

Reactions and textures in grossular–wollastonite–scapolite calc–silicate granulites from Maligawila, Sri Lanka: evidence for high-temperature isobaric cooling in the meta-sediments of the Highland Complex

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Abstract

Grossular–wollastonite–scapolite calc–silicate granulites from Maligawila in the Buttala klippe, which form part of the overthrust rocks of the Highland Complex of Sri Lanka, preserve a number of spectacular coronas and replacement textures that could be effectively used to infer their P – T –fluid history. These textures include coronas of garnet, garnet–quartz, and garnet–quartz–calcite at the grain boundaries of wollastonite, scapolite, and calcite as well as calcite–plagioclase and calcite–quartz symplectites or finer grains after scapolite and wollastonite respectively. Other textures include a double rind of coronal scapolite and coronal garnet between matrix garnet and calcite. The reactions that produced these coronas and replacement textures, except those involving clinopyroxene, are modelled in the $\text{CaO}–\text{Al}_2\text{O}_3–\text{SiO}_2–\text{CO}_2$ system using the reduced activities. Calculated examples of T – X_{CO_2} and P – X_{CO_2} projections indicate that the peak metamorphic temperature of about 900–875 °C at a pressure of 9 kbar and the peak metamorphic fluid composition is constrained to be low in X_{CO_2} ($0.1 < X_{\text{CO}_2} < 0.30$). Interpretation of the textural features on the basis of the partial grids revealed that the calc–silicate granulites underwent high-temperature isobaric cooling, from about 900–875 °C to a temperature below 675 °C, following the peak metamorphism. The late-stage cooling was accompanied by an influx of hydrous fluids. The calc–silicate granulites provide evidence for high-temperature isobaric cooling in the meta-sediments of the Highland Complex, earlier considered by some workers to be confined exclusively to the meta-igneous rocks. The coronal scapolite may have formed under open-system metasomatism. © 2001 Elsevier Science B.V. All rights reserved.

Keywords: Calc–silicate granulite; Reaction textures; Isobaric cooling, Highland Complex of Sri Lanka

Abbreviations: Alm, almandine; An, anorthite; Cc, calcite; Gros, grossular; Me, meionite; Pl, plagioclase; Qtz, quartz; Scp, scapolite; Wo, wollastonite.

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1. Introduction

Determination of P – T – t evolution and fluid regime during peak metamorphism of granulites is fundamental in formulating geodynamic models for the origin and evolution of the continental crust. Several recent studies have shown that calc–silicate granulites are very useful rocks to constrain P – T –fluid histories of granulites as they preserve a variety of reaction textures, high-temperature mineral parageneses, and fluid histories. These studies have come mainly from granulite terranes of Australia, Antarctica, and India (Warren et al., 1987; Motoyoshi et al., 1991; Harley et al., 1994; Bhowmik et al., 1995; Shaw and Arima, 1996; Satish-Kumar and Santosh, 1998 and the references cited therein). The interpretation of mineral parageneses and reaction textures in calc–silicate granulites, in terms of P – T –fluid evolution, is made possible by compilation of relevant thermodynamic data and the construction of a new set of reaction grids in the CaO–Al₂O₃–SiO₂–vapour (CASV) system (Holland and Powell, 1990, 1998; Moecher and Essene, 1990, 1991; Harley and Buick, 1992; Harley et al., 1994).

Petrological studies on calc–silicate granulites are of special interest to the granulite terranes of India and Sri Lanka, where the formation of incipient charnockites has been shown to be related to influx of CO₂, and in contrast, studies on calc–silicate granulites suggest low X_{CO₂} during peak metamorphism (Newton and Smith, 1980; Hansen et al., 1987; Santosh et al., 1990; Newton, 1992; Bhowmik et al., 1995; Shaw and Arima, 1996; Satish-Kumar and Santosh, 1998). The present study focuses on mineral chemistry, reaction textures, and P – T –fluid evolution in a suite of grossular–wollastonite–scapolite calc–silicate rocks from Maligawila, Sri Lanka and provides evidence for high-temperature isobaric cooling in the meta-sediments, previously considered by some workers to be confined exclusively to the meta-igneous rocks.

2. Geological setting

2.1. Background geology of Sri Lanka

The Proterozoic basement of Sri Lanka is made-up of three distinct crustal units identified on the basis of

distribution of Nd model ages of a variety of lithologies, viz., the Highland Complex (HC) (2.0–3.4 Ga), the Wannai Complex (WC) (1.1–1.8 Ga), and the Vijayan Complex (VC) (1.1–1.8 Ga) (Fig. 1) (Milisenda et al., 1988, 1994; Cooray, 1994). The centrally located HC comprises of granulite grade meta-sediments, orthogneiss, and meta-igneous charnockitic rocks. The meta-sediments, which include quartzite, marble, pelitic gneiss and migmatite, and calc–silicate granulite, are intimately associated with the charnockitic rocks in the field on all scales (Cooray, 1962, 1984). The WC, which lies west of the HC, consists of granulite and amphibolite grade rocks, somewhat similar to those of the HC but characterized by ‘younger’ Nd model ages. Spectacular exposures showing incipient charnockitization are found in several localities in both units.

The combined HC–WC unit represents a tilted section of the former lower–middle crust. The peak metamorphic pressure was in excess of 9 kbar in the eastern and south-eastern parts of the HC, whereas there is a marked decrease in paleo-pressure towards the west, and in the most western parts it was about 4–5 kbar (Schumacher et al., 1990; Faulhaber and Raith, 1991; Prame 1991; Raase and Schenk, 1994). The peak metamorphic temperature ranges between 700 and 900 °C in both units but the higher values are found near the eastern and south-eastern parts of the HC.

Two contrasting P – T – t paths, one for the meta-sediments and the other for the meta-igneous rocks, have been constructed for the HC rocks (Fig. 2). A clockwise P – T – t path has been inferred for the meta-sediments on the basis of (1) rare early kyanite and staurolite, (2) abundant peak sillimanite, (3) breakdown of garnet to cordierite-bearing assemblages, and (4) late andalusite (Perera, 1984; Sandiford et al., 1988; Hiroi et al., 1994; Ellis and Hiroi, 1997). The clockwise path suggested by Raase and Schenk (1994) and Kriegsman and Schumacher (1999) consists of a near isobaric prograde heating segment followed by a strong near isothermal decompression. In contrast, an isobaric cooling path followed by decompression with decreasing temperature, is inferred for the meta-igneous rocks based on garnet coronas around orthopyroxene and plagioclase and breakdown of garnet to orthopyroxene–plagioclase symplectites (Schumacher et al., 1990; Schenk et al., 1991; Prame, 1991). Hiroi et al. (1994) attributed

