

Role of CO₂ in Granulite Petrogenesis : Evidence from Fluid Inclusions

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(With 19 Figures and 5 Tables)

Abstract

As the exhumed sections of the deep crust, granulite facies rocks are central to the theme of fluid-rock interaction processes within the earth. Field evidences for deep-crustal fluid pathways are preserved in the gneiss-granulite reaction fronts in the incipient charnockite localities of South India and Sri Lanka, where patches and veins of granulites have formed along structurally-controlled zones of enhanced fluid flow. The dehydration is characterized by conversion of biotite and/or amphibole to orthopyroxene under reduced water activity, and is attended upon by distinct elemental abundances and mobility patterns. The incipient charnockites do not show LILE depletion, precluding the removal of a melt phase. Direct evidence for the role of fluids in effecting the gneiss-charnockite transformation comes from entrapped fluids within inclusions in various minerals. Microthermometric and micro-Raman laser spectroscopic investigations converge on near-pure carbon dioxide with only negligible amounts of dilutants as the ambient fluid species which buffered the system to low water activities. Quantification of the amount of CO₂, from stepped thermal decrepitation studies, indicate almost double the abundance in charnockite as compared to gneiss, with concomitant enrichment of the heavier carbon isotope, suggesting that the dehydration was induced by CO₂ influx from sub-lithospheric sources. Charnockitic rocks also form regional masses in the absence of intimately associated gneissic counterparts in various deep crustal segments. This study reports the occurrence of silicate-melt in association with CO₂ inclusions in massive charnockite samples from four critical crustal sections in South India, Sri Lanka, Antarctica and North America (Adirondacks). The results extend support for the recent experimental models on CO₂ enhanced melting in the deep crust. It is emphasized that regardless of the mechanism of granulite formation in different terrains, CO₂ plays a critical role in buffering the fluid composition and stabilizing the dry mineral assemblages in granulites.

Key Words: Charnockite, mineralogy, chemistry, carbonic inclusions, stable isotopes.

“...The story of granulite fluids is just beginning to be told...”

—Prof. Robert C. Newton

1. Introduction

1.1 The Granulite Problem Defined

Metamorphic rocks of the granulite grade represent exhumed sections of the lower

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portions of the earth's crust. Their study is hence important in understanding deep crustal and crust-mantle interaction processes. Although granulite facies rocks are sporadically exposed in younger terrains, largely by tectonic uplift along fault zones, their abundance in Precambrian shield areas indicates that the lower continental crust is predominantly of granulite grade. Deviating from the earlier notion that granulite formation occurs in zones of continent-continent collision, recent concepts take into account a variety of tectonic environments where granulites can develop (HARLEY, 1989).

Ever since the metamorphic facies concept was advocated, the petrogenesis of granulites remained a controversy among petrologists. Granulites are characterized by anhydrous mineral assemblages, typically the "dry" mineral orthopyroxene, and earlier studies ascribed its stability to high metamorphic temperatures. However, recent models identify the role of reduced water activity in stabilising the dry mineral assemblages. The granulite controversy stems principally from the mechanism involved in driving out the water from a "wet" amphibolite facies rock and generating a "dry" granulite. Current discussions on granulite genesis are essentially centred around the mechanism of dehydration (eg: NEWTON, 1989, VALLEY *et al.*, 1990).

1.2. Recent Models on Granulite Petrogenesis

The recognition that the amount of water plays a dominant role in the formation of granulites has triggered a series of heated debates over the last decade on the mechanism of generating low water activities in the deep crust. The model which has received much attention invokes the dilution and driving-out of water contained in the rock through the external influx of carbon dioxide (NEWTON *et al.*, 1980). Popularly known as "carbonic metamorphism", this model explains how concomitant reduction of water activity could be achieved to stabilize granulitic assemblages through flushing with non-aqueous fluids. Quite contrary to this, another mechanism was proposed by LAMB and VALLEY (1984) which envisages granulite formation as an essentially vapor-absent phenomenon, brought about by the metamorphism of already anhydrous rocks at the base of the crust. This model views that amphibolite grade protoliths totally lose their intergranular fluids by the breakdown of the hydrous minerals and metamorphism occurs by an increase in temperature, under fluid-absent conditions. The third major mechanism advocated is dehydration-melting involving the removal of water from amphibolite facies rocks through the extraction of water-enriched silicate melts, leaving behind a dry granulite residue (WATERS and WHALES, 1985). The above hypotheses of CO₂-streaming, dry metamorphism and partial melting all require the rocks to behave as open systems and some workers are of the view that all these mechanisms can occur in conjunction related to the passage of dry melts through the crust which will absorb pore water, lower the water activities, break the hydrous minerals and generate granulites (eg: FROST and FROST, 1987).

Although the source of fluids and the relative magnitudes of the contributions of fluid-rich and fluid-deficient metamorphism to the evolution of deep crust are yet to be clearly resolved, it is believed that the fluids that have acted on the deep crustal levels were

attendant upon specific thermal and deformational events, such as continental collision or rifting (NEWTON, 1989). The ultimate source of CO₂ is sought in the upper mantle and may have emanated from asthenospheric carbonatitic liquids liberated by deep crustal extension, from alkali basalts freezing at various lithosphere levels, or from subsolidus outgassing of CO₂ enriched deep lithosphere. Subduction of marine carbonate is also thought of as a possibility which functioned as a doubly efficacious mechanism in bringing about increased heat advection because of faster subduction rates in the Archaean (Newton, 1989).

1.3. Importance of Fluid Inclusions in understanding Granulite Genesis

Fluid movements and fluid-rock interaction processes exert a fundamental control on heat-flow, melting, mineral stability, mass transport and deformation style in the earth's deeper portions. Central to the theme of granulite controversy is the role of fluids in effecting desiccation. Metamorphic petrologists used to rely largely on the solid phase equilibria which characterize rock formation in order to make indirect deductions on fluid compositions, in conjunction with experimental thermodynamic information. However, in the recent years there has been an overwhelming pursuit of fluids which occur inside micron-sized cavities within minerals to derive direct information on the nature and role of fluids involved in metamorphism. The interpretation of CO₂-rich inclusions commonly found in granulite terrains has recently been the subject of heated debate, with LAMB *et al.* (1987) suggesting that they are from late fluids, whilst SANTOSH *et al.* (1991a) have shown that they were entrapped during incipient charnockite formation in southern India.

A rock is an aggregate of minerals and minerals crystallize from fluids; either a liquid phase or a vapor phase. During the crystallization history of a mineral, small portions of the mineral-forming media get entrapped within tiny cavities in the growing crystal face and in microfractures that develop in partially or completely grown crystals. The first category is known as "primary inclusions" which yield information on the early growth history of the mineral; the second category of "pseudosecondary inclusions" yield data on the processes which operated during the growth of the crystal and the third category of "secondary inclusions" tell the story of fluids which bathed the mineral after its complete crystallization, in the ideal case. Although several assumptions are involved in this technique, with a number of limitations, this branch of petrology has recently emerged as one of the most challenging tools in elucidating a variety of igneous, metamorphic and hydrothermal processes (eg: HOLLISTER and CRAWFORD, 1981; ROEDDER, 1984; SHEPHERD *et al.*, 1985; SANTOSH, 1988). High grade metamorphism and the powerful role which fluids play in the earth's crust invariably leave behind strong imprints of the different stages of fluid activity within minerals, resulting in an array of early, intermediate and late generations of fluid inclusions. They are thus true "petrologic fossils" which preserve direct evidence of fluid regimes in the deep crust, and constitute a unified theme for investigating the nature and role of fluids involved in granulite petrogenesis.

Fluid inclusion study is by no means a recent subject and has been in currency for

several decades, mostly among ore petrologists interested in tapping information on the temperature, pressure and depositional environments of economic mineral concentrations. The major leap forward in the application of fluid inclusion research in high grade metamorphism was made when TOURET (1971) reported a transition from water to carbon dioxide-dominated fluids within inclusions, while passing from amphibolite to granulite facies terrains. The NEWTON *et al* (1980) model on carbonic metamorphism and pervasive influx of CO₂ at the base of the crust derived much support from TOURET's (1971) observations. Subsequent studies identified the CO₂-rich fluid inclusions in granulite minerals as samples of infiltrated fluids that drove metamorphic dehydration reactions by lowering the water activity (SANTOSH, 1984, 1985, 1986) or even by transporting the heat required for metamorphism (HARRIS *et al.*, 1982).

1.4. Objectives of the Present Study

If indeed fluids played a dominant role in the petrogenesis of granulite facies rocks, the evidence for this has to come from fluid inclusions in minerals which were stabilized during granulite formation. Also, it should be possible to trace the nature and role of fluids which effected the transformation of hydrous mineral assemblages to anhydrous mineral assemblages. Finally, central to the theme of fluids is the question of their origin; whether they were internally generated or came from an external source and what really could be the source. In order to approach these problems, it is essential to concentrate in a classic granulite terrane where unbroken amphibolite-granulite transitions are exposed, identify the field characteristics, elucidate the mineral equilibria and pressure-temperature conditions and then evaluate the nature and role of fluids based on evidence from trapped fluid phases. It is also required to adjudge whether the results obtained are restricted to only the terrain of detailed study, or a more generalised picture can be evolved, commonly applicable to other granulite terrains also. The major objectives of the present work are hence to pursue the above aspects in their finer details.

2. Granulite Formation in South India

2.1. Geological Setting of South India

The Archaean crystalline rocks of South India comprise a granite-greenstone terrain in the north which grades into a granulite facies terrain in the south, through a narrow transitional zone (Fig. 1). Towards the southern tip of the Peninsula, a vast supracrustal sequence, known as the Kerala Khondalite Belt (KKB) is exposed, comprising dominantly of garnet-sillimanite-graphite granulites (termed as khondalites), garnet-biotite upper amphibolite facies gneisses (termed leptynites) and charnockites (hypersthene-bearing greasy green granulites of broadly granitic composition). Minor intercalations of basic granulites and calc-silicates also occur.

Peninsular India is transected by a series of E-W trending Proterozoic shear zones

(DRURY *et al.*, 1984). To the north of the Moyar shear zone (cf. Fig. 1) is an Archaean craton which underwent metamorphism, grading up to granulite facies at its southern margin, at about 2500 Ma. This event was associated with the accretion of a number of

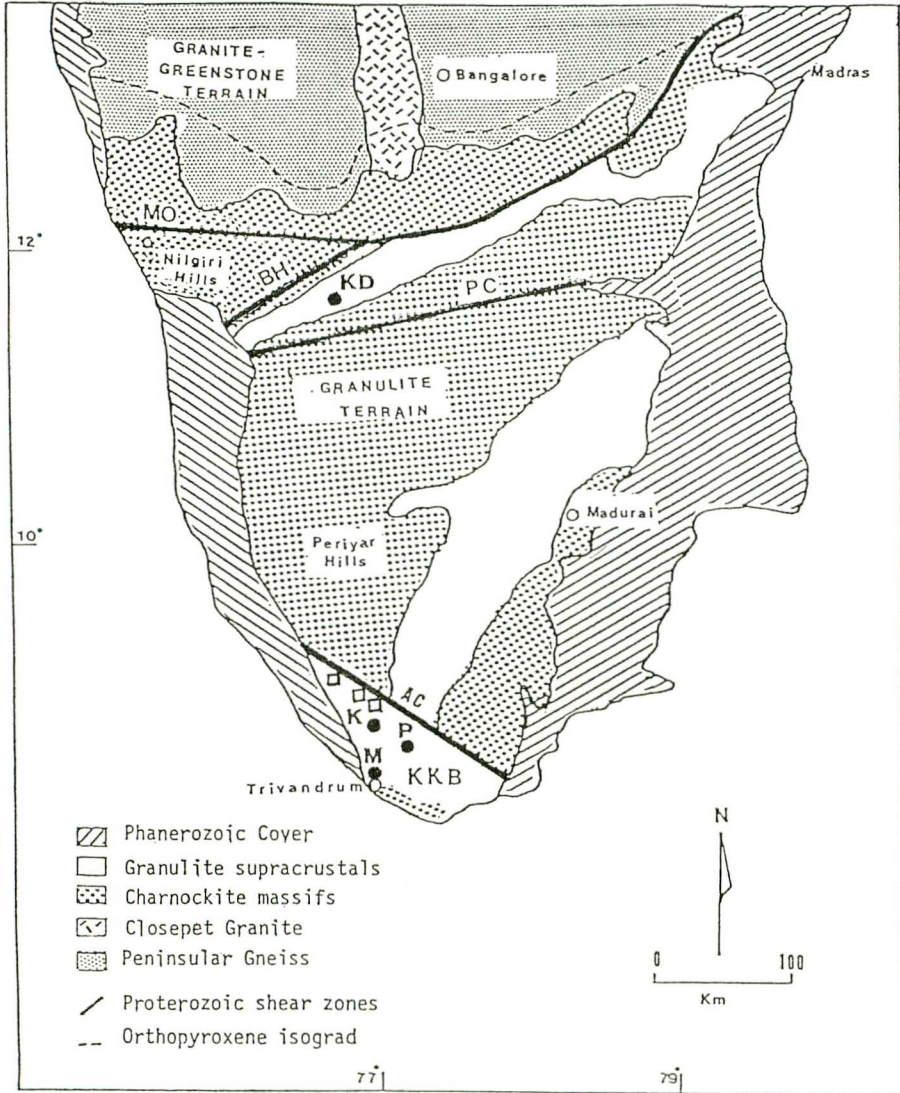


Fig. 1 Generalised geologic map of southern India showing the northern granite-greenstone terrain and the southern granulite terrain. Incipient charnockite localities of Kodakkad (KD), Kottavattom (K), Ponmudi (P) and Mannantala (M) are shown. The Proterozoic shear zones are: MO-Moyar; BH-Bhavani; PC-Palghat-Cauvery and AC-Achankovil. KKB-Kerala Khondalite Belt. The open squares near the AC shear zone represent cordierite-bearing charnockite localities.

blocks onto its southern margin. Although geochronological data are sparse, neither an Archaean history nor the 2500 Ma metamorphic event has been identified to the south of the Palghat-Cauvery shear zone (cf. Fig. 1). The whole of this southern region is metamorphosed to $700 \pm 100^\circ\text{C}$ and 5 ± 1 Kb, with no clear regional trends (HARRIS *et al.*, 1982; SRIKANTAPPA *et al.*, 1985; CHACKO *et al.*, 1987). There is increasing evidence for a charnockite forming event around 500 Ma, at least in the KKB (HARRIS *et al.*, 1990), which is in close correspondence to a similar event recognised recently in the adjacent Sri Lankan terrane (KAGAMI *et al.*, 1990), although an additional earlier high grade event cannot be ruled out, with repeated events of charnockite making, breaking and remaking (SANTOSH and YOSHIDA, 1986; YOSHIDA and SANTOSH, 1987).

2.2. The “massive” and “incipient” charnockites

Most of the granulite exposures the world over are generally massive (term used here to depict homogeneous nature) and occur as mappable units covering wide areas. In southern India, orthopyroxene-bearing charnockites cover extensive areas, comprising most of the upland regions of the Shield, particularly in the Nilgiri and Periyar Hills (cf. Fig. 1), and are commonly designated as massive charnockites. In contrast to this, charnockites also occur as veins and patches in intimate association with amphibole and/or biotite gneisses on a decimeter scale. They are considered to represent an arrested stage of transformation of the gneisses to granulites and are termed incipient charnockites. Incipient charnockites were found firstly along the transition zone between amphibolite and granulite facies in the northern Archaean blocks (PICHAMUTHU, 1960; JANARDHAN *et al.*, 1979) and secondly dispersed throughout the area south of the Palghat-Cauvery shear zone, particularly in the KKB (HOLT and WIGHTMAN, 1983; SRIKANTAPPA *et al.*, 1985). It has been recognised that incipient charnokite formation in southern India corresponds to two distinct categories: the one found in KKB has resulted by the dehydration of pelitic gneisses to charnockites which involved the reaction of biotite, quartz and garnet to orthopyroxene and K-feldspar (HANSEN *et al.*, 1987; SANTOSH *et al.*, 1988, 1990); the other found far north, at the amphibolite-granulite transition zone has resulted by the prograde transformation of tonalitic or granodioritic gneisses through breakdown of calcic amphibole by reaction with biotite and quartz producing orthopyroxene (STAHL *et al.*, 1987). HANSEN *et al.* (1987) termed former category as “Ponmudi type” and the latter as “Kabbal type”, following the locality names where they were first described. However, since a number of localities with such occurrences have been revealed by subsequent studies, it is opted to term the former as paracharnockites and the latter as orthocharnockites for clarity, based on the sedimentary or igneous precursor rocks for the two types of incipient charnockites. Although sufficient geochronological data are lacking on the south Indian granulite segment, it is now broadly agreed that there were at least two charnockite-forming events, one at ca. 2.5 Ga producing orthocharnockites in the north and the other at ca. 0.5 Ga producing paracharnockites in the south (RAITH *et al.*, 1989; HARRIS *et al.*, 1990).

