sillimanite inclusions in some of these garnets show crenulation lineation (Supplementary file 2g) which is oblique to the major lineation present in the matrix.

In the matrix, garnet, ribbon quartz (up to 5cm in length), needle shaped sillimanite (from 0.5 to cm in length), fine to medium grained (<0.2 - 0.5cm) plagioclase and K-feldspar which is present as recrystallized mineral phases occur as major mineral phases (Supplementary file 2h). Rutile needles (up to 1cm in length), tiny biotite flakes (up to 0.4 cm) and ilmenite are present as minor phases while zircon, apatite and monazite represent

accessories. Needle shaped sillimanite demarcate the major lineation in the matrix (Supplementary file 2h).

#### 3.4. Rock D

The khondalite contains various sizes of garnet (from ~0.25 - 3cm in diameter). Dharmapriya et al. (2014b) provided detailed description of the microtextures of this rock. Based on inclusion phases four garnet types can be distinguished. The first type of garnet (Type 1) contains sillimanite inclusions at core and kyanite inclusions at rim with staurolite at mantle (Supplementary file 2i,k). The core area of these garnets contains preferredly oriented quartz grains (Supplementary file 2j). The second type of garnet (Type 2) contains corundum, kyanite, spinel sillimanite, quartz and Fe-Ti oxides as inclusions. The third type of garnet (Type 3) has sillimanite, curved quartz and biotite inclusions (Supplementary file 2l) while the fourth type (Type 4) is fine grained garnet with inclusions of sillimanite, rutile and ilmenite (Supplementary file 2m).

Further, it was observed that rarely, the Type 1, Type 3 and Type 4 garnets could be identified in approximately 10 x 10 cm sized rock specimens while Type 2 was relatively less abundant and found only in some domains in the matrix where quartz was relatively less abundant compared to the rest of the rock. In this study we used Type 2 garnet (hereafter referred as  $Grt_4$ ) from this rock .

Grt<sub>4</sub> grains are coarse porphyroblastic (up to 3 cm in diameter), euhedral to subhedral in shape (Fig. 5a). The core and mantle of most of these Grt<sub>4</sub> (Fig. 5a) contain numerous isolated inclusions of anhedral, medium- to fine-grained corundum (0.5 - 4 mm), which frequently coexist with clusters of fibrolitic sillimanite (Fig. 5b, c). The sillimanite in theses clusters has probably grown as a pseudomorph after kyanite (Fig. 5b,c). Sometimes, Grt<sub>4</sub> contains relatively large (up to 0.5cm) prismatic sillimanite, which contains plenty of tiny

spinel inclusions and rare, tiny corundum inclusions (Fig. 5e). Close to these sillimanites, isolated, anhedral tiny staurolite grains (up to 0.3 mm) are also present (Fig. 5e,f). In some cases, the presence of spinel and sillimanite inclusions can be observed at the margins of corundum (Fig. 5e). In some  $Grt_4$  (Fig. 5h), coexistence of corundum and sillimanite clusters pseudomorphing kyanite are observed in the mantle part (Fig. 5i).

Towards the rim area of Grt<sub>4</sub>, isolated kyanite (Supplementary file 3b shows the Raman Spectrum of this kyanite) and quartz inclusions are present (Fig. 5d). In addition, coarse patches of pyrophyllite are present, in which relics of fibrolitic sillimanite could be observed (Fig. 5g). The shape of the pyrophyllite patch indicates the existence of fibrolitic sillimanite, pseudomorphing kyanite, which is a very common feature of Grt<sub>4</sub>.

Close associations of sillimanite clusters which represent pseudomorph kyanite and isolated quartz are present from core to rim of the other side in the same garnet (Fig.5j,k,m). Rare isolated staurolite grains are present close to quartz inclusions (Fig. 5m). However, there are no quartz inclusions in the corundum bearing portion of the particular garnet.

Hence, the presence of kyanite and/or kyanite pseudomorph + corundum  $\pm$  staurolite in core to mantle areas of Grt<sub>4</sub> indicates the prograde reaction:

$$St \Rightarrow Grt + Crn + Ky + V(5)$$

This may indicate that reaction (5) has taken place during prograde compression stage of the HC metasediments.

The inclusions of kyanite pseudomorphs + quartz  $\pm$  staurolites in the mantle to rim area and indicates that these inclusion bearing portions of the garnet have formed under quartz saturated conditions independent of reaction (5). Possible reactions in theses quartz saturated domains are referred in the discussion section.

Presence of sillimanite pseudomorphs after kyanite - quartz - staurolite from core to rim of one side in some Grt<sub>4</sub> indicate the reaction:

$$St + Qtz \Rightarrow Grt + Ky$$
 (6)

On the other side of  $Grt_4$ , the presence of corundum together with sillimanite containing spinel inclusions can be explained by the reaction,

$$Grt + Crn => Spl + Sil (7)$$

Dharmapriya et al, (2014b) concluded that reaction (7) has taken place close to peak metamorphic conditions.

During retrogression at much lower *P* and *T*, pyrophyllite has formed at the expense of fibrolitic sillimanite via the reaction,

$$Sil + Qtz + H_2O \Longrightarrow Prl(8)$$

The matrix of the studied khondalite consists of quartz saturated domains which is mainly composed of ribbon quartz, coarse-grained sillimanite, alkali-feldspars (from 0.3 up to 1 cm) as major mineral phases (Fig.S2n,o). Ilmenite is present as a minor constituent while zircon is present as an accessory mineral. Elongated quartz and coarse-grained sillimanite needles show a preferred orientation and define the major foliation and lineation of the rock (Supplementary file 2n). Coarse-grained, euhedral to subhedral alkali-feldspar is present within the pressure shadow of some porphyroblastic Grt<sub>4</sub>. The tiny sillimanite inclusion bearing anhedral quartz (Supplementary file 2o) could probably result from crystallization of a melt phase during the retrogression.

#### 4. Mineral Chemistry

Mineral compositions were analyzed using a JEOL JXA8530 Field Emission Electron Probe Microanalyzers (EPMA) installed at three laboratories: (1) Department of Environmental Changes, Faculty of Social and Cultural Studies, Kyushu University, Japan, (2) Indian Institute of Science, Bangalore, India and (3) National Consortium at the University of Utrecht, Netherlands. All analyses were carried out using an accelerating

voltage of 15 kV and 20 nA beam current, 1-3  $\mu$ m spot size in all three laboratories. Detailed descriptions of analytical procedures followed in the three laboratories are given in supplementary Document 1.

The following section contains the mineral compositions of studied porphyroblastic garnets and their inclusion phases.