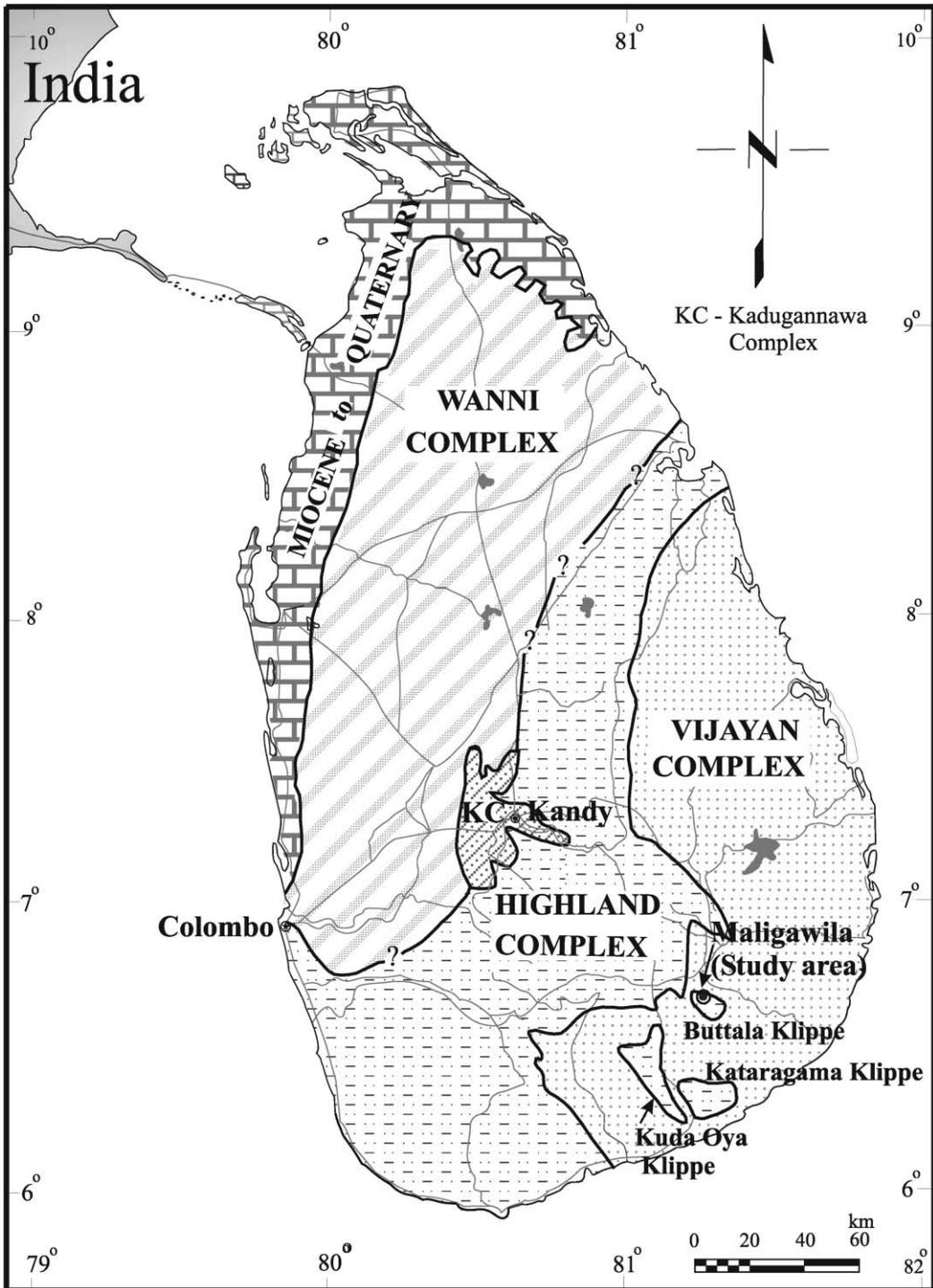


Fig. 1. Simplified geological map of Sri Lanka showing the Proterozoic crustal units (the Highland Complex, the Wannai Complex, and the Vijayan Complex and the three klippen of the Highland Complex within the Vijayan Complex. The Kadugannawa Complex is a very much smaller unit delimited on the basis of lithology.

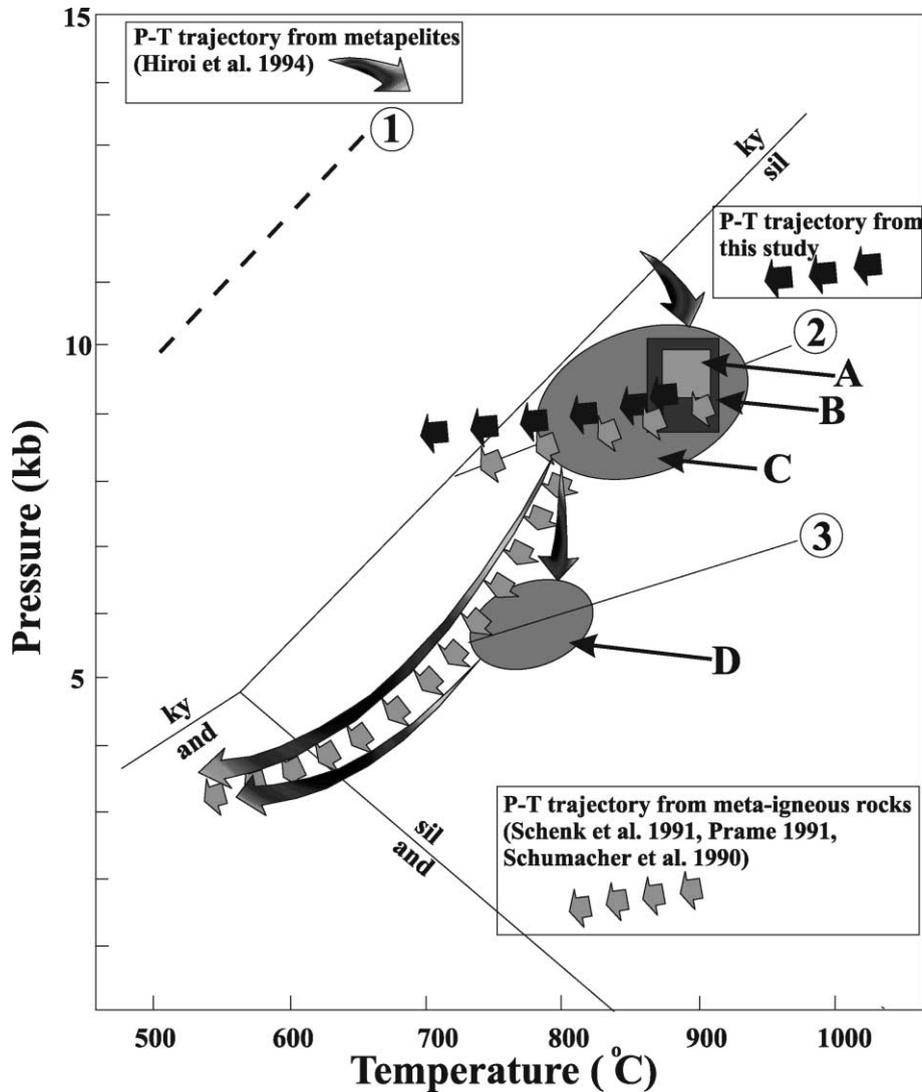


Fig. 2. Shows the P – T trajectories determined previously for the meta-igneous rocks and the meta-sediments of the Highland Complex (modified after Hiroi et al. (1994) and the P – T trajectory inferred for the calc–silicate granulites in the present study are shown. The P – T trajectories with a prograde heating segment determined for the metapelites by Raase and Schenk (1994) and Kriegsman and Schumacher (1999) are not shown. The univariant reaction curves that have been used to determine the trajectories are omitted for clarity. A=peak metamorphic condition determined for the calc–silicate granulites in the present study, B=estimated peak metamorphic condition for the rocks from south eastern HC including the klippen (Prame, 1991; Faulhaber and Raith, 1990), C=estimated peak metamorphic condition for the rocks from Kandy area, and D=estimated peak metamorphic condition of the rocks from the Colombo area. The dashed line 1 represents the equilibrium conditions between garnet ($X_{\text{Grs}}=0.08$), plagioclase ($X_{\text{an}}=0.1$), kyanite, and quartz. Reaction curves 2 and 3 show equilibrium conditions between garnet, rutile, sillmanite, ilmenite, and quartz (source of data—Hiroi et al., 1994).

diverse deep crustal processes, such as magmatic underplating of continental crust by sediments and magmas at or near convergent plate boundaries, for the inferred contrasting P – T – t paths.