The present study is principally concerned with the incipient charnockites, as they offer the best examples of decimeter-scale gneiss-granulite association to investigate the mechanism of transformation and the role of fluids. They post-date the regional granulites in both the southern and northern segments in South India, the latter belonging to an Archaean metamorphic event of possibly 2.6 Ga, with polyphased deformational history (cf. YOSHIDA and SANTOSH, 1987).

2.3. Incipient Charnockites: Field Evidence for Fluid Pathways

Intimately associated gneiss-granulite exposures are found in a number of large active quarries in the Kerala region of southern India, where freshly exposed rocks afford unique sections of the deep crust. The classic incipient charnockite localities in the KKB include those at Mannantala, Ponmudi and Kottavattom (cf. Fig. 1), all belonging to the paracharnockite type. Far north of KKB, in the Palghat-Cauvery shear zone is a ring of quarries near Kodakkad (bounded by BH and PC in Fig. 1), where orthocharnockites of the incipient variety are exposed. In all these localities, charnockite formation occurs as clots, patches, veins or dykes along oriented zones with compelling evidence of dehydration along structurally-controlled deep crustal fluid pathways. Examples from various localities are shown in Fig. 2 and are briefly summarised below.

The Mannantala quarry is a well-exposed example of structurally controlled dehydration and has been studied in detail by SANTOSH *et al.* (1990a). Graphite-bearing metasedimentary gneisses of upper amphibolite grade constitute about 80% of the exposure in the vertical quarry face. Dark green patches of incipient charnockite form veins that transgress and disrupt the dominant NW-SE striking foliation of the gneisses. The orthopyroxene-bearing charnockite patches are depleted in biotite and garnet relative to the gneiss and are characterised by coarsening of grain size and obliteration of the gneissic fabric. The transformation from gneiss to charnockite is seen to occur along shear zones within the metasediments (Fig. 2H, I). At Kalanjur, charnockite formation occurs along brittle fractures which served as pathways of enhanced fluid flow. In the quarry near Ponmudi hill resort, graphitic biotite-garnet acid gneisses are mottled and veined by dark green charnockite in large amounts which in places, can be seen to be a transgressive alteration which disrupts the dominant NW-SE foliation, with warping of foliation around charnockite-filled shears. While passing from the gneiss to charnockite, there is a remarkable coarsening of grain size and darkening. At Kottavattom, near the northern margin of the KKB, a large vertical quarry exposes fairly homogeneous light grey biotite-garnet gneisses. The streaky foliation of the gneisses is obliterated by a diffuse network of garnetiferous leucosomes which were possibly generated by biotite dehydration-melting reactions. The transformation of gneisses into coarse grained charnockites occurred along a system of conjugate fractures and foliation planes. In the charnockitized zones, the migmatitic structure of the precursor gneisses is completely extinguished due to thorough recrystallization and considerable coarsening.

Far removed from the khondalite belt, the Kodakkad locality of the orthocharnockite

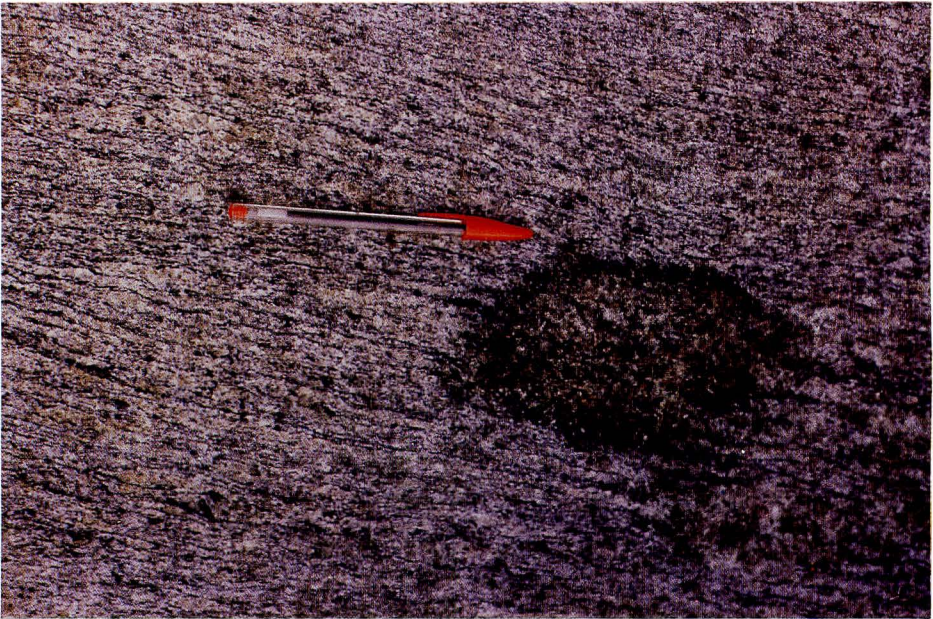
A**B**

Fig. 2 Field photographs showing gneiss-incipient charnockite fronts. **A:** Incipient charnockite formation along patches and veins at Kodakkad. **B:** Coarse patch of incipient charnockite within biotite-hornblende gneiss at Kodakkad. **C-E:** Stages of development of incipient charnockite at Kodakkad

C



D



E



Fig. 2 continued

from small dots and blebs to coarse patches. Note the warping of foliation about the charnockite patch in **E**. **F & G**: Field evidence for deep crustal fluid pathways at Kodakkad. In **F**, a wide zone of fluid influx is seen

F



G

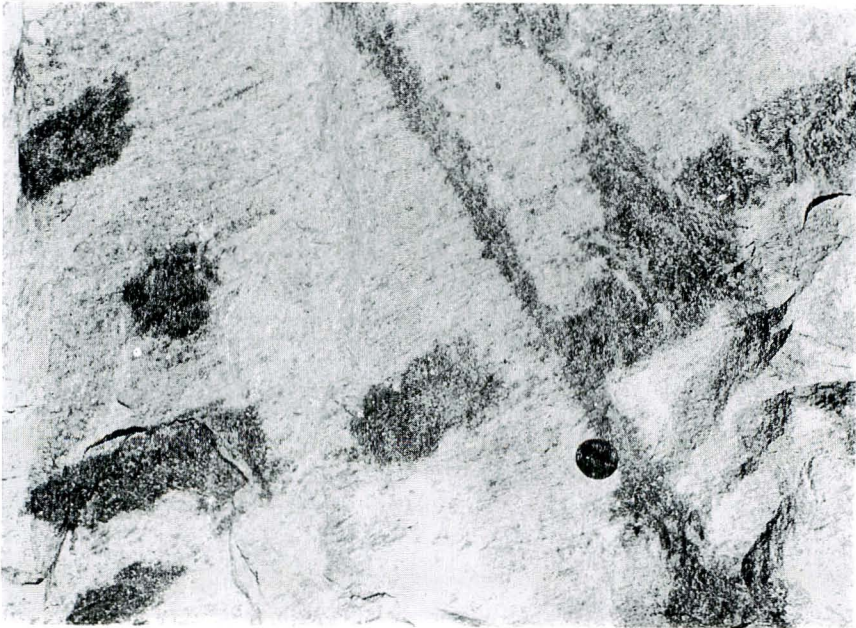


Fig. 2 continued

at the middle, which is completely converted to charnockite. Emanating from this zone are veins, dykes and patches of granulites, developed in all directions along which fluids influxed. The veins and patches are inter

H

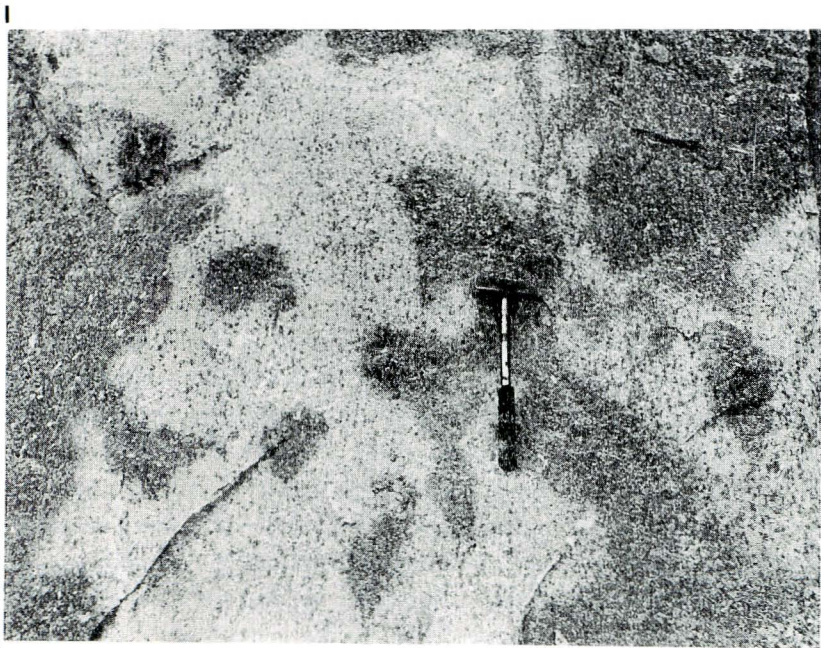
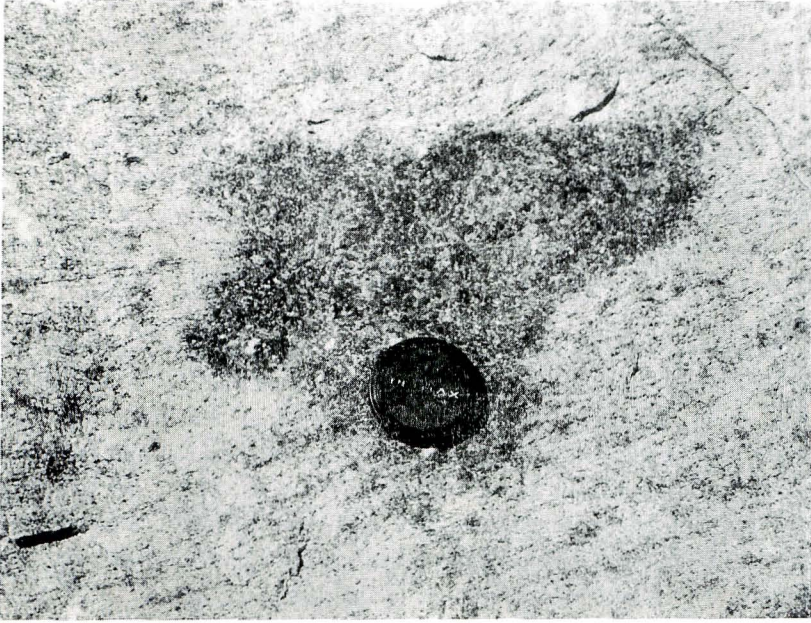


Fig. 2 continued

connected, as in **E**. **H** & **I**: Charnockite formation at the Mannantala quarry in KKB. See text for further details.

type is at the northern margin of the Palghat-Cauvery shear zone and just south of the Nilgiri massive charnockite block (cf. Fig. 1). Here, a chain of quarries exposes various stages of the local conversion of amphibole and biotite bearing gneisses with sporadic garnet into charnockite (Figs. 2 A-G). The occurrence of charnockites at KD varies from small clots or blebs to coarse patches and dendritic networks, with spectacular preservation of fluid channels. Charnockite formation is always a late phenomenon, post-dating the regional metamorphism and over-printing the penetrative deformational events. This locality is characteristically different from the paracharnockites of the KKB in that graphite is conspicuously absent and that the major mafic mineral constituent in the gneisses is hornblende.

The intimate association between gneiss and granulite in all these localities precludes a purely temperature controlled transformation from amphibolite to granulite and also argues against an origin by metamorphism of anhydrous lithologies. The compelling evidence for structurally-controlled desiccation along deep crustal fluid pathways suggest a fluid-controlled mechanism for charnockite formation. The incipient charnockites frequently cross-cut deformation fabrics such as gneissic foliation or migmatitic structures in the rock. The charnockite is generally coarser grained than the gneiss and has a granular texture with no foliation or alignment of the remaining biotite. Charnockites form either as irregular blebs with patches cross-cutting and obliterating the foliation as in Ponnudi and Kottavattom or as linear pattern or dendritic arrays around shear zones or brittle fractures as in Kodakkad. These features suggest that incipient charnockite can develop under a brittle regime as opposed to the earlier ductile-dominated gneiss forming conditions. Both microstructures and fabric analysis imply that incipient charnockite formation was a fluid-controlled process, generally occurring during uplift along deformationally enhanced zones of fluid flow (cf. SANTOSH *et al.*, 1991a).

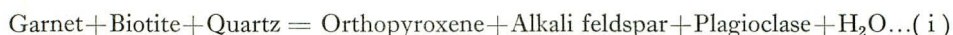
2.4. Cordierite-bearing Incipient Charnockites

Though the occurrence of cordierite in the gneisses and granulites of southern Kerala has been previously reported (eg: CHACKO *et al.*, 1987; SANTOSH, 1987), the incipient charnockite localities studied earlier were largely devoid of cordierite. Our recent field investigations revealed a number of new localities along the northern margin of the KKB where cordierite was found in association with orthopyroxene in charnockite dehydration fronts (SANTOSH *et al.*, 1991b; NANDA-KUMAR *et al.*, 1991, this volume). Here, garnet and biotite-bearing upper amphibolite facies gneisses with or without traces of cordierite transform to irregular patches or wide zones of incipient charnockites containing orthopyroxene crystals and gemmy violet cordierite, both of which are highly coarse grained. Original gneissic fabric is completely extinguished in most cases and the charnockitized zones are massive and coarse grained, disrupting the general NW-SE trending gneissic foliation. Although the field characteristics indicate a close similarity between these localities and the incipient charnockites reported earlier, narrow linear fluid pathways are rarely preserved and fluid channels have largely coalesced into wider zones of dehydration.

Considering the proximal relationship between the cordierite granulite locations and the Achankovil shear zone, a high degree of fluid influx to effect wider zones of dehydration is conceivable. The gneisses have undergone partial melting and migmatization, although charnockite formation clearly post-dates all penetrative events, including migmatization and the coarse greasy-green patches, which are the obvious results of increased reaction kinetics in the presence of a fluid phase, irregularly overprint the gneissic structures. The field features offer compelling evidence for a close genetic similarity between the cordierite-bearing granulites and incipient charnockites. They also appear to be related in time; a Rb-Sr whole rock isochron has yielded best fit age of 678 Ma (unpublished data of Santosh and Iyer, quoted in SANTOSH and DRURY, 1988). Rb-Sr and Sm-Nd mineral isochron age (SANTOSH and KAGAMI, unpublished data) also attribute an early Paleozoic age. Arrested charnockite formation with accessory cordierite has been recently reported from the adjacent Sri Lankan terrain, where radiometric ages assign a ca. 0.5 Ga event (KAGAMI *et al.*, 1990). Thus, a common genetic environment, largely aided by CO₂ flushing, is conceivable for the cordierite-granulites and the incipient charnockites in the southern India-Sri Lanka crustal segments.