#### 4.1. Garnet compositions

The studied garnets in Rock A (Grt<sub>1</sub>) are mainly almandine-pyrope solid solutions, with only small amount of grossular, spessartine and andradite components (Table 1). The  $X_{Alm}$  content increased from core (~0.500) to rim (~0.580) while the  $X_{pyr}$  content gradually decreased (~0.430 at cores and ~0.370 at rims). The  $X_{Mg}$  value and Ca content vary from ~0.430 to ~0.400 and 0.180 wt% to 0.110 wt% respectively, from core to rim.

The garnets in Rock B (Grt<sub>2</sub>) also exhibit mainly almandine-pyrope solid solutions (Table. 1). Both  $X_{Alm}$  and  $X_{Pyr}$  contents of Grt<sub>2</sub> show slight variations from core to rim. However, here the  $X_{Alm}$  content decreased (~0.460 and 0.440 respectively) and  $X_{Pyr}$  (~0.450 and 0.480 respectively) content increased from core to rim. There is a considerable zonation of Ca content from core to rim (~1.90 wt% and 1.00 wt% respectively).

Similar to previous garnets, the studied garnet in Rock C (Grt<sub>3</sub>) also exhibits mainly almandine-pyrope solid solutions (Table 1). In Grt<sub>3</sub> the  $X_{Alm}$  content gradually increased from core to rim (~0.460 to 0.510 respectively) while  $X_{Pyr}$  shows the opposite behaviour (~0.510 to 0.450 respectively). The Ca content is approximately the same from core to rim (~0.80 wt% and 0.70 wt% respectively).

The garnets in Rock D (Grt<sub>4</sub>) are mainly of almandine-pyrope solid solutions, with only small amounts of grossular, spessartine and andradite components (Table 1). The Grt<sub>1</sub> shows similar  $X_{Alm}$  content (~0.73) from core to rim. But the  $X_{Pyr}$  content increased (0.13 and

0.15 respectively) from core to rim. The cores of theses garnets display a relatively lower  $X_{Mg}$  value (~0.15) while rims display  $X_{Mg}$  value (~0.16). The MnO contents is ~3.6, wt- % in core compositions and ~3.2, wt-% in the rims while higher amount of Ca ~ 2.1 wt-% were in the core ~ 1.6, wt-% at the rim.

#### 4.2. Biotite

Biotite inclusions in  $\text{Grt}_1$ ,  $\text{Grt}_2$  and  $\text{Grt}_3$  show high Ti content (from ~ 5.00 to ~6.70 wt %, Table 2). All the biotite inclusions are Mg-rich and the  $X_{\text{Mg}}$  value is relatively higher in biotite inclusions within garnet ( $X_{\text{Mg}}$  0.740 – 0.780).

#### 4.3. Staurolite

Staurolites (Table 2) are enclosed by  $Grt_4$  are Zn-bearing (ZnO = 1.60 wt-%) and rich in Ti (TiO<sub>2</sub> = 1.6). The  $X_{Mg}$  value of the staurolite inclusions is ~0.29.

#### 4.4. Spinel

Spinel in Grt<sub>1</sub> (Table 2) show ZnO content of ~3.1 wt %. The  $X_{Mg}$  value of spinel inclusions are around 0.57. Spinel in Grt<sub>3</sub> has relatively higher Zn content (~5.6 wt %) while the  $X_{Mg}$  is 0.66.

#### 4.5. Sapphirine

The composition of sapphirine in  $\text{Grt}_1$  is very close to the end member 2:2:1 while those in  $\text{Grt}_2$  are in between the end member compositions of 2:2:1 and 7:9:3. The  $X_{\text{Mg}}$  value of sapphirine in  $\text{Grt}_1$  is around 0.76 while those in  $\text{Grt}_2$  have values of about 0.80. Sapphirine in  $\text{Grt}_3$  is also close to end member 2:2:1 with an  $X_{\text{Mg}}$  value of 0.74.

#### 5. P-T calculations of garnets

Dharmapriya et al. (2015b) evaluated the *P*-*T* evolution of Rocks A, B and C using textural observations coupled with conventional thermobarometry. Calculations show that these rocks were metamorphosed to ~11-12 kbar around 800-850  $^{\circ}$ C during prograde stage. The rocks subsequently underwent prograde decompression with increasing temperature up to peak UHT conditions. The peak metamorphic P-T conditions are 900-975 °C at ~ 9.5-10 kbar.

Experimentally calibrated, pressure independent sapphirine-spinel geothermometry (Das et al., 2006) yielded temperature range from 820 - 868 <sup>o</sup>C (average 837 <sup>o</sup>C) for coexisting spinel-sapphirine pairs in Grt<sub>1</sub>. However, it is difficult to determine the temperature conditions of the reaction (1) based on above Fe-Mg exchange thermometry, since mineral chemistry of these very tiny mineral inclusions such as sapphirne and spinel may have been undergone modification due to intense chemical diffusion under UHT peak metamorphic conditions and subsequent re-equilibration during the retrogression.

Dharmapriya et al., (2015b) showed that Ti-in biotite thermometer yielded the prograde temperature up to 845  $^{0}$ C for biotite inclusions in Grt<sub>1</sub>. From melting experiments of natural metapelites under fluid-absent conditions, Vielzeuf and Holloway (1988) pointed out that the reaction (2) has taken place under the minimal *T* of ~860 °C. Since the Ti content of the biotite inclusions in garnet of the Rock A is up to 7 wt %, the breakdown of the biotite could have taken place at very high temperature conditions. Hence, it is logical to speculate that reaction (1) had taken place prior to the reaction (2) during the prograde decompression with increasing temperature of the Rock A.

The P-T conditions of the inferred reaction (3) in Rock B can be evaluated by the existing experimental results. Based on experimental investigations, Schreyer (1988) suggests that pure Mg-staurolite is stable at P > 14 kbar and T > 710 - 760 °C in the MASH system. Later Fockenberg (1998) reported the stability of Mg-staurolite at P–T conditions of 12–66

kbar and 608–918 °C. Unfortunately the experimental results observed from pure Mgstaurolite are difficult to apply directly to the natural rocks (Kelsey et al., 2006; Tsunogae and Van Reenen, 2011). In contrast, Sato et al. (2010) reported high-*P*-*T*, experiments of staurolite with moderate  $X_{Mg}$  (= 0.7–0.5) at 12–19 kbar and 850–1050 °C. As mentioned in the section 3.2, due to occurrence of staurolite with  $X_{Mg}$  ~0.50 in the central HC close to sampling locality of Rock B, the reaction (3) could have taken place at *P*-*T* of ~11-12kbar and ~820-850 °C which is the derived prograde P-T condition of the Rock B (see Dharmapriya et al., 2015b). Hence the Ti-rich biotite (up to 5.00 wt %) could probably break down via reaction (2) just after the reaction (3) during the prograde decompression with increasing temperature of the Rock B.