The VC bounds the HC on the east and consists mainly of amphibolite grade granitic and basic gneisses and migmatite characterized by ‘younger’ Nd model ages similar to that of the WC.

2.2. Buttala klippe

Studies have shown that the combined HC–WC unit was overthrust after the peak metamorphism, on to the VC (Silva et al., 1980; Vitanage, 1985; Kröner et al., 1991; Kleinschrodt, 1994) and three klippen of the HC, viz., the Buttala klippe, the Kuda Oya klippe, and the Kataragama klippe occur within the VC (see Fig. 1). The Buttala klippe occurs in the form of two tongue-shaped rotated bodies and consists mainly of marble, pelitic rocks, quartzite, basic migmatite, calc–silicate rock, and minor pegmatite (Kallubandara, 1998). The studied calc–silicate granulite samples were collected from an isolated outcrop ($\sim 15 \times 10$ m), near Maligawila in the Buttala klippe. This outcrop was originally discovered by gem miners in 1997 and produced gem-quality grossular garnet (primary hessonite) but is now completely exhausted (see Mathavan et al., 2000). The ore is moderately weathered and the nearest outcrop, a hornblende–biotite migmatitic gneiss, is found about 100 m away.

3. Petrography

The calc–silicate body is fine- to medium-grained, dark-coloured, and generally massive with occasional centimeter-size mono-mineralic layers of garnet. It shows considerable mineralogical variation, particularly of garnet, scapolite, and clinopyroxene. Two distinct peak assemblages are recognised in the body: (I) grossular–wollastonite–scapolite–clinopyroxene±titanite and (II) grossular–wollastonite–scapolite–clinopyroxene–calcite–quartz±titanite. Both calcite and quartz are absent in assemblage I but locally quartz inclusion is found in wollastonite and scapolite. Primary plagioclase is absent in both assemblages. Rarely, garnet (matrix) occurs as small inclusion in scapolite. The grains in the two assemblages generally define a well-developed polygonal granoblastic texture, indicating grain boundary adjustments following or during peak metamorphism (Fig. 3a). Sometimes, garnet shows poikiloblastic habit and encloses scapolite, clinopyroxene, and wollastonite in assemblage I and additionally calcite, and quartz in assemblage II. Most of these enclosed grains, particularly cluster of scapolite grains, also show polygonal granoblastic texture. With the exception of quartz and calcite in assemblage

II, all other minerals including titanite, form coronal rims, besides forming the polygonal matrix grains (see below).

3.1. Retrograde coronas and textures

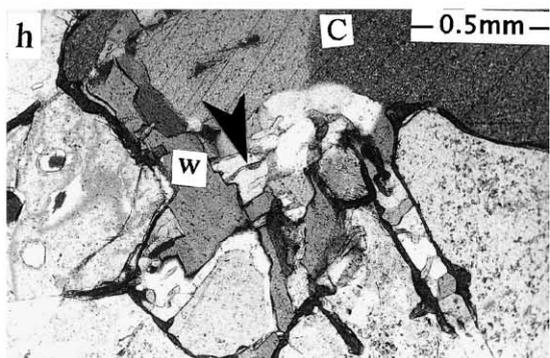
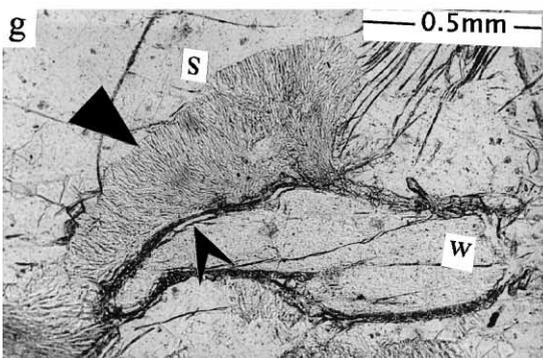
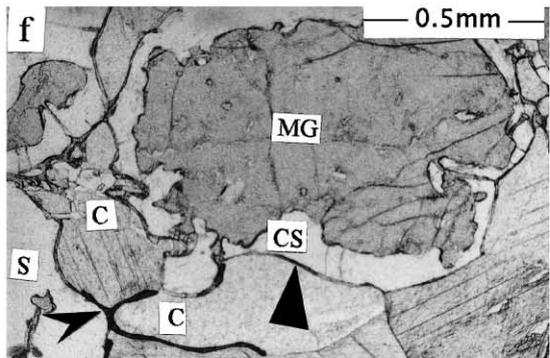
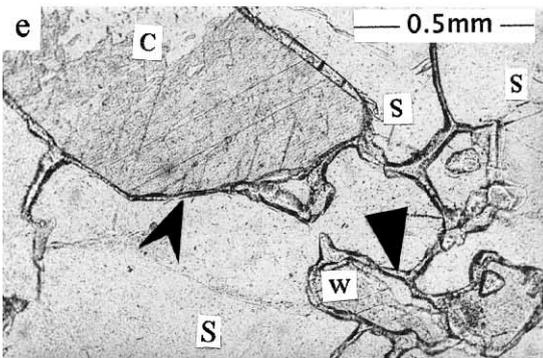
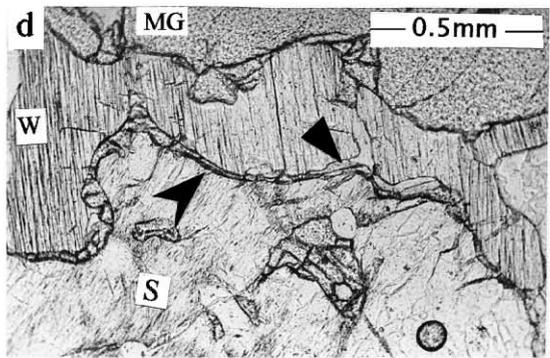
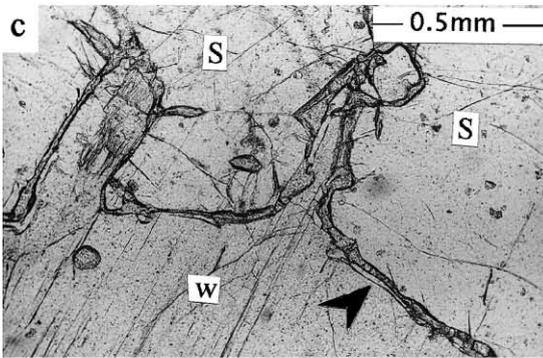
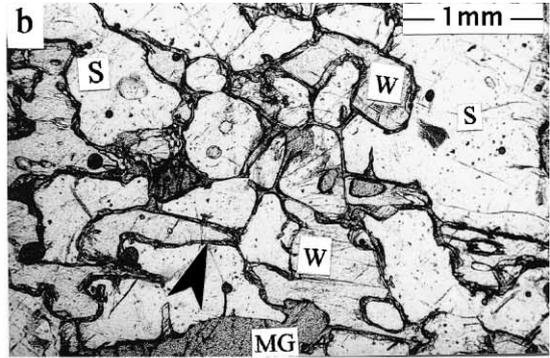
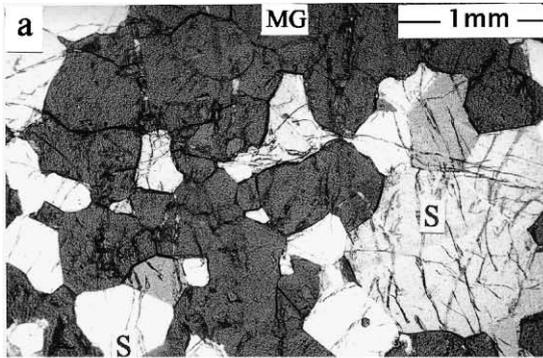
Several spectacular coronas and replacement textures, localized to grain boundaries or within individual grains, overprint the granoblastic texture defined by the peak assemblages (Fig. 3b and e). Five types of coronas and two types of replacement textures have been identified.