2.5. The Mineralogy of Transformation

In the paracharnockite localities, the charnockite is characterized by the mineral assemblage of alkali feldspar, plagioclase, quartz, garnet, biotite and orthopyroxene with accessory ilmenite and graphite. The gneiss samples have a similar assemblage, except that orthopyroxene is absent. In the orthocharnockite localities, orthopyroxene forms by the breakdown of calcic amphibole by reaction with biotite and quartz. The charnockite-forming reaction thus involves biotite or amphibole conversion to orthopyroxene (Fig. 4). Since this is a characteristic reaction for the transformation from amphibolite to granulite facies assemblages, these dehydration reactions have often been taken to represent the highest metamorphic "grade" reached. However, where the reaction is induced by fluid movement, such an assumption may not be well founded. Where garnet is present in the charnockite, adjacent garnet and biotite grains show irregular and sutured grain boundaries in contrast to textures within the gneisses where relatively large megacrysts of garnet are observed to have sharp linear contacts against subhedral laths of biotite (Fig. 4). This suggests that biotite and garnet are reactant phases in the charnockite. Representative modal mineral compositions of the gneisses and charnockite from a traverse across the gneiss-charnockite reaction front at Mannantala quarry (cf. Fig. 3) is given in Table 1. The decrease in the contents of garnet, biotite and quartz with a concomitant increase in orthopyroxene and feldspar in the charnockite as compared to the gneiss is a notable feature, which, along with the microstructural features described above, indicates that the charnockite formation is represented by the reaction (SANTOSH *et al.*, 1990a):



Since the biotites in the gneisses normally contain appreciable amounts of Ti and if

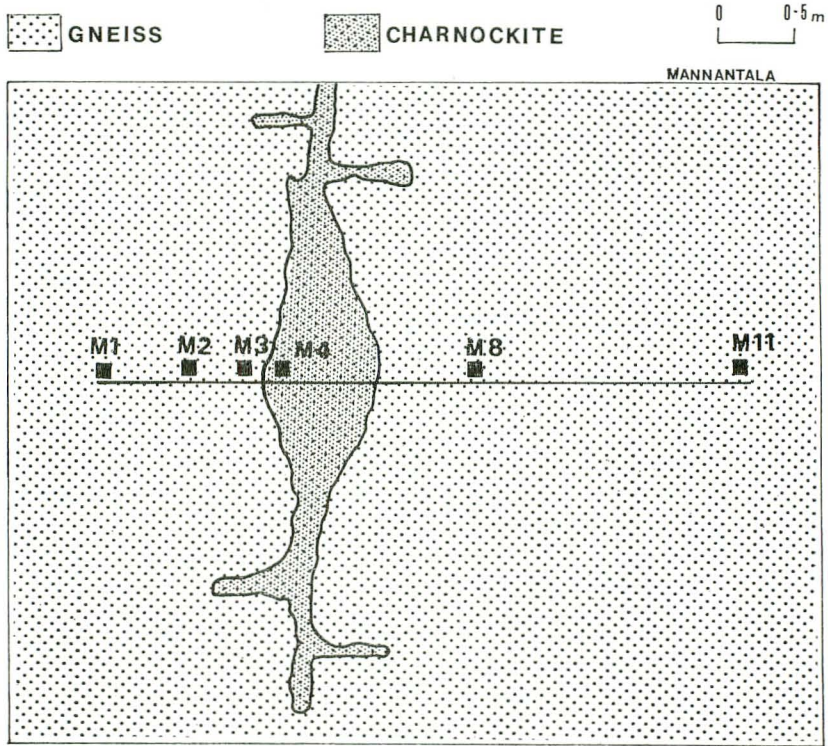


Fig. 3 Diagrammatic representation of a gneiss-charnockite reaction front at Mannantala quarry in the KKB. The sample traverse discussed in this study is also shown (M1 to M11).

Ti is added as a component, ilmenite will also be a product phase as commonly noted in the incipient charnockites (Fig. 4). The reactants are stable phases in the gneiss assemblage. Decrease in water activity will lead to the consumption of biotite, garnet and quartz and growth of pyroxene and alkali feldspar as reflected in the relative modal analyses of gneiss and charnockite (Table 1). Consequently, loss of garnet in the charnockite-forming reaction does not require a change in either temperature or pressure. It is also of importance to note that the garnet-consuming reaction involves a pronounced volume change and has a shallow slope in P-T space so that the reaction is initiated under a decompressional regime. This is quite in agreement with the brittle-dominated isothermal decompression history of incipient charnockite genesis. Further evidence for mineral reactions come from the cordierite-bearing lithologies (see NANDA-KUMAR *et al.*, 1991, this volume). Here the charnockite comprises an assemblage of cordierite-hypersthene-garnet-K-feldspar-plagioclase-quartz-magnetite-spinel. Cordierite forms colourless granoblastic polygonal grains, generally forming the matrix of a symplectitic reaction intergrowth with quartz and hypersthene. Large cordierite grains show typical lamellar twinning. Cordierite also occurs as coronal rims around garnet and spinel. Large

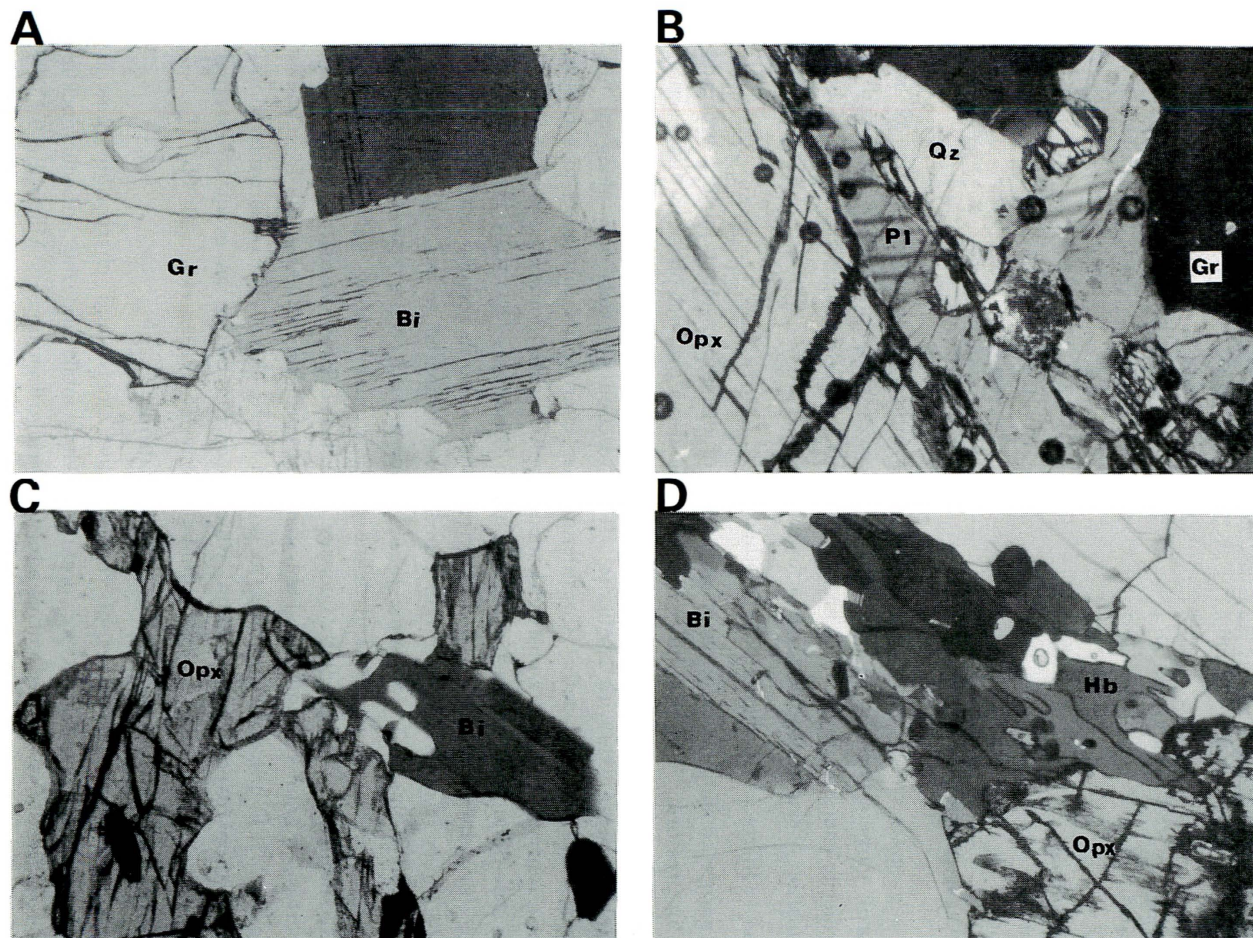


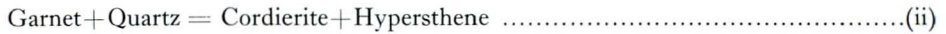
Fig. 4 Thin section photomicrographs of gneiss and incipient charnockite assemblages. **A:** Gneiss assemblage showing garnet megacryst and biotite. **B:** Charnockite showing orthopyroxene-plagioclase-quartz-garnet assemblage. **C:** Charnockite formation in paracharnockite localities by the consumption of biotite. **D:** Charnockite formation in the orthocharnockite localities by the consumption of hornblende. All photographs in 40X. A & C in parallel nicols; others in crossed nicols.

Table 1 Modal analyses (%) of Gneiss and Charnockite Samples.

	M-1 (Gneiss)	M-2 (Gneiss)	M-3 (Gneiss)	M-4 (Charn.)	M-8 (Gneiss)	M-11 (Gneiss)
Quartz	32.3	29.6	27.7	20.4	29.8	31.7
Plagioclase	26.8	27.8	29.6	30.5	28.3	25.6
K-feldspar	22.8	20.4	22.2	34.7	23.5	21.6
Garnet	8.8	12.3	11.8	6.8	9.3	9.8
Biotite	8.4	8.7	7.6	2.2	7.9	9.5
Orthopyroxene	—	—	—	4.6	—	—
Graphite	0.8	1.0	0.9	0.4	1.0	1.7
Ilmenite	0.1	0.2	0.2	0.4	0.2	0.1

Data Source: SANTOSH *et al.* (1990)

cordierite aggregates embay garnet grains and fine symplectitic intergrowths replace and pseudomorph garnets partially to completely. These textures are widely developed in all the cordierite charnockite localities investigated and represent the key reaction involved in the formation of cordierite-bearing charnockites:



The garnet consuming reaction is essentially of decompression type and proceeds by a drop in pressure. The possibility that spinel is also a product phase in this reaction merits consideration because composite grains of magnetite+hercynite sometimes show coarse graphic intergrowth with cordierite. This reaction proceeds only along an unusually steep decompression path. Considering that charnockite formation in southern Kerala was essentially linked to a post-peak metamorphic brittle-dominated regime following the ductile-dominated gneiss-forming rheologic conditions (SANTOSH *et al.*, 1991a), such a steep decompression path is not abnormal, and is also in keeping with the density decrease of fluids trapped within product phases as compared to reactant phases (see section on fluid inclusions). In some localities, cordierite occurs sporadically in the gneisses. They are small grained and define compositional banding along with biotite, garnet and sillimanite. Where both gneisses and charnockites coexist, there is a significant increase in grain size and modal abundance of cordierite while passing from gneiss to charnockite. It is inferred that cordierite growth in the charnockites was contemporaneous with orthopyroxene crystallization, which occurred during an event of external CO₂ influx along structurally-controlled fluid pathways (SANTOSH *et al.*, 1990b).

2.6. Mineral Chemistry and P-T-X_{H₂O} Computations

Electron microprobe analyses of representative gneiss and charnockite mineral assemblages from the paracharnockite locality of Mannantala and the orthocharnockite locality of Kodakkad were carried out using a Cambridge Instruments Microscan 9 (M9) wavelength dispersive system at the Department of Earth Sciences, Open University (U.K.), which operates at an accelerating potential of 20 kV and a probe current of 30 nA. The analytical results are presented in Tables 2 and 3 and a summary of temperature and

Table 2 Representative Microprobe Analyses from Mannantala

SAMPLE NO.	M-2 (gneiss)					M-3 (gneiss)					M-4 (chamocklite)						
	GAR(C)	GAR(R)	PLAG(C)	PLAC(R)	BIOT	GAR(C)	GAR(R)	PLAG(C)	PLAG(R)	BIOT	GAR(C)	GAR(R)	OPX	OPX	PLAG(C)	PLAG(R)	BIOT
W%	38.22	38.40	63.49	62.73	36.49	38.12	38.33	63.49	63.28	36.81	38.45	38.36	48.86	49.33	64.46	64.03	37.04
SiO ₂	0.05	0.07	0.022	0.02	5.58	0.05	0.05	0.02	0.04	5.47	0.05	0.05	0.22	0.17	0.04	0.02	5.12
TiO ₂	21.85	21.82	2.07	22.30	14.64	21.81	21.85	22.07	22.32	14.76	21.71	21.72	2.67	2.42	22.12	22.32	14.64
Al ₂ O ₃	—	0.02	—	—	0.02	0.02	0.03	—	—	0.02	0.02	0.02	0.02	—	—	—	0.02
FeO*	33.62	33.83	0.03	0.03	16.72	33.71	33.82	0.03	0.03	16.76	33.93	33.89	34.48	33.72	0.04	0.04	16.20
MnO	0.59	0.62	0.02	—	0.02	0.60	0.62	0.02	—	0.05	0.62	0.60	0.30	0.29	0.02	0.02	0.02
MgO	6.15	6.04	—	—	11.41	6.12	6.12	—	—	11.45	5.86	5.77	13.34	13.91	—	—	12.83
CaO	0.97	1.01	3.91	4.17	0.03	1.05	1.01	3.91	4.11	0.01	1.06	1.04	0.22	0.25	3.81	4.05	0.01
Na ₂ O	—	—	8.84	8.75	0.04	—	—	8.84	8.72	0.05	—	—	0.02	0.02	9.10	9.13	0.05
K ₂ O	—	—	0.70	0.64	9.35	—	—	0.70	0.57	9.29	—	—	—	—	0.45	0.37	9.66
Total	101.45	101.81	99.08	98.64	94.30	101.48	101.83	99.08	99.07	94.67	101.70	101.45	100.13	100.11	100.04	99.98	95.59
Atoms	12(0)	12(0)	8(0)	8(0)	22(0)	12(0)	12(0)	8(0)	8(0)	22(0)	12(0)	12(0)	6(0)	6(0)	8(0)	8(0)	22(0)
Si	2.98	2.98	2.83	2.82	5.56	2.97	2.98	2.83	2.82	5.58	2.99	2.99	1.93	1.94	2.84	2.83	5.56
Ti	0.00	0.00	0.00	0.00	0.64	0.00	0.00	0.00	0.00	0.62	0.00	0.00	0.01	0.01	0.00	0.00	0.58
Al	2.01	2.00	1.16	1.19	2.63	2.01	2.00	1.16	1.18	2.64	1.99	2.00	0.12	0.11	1.15	1.16	2.59
Cr	—	0.00	—	—	0.00	0.00	0.00	—	—	0.00	0.00	0.00	0.00	—	—	—	0.00
Fe	2.19	2.20	0.00	0.00	2.13	2.20	2.20	0.00	0.00	2.13	2.21	2.21	1.24	1.11	0.00	0.00	2.03
Mn	0.04	0.04	0.00	0.00	0.00	0.04	0.04	0.00	—	0.01	0.04	0.04	0.01	0.01	0.00	0.00	0.00
Mg	0.72	0.70	—	—	2.60	0.71	0.71	—	—	2.59	0.68	0.67	0.79	0.82	—	—	2.87
Ca	0.08	0.08	0.19	0.20	0.01	0.09	0.08	0.19	0.20	0.00	0.09	0.09	0.01	0.01	0.18	0.19	0.00
Na	—	—	0.77	0.76	0.01	—	—	0.77	0.76	0.02	—	—	0.00	0.00	0.78	0.78	0.01
K	—	—	0.04	0.04	1.82	—	—	0.04	0.03	—	—	—	—	—	0.02	0.01	1.85

C-Core; R-Rim; *All Fe calculated as FeO

Data source: SANTOSH *et al.* (1990)