The biotite dehydration reaction (2) and (4) in Rock C could also have taken place contemporaneously during the prograde decompression. Dharmapriya et al., (2015a) have shown evidence for formation of garnet via the prograde dehydration reaction (2) in a corundum bearing pelitic granulite. Core area of some of those garnets contains biotite, sillimanite and quartz inclusions. Due to the presence of spinel inclusions at the rim of the same garnet and absence of quartz at the garnet rim and in the matrix of the rock, these authors argued that the rim area of the garnet could have formed after the complete consumption of quartz via reaction Bt+Sil => Grt+Spl+Kfs+melt during the prograde decompression with increasing temperature. However, in the Rock C, formation of a vast area of  $Grt_3$  (from core to rim) is a result of the reaction (2). The reaction (4) represents only at the mantle area in one side of the  $Grt_3$ .

Authors have further evaluated the *P*-*T* evolution of Rock D using textural observations coupled with pseudosections, petrogenetic grids and conventional thermobarometry and found that the Rock D has reached ~11 kbar at 780  $^{0}$ C during prograde metamorphism. Subsequently, it has undergone a decompression stage with increasing

temperature, reaching peak T of ~900 °C at ~9 kbar. Using petrogenetic grids, numbers of workers (e.g Hiroi et al., 1994) have shown that reaction (5) had taken place prior to reaction (6).

The prograde conditions existed in minor silica-deficient domains of Grt<sub>4</sub> of Rock D were evaluated using pseudosections (using Perplex\_X\_07 after Connolly, 2005) based on calculated local bulk rock compositions for the silica-deficient domain of Grt<sub>4</sub> using estimated ratios among garnet, kyanite, corundum and staurolite (Table 3). Based on textural observations (Fig. 5.), it is inferred that all corundum grains enclosed by garnet are produced as a result of staurolite breakdown via reaction (5). This is a reaction with changes in minor elements (e.g., minor Ti enrichment in staurolite and some Mn and Ca dilution in garnet). The modal abundances of Grt<sub>4</sub>, staurolite and kyanite prior to the corundum formed by breakdown of staurolite, were recalculated (Table 3). The results suggest that some kyanite was already present before staurolite started to breakdown. The amount of H<sub>2</sub>O was added stoichiometrically equivalent to that ideally resided in the staurolite, which is likely to have escaped the system along the prograde path. Pseudosections were calculated in FMAS and FMAS systems at  $X_{Mg}$  = 0.15 which was the  $X_{Mg}$  of Grt<sub>4</sub>. Though staurolite has  $X_{Mg}$  = 0.29, the effect of  $X_{Mg}$  of staurolite was neglected as the obtained staurolite volume in Grt<sub>4</sub> is ~1%. The used composition have labeled in each pseudosection. Figure 7a shows the modelpseudosection (where  $SiO_2$  mol% is 38.01) for this small reaction domain, assuming local equilibrium (e.g. Stüwe, 1997) within the Grt<sub>4</sub> porphyroblast hosting the reaction domain. This shows that the P-T field for the inferred staurolite breakdown reaction is largely consistent with the FASH end-member reaction, but has shifted to higher P and T. Thus, the results indicate that staurolite broke down to Grt + Ky + Crn + V (bold field) between 8 and 12 kbar at temperatures between 730 and 800 °C (Fig. 6a). It is likely that H<sub>2</sub>O was lost from the assemblage at this stage.

Since the assemblage outside the Grt<sub>4</sub> porphyroblast contains abundant quartz and it lacks corundum, it is deduced that garnet originally enclosed some quartz. Hence, another two pseudosections (Fig.6b, c) were calculated after adding some SiO<sub>2</sub> (SiO<sub>2</sub> mol% at 38.5 and 40.08) to investigate a possible earlier path leading to quartz depletion. It turns out that, above 6 kbar, the resulting diagram (Figure 6b and c) do not change even more SiO<sub>2</sub> is added. Thus the results show that reaction (5) operated between 8 and 12 kbar at temperatures between >730 °C. These calculations are typical with the obtained result from pseudosections constructed under the pre-melting, of which original rock composition was calculated through stepwise re-integrations of melt into the residual XRF composition, showing that the rock has reached > 10 kbar at 750-775 <sup>0</sup>C during prograde metamorphism.

#### 6. Discussion

6.1 A model for the incorporation of silica-saturated and silica-deficient microdomains in single garnet grains

Silica-saturated and silica-deficient portions in the same garnet can result in different ways. One possibility is the consumption of quartz during the progress of garnet formation reactions, which could lead to quartz depletion (e.g., Alvarez-Valero and Kriegsman 2010; Dharmapriya et al. 2015a). Subsequent garnet-producing reactions may lead under the silica-deficient garnet rims (e.g. Dharmapriya et al., 2015a). This mechanism generally leads to a concentric zonation pattern with silica-saturated domains in the garnet core and silica-deficient domains in the garnet mantle to rim.

In a second mechanism, quartz can be incorporated into garnet by crystallization of a melt inclusion (Sawyer, 2001; Harley, 2008). Even a dehydration melting reaction under silica-deficient conditions can produce a melt phase that later precipitates forming quartz.

This mechanism can produce a random distribution of quartz-bearing and quartz-deficient domains in garnet.

Here, we discuss an alternative mechanism to form silica-saturated and silicadeficient domains in single garnet grains in an alternating pattern that may explain some contrasting observations. In addition, it is essential to evaluate the P-T conditions at which the particular silica-saturated and silica-deficient mineral reactions were taken place, due to the fact that silica-saturated granulites are generally indicative of high to extreme temperature metamorphism (Kelsey and Hand 2015). Such silica-saturated granulites may contain mineral assemblages, which are diagnostic to ultrahigh temperature conditions (e.g. Harley, 2008; Kelsey and Hand 2015). In contrast, the stability of silica-deficient granulites usually expands to lower temperatures than that for silica-saturated assemblages. Further, there are only few localities of silica-deficient granulites globally, which record temperatures less than 900 °C (e.g. Sharma and Prakash, 2008; Nasipuri et al. 2009; Prakash and Sharma 2011; Prakash and Sharma 2008; Prakash et al. 2013; Prakash and Singh 2014).