(1) In both assemblage I and II, composite garnet–quartz coronas are developed, locally, between wollastonite and scapolite (Fig. 3c, e and g). Quartz in the coronas is always found next to wollastonite and occurs commonly as isolated blebs but occasionally as a thin lamina. The modal amount of the coronal garnet is very low, 1–2%, while that of the quartz is even lower.

(2) Rare, composite garnet–quartz–calcite coronas are found locally at the grain boundary between scapolite and wollastonite in assemblage II (Fig. 3d). Again the modal amount of coronal garnet is low and the volume ratio of garnet/quartz/calcite in the corona is about 20:2:1. Blebs of calcite associated with this type of composite coronas differ morphologically from the vermicular calcite found in the breakdown textures of scapolite and the fine granular grains produced during the breakdown of wollastonite (see below).

(3) Garnet coronal rims, the commonest of the garnet corona types in the two assemblages, are found between calcite and scapolite, scapolite and wollastonite, and scapolite–scapolite grain boundaries (Fig. 3b and e). In some rare instances, a rim of titanite occurs between calcite and scapolite in assemblage II, instead of or continuous with the coronal rim garnet (Fig. 3f).

(4) Textural relations suggest that subsequent to the development of both composite and single garnet coronas, both scapolite and wollastonite in assemblage I and II be partially or completely replaced by retrograde products. Scapolite breaks down to a symplectic intergrowth of vermicular calcite and plagioclase (Fig. 3g). The intergrowth has a lobate shape in partially replaced scapolite grains and may occur next to the coronal garnet, suggesting that its development post-dates the growth of coronal garnets (cf. Fig. 3g).



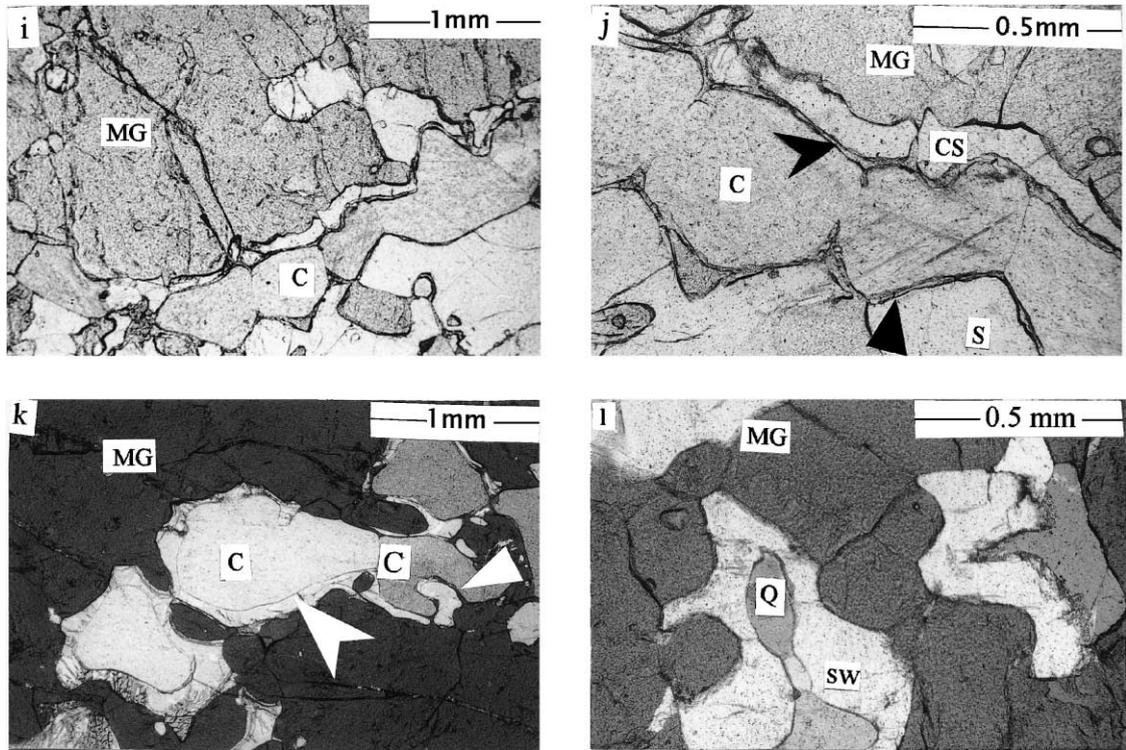


Fig. 3. Photomicrographs of the calc–silicate granulites. (a) Polygonal granoblastic texture defined mainly by scapolite (S) and matrix garnet (MG) (oblique polars). (b) Coronal garnet (arrow-head) overprinting polygonal granoblastic texture defined by wollastonite (W), scapolite (S), and matrix coronal garnet (MG). Note quartz inclusions in wollastonite, and scapolite (single polar). (c) Composite garnet–quartz corona (arrow-head) between scapolite (S) and wollastonite (W) (single polars). (d) Composite garnet–quartz–calcite (triangles) corona between wollastonite (W) and scapolite (S). Note (1) small calcite–quartz grains near the triangles; (2) the large matrix garnet (GM) and inclusions in scapolite (single polar). (e) Coronal garnet (arrow-head) between calcite (C) and scapolite (S) and scapolite–scapolite grain boundaries (upper right-hand corner). Note garnet–quartz corona (triangle) between wollastonite (W) and scapolite (single polar). (f) Partial rim of titanite (arrow-head) at the grain boundaries of calcite(c)–calcite–scapolite (S). Note (1) embayed matrix garnet (MG) and the development of coronal scapolite (CS) between matrix garnet and calcite (C); (2) coronal garnet (triangle) between coronal scapolite (CS) and calcite (oblique polars). (g) Partial breakdown of scapolite (S), which is next to garnet–quartz corona (arrow-head), to calcite–plagioclase symplectite (triangle) (oblique polars). (h) Breakdown of wollastonite (W) to calcite–quartz symplectite (arrow-head) and finer grains of calcite–quartz (triangle). Note the rim of coronal garnet on the pseudomorphed wollastonite grain (triangle) (oblique polars). (i) Coronal scapolite (arrow-head) between matrix garnet (MG) and calcite (C) and coronal garnet (oblique polars). (j) Enlarged central part of Fig. 3(i). Note (1) the double rind of coronal scapolite (CS) and coronal garnet (arrow-head) between matrix garnet (MG) and calcite (C); (2) coronal garnet (triangle) between matrix scapolite (S) and calcite (C) (single polar). (k) Coronal scapolite (arrow-head) within matrix garnet (MG) and enclosed calcite grains (C). Note thin coronal garnet (triangle) between calcite and coronal scapolite. (l) Secondary wollastonite (SW) within matrix garnet (MG) and enclosed quartz (oblique polars).

Scapolite grains enclosed within the poikiloblastic garnet as well as the coronal scapolite (see below) formed between matrix garnet and calcite also has broken down to calcite–plagioclase symplectites (Fig. 3k).

(5) Finer grains of or vermicular intergrowth of calcite and quartz replace prismatic blades of wollastonite in both assemblages. (Fig. 3h). Hapuarachchi

(1968) described similar textures from other localities in the HC. Wollastonite grains rimmed by coronal garnet also have broken down, indicating that the replacement of wollastonite occurred later than the growth of coronal garnet as is in the case of scapolite (cf. Fig. 3g).