Table 3 Microprobe Analyses of Minerals in Kodakkad Incipient Charnockite

Sample No: Mineral	KKD-15B Garnet (C)	Garnet (R)	Biotite	Biotite	Opx	K-Spar	Plag (R)	KKD-15A Garnet	Garnet	Biotite	Biotite	H'Blende
SiO ₂	37.00	36.98	34.62	34.62	47.13	65.35	64.31	37.21	37.33	33.16	32.84	40.00
TiO ₂	0.03	0.02	4.331	4.19	0.10	0.20	0.02	0.03	0.03	2.86	2.58	1.53
Al ₂ O ₃	20.2	20.30	3.04	13.11	0.41	18.53	21.80	20.54	20.44	12.77	12.65	10.37
Cr ₂ O ₃	60.02	0.02	0.03	0.03	0.48							
FeO	29.61	28.86	29.37	29.47	43.70	0.07	0.10	33.49	33.25	32.44	31.88	27.35
MnO	4.86	5.13	0.32	0.32	1.36	0.02	0.02	2.92	2.95	0.33	0.29	0.59
MgO	1.00	0.92	4.70	4.87	5.61			1.11	1.09	4.12	4.17	3.91
ZnO												
CaO	7.01	7.29	0.04	0.04	0.80	0.43	3.37	5.58	5.79	0.04	0.06	10.48
Na ₂ O			0.06	0.04	0.02	2.70	9.76	0.04	0.06	0.04	0.08	1.95
K ₂ O			8.88	8.89		12.61	0.57			9.25	9.03	1.42
Total	99.79	99.52	95.39	95.58	99.6	99.73	99.95	100.92	100.94	95.01	93.58	97.60
Atoms on	12(0)	12(0)	22(0)	22(0)	16(0)	8(0)	8(0)	Atoms on 12(0)	Atoms on 22(0)	Atoms on 23(0)		
Si	3.004	3.007	5.582	5.572	1.983	2.993	2.847	2.998	3.006	5.496	5.518	6.395
Ti	0.002	0.001	0.525	0.507	0.003	0.001	0.001	0.002	0.002	0.356	0.326	0.183
Al	1.939	1.946	22.478	2.488	0.020	1.000	1.137	1.951	1.940	2.494	2.505	1.954
Cr	0.001	0.001	0.004	0.004	0.016							
Fe	2.011	1.963	3.960	3.997	1.537	0.003	0.004	2.257	2.239	4.497	4.479	3.657
Mn	0.334	0.354	0.043	0.043	0.048	0.001	0.001	0.199	0.201	0.047	0.041	0.080
Mg	0.121	0.112	1.130	1.168	0.352			0.133	0.131	1.017	1.044	0.932
Zn												
Ca	0.610	0.635	0.007	0.007	0.036	0.021	0.160	0.482	0.499	0.007	0.010	1.796
Na			0.019	0.012	0.002	0.240	0.838	0.006	0.010	0.013	0.027	0.604
K			1.827	1.826		0.737	0.032			1.956	1.936	0.289
Σ	8.023	8.018	15.574	15.594	3.997	4.995	50.19	8.028	8.027	15.884	15.884	15.890

C-Core; R-Rim All Fe calculated as FeO

Table 4 Summary of Temperature and Pressure Estimates in Gneisses and Incipient Charnockites Based on Mineral Phase Equilibria Computations.

Locality	Mannantala		Kodakkad	
	Gneis	Charnockite	Gneiss	Charnockite
Thermometry				
<i>Garnet-Biotite</i>				
Thompson	645–721		657–669	
Ferry & Spear	642–746		646–663	
Hodges & Spear	654–758		714–733	
Perchuk & Lavrent'eva	626–678		611–620	
<i>Garnet-Orthopyroxene</i>				
Powell		758–793		637–648
Barometry				
<i>Gar-Opx-Plag-Otz</i>				
Perkins & Newton		5.52–5.96		7.0–7.40

Thermometers in degree centigrade quoted at 5 Kbar for Mannantala and 7 Kbar for Kodakkad. Barometer in kilobars quoted at 700°C. Full references of geothermobarometers given in the text.

pressure computations are given in Table 4. As seen from the tables, analyses of core and margin of individual garnet and plagioclase grains do not show any significant or systematic variation, as also observed in the previous studies in the KKB (eg: SRIKANTAPPA *et al.*, 1985). Since all the analyses listed represent minerals in mutual grain-contact, thermobarometric computations can be applied by considering them as equilibrium assemblages.

The Powell (1978) thermometer based on the distribution of Fe and Mg between coexisting garnet and orthopyroxene is applied here to estimate the temperature of equilibration of charnockite assemblage. While in Mannantala a range of 758–793° is obtained, the Kodakkad sample yields a lower value, 637–648°C. However, this is in good agreement with the phase equilibria predictions that biotite to orthopyroxene transformation requires higher temperatures than hornblende to orthopyroxene conversion (HANSEN *et al.*, 1987), the present results ascribing a difference of more than 100°C.

The analyses of coexisting garnet-biotite pairs in the gneisses afford a geothermometer for the upper amphibolite facies assemblage. This thermometer has been calibrated by different workers based on different approaches on the Fe-Mg exchange reactions. Here, four calibrations have been applied (cf. Table 4). THOMPSON'S (1976) thermometer is a semi-empirical calibration which makes no allowance for the effects of non-ideal mixing in either phase and does not allow for variations in non-ferric components between the phases (eg: Ti, Al, Ca). The FERRY and SPEAR (1978) method provides an experimental study of the Fe-rich system for the range of temperatures between 550–800°C. The quoted non-ideality of biotite is a measure of deviation of how far from ideal the biotite is. This thermometer has been updated by HODGES and SPEAR (1982) by making empirical

adjustments for non-ideal mixing in garnet, resulting in a slight upward revision of T of less than 30°C . PERCHUK and LAVRENT'eva (1983) provide another experimental study on natural assemblages in the range of $600\text{--}1000^{\circ}\text{C}$, assuming ideal mixing, but based on more Mg-rich, and hence more realistic, phases. In the present case, the computations from all these four calibrations lie broadly within an overlapping range, $626\text{--}758^{\circ}\text{C}$ for the Mannantala gneisses and $611\text{--}733^{\circ}\text{C}$ in the Kodakkad gneisses.

The most useful barometer for charnockites comes from the continuous reaction in the assemblage, garnet-orthopyroxene-plagioclase-quartz. PERKINS and NEWTON (1981) computed pressures from the Mg-end member reaction, the quoted uncertainties are of the order of 1 Kbar. In the present case, the Mannantala charnockite yields a pressure of $5.52\text{--}5.96$ Kbar, while the Kodakkad charnockite reads $7.00\text{--}7.40$ Kbar, almost 2 Kb rise in the orthocharnockite as compared to the paracharnockite. Although these pressures are indeed high as compared to the estimates from paracharnockite localities, the Kodakkad is located within the northern segment, which is considered to represent a more deeper section of the crust than the south. Previous studies on regional granulites have also detected this difference in pressure and attributed a tectonic relationship between the

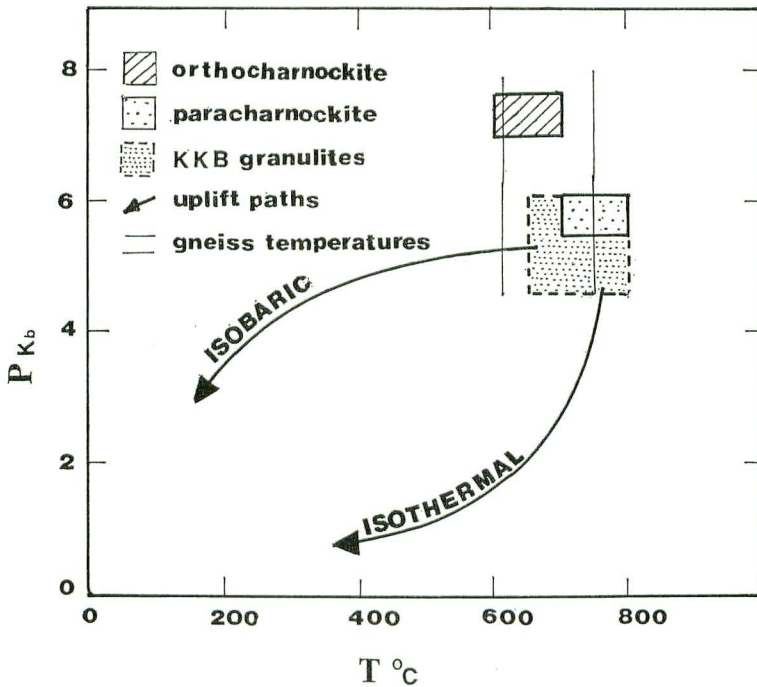


Fig. 5 Pressure-temperature diagram showing the fields of thermobarometric estimates from mineral phase equilibria for paracharnockites and orthocharnockites (this study), and the granulites of KKB (various studies mentioned in the text). The thin vertical lines representing gneiss temperature range is from garnet-biotite thermometry. Two types of uplift paths, isothermal and isobaric, are also shown. See text for details.

northern and southern segments, with the P-C shear zone as the boundary (HARRIS *et al.*, 1982; DRURY *et al.*, 1984). The P-T estimates from both the localities are shown in Fig. 5. Average P-T conditions of 750°C and 5.5 Kbar for the paracharnockite formation and 650°C and 7 Kbar for the orthocharnockites are deduced. Fig. 5 also shows two types of idealized metamorphic uplift paths; one concave towards the temperature axis, resulting by isobaric uplift tectonics and the other convex towards the temperature axis, resulting by isothermal uplift tectonics. The different tectonic blocks in South India are believed to have undergone different uplift histories. TOURET and HANSTEEN (1988) provided mineralogic and fluid inclusion evidence which suggest an isobaric uplift mechanism for the high pressure-type massive charnockites of the Nilgiri Hills in the northern part of the south Indian high grade segment. The incipient charnockites (from both the northern and southern segments) provide compelling field and microstructural evidences for an isothermal uplift history, aided by decompression tectonics. The decrease in fluid densities trapped within late crystallizing minerals also supports this notion, as will be discussed later.

Fig. 6A shows the water contents of fluids in equilibrium with incipient charnockite based on biotite-orthopyroxene-K-feldspar-fluid equilibrium (after HANSEN *et al.*, 1987).

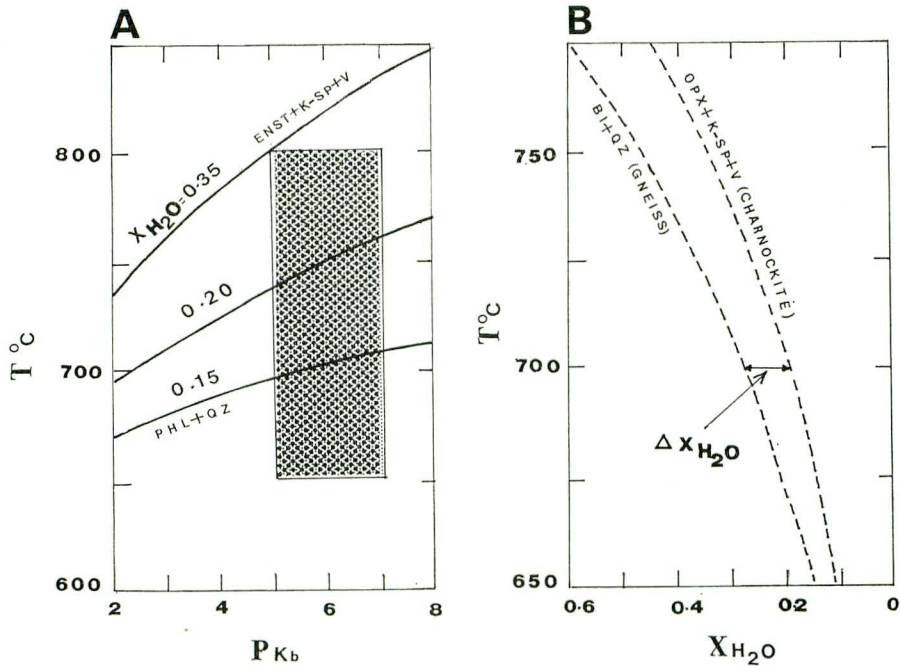


Fig. 6 Estimation of water activities during charnockite formation. The ornamented region in **A** represents the range of temperatures under which the S. Indian granulites equilibrated. The shift in mol fraction of water required to dehydrate a gneiss to charnockite in incipient charnockite localities (after SANTOSH *et al.*, 1990) is shown in **B**. See text for discussion.

From the figure, for P-T conditions generally recorded by the incipient charnockites in the region (shown as hatched box in the figure), the mol fraction of water in vapor phase in equilibrium with these rocks should have been less than ca. 0.35. For the biotite breakdown reaction, the equilibrium can be represented in the KMASH system by the equation:



However, it is unlikely that equilibrium (iii) above was responsible for generating all the pyroxene in the charnockite. It is seen from mineral reaction textures and modal analyses discussed before that garnet, biotite and quartz are consumed during the orthopyroxene-producing event, represented by reaction (i) discussed before. If all observed pyroxene results from equilibrium (iii), then 1.5 moles of pyroxene will result from the loss of 1 mol of biotite. This suggests that 6.5 cc loss of biotite seen in modal analyses of M2 and M4 samples (cf. Table 1) results in production of 1.8 cc of pyroxene. In fact nearly three times this volume of pyroxene is observed in the charnockite indicating that it results from garnet as well as biotite breakdown. If the dehydration is driven by change in $X_{\text{H}_2\text{O}}$, then it should be possible to constrain $\Delta X_{\text{H}_2\text{O}}$ across the reaction boundary. SANTOSH *et al* (1990) gives the details of determining the shift in water activity required to drive the orthopyroxene-forming reaction. The marked increase in Mg content in the biotite between gneiss and charnockite requires an increase in $X_{\text{H}_2\text{O}} > 0.05$, at 700°C, to > 0.1 at 750°C (Fig. 6B). Following these, it was estimated that charnockite formation occurred at $X_{\text{H}_2\text{O}}$ of about 0.2 (SANTOSH *et al.*, 1990). Such low water activities require that the water in the gneisses were driven out, most possibly by an influxing carbonic fluid.

2.7. Geochemical Characteristics

The open-system behaviour of rocks during any dehydration mechanism would result in the distinct chemical transport and exchange of elements. Hence knowledge of element mobility in granulite formation is fundamental to the understanding of deep crustal processes. It is well established that distinct mineralogical and geochemical changes attend regional metamorphism, the depletion in large ion lithophile elements (LILE) being one of the more characteristic features (eg: TARNEY, 1976; ROLLINSON and WINDLEY, 1980; SHERATON, 1985). In order to detect the elemental abundances and to characterize their mobility in gneiss-granulite reaction fronts of present study, whole rock geochemical analyses of major and trace elements were carried out in gneiss-charnockite pairs from a number of localities. The analyses were done at the Muroran Institute of Technology, Muroran (Japan) using a combination of wet chemical techniques, Atomic Absorption Spectrometry and X-ray Fluorescence Spectrometry. The analytical procedures and detailed results will be discussed elsewhere (YOSHIDA *et al.*, 1991). Representative analyses from a paracharnockite locality (Kottavattom) and an orthocharnockite locality (Kodakkad) are given in Table 5. Along with these were also analysed a massive charnockite sample from Ooty in the Nilgiri hills and a khondalite sample from

Chittikara in the KKB. These two samples were included to compare the regional granulite geochemistry with the local granulite (incipient charnockite) geochemistry. The analytical results of these samples are also listed in Table 5.