#### 6.1.1 Formation of Grt<sub>1</sub>

The distribution of inclusion phases in Grt<sub>1</sub> reveals that high Al/low-Si reaction (1) has contributed to the growth of core areas of some garnet grains and rim areas of some other grains. Similarly, high-Al/high-Si reaction (2) produced mantle to rim areas of some garnet grains, but core to mantle areas of some other grains. Hence, we consider that Grt<sub>1</sub> grains have grown across alternating microdomains with different effective bulk compositions, namely high-Al/low-Si versus high-Al/high-Si microdomains.

However, P-T estimations indicated that the reaction (1) could probably have taken place prior to the reaction (2) during the prograde decompression. Thus, it is more logical to explain the high Al/low-Si reaction (1) around the core and high-Al/high-Si reaction (2) from

mantle to rim of the same garnet grain. Nevertheless, the presence of high-Al/high-Si reaction (2) from core to rim of one side of the garnet and high Al/low-Si reaction (1) in the rim area of other side of the same garnet revealed that the core and mantle area of the particular garnet is younger than the rim area of the same garnet represented by the reaction (1).

#### 6.1.2. Formation of Grt<sub>2</sub>

Inclusions of kyanite+sapphirine in the core areas of Grt<sub>2</sub>, indicated that garnet started growing via reaction (3) in high-Al/low-Si domains, followed by reaction (2) in high-Al/low-Si mantle to rim areas.

#### 6.1.3. Formation of Grt<sub>3</sub>

The inclusion phases indicate that most of Grt<sub>3</sub> has formed via reaction (2) which is typical for high-Al/high-Si domains. However the presence of sillimanite+sapphirine and spinel+sillimanite without quartz as inclusions indicates that some high-Al/low-Si microdomains were present where garnet grew via reaction (4). Hence, Grt<sub>3</sub> has also grown across microdomains with different compositions during prograde metamorphism.

#### 6.1.4. Formation of Grt.

Inclusion phases of  $Grt_4$  reveal that this garnet type also grew across microdomains with different bulk compositions. The incorporation of sillimanite pseudomorphs after kyanite accompanied by quartz ± staurolite in core to rim on one side of a garnet (Fig. 51,m,n) indicates that this side of the garnet has formed in a high-Al/high-Si microdomain via reaction (6). Sillimanite pseudomorphs after kyanite + corundum toward the mantle on the

other side in the same garnet (Fig. 5k) indicated that the particular portion of the garnet has formed via reaction (5) in a high-Al/low-Si micro domain. In the FASH chemical system (e.g. Hiroi et al., 1994), reaction (6) operates at lower P-T conditions than reaction (5).

In contrast, core to mantle areas of some other  $Grt_4$  with corundum and sillimanite psudomorphs after kyanite  $\pm$  staurolite indicated that the evolution of those garnets had taken place via the reaction (5) at aluminum-rich, silica-deficient microdomains. However, the presence of quartz, kyanite and/ or kyanite-pseudomorphs  $\pm$  staurolite  $\pm$  K-feldspar towards the mantle to rim area of some  $Grt_4$  (where corundum, kyanite-pseudomorph  $\pm$  staurolite present at the core) indicates that the mantle to rim area of these garnets had formed at Alrich, silica-saturated microdomains and hence independent from reaction (5).

One possibility is that the quartz inclusion bearing portions of Grt<sub>4</sub> could have formed via reaction (6). Though, in the FASH chemical system (e.g. Hiroi et al., 1994) the reaction (6) takes place relatively at lower *P*-*T* conditions prior to reaction (5). However, Fockenberg (1998) suggested that end-member Mg-staurolite is stable at very wide range of *P*-*T* conditions of pressures from 12 to 66 kbar and temperatures ranging from 608 to 918 °C. Ti content can also shift the stability field of staurolite to relatively higher temperature conditions (e.g. Rasse and Shenk, 1994). However, in the present case, the X<sub>Mg</sub> content of staurolite in both silica saturated and deficient domains are approximately similar (~0.30). Available experimental and thermodynamic studies of staurolite suggest that typically Fe-rich staurolite becomes unstable above the amphibolite facies resulting the progress of the ((K)FMASH) continuous reactions  $St+Ms+Qtz => Grt+Bt+Ky+H_2O$  or the reaction (6) at ~700 °C (e.g., Spear and Cheney, 1989: Tsunogae and Van Reenen, 2011). However, both reactions take place prior to the reaction (5) in *P*-*T*-*t* space. Hence the possibility is that the garnet formation could initiate in Al-rich but quartz-deficient microdomains and, then continue into Al-rich and quartz-saturated microdomains. Further, the core area of the garnet

may also be formed in Al-rich and quartz-deficient micodomains. In such a case, the quartz inclusion-bearing mantle to rim area of the garnet is relatively older than the corundum inclusion bearing core.

# 6.2. Existence of silica-saturated and silica-deficient microdomains in metasediments during the prograde metamorphism

Locally existing of macro to micro scale silica-saturated and silica-deficient bulk compositional domains are reported in metasediments from numerous medium to high grade metamorphic terrenes (e.g. Powell and Vernon, 1979; Adjerid et al., 2008, 2013; Jiao, 2013; Harley, 2008; Dharmapriya et al., 2015a). For an example, Dharmapriya et al., (2015a) reported macro-scale compositional differences between silica-saturated and silica-deficient rock domains from the HC. Authors argued that these compositional domains may reflect a primary heterogeneity in the sediment during deposition. A number of workers reported evidence for the presence of closely associated silica-saturated and silica-deficient microdomains in medium grade to HT/UHT metamorphic rocks (Powell and Vernon, 1979; Adjerid et al., 2008, 2013; Jiao, 2013; Kelsey and Hand, 2015). Jiao et al., (2013) reported existence of silica-saturated and silica-deficient microdomains in HT granulites of Khondalite belt, North China Craton. Those authors showed textural evidence for various inclusion bearing garnets and different garnet break down reactions at silica-saturated and silicadeficient microdomains in the same rock. Further, Adjerid et al., (2013) reported textural evidence for silica-saturated and silica-deficient reactions occurring in close microdomains at UHT granulites of Khanfous area in Ouzzal metacraton, Hoggar, Algeria. Silica-saturated and silica-deficient domains can result in metasedimentary rocks by processes such as metamorphic differentiation (e.g. Fletcher, 1977), formation of differentiated crenulated cleavages (e.g. Williams, 1972; Granath, 1980), crystallization of silica saturated melt pods in

silica-deficient rocks (e.g. Harley, 2004, 2008), local influxes of silica rich fluid phase into silica deficient rock (e.g. Leite et al., 2009) and existence of original compositional heterogeneity in protolith sedimentary rocks which represent silica-saturated and silicadeficient domains (e.g. Dharmapriya et al., 2015a). Textual evidence indicated that genesis of all studied garnets incorporate inclusion phases representing distinct silica saturated and silica-deficient dehydration reactions which could contribute to progress of garnet formation around upper amphibolite (e.g. Grt<sub>4</sub>) to amphibolite to granulite facies transition (e.g. Grt<sub>1</sub>, Grt<sub>2</sub> and Grt<sub>3</sub>). In other words, garnets have grown across different effective bulk compositional microdomains which probably existed up to amphibolites facies conditions during the prograde evolution. Therefore, one possibility is to consider that the silicasaturated and deficient microdomains in Grt<sub>1</sub> to Grt<sub>4</sub> probably represent heterogeneous compositional layers such as paleobeddings/laminations which perhaps have occurred as few centimeter to millimeter in thickness in the precursor sediments. Conversely, differentiated crenulated cleavages during prograde metamorphism may also cause to existence of such silica-saturated and silica-deficient domains.