(6) In assemblage II, matrix garnet that is in contact with calcite is locally embayed and a double rind

consisting of coronal scapolite and rim garnet occurs between them (Fig. 3f, i and j). It is evident that the double rind was formed subsequent to the peak metamorphism and the development of the granuloblastic texture. Coronal scapolite is also developed within poikiloblastic garnet between garnet and the enclosed calcite grains. Here, too, the coronal rim garnet occurs between the calcite and the coronal scapolite (Fig. 3k). The coronal scapolite usually has a larger width than the coronal garnet and it occurs always next to the granuloblastic garnet. As mentioned earlier, the coronal scapolite, too, is partially replaced by calcite–plagioclase intergrowth (cf. Fig. 3k). In some instances, a thin clinopyroxene rim occurs next to the coronal scapolite, instead of coronal garnet.

(7) Also in assemblage II, secondary wollastonite is locally developed within poikiloblastic garnet between the garnet and the enclosed quartz grains (Fig. 3l).

3.2. Retrograde reactions

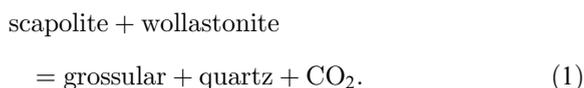
The textures observed in the calc–silicate rocks are modeled in the CASV system following Harley et al. (1994) and Fitzsimons and Harley (1994), while recognizing that the involvement of clinopyroxene and possibly titanite will affect mineral compositions and stoichiometries. The model reactions in the CASV system relevant to the present study are shown in Table 1.

Table 1

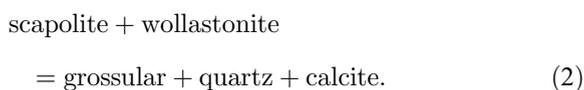
Model reactions in the CaO–Al₂O₃–SiO₂–CO₂ (CASV) system relevant to the present study

5 Wollastonite + scapolite = 3 grossular + 2 quartz + CO ₂	(1)
Scapolite + 6 wollastonite = 3 grossular + calcite + 3 quartz	(2)
3 Wollastonite + scapolite + 2 calcite = 3 grossular + 3 CO ₂	(3)
Scapolite + 5 calcite + 3 quartz = 3 grossular + 6 CO ₂	(4)
Rutile + calcite + quartz = titanite + CO ₂	(5)
Scapolite = calcite + 3 anorthite	(6)
Wollastonite + CO ₂ = calcite + quartz	(7)
3 Grossular + calcite + 3 quartz = scapolite + 6 wollastonite	(8)
Grossular + 2 anorthite + CO ₂ = scapolite + wollastonite	(9)
Quartz + anorthite + 2 calcite = grossular + 2 CO ₂	(10)
5 Anorthite + grossular + 2 CO ₂ = quartz + 2 scapolite	(11)
Grossular + quartz = anorthite + 2 wollastonite	(12)
Scapolite + quartz = wollastonite + 3 anorthite + CO ₂	(13)

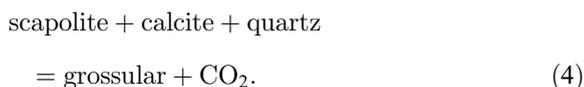
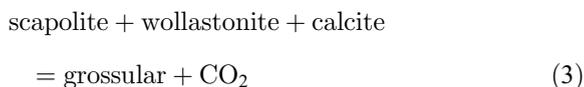
Garnet–quartz composite coronas between wollastonite and scapolite in assemblage I and II (cf. Fig. 2c, f and g) suggest the progress of the model decarbonation reaction,



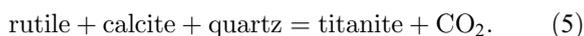
Similarly, garnet–quartz–calcite composite coronas developed, locally, between wollastonite and scapolite in assemblage II corresponding to the vapour absent-reaction,



Single garnet coronas, commonly separating calcite–scapolite–wollastonite grain boundaries, and the presence of nearby quartz in assemblage II (cf. Fig. 2e and b) suggest the model decarbonation reactions,

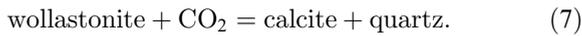


Coronal garnet is not seen between clinopyroxene and other minerals but frequent association of clinopyroxene with calcite, wollastonite, and scapolite suggests that clinopyroxene may have been a reactant in the formation of coronal garnet. Several garnet-forming reactions, involving clinopyroxene as a reactant have been listed by Harley et al. (1994), Fitzsimons and Harley (1994), and others. Involvement of clinopyroxene and other non-CASV components such as Na₂O and MgO transform the above univariant reactions into multivariant reactions. The reaction that produces a partial titanite rim around calcite and scapolite is tentatively related to the model reaction,



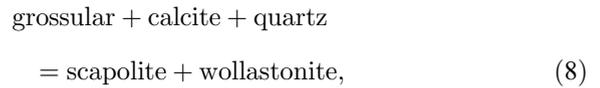
Partial or complete breakdown of scapolite (cf. Fig. 2f) corresponds to the vapour-absent reaction,
 scapolite = calcite + anorthite. (6)

And the replacement of wollastonite is related to the vapour-present reaction,



The double rind of coronal scapolite and rim garnet between matrix garnet and calcite in assemblage II must have formed in two stages; the coronal scapolite

in the first stage and the rim garnet subsequently. However, the origin of the coronal scapolite is problematic. One of the possibilities is that it was formed as result of breakdown of matrix garnet, via reaction,



which is the reverse of reaction (2), forming coronal garnet. The formation of secondary wollastonite within poikiloblastic garnet, between the garnet and the

Table 2(a)
 Representative garnet and pyroxene analyses

Mineral Type	Garnet Matrix Core	Garnet Matrix Rim	Garnet Matrix Core	Garnet Matrix Rim	Garnet Coronal	Garnet Coronal	Pyroxene Matrix	Pyroxene Matrix
Sample number	BU9702	BU9702	BU9701	BU9701	BU9701	BU9701	BU9701	BU9701
Analysis number	5	4/2	6	3/2	4	54	7	4/1
SiO ₂	38.32	38.57	39.94	40.02	38.66	38.70	53.16	52.44
TiO ₂	0.37	0.35	0.16	0.28	0.20	0.00	0.00	0.03
Al ₂ O ₃	20.42	20.83	21.46	21.55	19.79	19.61	0.95	0.78
Cr ₂ O ₃	0.00	0.00	0.00	0.00	0.02	0.01	0.00	0.00
Fe ₂ O ₃ ^a	6.12	4.02	2.30	2.28	4.70	5.57	2.68	1.58
FeO	1.46	2.55	2.34	1.45	1.14	0.75	8.15	10.20
MnO	0.45	0.41	0.34	0.20	0.47	0.52	0.19	0.25
MgO	0.11	0.08	0.38	0.21	0.07	0.02	11.96	11.46
CaO	34.37	33.74	35.10	36.03	34.80	34.85	24.51	24.41
Na ₂ O	0.00	0.00	0.00	0.00	0.02	0.12	0.12	0.13
K ₂ O	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Total	101.61	100.40	101.73	101.95	99.87	99.99	101.73	101.29
Formula for 24 oxygens						Formula for 6 oxygens		
Si	5.792	5.820	5.965	5.953	5.921	5.934	1.966	1.964
Ti	0.042	0.029	0.018	0.032	0.023	0.000	0.000	0.001
Al	3.637	3.738	3.777	3.774	3.572	3.544	0.041	0.035
Cr	0.000	0.000	0.000	0.000	0.004	0.000	0.000	0.000
Fe ⁺³	0.696	0.461	0.259	0.254	0.542	0.643	0.074	0.044
Fe ⁺²	0.184	0.326	0.279	0.180	0.147	0.097	0.252	0.319
Mn	0.058	0.053	0.043	0.025	0.061	0.067	0.006	0.008
Mg	0.024	0.019	0.048	0.047	0.016	0.005	0.659	0.639
Ca	5.566	5.504	5.617	5.735	5.701	5.656	0.991	0.979
Na	0.000	0.000	0.000	0.000	0.006	0.052	0.009	0.009
K	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Total	16.000	15.999	16.000	16.000	15.994	15.997	4.000	3.999
Garnet end members								
Almandine	5.50	3.15	4.67	3.00	2.48	1.65	–	–
Andradite	11.24	16.70	6.48	6.34	13.42	15.61	–	–
Grossular	82.05	78.65	87.33	89.45	82.72	81.50	–	–
Spessartine	0.89	0.99	0.71	0.41	1.02	1.14	–	–
X _{Mg}	–	–	–	–	–	–	0.72	0.67