Broadly, incipient charnockites from both the localities have comparable major and trace element abundances, but show significant deviation from the massive charnockite. More pronounced are the enrichment in silica, sodium and potassium and the depletion in calcium and iron. One of the consistent and common features for the incipient charnockite localities is the pronounced decrease in FeO and increase in Fe₂O₃ while passing from gneiss to charnockite, suggesting high oxidation states during charnockite formation,

Table 5 Major and Trace Element Geochemistry of Gneiss-Incipient Charnockite Pairs and Regional Granulites from South India. (KTV-Kottavattom paracharnockite; KKD-Kodakkad orthocharnockite; OTY- Ooty massive charnockite from Nilgiri Hills; CHT- khondalite from Chittikara locality in the Kerala Khondalite Belt)

	KTV		KKD-3		OTY	CHT
	G	C	G	C		
Major elements wt%						
SiO ₂	69.01	69.52	72.09	71.93	65.45	73.09
TiO ₂	0.53	0.82	0.431	0.21	0.59	0.40
Al ₂ O ₃	15.43	14.98	3.19	14.42	15.10	14.11
Fe ₂ O ₃	1.06	1.37	1.12	0.99	1.50	1.06
FeO	3.64	1.24	2.68	1.48	6.80	3.42
MnO	tr	tr	tr	tr	tr	tr
MgO	0.58	0.43	0.48	0.22	2.71	0.88
CaO	1.76	2.22	1.84	1.14	4.53	2.24
Na ₂ O	2.17	2.49	3.42	3.43	2.06	3.45
K ₂ O	5.15	5.25	4.16	5.44	0.29	0.98
P ₂ O ₅	0.24	0.33	0.14	0.06	0.11	0.03
H ₂ O ⁺	0.52	0.79	0.48	0.27	0.54	0.28
H ₂ O ⁻	0.1	0.67	0.18	0.12	0.12	0.06
Total	100.23	100.11	100.10	99.71	99.80	100.00
Trace elements ppm						
Cu	8.7	10.2	1.5	1.0	22.3	2.5
Pb	37.6	41.2	37.5	18.8	5.6	9.2
Zn	34.1	122	67.8	33.7	72.6	39.7
Ni	25.7	22.9	22.2	23.2	86.5	36.5
Co	6.2	9.1	5.1	2.8	25.2	8.9
Cr	60.5	37.6	35.4	20.5	137	60.6
Ba	1032	1024	666	980	73.7	212
Sr	125	140	110	105	149	117
Li	40.9	29.0	26.5	12.1	15.0	25.1
Rb	237	231	165	190	0.8	21.7

which is in keeping with a genetic model involving the influx of CO_2 rich fluids. The trace element abundances, especially the large ion lithophile elements (LILE) in the massive charnockite are in accordance with those expected during regional high grade metamorphism in that they show pronounced depletion of Pb, Ba, Rb and K values. However, interestingly, no such depletion is observed in the incipient charnockite, deviating from the predicted behaviour of LILE during granulite genesis. It should be cautioned that LILE depletion cannot be applied as a universal phenomenon attending granulite formation and regional studies have shown a co-variance relationship between K/Rb ratios and K-contents, implying that it is K-poor granulites that are strongly depleted in the LILE (TARNEY and WINDLEY, 1977; RUDNICK *et al.*, 1985), which makes the comparisons of lithologies with different precursors rather meaningless. However, the LILE behaviour is more pronounced in drawing a distinction between the ortho- and para-charnockite types as shown in Fig. 7. In the paracharnockites, granulite formation

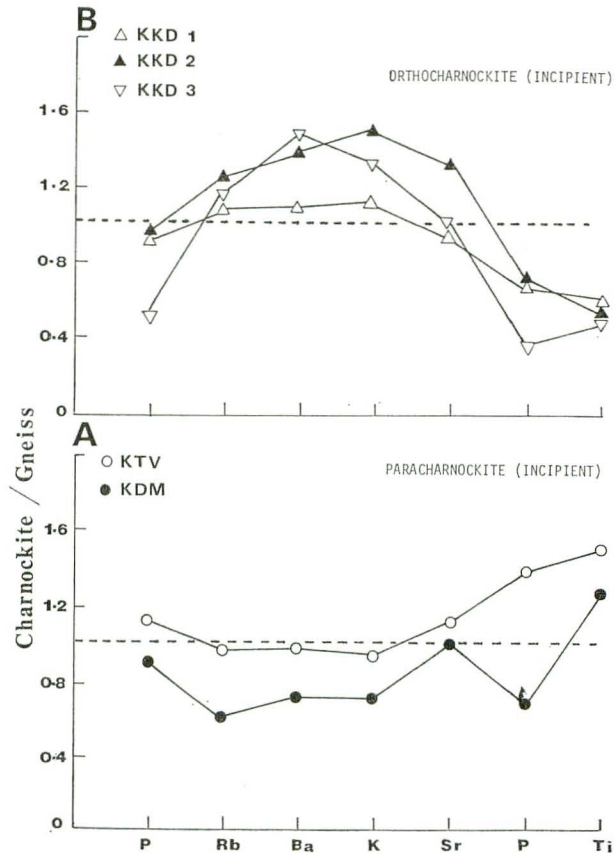


Fig. 7 Spidergrams showing elemental variations in ortho- and para-charnockite localities. The incipient charnockite in each locality has been normalised by its gneiss precursor. The dotted lines indicate charnockite/gneiss=1.

appears to have been accompanied by slight depletion in Pb, Rb, Ba and K, whereas the orthocharnockites show a completely reversed trend with marked enrichment of LILE. The heavier incompatible elements like P and Ti are enriched in the paracharnockite while they show depletion in the latter case. The other elements like Ni, Cr, Co and Zn also show variation from gneiss to charnockite in individual localities, though no common criterion to link these is observed, except the modal abundance of mafic accessories into which these elements are preferentially partitioned.

The low Ba, Rb and Pb values and the significantly high K/Rb ratios of the massive charnockites are well in accord with a model involving pervasive granulitization by carbonic metamorphism at the base of the crust (NEWTON *et al.*, 1980; TARNEY and WINDLEY, 1977; SANTOSH, 1986). Though the incipient charnockites which preserve evidence for arrested granulite formation have long been considered to be the intermediate stage of granulite development in the deep crust, their distinct geochemical characters including undepleted nature require different mechanism of granulite formation to derive such locally desiccated assemblages, repeatedly at different time intervals and involving precursor rocks with different mineralogy and bulk chemistry. Of particular importance is their undepleted nature, implying that melt extraction did not occur, or was insignificant. The arrested charnockites from two different regions of the same high grade terrain, representing two temporally different events, are also broadly contrasted in their trace element patterns. While the paracharnockite formation in the khondalite belt has less pronounced element mobilities and is characterised by only a slight depletion of the lighter incompatible elements, the incipient charnockites of the orthocharnockite segment show well-defined enrichment in LILE. The loss of Rb and Ba at Kottatavtom is correlatable with the instability of biotite during the dehydration reaction. However, since K-feldspar is one of the main product phases in the charnockite, these elements are accommodated, resulting in the relatively low K/Rb values and undepleted nature. There is a marked enrichment in the incompatible elements like Ti and P in this locality. The charnockite forming reaction here consumes garnet, biotite and quartz to produce orthopyroxene, K-feldspar and ilmenite. The trends for Ti could be a reflection of their accommodation in ilmenite. It is presumed that the increased abundance of P in this sample might only be a reflection of the higher modal abundance of apatite in the rock. In contrast, gain of K, Rb, Ba, accompanied by a loss of CaO as observed in KD agree well with similar observations of STAHL *et al.* (1987) from the Kabbaldurga orthocharnockite locality in Karnataka and suggest extensive replacement of plagioclase in the gneisses by K-feldspar in the charnockite through Na-Ca-K exchange reactions with influxing carbonic fluids. The marked depletion in Fe, Mg, Ti and P in this locality correlates with the progressive dissolution of hornblende, biotite, magnetite and accessory minerals like apatite. The near-constant Sr content could be explained by the similar modal amounts of feldspars in both gneiss and charnockite. One of the major recommendations of this study is systematic geochemical investigations in critical gneiss-incipient charnockite localities and representative regional granulite samples in order to resolve these aspects further.

2.8 Fluid Inclusion Studies

As fluid inclusions represent microgeochemical systems containing the remnants of mineral-forming fluids, a detailed study of the inclusion types, their distribution pattern, composition and densities yield valuable information on metamorphic fluid characteristics. Trapped fluid phases were examined in doubly polished wafers (ranging in thickness from 0.2 to 0.8 mm) of rocks containing representative gneiss and charnockite assemblages from number of localities. Inclusion petrography was done using a petrologic microscope under varying magnifications to document the various genetic and phase categories. Heating-freezing experiments for microthermometric observations were carried out using a temperature-calibrated CHAIXMECA microthermometry apparatus. High temperature experiments for visual decrepitation were carried out in a LEITZ-1350 Heating Stage. The composition of representative inclusions were ascertained by laser-induced Raman spectra analysis using Raman microprobe (Model RAMANOR U-1000 of Jobin Vyon) at the RIKEN, Tokyo. The abundance and carbon isotopic composition of the trapped fluids were analysed using a stepped thermal decrepitation method (SANTOSH *et al.*, 1988; JACKSON *et al.*, 1988a, b) at the Planetary Sciences Laboratory of The Open University, U.K. Results on the salient fluid characteristics are briefly discussed in the following sub-sections.

2.8.1. Fluid Phase Petrology

Fluid inclusions occur in most of the minerals in the gneiss and charnockite assemblage, like quartz, garnet, feldspars, cordierite and orthopyroxene. They range from less than 10 microns up to 50 microns in size and show a variety of cavity shapes including rhombohedral (negative-crystal or diamond-shape,) polygonal, elongate, ovoidal, tabular or irregular. Although garnet, feldspars and orthopyroxene contain dominantly single generation fluids, inclusions in quartz belong to more than one generation as indicated by cross-cutting arrays. The most dominant category as well as the earliest generation in all the minerals is characterized by inclusions which are completely filled at room temperature, containing a dense, dark fluid phase (Fig. 8). These are the "carbonic" inclusions, filled with near-pure CO₂ (see below). Although they represent the earliest generation fluids in all the minerals, they are commonly distributed along linear arrays which pinch out across individual grains. This array-bound character need not necessarily indicate that they are late fluids because in a stable crust, carbon dioxide-rich fluids will not form a continuous interconnected grain boundary network and will move only by hydraulic fracture, along cracks (WALTHER and ORVILLE, 1982), as against pure water which can form a continuous fluid network. Hence in most high grade metamorphic rocks, carbon dioxide-rich inclusions are found to occur along sub-linear to linear trails, especially in quartz. However, isolated carbonic inclusions were found in garnet and plagioclase. The inclusions in cordierites appear to be distinctly contrasted from the common ovoidal or diamond-shaped cavities for those in quartz and feldspars (Fig. 8C, D). The larger inclusions in cordierites have perfectly rectangular cavities while the smaller ones are tubular. The uniquely regular geometric shapes for cordierite-bound fluid inclusions

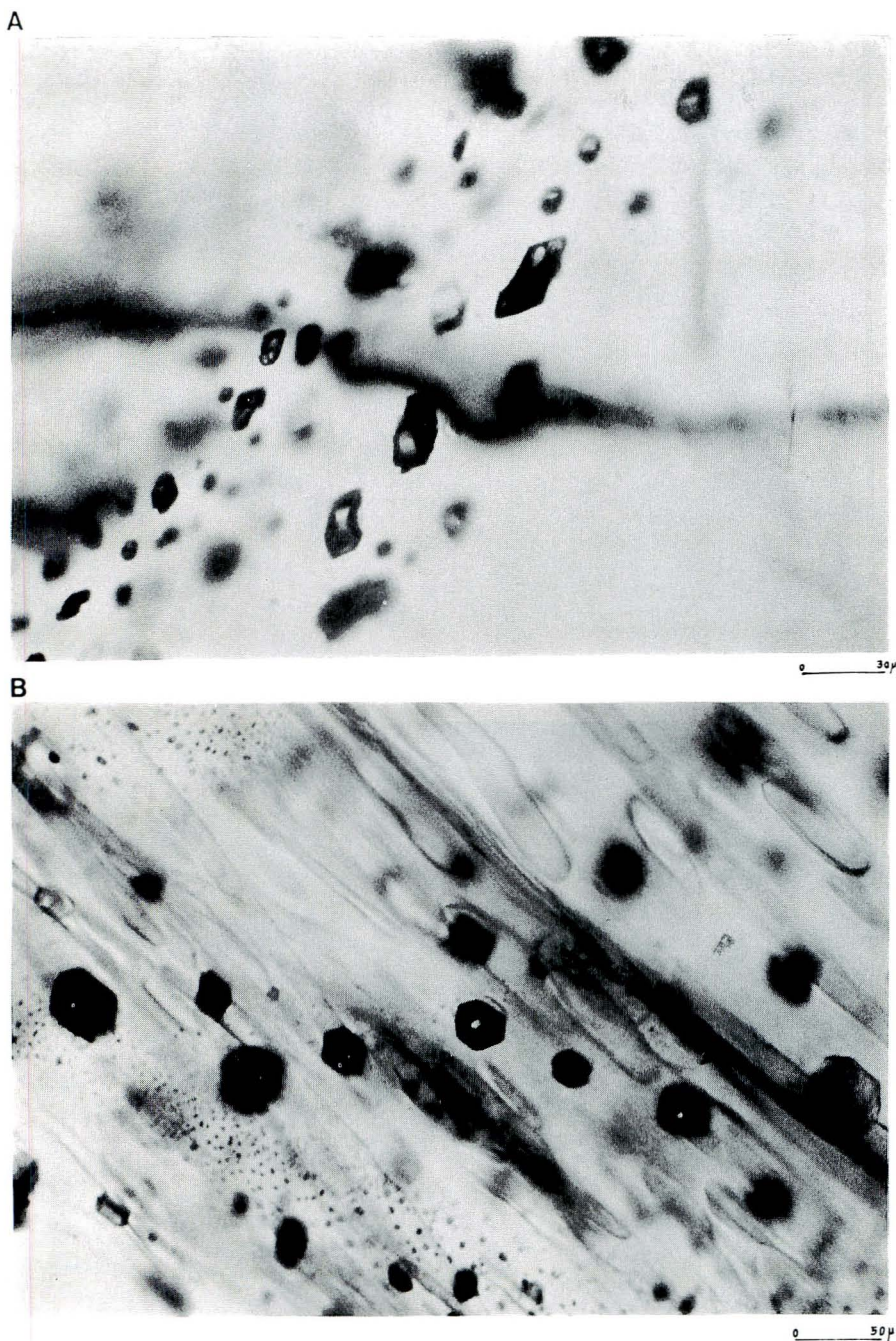
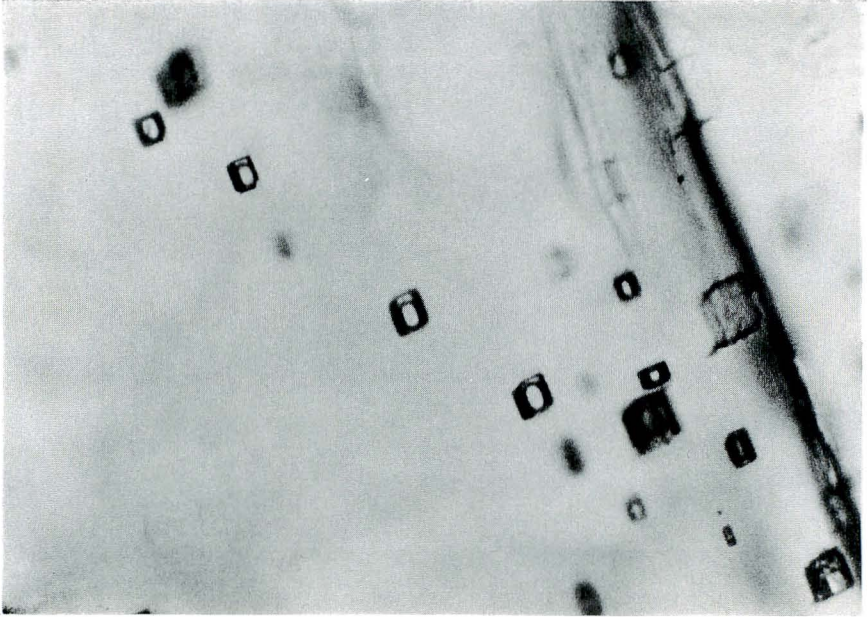


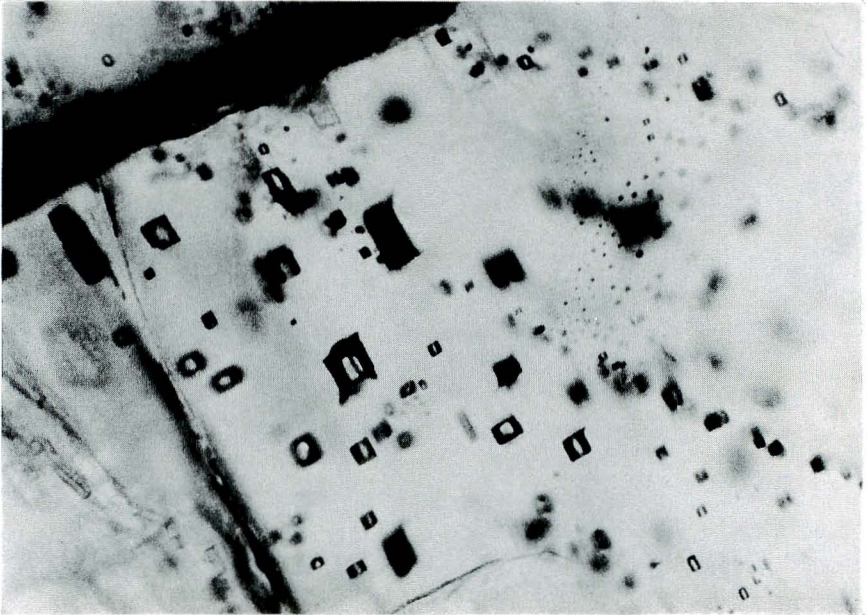
Fig. 8 Photomicrographs of polished rock wafers showing fluid inclusions in various granulite minerals. **A:** quartz, **B:** K-feldspar (perthite) and **C & D:** cordierite. Some of the cordierite-bound inclusions contain low density carbon dioxide so that they already show two phases at room temperature as in **C**, but the phase boundary disappears on slight warming.