Existence of various millimeters to few centimeter scale alternate compositional lamellae due to sedimentary bedding or laminations is a common feature of sedimentary rocks. The bulk chemical compositions (such as Si, Fe and Al content) of those individual lamella can vary according to the nature of deposited sediments. As an example, Emiliani (1992) reported banded iron formations consisting of laminated layers of iron-rich/ silica-poor and iron-poor/silica-rich intervals. Sedimentary lamination, bedding and typical structures of sedimentary rocks such as cross-bedding and graded bedding may be preserved in low to medium grade metasedimentary rocks (Hacker and Goodge, 1990; Bucher and Grapes, 2011). Unfortunately, it is often impossible to recognize paleobedding of protolith sediments in high grade metamorphic rocks (e.g. Passchier and Trouw, 1998). Occasionally,

preservation of relict paleobedding of protolith sediments in highly deformed granulite facies rocks has also rarely been reported (e.g. Touret, 1965; Perera, 1994; Kehelpannala, 1997, 2003). Perera, (1994b) reported evidence for preservation of paleo cross-bedding in granulite facies rocks in the HC. Kehelpannala (1997, 2003) reported evidence for preservation of paleo bedding in metasediments from the WC close to HC-WC inferred tectonic contact. Further, Kehelpannala (2003) argued that some of the metasedimentry rocks of the HC contain modified primary- sedimentary beddings (paleobedding), which may have resulted from strong deformation and metamorphism of a sequence of sandstone, pelites and semipelite of protholith sediments. Touret (1965) reported preservation of extremely delicate structures such as flysch-type banding or cross-bedding in southern Norway granulite domain.

There are number of workers (e.g. Voll and Kleinschrodt; 1991; Kleinschrodt; 1994; Kröner et al., 1994b; Kehelpannala, 1997, 2003) who suggested a phenomena of strong flattening of original sedimentary sequence of the HC at the lower crustal level. Kröner et al., (1994b) argued that original supracrustal sequence in the Highland sedimentary basin were subjected to folding, thrusting and strong flattening while transporting into the lower crust during the final stages of continental collision around Sri Lanka, simultaneous with assembly of Gondwana. Hence, it is reasonable to argue that the local existence of millimeter to micrometer scale Al-rich silica-saturated and Al-rich silica-deficient lamella during the prograde evolution, which were present as few centimeter to millimeter scale bedding or laminations prior to strong flatting event of original sedimentary prololiths.

Alternatively, such Al-rich silica-saturated and Al-rich silica-deficient microdomains could be present during prograde metamorphism due to existence of differentiated crenulation cleavage which is a common type of cleavage in multiply-deformed, intermediate to high-grade metapelitic rocks (Bell and Cuff, 1989; Williams et al., 2001). Several workers

(e.g. Schoneveld, 1977; Bell and Cuff, 1989; Kim and Bell, 2005; Sharib and Bell, 2011) have reported evidence for genesis of garnet porphyroblasts across the compositionally different, differentiated crenulation cleavages. Further, existence of quartz-rich and quartz-deficient micro domains across the differentiated crenulation cleavages have also reported in the literature (e.g. Williams, 1972; Fletcher, 1977; Granath, 1980).

The next questions are, if the above garnets  $(Grt_1 - Grt_4)$  have grown across different effective bulk compositional micro-domains: (1) why is the evidence for continuation of theses different effective bulk compositional micro-domains is absent in the matrix? (2) why do the garnets not show chemical zonation at silica-saturated and silica-deficient minerals bearing domains?

#### 6.3. Disappearance of compositional domains in the matrix

The answer to the first question above is the high temperature associated multiple deformation history of the Highland Complex, which could probably obliterate the evidence of the existence of different compositional lamellae in the rock. Number of workers (e.g. Berger and Jayasinghe, 1976; Yoshida et al., 1990; Kriegsman, 1991, 1994; Kehelpannala, 1991, 1997) suggested that the Sri Lankan basement suffered multiple deformation events.

Most of above workers (Berger and Jayasinghe, 1976; Kriegsman, 1991, 1994; Kehelpannala, 1997) have suggested that the present major gneissic foliation and major lineation ( $S_2$  and  $L_2$  respectively) in the HC granulites have been resulted during the  $D_2$ deformation event simultaneously with the peak metamorphism of HC during assembly of Gondwana (e.g Kehelpannala, 2004; Kröner et al., 2003). The evidence for early foliation ( $S_1$ ) represented in some of mineral inclusions as trails indicating a well developed crenulation, resulted during  $D_1$  deformation event in the host garnets (Yoshida et al., 1990; Kehelpannala, 1991, 1997; Kreigsman, 1991) and some others, mainly sillimanite needles,

display an internal folding or crenulations (Kehelpannala, 1991; Kriegsman, 1991; Kröner et al., 1994b). Presence of mineral lineation, which is oblique to major matrix lineation  $L_2$  in some garnets, could represent the early mineral lineation  $L_1$  (Yoshida, 1990; Kriegsman, 1991, 1994; Kehelpannala, 1997).  $D_2$  deformation event may have had two main phases (Kehelpannala, 1997): the first phase is less strong and resulted an early phase producing crenulation folds (some of which are now included in garnet porphyroblasts) and small scale crenulation fold hinges found in metapelites, and later the most strong ductile deformation caused the formation of strong stretching lineation ( $L_2$ ) and prominent foliation ( $S_2$ ).

Suggested  $D_1$  deformation, which predates the peak granulite facies metamorphism in the HC, has been interpreted as being due to some early tectonic event. Some rare textural evidences preserved in garnet porphyroblasts indicate that the HC metasediments have entered from sillimanite stability field to kyanite stability field during prograde evolution probably due to the crustal thickening (Raase and Schenk, 1994; Malaviarachchi and Takasu, 2011a; Dharmapriya et al., 2014b, 2015b).