^a Calculated using the method of Droop (1987).

enclosed quartz may provide evidence for the involvement of wollastonite as a product in the above reaction. Also, clinopyroxene may be a product in the breakdown of garnet, as it occurs in places, next to the coronal scapolite. However, the coronal scapolite always occurs alone, exclusive of wollastonite, in the double rind and suggests that it could possibly have formed under 'open system' condition by partial metasomatic replacement of matrix garnet (also see below).

The second stage in the formation of the double rind involves the development of coronal garnet between the calcite and the coronal scapolite, and is related to reaction (4).

4. Mineral chemistry

Mineral analyses were carried out on selected samples containing the above described reaction textures. The analyses were performed on carbon-coated thin sections at the University of Mainz, Germany, using the JEOL Superprobe (JEOL JXE 8900 RL) with operating conditions of 15-kV accelerating voltage, 12-nA beam current, and 2- μ m beam diameter. Elements were calibrated against synthetic and natural standards. Halides were calibrated with Tugtupite, $\text{Na}_4\text{AlBeSi}_4\text{O}_{12}(\text{Cl}, \text{S})$ (7.58 wt.% Cl) and F-phlogopite (9.20 wt.% F). An online ZAF correction was used

Table 2(b)
Representative scapolite and plagioclase analyses

Mineral Type	Scapolite Matrix	Scapolite Matrix	Scapolite ^a	Scapolite ^a	Scapolite Coronal	Scapolite Coronal	Plagioclase Symplectic	Plagioclase Symplectic
Sample number	BU9701	BU9701	NBU9701	NBU9701	BU9701	BU9701	BU9701	NBU9701
Analysis number	1	54	6	7	3	11	49	16
SiO ₂	45.97	44.72	45.35	44.46	45.81	45.65	44.61	41.67
TiO ₂	0.02	0.01	0.00	0.00	0.00	0.00	0.00	0.00
Al ₂ O ₃	28.16	27.34	29.15	29.26	28.43	28.80	33.24	36.06
FeO ^b	0.08	0.19	0.00	0.00	0.00	0.11	0.14	0.03
MnO	0.01	0.00	0.00	0.00	0.00	0.00	0.00	0.02
MgO	0.01	0.07	0.00	0.03	0.00	0.00	0.02	0.00
CaO	19.57	19.22	19.31	19.52	19.41	19.04	17.82	19.87
Na ₂ O	2.46	2.66	2.52	2.50	2.46	2.42	1.66	0.92
Cr ₂ O ₃	0.01	0.02	0.00	0.00	0.00	0.00	0.05	0.00
K ₂ O	0.19	0.21	0.08	0.11	0.21	0.20	0.16	0.09
Cl	0.05	0.07	0.03	0.03	0.03	0.02	–	–
Total	96.53	94.51	96.44	95.91	96.35	96.24	97.70	98.66
Formula for Si+Al=12							Formula for 32 oxygens	
Si	6.971	6.977	6.828	6.792	6.931	6.882	8.446	7.878
Al	5.029	5.023	5.172	5.201	5.069	5.117	7.412	8.030
Ti	0.003	0.001	0.000	0.000	0.000	0.014	0.000	0.000
Fe	0.010	0.025	0.000	0.000	0.000	0.004	0.023	0.005
Mn	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.003
Mg	0.003	0.016	0.000	0.006	0.000	0.000	0.005	0.000
Ca	3.180	3.213	3.114	3.160	3.192	3.075	3.615	4.021
Na	0.723	0.805	0.736	0.732	0.727	0.707	0.609	0.338
K	0.028	0.042	0.014	0.022	0.036	0.036	0.039	0.021
Cr	0.001	0.002	0.000	0.000	0.000	0.000	0.008	0.000
Sum	15.959	16.104	15.865	15.934	15.955	15.830	20.150	20.290
Cl	0.01	0.013	0.007	0.007	0.007	0.005	–	–
CO ₂	0.99	0.987	0.993	0.993	0.993	0.995	–	–
%Me	82	80	81	78	81	81	–	–
Eq An	68	67	72	73	70	70	–	–
An	–	–	–	–	–	–	85	95

^a Scapolite partly broken down to calcite–plagioclase symplectite.

^b FeO Total CO₂ by stoichiometry.

in data processing (Armstrong, 1988). Representative analyses are presented in Tables 2(a) and 2(b).

Both the granoblastic garnets and the coronal type garnets have similar compositions and are rich in grossular end-member, X_{Grs} being 78–86% and 81–86% respectively, (Tables 2(a)). Also, they have considerable amount of andradite component, X_{And} , being 11–15 % and 6–16 % in granoblastic and coronal garnets, respectively, but very little spessartine and almandine components (averaging 1–5%). Matrix garnets show minor rim–core compositional variations with respect to andradite component, indicating partial re-equilibration, possibly after the growth of coronal garnet. Clinopyroxenes are essentially diopside–hedenbergite solid solutions with X_{Mg} ranging between 0.72 and 0.64 (Tables 2(a)). The other components like CaTs (0–4%) and the essenite components ($\text{CaFe}^{+3}\text{AlSiO}_6$) (based on calculated Fe^{+3}) (4–8%) are low and contribute little to the chemistry of clinopyroxene. Scapolites have ‘equivalent anorthite content’, EqAn, ranging from 67% to 73% calculated on the basis of $\text{Si}+\text{Al}=12$. The meionite content ranges between 76% and 82%. As like the two types of garnets, both the matrix and the coronal scapolites as well as those scapolite grains partially breaking down to calcite–plagioclase symplectite have similar compositions (Table 2(b)). The amount of X_{Cl} ($X_{\text{Cl}}=\text{Cl}/\text{Cl}+\text{CO}_3+\text{SO}_4$) is negligible and the site occupancy is interpreted to be taken by CO_3 as S was not detected in the microprobe reconnaissance surveys. Thus, X_{CO_3} could be directly calculated on the basis of $X_{\text{CO}_3}=(1-X_{\text{Cl}})$. Both the EqAn and the Me% contents of the scapolites reported here are slightly lower than those reported from high-temperature calc–silicate rocks from India (cf. Satish-Kumar and Santosh, 1998; Shaw and Arima, 1996) and suggest that the compositions of scapolites may have been reset. Plagioclase associated with symplectic breakdown of scapolite is anorthite-rich, An_{83-95} . Wollastonite and calcite have nearly pure compositions.