C



0 30μ

D



0 50μ

Fig. 8 continued

are evidently linked to the crystallographic properties and crystal-growth mechanism of the mineral, although from the present level of information it is not possible to make any conclusive deductions.

Some of the array-bound carbonic inclusions, particularly from the graphite-bearing (para-type) incipient charnockite localities contain traces of additional fluid species, apart from carbon dioxide. This is brought out by the depression in melting temperatures below that for pure CO_2 (see below). Methane and nitrogen are the likely candidates which commonly occur as dilutants in metamorphic fluids. However, their quantity is in most cases far too little (less than 10 mol per cent) to significantly affect any interpretations of fluid densities.

Some of the carbonic inclusions are bi-phase at room temperature and develop a third phase on slight cooling, consistent with the composition, CO_2 (liquid) + CO_2 (gas) + H_2O (liquid). These mixed carbonic-aqueous inclusions are more abundant in the gneisses than the charnockites. Generally, they are restricted to late fracture-bound arrays where they co-exist with aqueous inclusions and probably result from mixing of late water-rich fluids with previously trapped CO_2 in the charnockite.

The fourth category consists of H_2O -rich inclusions which generally lack any daughter crystals at room temperature and have low salinities. With polygonal, tabular or elongate cavities of small dimensions (ca. 5 microns), they occur along rehealed fractures that normally traverse several grains. Aqueous inclusions are common in quartz from all rock types, though their low abundance and distribution pattern as "secondary" indicate them to be traces of only a late-stage aqueous regime.

A careful examination revealed another rare category of inclusions in the gneisses which show large, irregular decrepitated cavities. The fluid inside these inclusions has normally leaked out and the inclusion is characterized by a trapped cubic crystal of possibly halite. These transposed brine inclusions are restricted to the gneissic quartz from the paracharnockite localities and might indicate an earlier regime of saline brines in the pelitic gneisses.

The various inclusion types in gneiss and charnockite are schematically summarised in Fig. 9. Carbonic inclusions constitute the dominant category in all samples, either forming azonal clusters or planar arrays which pinch out within individual grains. In the graphite-bearing paracharnockite localities, although the earlier regime is characterized by pure CO_2 , a slightly later generation contains possible traces of nitrogen and methane (see below). Late secondary trails are characterized by mixed carbonic aqueous and low salinity aqueous inclusions. Gneisses in the close-pair also contain some carbonic inclusions, but the dominant category has two immiscible fluid phases and corresponds to a mixed carbonic-aqueous type.

From inclusion petrography it is deduced that: (1) the gneiss to charnockite conversion involved addition of CO_2 -rich fluids because the gneisses contain mixed carbonic-aqueous inclusions as the dominant category while the charnockites contain abundant pure carbonic inclusions, and (2) the fluid evolution in charnockite formation was characterized by an early pure carbonic regime, followed by an intermediate mixed carbonic-aqueous regime

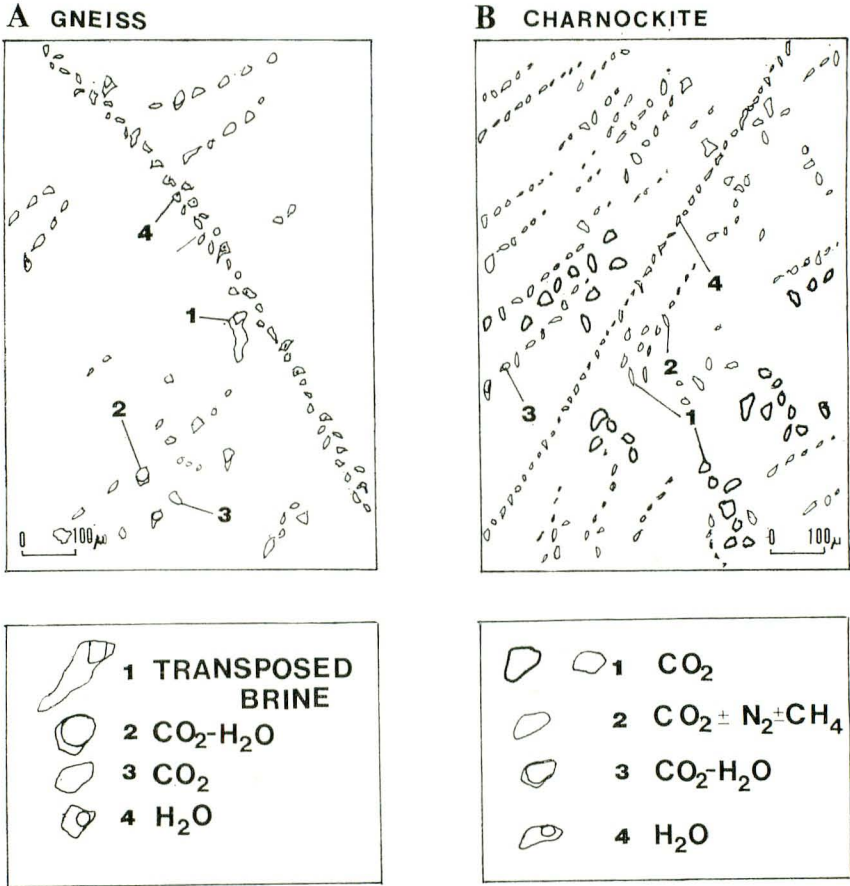


Fig. 9 Sketches from photomicrographs showing the distribution pattern and phase composition of fluid inclusions in gneiss and incipient charnockite. Note the increased abundance of carbonic inclusions in the charnockite as compared to gneiss. See text for discussion.

and a late aqueous regime. Further details for these inferences are presented in SANTOSH *et al.* (1991b).

2.8.2. Composition and Density of Fluids

Fluid composition and densities were estimated from phase changes occurring within inclusions during heating-freezing experiments. Most of the carbonic inclusions freeze on deep cooling and on slow reheating, melting occurs close to -56.6°C (Fig. 10A). Since this is the triple point for pure carbon dioxide, it is deduced that the trapped fluids have a composition approximating to pure CO₂. The slight depression in melting temperatures in a few inclusions below -56.6°C as seen from the histograms in Fig. 10A corresponds to the melting of carbonic inclusions which contain traces of other volatile species, mainly nitrogen, because addition of impurities into the pure CO₂-system would cause a depres-

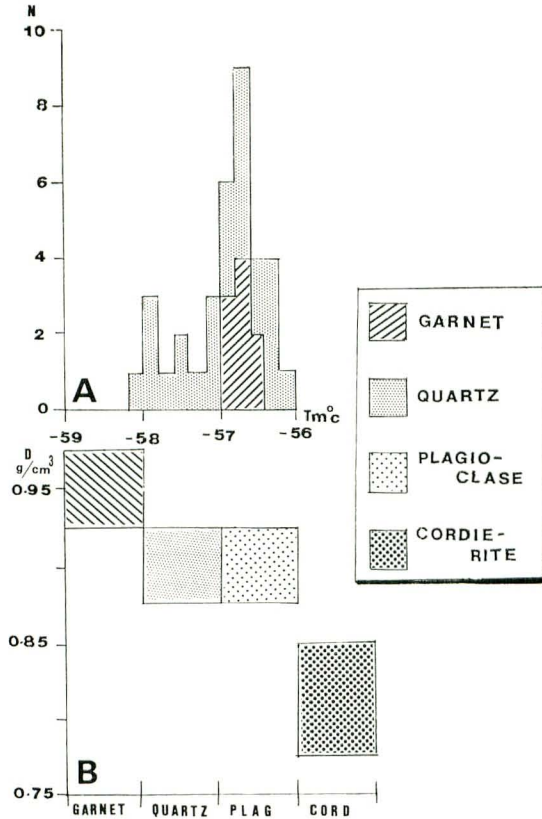


Fig. 10 Microthermometric data on carbonic inclusions in incipient charnockites from southern Kerala. **A:** Histograms showing the melting temperatures. **B:** Density data for carbonic inclusions in different granulite minerals obtained from inclusion homogenization temperatures. Note the pronounced decrease in density from garnet to cordierite. See text for discussion.

sion of temperatures at which melting occurs. KLATT *et al.* (1988) estimated that some of the CO₂ inclusions in Kottavattom incipient charnockite contain upto 14 mol per cent nitrogen. However, in this study, most of the early carbonic inclusions investigated showed a near-pure CO₂ composition, as also ascertained from the spectral positions of Raman shift obtained from laser microprobe analyses of representative inclusions (Fig. 11). No peaks were obtained during scanning in the N₂ and CH₂ regions in these inclusions.

After melting, on further warming of the carbonic inclusions, the vapor carbon dioxide homogenizes into the liquid phase in all the minerals. There is a significant range in the homogenization temperatures of CO₂ in the different minerals. The lowest homogenization temperatures are recorded from garnet and the highest from cordierite. Plagioclase and quartz have inclusions with homogenization temperatures lying intermediate. When

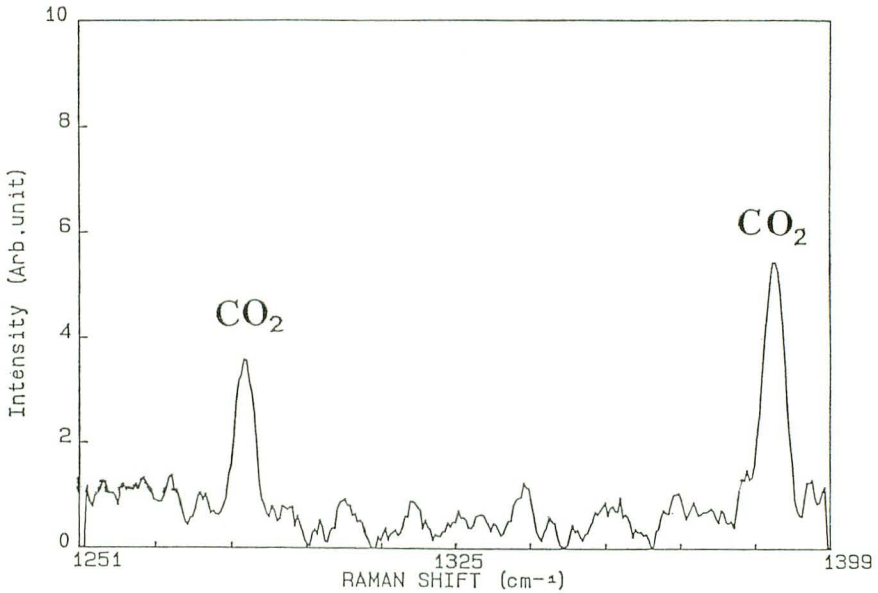


Fig. 11 Laser-induced Raman spectra of a carbonic inclusion from the Mannantala incipient charnockite quarry. The peaks are characteristic for pure carbon dioxide.

translated into densities, these temperatures indicate that the granulite minerals trapped moderate to high density carbon dioxide. Garnet entrapped the highest density fluid, while cordierite entrapped the lowest density fluid. As seen from the microstructural characteristics, garnet and quartz are reactant phases and cordierite is a product phase in these rocks. The density contrast between earlier minerals and the ones which crystallized later is hence important and signifies that the rocks intersected isochores (equal density lines) of progressively lower densities during crystallization and uplift. This has significant bearing on reconstructing the piezothermic array of metamorphism and the tectonic history of the region. A combination of data from solid phase equilibria and fluid phases are shown in a P-T grid in Fig. 12. The isochores for mixed carbonic-aqueous and aqueous inclusions yield a unique determination of the low temperature P-T region where these inclusions were simultaneously entrapped. The combined solid and fluid data define a piezothermic array with its convexity towards the temperature axis, similar to the one deduced from fluid inclusion studies of the low-pressure type massive charnockites in southern India (SANTOSH, 1985). The convexity of the array implies that the rate of uplift exceeded the rate of heat transfer process. The uplift path is in consonance with the field and microstructural characteristics suggesting an isothermal history, aided by decompression during extensional tectonics.

2.8.3. Abundance and Isotopic Composition of CO₂

Stepped thermal decrepitation experiments provide quantitative estimates of the abundance of CO₂ entrapped in inclusions and this has critical implications for the conditions

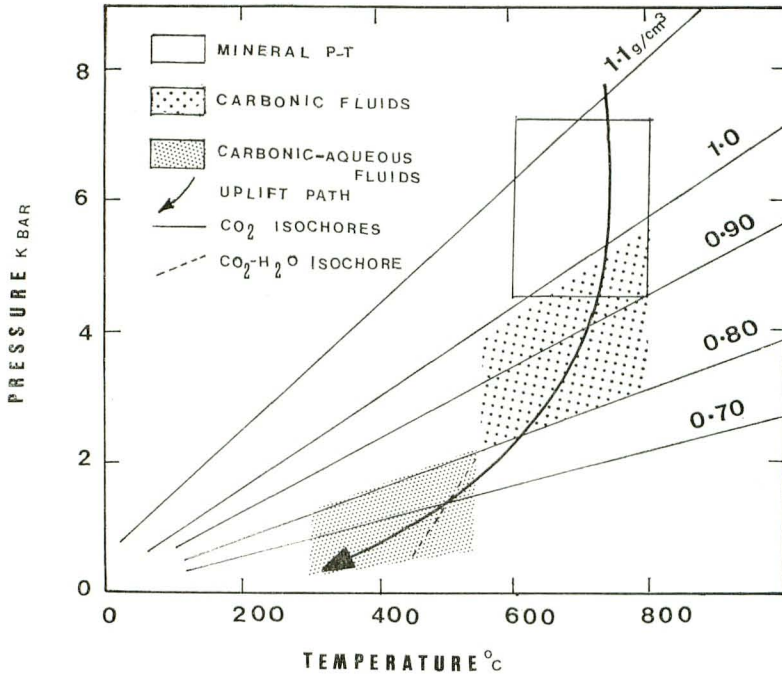


Fig. 12 Combined P-T-X data from solid and fluid phases in incipient charnockites showing carbon dioxide isochores (derived from fluid inclusion data) and P-T box (derived from solid phase equilibria). The fluid evolution from early pure carbonic to mixed carbonic-aqueous is shown by ornamentation. The uplift path defined is typically temperature-convex, indicating an isothermal uplift history for the incipient charnockites.

of fluid entrapment in the incipient charnockite formation (SANTOSH *et al.*, 1988; JACKSON *et al.*, 1988a, b; SANTOSH *et al.*, 1990). These experiments which extract the contents of fluid inclusions at different temperature steps from mineral separates provide information on (a) the actual contents of the inclusions and the abundance of fluids trapped in the different rock types and (b) the stable isotope composition by analysing the carbon isotopes in the extracted fluids using a sensitive mass spectrometer attached to the gas extraction line. Full analytical results of a large set of mineral grains in gneiss-charnockite pairs and cordierite charnockites are presented elsewhere (SANTOSH *et al.*, 1990, 1991b). A few typical examples are discussed here.