All of our studied garnets are formed by dehydration reactions during prograde metamorphism. Hence growths of theses garnets have taken place under  $D_1$  and early stage of  $D_2$  events, which took place prior to peak metamorphism. Therefore, during the  $D_1$  event, the crustal thickening may have caused strong flattening and transport of metasediments into the lower crust. During this crustal thickening event the original sedimentary sequence in the HC have probably been continuously destroyed (Kröner et al., 1994b; Kehepannala, 1997, 2003). Although the original sequence of the sedimentary units could be destroyed by tectonic stresses, there is a tendency to preserve original palaeobedding/laminations of the sediment sequence in the macro to micro scale (e.g. Perera, (1994) and Kehelpannala, (1997, 2003).

There is sufficient evidence in studied Rock A, C and D for rotation of the foliation during the prograde evolution. The mantle area of Grt<sub>1</sub> contains rotated crenulation lineation

demarcated by biotite and tiny sillimanite needles (Fig. 2b, c, d), which is oblique to the major matrix lineation of the rock. In Rock C as well, some of the garnet contains rotated crenulation lineation demarcated by sillimanite needles (Supplementary file 2g), which is oblique to the major matrix lineation of the rock. In Rock D, Type 1 garnet contains mineral lineation, which is demarcated by preferred oriented quartz grains are now oblique to major lineation  $L_2$  in the rock matrix (Supplementary file 2i, j). Further, existence of curved quartz grains in core to mantle of garnet (Type 3; Supplementary file 2l) also provides evidence for rotation of the foliation during prograde metamorphism.

However, at the late stage of the  $D_2$  deformation (Kehepannala, 1997), the original sedimentary sequence probably have been destroyed and reworked due to strong tectonic stresses and associated granulite facies metamorphism. Estimated peak metamorphic *P-T* conditions of all of above studied rocks are 9-10 kbar and 900 – 975 <sup>o</sup>C. However, even under such higher temperature conditions a considerable amount of melt can be produced, as clearly seen by field and textural evidence (e.g. existence of in-situ melts in the UHT granulites in the HC) coupled with pseudosection calculations (e.g. Dharmapriya et al., (2015a, b, 2016)). Some of these generated melt fraction could escape from the system making considerable changes in the bulk chemical composition of the ordinal rock. Remaining fraction of melt phases may have contributed to rework the compositions of the original rock in macro to micro scales. Further, loss of some melt phases from the system and crystallizations of new minerals from the existing melt phases during cooling after peak metamorphism could also change the original matrix structures and compositions in the studied pelitic granulites. In this context, the representative illustration interpreting the formation of various garnet grains from Grt<sub>1</sub> to Grt<sub>4</sub> is shown in Fig. 7.

#### 6.4. Chemical Homogenization of the garnets

The absence of compositional difference across the garnet in silica-saturated and silica-deficient mineral bearing domains could be due to the self-diffusion of the garnet at high to UHT metamorphism. Further, growth zoning of garnet are frequently obliterated in high-grade garnet due to volume diffusion (Blackburn, 1969, Grant and Weiblen, 1971, Tracy et al. 1976, Yardley, 1977, Tuccillo et al., 1990, Fernando et al, 2003). Since the rates of self-diffusion of cations in silicates commonly increase exponentially with temperature (Yardley, 1977), the studied garnets could lead to chemical homogenization across the garnet grain-profile in quartz saturated and quartz deficient mineral bearing domains.

#### 7. Conclusion

The textural evidence of the studied garnets indicates that their evolution has taken place via different dehydration reactions during prograde metamorphism. These dehydration reactions have taken place in silica-saturated and silica-deficient microenvironments of the studied rocks. Due to incorporation of mineral inclusions, which represent both silicasaturated and silica-deficient domains in same garnets, we speculate that these garnets have continued to grow across different effective bulk compositional microdomains. Due to strong ductile deformation-associated UHT metamorphism, metamorphic differentiation and crystallization of in-situ melt, the existing compositional microdomains may have disappeared and modified resulting in more homogeneous matrix within the rocks. The absence of compositional difference across the garnet grown in silica-saturated and silicadeficient domains could be due to the chemical homogenization via self diffusion of elements at high to UHT metamorphism.

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#### **Figure and Table Captions**

#### **Figure captions**

**Fig. 1.** Geological map of Sri Lanka showing the lithotectonic subdivision (after Cooray, 1994). A star shows sampling locality and circles show UHT localities reported so far: 1. Osanai (1989): T = 900 °C; 2 & 3. Kriegsman and Schumacher (1999): T = 830 °C (here it is corrected as 950 °C); 4. Sajeev and Osanai (2004a): T = 950 °C: 5. Sajeev and Osanai (2004b): T = 1150 °C; 6. Osanai et al. (2006): T = 1000 °C and 7. Sajeev et al. (2007): T = 925 °C. 8 Dharmapriya et al. (2015a): 950-975 °C, (b) Detailed geological map (modified after Kröner et al., 2003) and the sampling localities.

**Fig. 2.** Microtextures of Grt<sub>1</sub> in Rock A. (b-h: Microtextures of the first Grt<sub>1</sub> grain shown in image (a) in garnet rich layer, i-l Microtextures of the second Grt<sub>1</sub> grain shown in the garnet rich layer, m-o: Microtextures of the third Grt<sub>1</sub> grain sown in image (n) in the garnet rich layer). (a) a petrographic thin section representing the garnet rich layer, (b) a mosaic of BSE images of a Grt<sub>1</sub> in which the quartz-deficient core domain and quartz saturated mantle to rim domains are shown, (c) CPL image showing the inclusion phases at the boundary of silica-saturated and silica-deficient domains, (d) closer view of the core area of the garnet in image (c), (e) inclusions of sapphirine, spinel, silimanite and quartz, (f) tiny inclusion phases of garnet coexisting with sapphirne+spinel (g) coexistence of spinel and sillimanite. Gedrite is also present, (h) formation of biotite+plagioclase corona around garnet, (i) a mosaic of rim of one side to rim of opposite side (cross the core) of a garnet, (j) the image representing the inclusion phases close to the boundary of silica-saturated and silica-deficient domains, (k) BSE image of core area of the garnet, spinel. Tiny sillimanite are also present, (l) closer view of the center part of the image (k) where coexistence of gedrite+sapphirine and gedrite+spinel

are present, (m) presence of clusters of spinel-sapphirine-sillimanite toward the rim area of garnet, (n) a thin section representing the garnet of image (o) a mosaic of the garnet in which the rim area of one side contains a silica-deficient domain and the rest of the portion representing a silica saturated domain.