5. Pressure–temperature–fluid history

The reactions and textures in the calc–silicates, except those involving clinopyroxene, and possibly titanite, are modelled in the CASV system by con-

structing partial grids of $T-X_{\text{CO}_2}$ and $P-X_{\text{CO}_2}$, using the activities derived from analysed compositions of the minerals. Harley et al. (1994) and Fitzsimons and Harley (1994) have outlined and discussed in detail the limitations of the activity corrected partial grids. Clinopyroxene was treated as an excess phase and the non-CASV components in garnet, scapolite, and plagioclase were accommodated using reduced activities for grossular, meionite, and anorthite. The Schreinemakers diagrams of the CASV system involving grossular, calcite, wollastonite, scapolite, quartz, and anorthite were constructed with the help of the PeR-pLeX software of Connolly (1990, updated in 1995 and 1999). The stable univariant reactions in the CASV system relevant to the present study are shown in Table 1.

5.1. Activity models

In our calculations, we used the internal consistent thermodynamic database of Holland and Powell (1998) together with the following activity models. The equation of state used for H_2O and CO_2 is that of Holland and Powell (1991). The activity–composition relations given by Holland and Powell (1998) were used for garnet ($a_{\text{Grs}}=0.71$) while Holland and Powell’s (1992) model was used for plagioclase ($a_{\text{An}}=0.88$; $a_{\text{Ab}}=0.12$). The experimentally based, disorder scapolite model of Baker and Newton (1995) and the empirical model of Moecher and Essene (1990, 1991) are currently available, in addition to previous models, for the calculation of the activity of scapolite. The former model gives an average scapolite activity, a_{Me} , 0.119 for the analysed scapolite samples while the latter model gives an average value a_{Me} of 0.40. The model of Baker and Newton (1995), which is applicable for temperatures above 750 °C and gives compatible apparent CO_2 activity values with that of Moecher and Essene (1990) for some Grenville granulites (see Baker and Newton, 1995), is considered as a reasonable model for calc–silicate granulites and is used in our calculations.

5.2. $T-X_{\text{CO}_2}$ and $P-X_{\text{CO}_2}$ relations

Because there is no relevant geobarometer to constrain paleo-pressure in calc–silicate granulites, the peak metamorphic pressure of 9 kbar obtained

from the metabasites in the nearby Kataragama klippe (Faulhaber and Raith, 1991; Prame 1991) is used as the reference pressure to construct the partial $T-X_{\text{CO}_2}$ diagram (Fig. 4). The topology of this diagram is similar to those constructed by Harley et al. (1994) and Fitzsimons and Harley (1994) and includes the isobaric invariant points of [Wo], [Cc], and [An]. As pointed by Harley et al. (1994) and Fitzsimons and Harley (1994), these are not true invariant points because their positions change with change in compositions of the phases (e.g. change in scapolite composition as the coronal garnet forming reaction progress). However, the activity corrected approach will provide an approximation to the true grids as (1) the extents of the progress of the reactions of the peak assemblage are small—only small coronal rims

have been formed, (2) the breakdown of scapolite and wollastonite does not change the compositions of the phases (Fitzsimons and Harley, 1994; Satish-Kumar and Santosh, 1998). The presence of electrolytes (halogens) can have a considerable effect on the reaction topologies in the CASV system (cf. Satish-Kumar and Santosh, 1998). The chlorine content of scapolite is considered as a good indicator of salinity in coexisting fluids (Ellis, 1978 and others). However, the analysed scapolite samples have low chlorine content (Table 2(a)) and thus the reaction topology based on pure $\text{H}_2\text{O}+\text{CO}_2$ fluids is probably a good approximation (cf. Satish-Kumar and Santosh, 1998).

The metamorphic conditions under which the peak assemblages I and II are stable, is shown in the

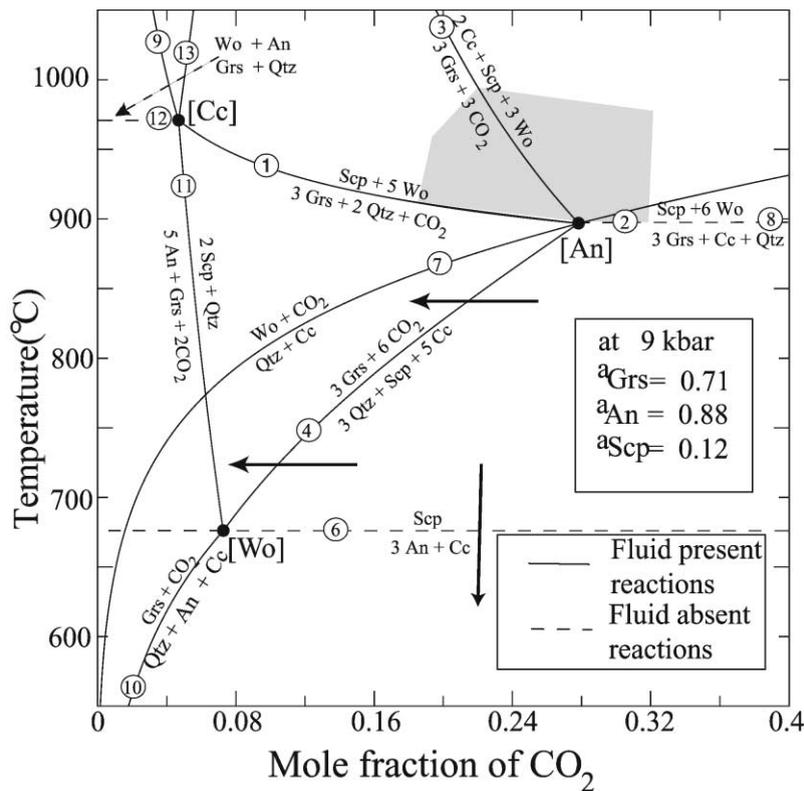


Fig. 4. An isobaric (9 kbar) $T-X_{\text{CO}_2}$ grid for the phases calcite, grossular, wollastonite, quartz, anorthite, and CO_2 in the CASV system, calculated using Holland and Powell's (1998) internally consistent database for the activity corrected mineral phases. The hatched area represents the probable peak metamorphic conditions for the studied calc-silicate rocks. The vertical arrows indicate the cooling path inferred from the reaction textures and the horizontal arrow indicates the conditions, under which coronal garnet could form due to influx of hydrous fluids (see text for discussion).

hatched area in Fig. 4, at $T > [\text{An}]$ invariant point. The T - X_{CO_2} grid suggests that the minimum peak metamorphic temperature of the calc–silicate granulites is about 900 °C (at the position of the anorthite invariant point) at the assumed pressure of 9 kbar, and that the peak metamorphic fluid composition is buffered at low X_{CO_2} (0.08–0.30). This specific fluid composition is considered to reflect internal buffering by mineral–fluid equilibria.