Quantitative estimates of the abundance and isotopic compositions of fluid inclusions in five quartz samples from the gneiss and one quartz sample from the incipient charnockite at Mannantala are plotted in Fig. 13. The isotope traverse was done across a representative gneiss-charnockite reaction front discussed earlier in Fig. 3. Although the gas released at 400°C combustion step is normally dominated by isotopically light (down to -26.7%), surficial contamination, a low temperature fluid inclusion release is superimposed on this, resulting in heavier isotope values in some samples (-16 to -22%). The CO₂ yield increases rapidly to a peak release in the 600–800°C interval. This peak release is not only

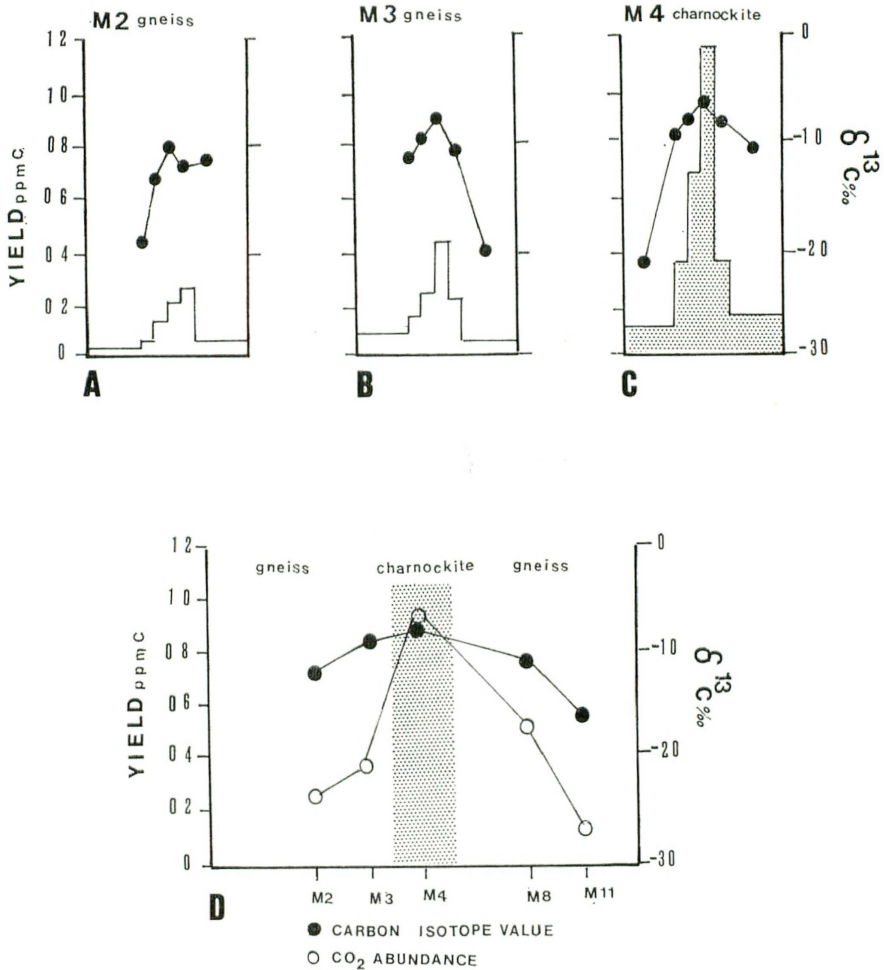


Fig. 13 **A-C**: Stepped thermal decrepitation results of carbonic inclusions in Mantantala rocks (gneisses and incipient charnockite) showing the abundance (histograms) and isotopic composition (solid circles with tie lines) of the released fluids. **D**: Pattern of variation in the abundance of carbon dioxide and its isotopic composition across the reaction front.

characterised by a high carbon abundance, but also by the isotopically heaviest carbon given off the sample (-6 to -8‰ in charnockite and -9 to -15‰ in gneiss). The carbon yield drops at temperatures above 800°C with essentially all carbon released from the sample by 1200°C .

There is a strong correlation between the temperature interval of peak release of CO_2 during stepped heating (600 – 800°C) with the temperature at which carbonic fluid inclusions are observed to explode during visual decrepitation experiments using a heating stage and microscope attachment. In general, mixed carbonic-aqueous inclusions were observed

to decrepitate at $T < 600^{\circ}\text{C}$. The gas released between 600 and 800°C comes predominantly from the rupture of carbonic fluid inclusions. Although carbonate breakdown should be considered as a possible alternative source for CO₂ released at these temperatures, a number of lines of evidence argue against this (cf. JACKSON *et al.*, 1988a, b). All the samples were acid washed to remove any carbonate and a reaction was seldom observed. All the mineral grains were carefully examined under the microscope to check for lack of green veining. Furthermore, fluid inclusions are abundant whereas carbonate is rarely observed.

The most important aspect of the stepped heating results is the significantly higher abundance of CO₂ in the charnockite quartz compared to the gneissic quartz (cf. Fig. 13) over the peak release interval. This is another line of evidence which suggests that charnockite formation and fluid entrapment were broadly synchronous events and the fluid inclusions preserve the remnants of the ambient fluid species which was present during the peak P-T conditions of charnockite formation (cf. SANTOSH *et al.*, 1991b).

The carbon isotope value obtained from the charnockite (-8.2%) is heavier as compared with the values from the gneisses between -9.8 and -12.1% , with progressively lighter values recorded away from the charnockite reaction front. As the charnockite is approached (cf. Fig. 3), the abundances increase and the isotope values become heavier (Fig. 13D). The pattern of variation in the isotopic composition of CO₂ over the peak release range from quartz grains along the traverse is plotted in Fig. 14. Carbon isotope values appear to decrease from the reaction front into the gneiss over a distance of ca. 0.5 m. At distances over 0.5 m from the charnockite, the isotope values are apparently unaffected (-11 to -12%). If the shape of the carbon isotope front between sample M1 and M4 results from diffusion of CO₂, then the data will lie on a sigmoidal curve with a point of inflection along the reaction boundary (SANTOSH *et al.*, 1990) as shown by curve D in Fig. 14. This would imply that the carbon isotope value within the charnockite prior to diffusion is about -5.6% . Although this possibility cannot be totally excluded, it is quite unlikely because the implied carbon isotope value within the charnockite is significantly heavier than has been observed from quartz inclusions in several incipient charnockite localities. Alternatively, if the isotope front results from fluid advection, the isotope values will define a sigmoidal curve with the point of inflection displaced from the charnockite/gneiss boundary (curve A in Fig. 14). Thus, an advective fluid flow mechanism fits well with the observed fluid characteristics. The data also show that the orthopyroxene-in reaction front and the carbon isotope front were decoupled, the carbon isotope front penetrating about 15 cm into the gneiss beyond the charnockite/gneiss boundary. The position of the carbon isotope front is strongly dependent on the graphite content. The isotopic analyses show a shift towards heavier carbon in the charnockite graphite as compared to the graphite in the gneiss (SANTOSH *et al.*, 1990). An external source of CO₂ is required in order to account for the shift to heavier carbon isotope values observed in both graphite and entrapped CO₂ within the charnockite.

The source of carbon dioxide is a much debated topic. For the origin of externally derived CO₂ in metamorphic rocks, three possible sources are envisaged, namely, (1)

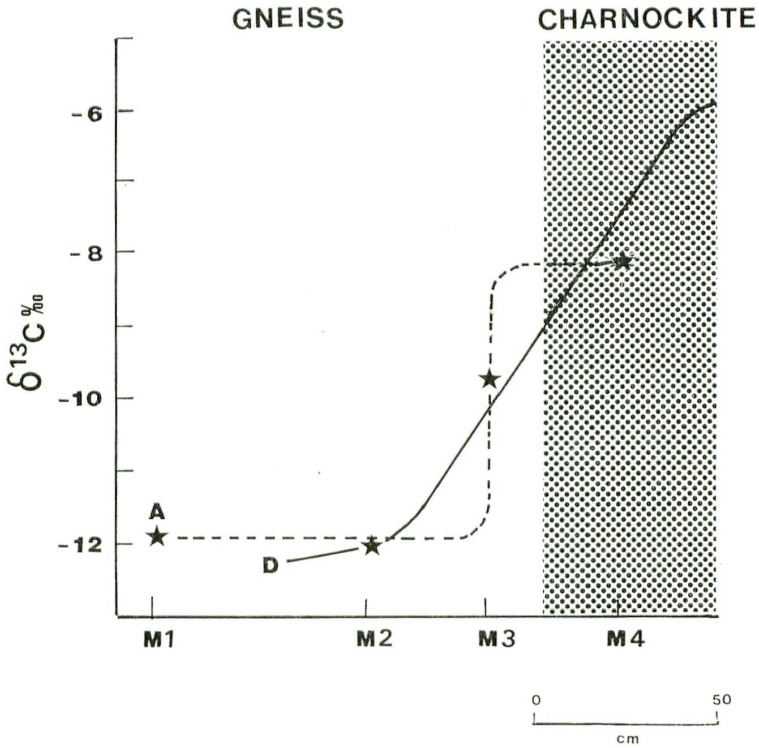


Fig. 14 Carbon isotope traverse at Mannantala charnockite front, showing the profiles for two types of fluid transport, namely advection (A) and diffusion (D). An advective fluid flow mechanism is indicated by the present data. See text for details.

decarbonation reactions, (2) oxidation of organic matter in the rocks and (3) juvenile CO_2 . In these different processes, the CO_2 generated will have broadly distinct isotopic signatures, although a combination of more than one of the above processes is also possible in the case of granulites (cf. SANTOSH *et al.*, 1988; JACKSON *et al.*, 1888a, b). Carbonate rocks yield CO_2 through decarbonation during metamorphism. Under conditions of isotopic equilibrium, the CO_2 thus generated at granulite facies temperatures would be enriched by 2 to 3 per mil as compared to carbonate (BOTTINGA, 1969). The distinctly low carbon isotope values obtained for charnockites which coexist with calc-silicates in south Kerala (SANTOSH *et al.*, 1988) preclude such an origin. The other mechanism is the generation of carbon dioxide by oxidation of graphite. The shift towards heavier carbon levels in the graphite from charnockite as compared to the adjacent gneiss, discussed above, precludes an internal generation of CO_2 . Also, SANTOSH *et al.* (1990) have shown that if such a mechanism operated, then it would produce unrealistically large volumes of methane. The third possibility is a deep-seated source for CO_2 , representing either juvenile CO_2 derived from the mantle by degassing or homogenized crustal carbon (sub-lithospheric source). Large scale CO_2 streaming could occur from the mantle under doubly thickened Archaean

crust, without causing significant melting. The isotopic composition of mantle carbon is variable and it is not strictly possible to attribute a single composition for "deep-seated" carbon. However, despite the fact that the carbon isotopes in diamonds vary from +3 to -35‰, the dominant flux of mantle carbon is outgassed at mid-oceanic ridges (MOR) and has a relatively uniform composition averaging -7.5‰ and ocean ridge basalt (ORB) asthenospheric mantle is characterized by slightly heavier isotope values, averaging -5.5‰. As a first approximation, a carbon flux from subcontinental mantle is likely to be dominated by MOR or ORB source carbon (i.e., between -7.5 and -5.5‰). In the case of incipient charnockites, as in the Mannantala example, the data imply an external source and introduction of CO₂ with carbon isotope value heavier than -8.2‰ (the isotopic ratio CO₂ entrapped in quartz from the charnockite), which is essentially indicative of a juvenile component, either from the mantle or sublithospheric source.

3. Nature of Fluids in other Granulite Terrains

Granulite segments in Sri Lanka, Antarctica and Adirondacks (U.S.A.)

Among the deep crustal segments of granulite grade the world over, those in Sri Lanka, Antarctica and the Adirondack mountains of New York State, U.S.A., have received particular attention in discussions pertaining to granulite petrogenesis (HANSEN *et al.*, 1987; LAMB *et al.*, 1987; YOSHIDA, 1978; SANTOSH *et al.*, 1991c). Of particular relevance in this study is the occurrence of charnockitic rocks in all these terrains. However, apart from South India and Sri Lanka, incipient charnockites showing arrested gneiss-granulite transformation have not been documented from the other two terrains, where the charnockites are generally monotonous and massive in nature.

In Sri Lanka, incipient charnockites occur in a number of localities, mainly restricted to the central and western parts of the Precambrian crystalline segment. Incipient charnockitisation takes place as pods, veins or branching out "trees", regardless to the foliation of the country rocks (SANTOSH *et al.*, 1991c). In many cases, the foliation of the country rocks get warped and pulled into the shears which are filled with charnockitic pods and veins. In the quarry faces, the pods and veins are seen as either scattered, or inter-linked, both representing an intricate network of fluid channels at depth. In a few cases, charnockitization proceeds along mylonitic shear bands. Pink granitic pegmatites, sometimes containing cordierite, and rarely andalusite, are found to be cut by charnockitic veins. Field and microstructural evidences offer a clear case for incipient charnockite formation postdating the major tectonothermal events in the region, including the regional granulite-amphibolite facies metamorphism (YOSHIDA *et al.*, 1990). The age of incipient charnockite formation here has been recently constrained at 430 Ma, based on radiometric dating by KAGAMI *et al.* (1990). The age of regional metamorphism (which produced massive charnockites), on the other hand, has been detected at ca. 1000 Ma based on U-Pb zircon geochronology (KRONER *et al.*, 1987), suggesting at least two distinct charnockite formation events, with the incipient charnockites belonging to the younger event, closely com-

parable with their south Indian counterparts. The dehydration reaction which produced incipient charnockites is similar to that in the south Indian incipient charnockites and is characteristically a low pressure phenomenon (3–5 Kbars at 650–800°C). In localities where garnet and cordierite are present, several reaction textures are observed including cordierite coronae around garnet, cordierite/hypersthene symplectites etc. (SANTOSH *et al.*, 1991c). Consequently, a decompression-related petrogenesis and isothermal uplift history are characteristic of the Sri Lankan incipient charnockites also.

Like Sri Lanka, the Lutzow-Holm Bay region in East Antarctica also has a predominantly Proterozoic geologic history and the comparable geologic, structural and metamorphic parameters, together with detailed consideration of paleomagnetic data have been used to demonstrate the juxtaposition of Sri Lanka, Antarctica and India in the Gondwanian continental fit (YOSHIDA *et al.*, 1990). Charnockites in Lutzow-Holm Bay region are found dominantly in the Skallen and Ongul Group of rocks, where they represent the products of regional granulite facies metamorphism of orthogneisses (with only sporadic garnet) and garnet and biotite bearing paragneisses. YOSHIDA (1978) summarised the tectonometamorphic history of the Lutzow-Holm Bay region where he identified four events. The first event at ca. 1900 Ma is represented by high grade metamorphism and emplacement of igneous plutons which served as the precursors for later charnockite formation. The second event took place at ca. 1100 Ma. during which, intermediate pressure granulite facies rocks developed, dominantly represented by hypersthene-bearing charnockitic rocks. The third event at ca. 900 Ma is characterized by upper amphibolite facies metamorphism and the culmination at ca. 500 Ma marks the fourth event, which was accompanied by the intrusion of granite-pegmatites and associated low pressure-type lower amphibolite facies overprinting, although controversy exists in that all the events from the first to the third category could belong to a single metamorphism during the late Proterozoic. Incipient charnockites of the characteristic patch and vein patterns have not been recorded as yet from Antarctica, probably due to the lack of blasted quarry faces in that remote continent (it is well known that the common presence of fresh quarries in southern India and Sri Lanka aided in the identification of incipient charnockites). Hence, the existence of incipient charnockite in Antarctica cannot be ruled out. YOSHIDA (1978) reported an example of *in situ* charnockite formation from Skallen, where gneisses grade into massive charnockites along a reaction boundary. A postulation based on the knowledge from the south Indian and Sri Lankan terranes would allow to predict the possibility that incipient charnockites may occur in Antarctica associated with either the third or fourth phase of the tectono-metamorphic history as discussed above. These events are characterized by upper amphibolite facies metamorphism and has the right setting to generate granulites along minor shear zones or faults of decompression and enhanced fluid flow. However, this remains an important problem for future field investigations in Antarctica. The present study hence mainly concerns the data from massive-type charnockites in Skallen and Ongul areas. A complex P-T history has been attributed to the Lutzow-Holm Bay region based on detailed field studies and experimental calibrations

of mineral thermometers (YOSHIDA, 1979), with an early high pressure metamorphic episode constrained at 880°C and 13.5 Kbar, followed by an intermediate pressure regime at 880°C and 8.8 Kbar and a late low pressure event around 680°C and 5.5 Kbar. Although there is considerable debate over the correlation of various metamorphic events in Antarctica among different workers, the general picture is that charnockitic rocks formed around 5–8 Kbar pressure and 700–800°C. The interesting aspect to note is that like the south Indian and Sri Lankan terrains, the Lutzow-Holm Bay area also appears to have undergone a decompression-related isothermal uplift history, with drastic decrease in pressure at an almost constant temperature. The piezothermic array for the Antarctic charnockites is hence quite similar to the intermediate pressure massive charnockites of South India (SANTOSH, 1985), characterized by high convexity towards the temperature axis.