**Fig. 3.** Microtextures of  $Grt_2$  in Rock B, (a) mineral inclusions from core to rim (b) BSE image of the image (a), (c) BSE image representing mineral inclusions in the core area, (d) tiny isolated sillimanite inclusion at the rim area.

**Fig. 4** Microtextures of Grt<sub>3</sub> in Rock C, (a) the whole view of the studied garnet, (b) BSE image showing the inclusion phases of core to rim area, (c) the sapphirine and spinel bearing inclusion domain, (d) BSE image showing the sapphirine and spinel bearing domain, (e) BSE image showing coexistence of sapphirine+sillimanite, (f) BSE image showing coexistence of spinel+sillimanite

**Fig. 5.** Petrography of  $Grt_4$  in Rock D (images b-d show the textures of the  $Grt_4$  of the image (a); images i-m shows the textures of the  $Grt_4$  of the image (h), (a) a petrographic thin section showing a  $Grt_4$  (b) a mosaic showing anhedral corundum and sillimanite clusters which pseudomorph kyanite in the core area, (b) aggregate of fibrolitic sillimanite pseudomorphing kyanite, (c) isolated kyanite grain towards the rim are. An isolated quartz grain also present close to kyanite, (e) tiny spinel inclusions and corundum inclusions in coarse prismatic sillimanite; a staurolite grain is also present in the bottom right hand side of the image, (f) an isolated staurolite grain, (g) formation of pyrophyllite towards the rim, (h) petrographic thin section of another phorphyroblastic garnet grain (i) core existence of corundum and sillimanite clusters which pseudomorph kyanite in the mantle area, (j) tiny inclusion of quartz

close to kyanite pseudomorph after sillimanite, (k) isolated cluster of sillimanite pseudomorphing kyanite (l) close association of isolated quartz and cluster of sillimanite pseudomorphing kyanite, (m) isolated quartz, staurolite and cluster of sillimanite pseudomorphing kyanite.

**Fig. 6.** *P-T* pseudosection represented in FMASH system using local rock compositions calculated from mineral analyses and estimated abundances for specific garnet  $Grt_4$  in Rock D under different bulk SiO<sub>2</sub> content: (a) at SiO<sub>2</sub> = 38.01 mole %, (b) SiO<sub>2</sub> = 38.5 mol %, (c) SiO<sub>2</sub> = 40.8 mol %

**Fig. 7.** A cartoon illustrating the evolution of porphyroblastic garnets across alternating silica-saturated and silica-deficient microdomains. See the rotation of the foliation of each rock during the progress of the garnet growth.

#### **Table captions**

**Table 1.** Representative EPMA data of the studied garnets

**Table 2.** Representative EPMA data of biotite, staurolite, spinel, sapphirine and corundum inclusions in the studied garnets

Table 3. Reaction and mass balance of staurolite breakdownin FASH system.

#### Appendices

**Supplementary file 1.** Field relations of studied rocks: (a) The quarry from which Rock A and C collected, (b) a hand specimen of Rock A, (c) Thin section representing garnet rich layer of Rock A, (d) The quarry from which Rock B was collected, (e) a hand specimen of

Rock B, (f) a large rock fragment of Rock C, (g) a hand specimen of Rock C, (h) the road exposure form which the Rock D was collected, (i) a hand specimen of Rock C.

**Supplementary file 2**. Images a & b from rock A; c-f from rock B, g & h from rock C; ifrom rock D, (a) a garnet grain away from the garnet rich layer, (b) the matrix of the rock A, (c) the matrix of the rock B, (d) orthopyroxene+sillimanite corona and orthopyroxene+cordierite symplectites around the garnet, (e) exsolved hematite-ilmeniterutile, (f) a sapphirine grain surrounded by plagioclase in the matrix, (g) a garnet grain free of any symplectite. See the orientation of sillimanite and rotated crenulations lineation inside the garnet, (h) the matrix of the rock C, (i) a sketch diagram showing inclusion phases, distributed from rim to core of garnet Type 1, (j) oriented quartz grains in the core area of the garnet Type 1, (k) distribution of inclusion phases from rim to core under PPL, (l) snowball garnet (Type 3), (m) inclusion phases of garnet Type 4.

**Supplementary file 3.** Raman spectra with characteristics peaks. (a) Kyanite enclosed by Grt<sub>2</sub>; (b) kyanite enclosed by Grt<sub>4</sub>