According to the T - X_{CO_2} diagram, both the breakdown of scapolite by vapour-absent reaction (6) and the formation of coronal garnet by vapour-absent reaction (2) are consistent with cooling following peak metamorphism. The coronal garnet producing vapour-present reactions (1) and (3) have negative slopes, dt/dx_{CO_2} , in the T - X_{CO_2} diagram. Hence, the

development of coronal garnets by reactions (1) and (3) as well as the breakdown of wollastonite via reaction (7), are also consistent with cooling. In contrast, formation of coronal garnet by reaction (4), which has a positive slope, requires influx of $\text{H}_2\text{O} \pm \text{CO}_2$ -rich fluid. The P - X_{CO_2} grid (Fig. 5), on the other hand, implies that reactions (1) and (3) could progress to produce garnet coronas by either compression or influx of $\text{H}_2\text{O} \pm \text{CO}_2$ -rich fluid. Furthermore, the grid also indicates that reaction (4) would form garnet coronas with influx of fluids but the vapour-absent reaction (2) could form garnet coronas by compression alone. There is no petrological evidence for compression following the peak metamorphism but evidence for isobaric cooling from 900 to 750 °C is well documented in the metabasites and the

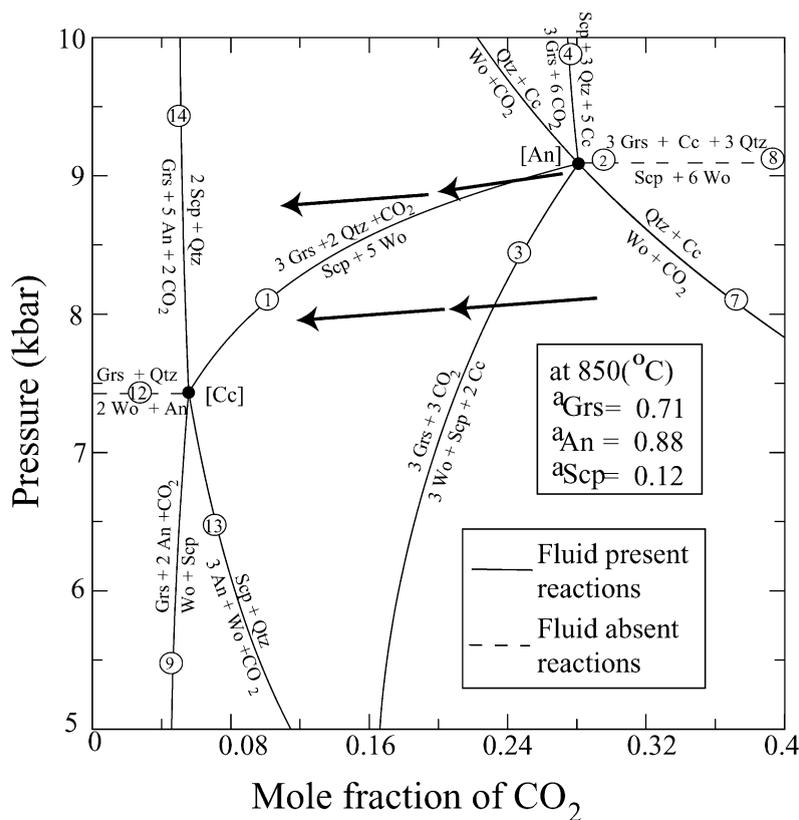


Fig. 5. An isothermal (850 °C) P - X_{CO_2} grid for mineral phases in the CASV system calculated using Holland and Powell's (1998) internally consistent database for activity corrected grossular, calcite, wollastonite, scapolite, and anorthite. The vertical and the horizontal arrows respectively indicate the possible conditions under which retrograde coronal garnets and coronal scapolite could form (see text for discussion).

charnockitic rocks in the south-eastern HC and the Kataragama klippe (Schumacher et al., 1990; Schenk et al., 1991; Prame, 1991). Therefore, as indicated by the T - X_{CO_2} grid, the development of coronal garnets in the calc–silicate granulites is ascribed to cooling and late stage influx of H_2O -rich fluids that have reacted with the calc–silicates.

The T - X_{CO_2} (Fig. 4) grid also implies that the breakdown of scapolite (reaction (6)) occurred at a lower temperature, following the growth of coronal garnet around calcite, wollastonite, and scapolite via reactions (1), (2), (3) and (4). This temporal relation is consistent with the time relations inferred from petrographic study (cf. Fitzsimons and Harley, 1994).

It was noted earlier that the origin of the coronal scapolite observed between matrix garnet and calcite is problematic. If it was produced by partial breakdown of matrix garnet, via reaction (8), then, the P - T_{CO_2} grid suggests an early decompression, whereas the T - X_{CO_2} implies heating during its formation. Neither of these suggestions is convincing because (1) other critical textures such as scapolite–wollastonite and or plagioclase–wollastonite symplectite after garnet, indicating decompression, have not been recorded in the studied rocks, and (2) heating following the peak metamorphism is unlikely.

5.3. P - T trajectory inferred for the Maligawila calc–silicate granulites

The peak metamorphic conditions and the retrograde P - T trajectory inferred for the Mailgawila calc–silicate granulites together with the previously determined P - T trajectories for the rocks of HC are shown in Fig. 2. The peak metamorphic conditions in the Maligawila area are broadly constrained to be 900–875 °C and 10–9 kbar and these conditions are consistent with independent estimates for the south-eastern Sri Lanka (Faulhaber and Raith, 1991; Prame, 1991). Following the peak metamorphism, the rocks underwent high-temperature isobaric cooling, at deep crustal conditions. The high-temperature isobaric cooling segment inferred for the studied rocks is similar to that of the meta-igneous rocks, except that lower temperature limit that extends below 675 °C. This lowest temperature limit is constrained by the thermal stability of scapolite determined by reaction (6).

6. Summary and discussion

Modelling in the CASV system indicates the peak metamorphic temperature of grossular–wollastonite–scapolite calc–silicate rocks from the Buttala klippe to be about 900–875 °C at a pressure of 9 kbar, and the near peak metamorphic fluid composition is internally buffered by mineral–fluid equilibria to be low in X_{CO_2} ($0.1 < X_{\text{CO}_2} < 0.30$). The latter inference together with abundance of orthopyroxene-bearing charnockitic rocks, which require low $a_{\text{H}_2\text{O}}$ for their genesis, favours a fluid-absent or internally buffered fluid condition during peak granulite facies metamorphism in the HC.

The topologies of the T - X_{CO_2} and the P - X_{CO_2} partial grids and the reaction textures preserved in the calc–silicate rocks indicate that the coronal garnets have formed initially by isobaric cooling and later, by cooling and or influx of hydrous of fluids at deep crustal condition. The cooling was from about 900–875 °C to temperature below 675 °C. These calc–silicates, which are thought to be metamorphosed marls composed originally of a mixture of calcareous and pelitic sediments (Mathavan et al., 2000), provide evidence for a phase of high-temperature isobaric cooling in the meta-sediments of the HC and provide evidence for a common high-temperature isobaric cooling event for both in the meta-igneous and meta-sediments of the HC. This conclusion is consistent with the field data showing an intimate association of meta-sediments and meta-igneous rocks in the HC. Thus, the previously held idea that the isobaric cooling phase was confined exclusively to the meta-igneous rocks (e.g. Hiroi et al., 1994; Kriegsman and Schumacher, 1999) is rejected. This study also emphasizes the importance of studying several lithologies in order to establish the P - T evolution history of metamorphic terranes.

7. Conclusions

The grossular–scapolite–wollastonite calc–silicate rocks from the Buttala klippe, which represent a part of the overthrust HC rocks within the VC, preserve a number of reaction textures, similar to those reported from other granulite facies terranes. Modelling in the CASV system indicates that the near

peak metamorphic fluid composition is internally buffered and it is constrained to be low in X_{CO_2} ($0.1 < X_{\text{CO}_2} < 0.30$) at a temperature of 900–875 °C and a pressure of 9 kbar. The formation of garnet, garnet–quartz, and garnet–quartz–calcite coronas around calcite, scapolite, and wollastonite and the breakdown of wollastonite and scapolite to symplectites and/or finer grains of calcite–quartz and calcite–plagioclase respectively, record evidence for near isobaric cooling from about 900 °C to a temperature below 675 °C following the peak metamorphism. The late-stage cooling was probably accompanied by influx of hydrous fluids. Thus, the calc–silicate granulites, which are considered to be metamorphosed marls, provide evidence for high-temperature isobaric cooling in the meta-sediments of the HC, previously thought to be confined exclusively to the meta-igneous rocks by some workers. In the light of the data presented in this study, the P – T – t paths and the tectonic models suggested for the HC of Sri Lanka have to be reconsidered.

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