#### Supplementary file 4. Analytical procedures at three EPMA laboratories

	-	Rock A			Rock B			Rock C		_	Rock D	
	Core	Mantle	Rim	Core	Mantle	Rim	Core	Mantle	Rim	Core	Mantle	Rim
SiO <sub>2</sub>	39.98	39.50	39.50	40.61	40.08	40.30	40.58	39.29	39.73	36.82	36.24	36.73
TiO <sub>2</sub>	0.05	0.00	0.05	0.05	0.00	0.02	0.02	0.02	0.00	0.04	0.03	0.03
Al <sub>2</sub> O <sub>3</sub>	22.28	22.43	22.03	22.64	22.89	22.46	22.69	22.43	22.43	20.98	21.09	21.10
Cr <sub>2</sub> O <sub>3</sub>	0.00	0.02	0.00	0.04	0.04	0.00	0.04	0.01	0.00	0.01	0.01	0.02
FeO	23.97	25.37	26.26	21.97	22.28	22.02	22.18	23.27	24.80	33.77	34.29	34.43
MnO	0.58	0.65	0.71	2.02	1.93	2.32	0.51	0.65	0.89	3.51	3.34	3.11
MgO	11.53	10.93	9.83	12.25	13.16	12.99	14.00	13.27	12.07	3.35	3.49	3.65
CaO	2.29	1.95	1.36	1.87	1.07	1.03	0.78	0.73	0.68	2.18	1.63	1.52
Na <sub>2</sub> O	0.03	0.04	0.02	0.00	0.01	0.02	0.00	0.01	0.00	0.03	0.01	0.00
K <sub>2</sub> O	0.00	0.00	0.02	0.00	0.00	0.00	0.00	0.01	0.00	0.00	0.00	0.00
Total	100.71	100.89	99.78	101.5	101.45	101.16	100.80	99.69	100.60	100.69	100.13	100.59
0	12	12	12	12	12	12	12	12	12	12	12	12
Si	3.005	2.984	3.024	3.013	2.974	3.000	3.005	2.968	2.992	2.958	2.933	2.952
Ti	0.003	0.000	0.003	0.003	0.000	0.001	0.001	0.001	0.000	0.002	0.002	0.002
Al	1.974	1.997	1.988	1.980	2.002	1.970	1.981	1.997	1.991	1.987	2.012	1.999
Cr	0.000	0.001	0.000	0.002	0.002	0.000	0.002	0.001	0.000	0.001	0.001	0.001
Fe	1.507	1.603	1.682	1.363	1.383	1.371	1.374	1.470	1.562	2.269	2.321	2.314
Mn	0.037	0.042	0.046	0.127	0.121	0.146	0.032	0.042	0.057	0.239	0.229	0.212
Mg	1.292	1.230	1.122	1.355	1.455	1.441	1.545	1.494	1.355	0.401	0.421	0.437
Ca	0.184	0.158	0.112	0.149	0.085	0.082	0.062	0.059	0.055	0.188	0.141	0.131
Na	0.004	0.006	0.003	0.000	0.001	0.003	0.000	0.001	0.000	0.005	0.001	0.000
K Tet	0.000	0.000	0.002	0.000	0.000	0.000	0.000	0.001	0.000	0.000	0.000	0.000
cation	8.007	8.020	7.981	7.993	8.024	8.015	8.002	8.033	8.012	8.049	8.060	8.047
Fe <sup>3+</sup>	0.021	0.060	0.000	0.000	0.072	0.045	0.006	0.100	0.036	0.048	0.178	0.140
Fe <sup>2+</sup>	1.486	1.542	1.682	1.355	1.311	1.326	1.367	1.370	1.526	2.220	2.142	2.174
Alm	0.496	0.519	0.576	0.459	0.441	0.443	0.455	0.462	0.510	0.728	0.730	0.736
Spe	0.012	0.014	0.015	0.042	0.041	0.049	0.011	0.014	0.019	0.078	0.078	0.072
Pyr	0.431	0.414	0.372	0.449	0.490	0.481	0.514	0.504	0.453	0.132	0.143	0.148
Grs	0.052	0.034	0.138	0.061	0.013	0.015	0.017	0.006	0.009	0.044	0.017	0.017
$X_{Mg}$	0.46	0.43	0.40	0.50	0.51	0.51	0.53	0.50	0.46	0.15	0.15	0.16
	R R											

## Table 1. Representative EPMA data of studied garnets

		Bt		St	Sp	ol		Spr		Crn
	Rock	Rock	Rock	Rock	Rock	Rock	Rock	Rock	Rock	Rock
	Α	В	C	D	Α	C	Α	В	C	D
SiO <sub>2</sub>	37.42	36.78	36.72	23.29	0.12	0.09	12.16	13.24	12.60	0.01
TiO <sub>2</sub>	6.77	5.01	6.39	1.55	0.02	0.01	0.03	0.07	0.07	0.01
$Al_2O_3$	16.61	16.34	16.76	57.87	63.20	63.70	64.65	60.45	61.51	100.62
$Cr_2O_3$	0.00	0.03	0.00	0.13	0.08	0.04	0.02	0.11	0.00	0.04
FeO	9.13	9.02	9.88	11.23	19.13	14.59	7.01	9.48	9.78	0.43
MnO	0.03	0.07	0.05	0.05	0.08	0.00	0.06	0.04	0.05	0.00
MgO	15.69	18.01	16.02	2.55	14.43	15.52	16.01	16.61	15.23	0.00
CaO	0.01	0.00	0.02	0.05	0.03	0.00	0.01	0.01	0.04	0.00
Na <sub>2</sub> O	0.47	0.09	0.42	0.00	0.02	0.00	0.03	0.04	0.00	0.00
$K_2O$	9.45	9.78	9.56	0.00	3.14	0.01	0.02	0.00	0.00	0.00
ZnO	0.00	0.00	0.00	1.56	314	5.59	0.00	0.00	0.00	0.00
Total	95.58	95.12	95.82	98.28	100.26	99.54	100.01	100.05	99.29	101.12
0	22	22	22	23	4	4	10	10	10	3
Si	5.419	5.367	5.336	3.230	0.003	0.002	0.722	0.796	0.763	0.000
Ti	0.737	0.549	0.698	0.162	0.000	0.000	0.001	0.003	0.003	0.000
Al	2.835	2.809	2.871	9.461	1.960	1.972	4.521	4.283	4.393	1.995
Cr	0.000	0.003	0.000	0.014	0.002	0.001	0.001	0.005	0.000	0.001
Fe	1.106	1.101	1.201	1.303	0.421	0.320	0.348	0.477	0.496	0.006
Mn	0.004	0.009	0.006	0.006	0.002	0.000	0.003	0.002	0.003	0.000
Mg	3.386	3.916	3.469	0.527	0.566	0.607	1.416	1.488	1.376	0.000
Ca	0.002	0.000	0.003	0.007	0.001	0.000	0.001	0.001	0.003	0.000
Na	0.132	0.025	0.118	0.000	0.000	0.000	0.003	0.005	0.000	0.000
Κ	1.746	1.820	1.772	0.000	0.001	0.000	0.002	0.000	0.000	0.000
Zn	0.000	0.000	0.000	0.160	0.061	0.108	0.000	0.000	0.000	0.000
Total	15 266	15 600	15 176	14.970	2.055	2 002	7.019	7.050	7 027	2 002
cation	15.300	15.600	15.470	14.870	2.955	2.903	/.018	7.059	7.037	2.002
ХMg	0.75	0.78	0.74	0.29	0.57	0.66	0.80	0.76	0.74	
			7							

**Table 2.** Representative EPMA data of biotite, staurolite, spinel, sapphirine and corundum inclusions in studied garnets

Table 3 Reaction and mass balance of staurolite breakdown reaction in FASH

	Grt	Ку	Crn	H <sub>2</sub> O	St
FASH end-member system (molar)					
FeO	3	0	0	0	4
$Al_2O_3$	1	1	1	0	9
SiO <sub>2</sub>	3	1	0	0	7.5
H <sub>2</sub> O	0	0	0	1	2
Reaction coefficient	1.333	3.5	4.167	2	-1
molar V	11.526	4.404	2.545		44.88
Observed mode (V %) Observed mode	60	25	14		1
(molar)	5.2	5.7	5.5		0.02
Original mode (molar)	3.45	1.06	0		1.34
Normalised	0.23	0.07	0		0.09



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# Highlights

- Incorporation of alternating silica-saturated and silica deficient inclusion phases in single grain garnet porphyroblast are reported
- Independent silica-saturated and silica-deficient dehydration reactions which govern the growth of such complex garnets during prograde metamorphism are described
- A model to explain the occurrences of such complex garnets has been discussed.

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