The Proterozoic deep crust in the Adirondack Mountains in New York State is part of the Grenville province and were metamorphosed during the Grenville orogeny at 1100 Ma (BOHLEN *et al.*, 1985). Metamorphic grade here increases from upper amphibolite facies in the lowlands to granulite facies in the highlands. The pressures and temperatures show a range of 6–7.6 Kbar and 650–775°C, with the highest P-T located in the central highlands (BOHLEN *et al.*, 1985). The upper amphibolite facies gneisses in the lowlands are dominantly metasedimentary rocks while the granulite facies assemblages in the highlands comprise a large proportion of metaigneous rocks. The highland peak zone is underlain by a large composite body of metanorthosite known as the Marcy massif. The charnockitic rocks occur in the highland series and comprise mangeritic, charnockitic and hornblende granitic to alaskitic varieties. The pressure-temperature-time path of evolution of Adirondack granulites is anti-clockwise and is different from those of the other three terrains of present discussion. The fluid entrapment history, with higher density inclusions captured in late crystallizing minerals also ascribe an isobaric uplift path (see next section). The Adirondack Mountains have been an area of intense research in recent years on the topic of granulite genesis and one of the principal models of granulite formation under vapor-absent conditions has been proposed based on the studies in this region (LAMB and VALLEY, 1984). Adirondacks have also been central to the recent discussions on post-metamorphic fluid infiltration (eg: LAMB *et al.*, 1987; VALLEY *et al.*, 1990), triggering heated debates over the role of CO₂ in granulite genesis.

3.2. Fluid Phase Petrology

Fluid inclusion studies in the gneiss-incipient charnockite reaction front at the classic Kurunegala quarry in Sri Lanka reveal abundant CO₂ inclusions in the charnockite with a melting temperature close to that for pure carbon dioxide. The mean density (0.87 g/cm³) is slightly lower than their equivalents in the South Indian terrain, and correlates with the lower pressures for charnockite equilibration in Sri Lanka (SANTOSH *et al.*, 1991c). Visual decrepitations of carbonic inclusions in polished wafers of gneiss and charnockite indicate that the latter contains almost double the abundance of CO₂ as compared to the

former, as depicted by the increased number of decrepitation events. Fluid evolution is traced from early carbonic through intermediate mixed carbonic-aqueous to late aqueous regimes. The carbonic-aqueous inclusions with varying vapor:liquid ratios suggest immiscibility between the two fluid phases during uplift, constrained at ca. 485°C and 1.8 kbar, either by unmixing of an originally homogeneous carbonic-aqueous fluid or by mixing of late aqueous fluids with the early pure carbonic fluids (SANTOSH *et al.*, 1991c). Microthermometric measurements attribute a salinity of ca. 6 wt. per cent NaCl equivalent for the late aqueous fluids. The fluid characteristics strongly suggest that dehydration and incipient charnockite formation was effected by externally influxed carbon dioxide-rich fluids.

The charnockites from Lutzow-Holm Bay region in Antarctica also commonly contain carbonic inclusions. They are generally distributed along linear arrays which pinch out within individual grains in quartz. However, scattered, azonal carbonic inclusions were observed in garnet and plagioclase. From fluid phase petrology, it has been estimated that there is a pronounced increase in the content of monophase carbonic inclusions while passing from the amphibolite grade gneisses to granulite grade charnockites (SANTOSH and YOSHIDA, 1991). The gneisses are dominated by mixed CO₂-H₂O inclusions. The carbonic inclusions show melting temperatures close to -56.6°C, indicating a pure carbon dioxide composition. Homogenization occurs into the liquid phase at considerably low temperatures (-20°C), indicating very high density (up to 1.0 g/cm³) for the trapped carbon dioxide fluid. This correlates well with the higher pressures of equilibration for the Antarctic charnockites as estimated from mineral phase equilibria. From garnet through plagioclase to quartz, there is a progressive decrease in the density of the entrapped fluid and some of the carbonic inclusions in late secondary trails contain low density CO₂ ca. (0.80 g/cm³). This density decrease is in correspondence with a T-convex uplift path for these rocks whereby isochores of lower densities were intersected during uplift.

Detailed fluid inclusion studies in the Adirondack granulites are reported by LAMB *et al* (1987), and the results have been synthesized by them and by VALLEY *et al* (1990) to argue that the fluids were post-metamorphic. Abundant CO₂-rich inclusions characterize the Adirondack granulite assemblage. In this study, fluid inclusions were examined in charnockite, mangerite and anorthosite samples from the highland area and all the samples showed at least two generations of fluids (SANTOSH, in prep.). The earlier generation is high density pure carbonic, while the cross-cutting later generation contains low density CO₂ or mixed CO₂-H₂O fluids, coexisting with H₂O rich inclusions. Microthermometric measurements indicate that quartz entrapped fluids of higher density (up to 1.1 g/cm³) than garnet and pyroxene (0.80-0.95 g/cm³). This indicates that isochores of progressively higher densities were intersected during uplift and the uplift mechanism could thus have been of isobaric-type, as against the isothermal histories for the other granulite segments considered in this study. Although one school of thought considers the Adirondack inclusions to be the result of post-metamorphic fluid infiltration (LAMB *et al.*, 1987), it is felt that there is ample evidence to suggest a peak-metamorphic entrapment history of the high

density carbonic fluids (SANTOSH, in prep.). One of the key arguments to invoke post-metamorphic entrapment is the array-bound nature of the inclusions and the variable densities along the same arrays (LAMB *et al.*, 1987). In fact, both of these features need not necessarily indicate that the fluids are "secondary" in origin. As mentioned in an earlier section, carbon dioxide seldom forms an interconnected grainboundary network in a stable crust undergoing deformation and hence the movement of CO₂ occurs dominantly by hydraulic fracturing, mostly along cracks in minerals. Rehealing of these cracks would result in arrays of carbonic inclusions, which on first look may be misinterpreted as secondary. Recent experimental studies on synthetic fluid inclusions by STERNER and BODNAR (1989) indicate that if inclusions in the size range from 3 to 50 microns are subjected to a pressure differential of 1.0 to 1.5 Kbar at a fixed temperature, they will change both their shape and volume. Consequently, the internal fluid pressure would revert to within 1 Kbar of the external pressure. But most important, in the course of these experiments, majority of the inclusions remained as closed systems with no change to their original composition, in spite of externally applied compositional gradients. It is thus possible to model all the Adirondack inclusions with variable densities in terms of peak entrapment. The fact that production of large volumes of carbon dioxide by any mechanism is most likely to occur during the highest thermal anomaly attending granulite formation itself testifies to the entrapment of CO₂-rich inclusions during charnockite formation. These aspects, together with the new finding of melt inclusions in the Adirondack granulites presented in the next section, require a reconsideration of the timing of fluid entrapment in the Adirondack granulites.

3.3. Silicate Melt-CO₂ Inclusions and Their Implications

In this study, in addition to the detailed investigations in incipient charnockite localities of southern India, a few massive charnockite samples from southern India, Sri Lanka, Antarctica and the Adirondacks were examined in order to assess the role of fluids in their petrogenesis. A detailed account of the results will be synthesized elsewhere, but the most salient finding is the occurrence of silicate melt and CO₂ inclusions. In Figs. 15–18 are presented the photomicrographs of inclusions in massive charnockites from four crustal segments. Indeed, in all cases, carbonic inclusions invariably occur. But what is more significant is the co-existing inclusions which contain several solid phases, microcrysts or devitrified silicate glass. Previous studies have either not identified them or ignored them. These inclusions were mainly found in garnet and plagioclase, although some occur in quartz as well. They occur in a variety of shapes from geometrically regular to highly irregular patterns. Most of the cavity is occupied by an intricate aggregate of microcrystalline grains; some of them contain well-developed solid phases with perfect crystal outlines as in the large melt inclusion from Adirondacks shown in Fig. 18 D. These inclusions also contain a gas bubble, normally in a compressed state, which occupies less than one fourth of the cavity volume. In all characteristics, they correspond to recrystallized melt inclusions described from varied igneous rocks including granites (cf. ROEDDER, 1984). Identification of the individual daughter crystals is not easy by simple optical

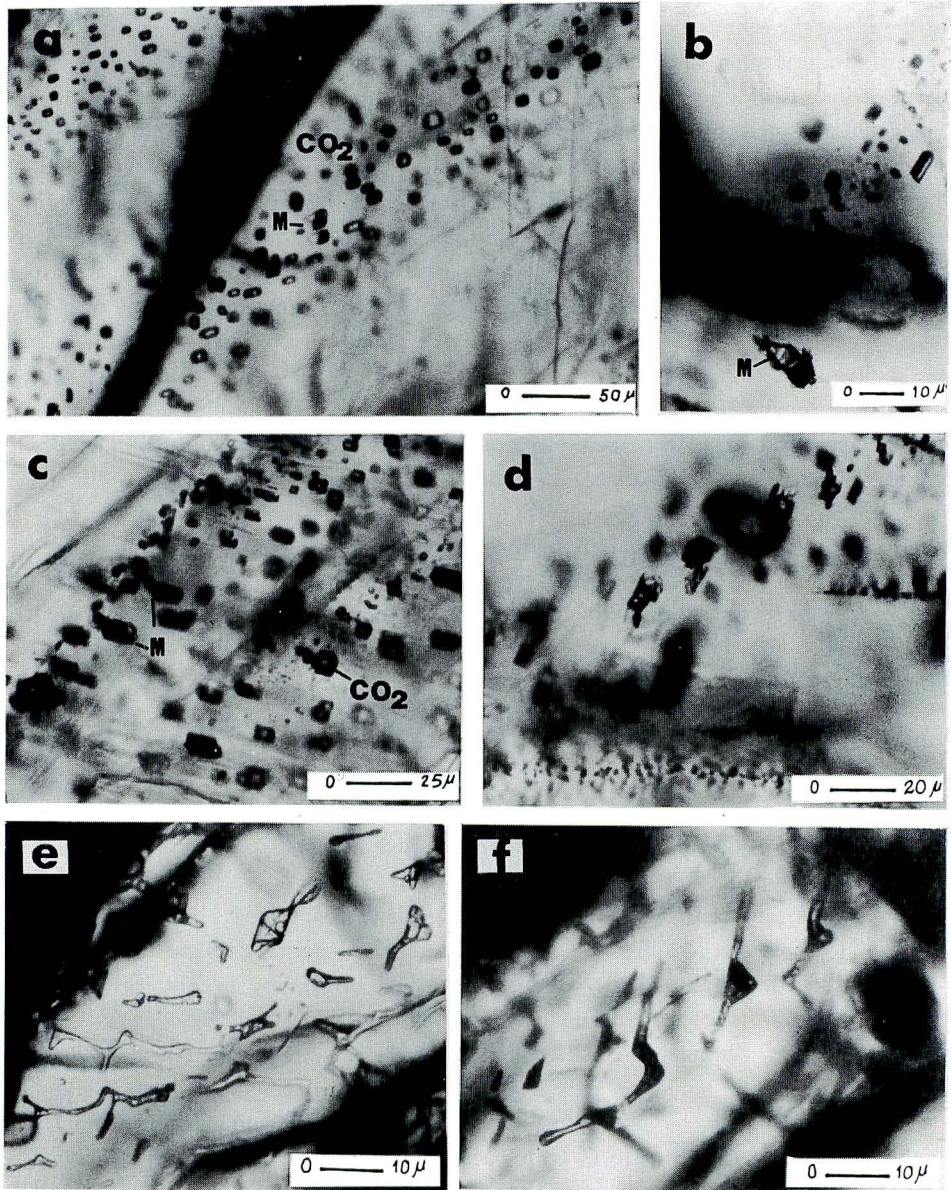


Fig. 15-18: Silicate-melt and CO_2 inclusions in granulite minerals of massive charnockites from southern India, Sri Lanka, Antarctica and Adirondacks respectively. **e** & **f** in Fig. 15 and **d** in Fig. 17 show microcrystalline aggregate of solid phases within garnet-bound inclusions. M-indicates melt inclusion.

techniques due to their extremely small size, especially those occurring as microcrysts in garnet-bound inclusions. This aspect remains to be resolved by the ongoing studies. It is interesting to note that in most cases, if not all, the melt inclusions coexist with monophase carbonic inclusions of high density (Fig. 15–18). The inclusion petrography thus suggests that the silicate melt and carbonic inclusions in these rocks are primary and were entrapped contemporaneously.

High temperature heating experiments were carried out on representative melt inclusions in garnet, plagioclase and quartz (Fig. 19). On heating, the daughter crystals within the inclusions started dissolving around 650°C and almost all of the solids melted by 800–1000°C. There is a considerable overlap in the dissolution temperatures of silicate-melt inclusions in various minerals from the different terranes and lie in the range of 750–1000°C. Freezing experiments indicate that the compressed fluid bubbles inside these inclusions are CO₂, although only limited observations could be made due to the small size of the bubble within the tiny inclusions, making phase changes difficult to document.

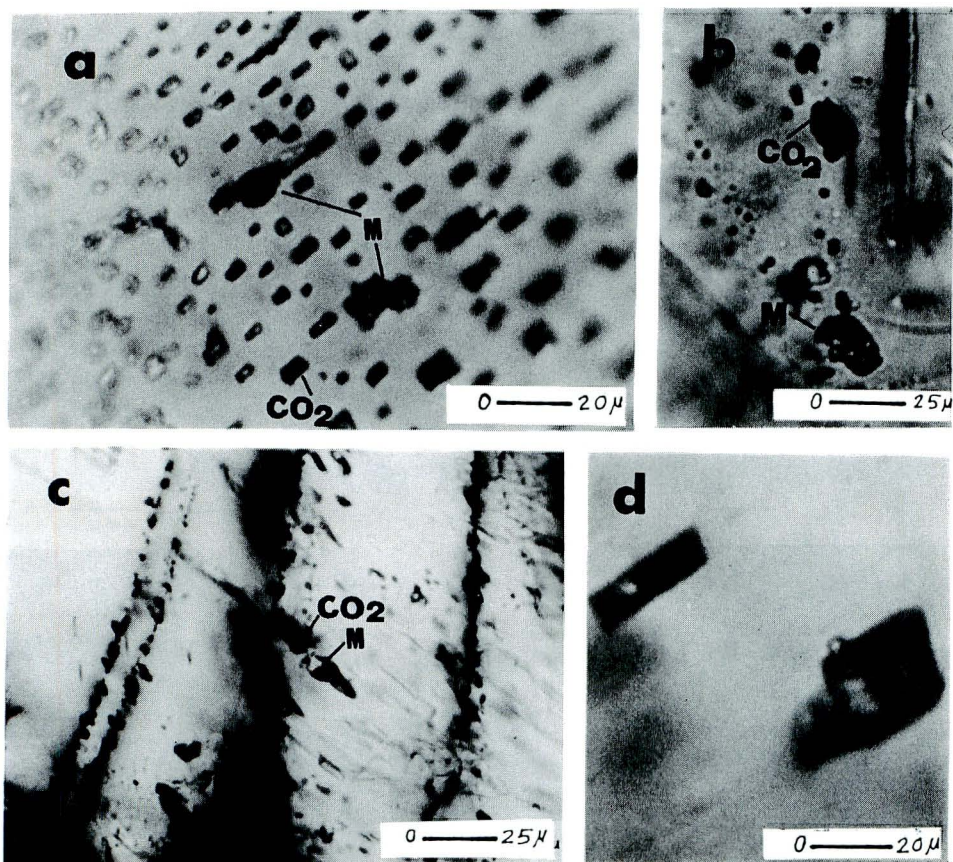


Fig. 16

In the coexisting fluid-rich inclusions, phase-changes were vividly observed and in all cases, they turned out to be pure carbon dioxide of the highest density among the varying density populations in each sample. A combination of the carbon dioxide densities and melt homogenization temperatures serve as a unique barometer for estimating their entrapment pressures. Thus the high density inclusions (av. 1.0 g/cm^3) and the higher melt homogenizations (1000°C) yield a pressure of ca. 7 Kbar for Antractic and Adirondack samples where as the intermediate densities ($0.80\text{--}0.95 \text{ g/cm}^3$) and homogenization

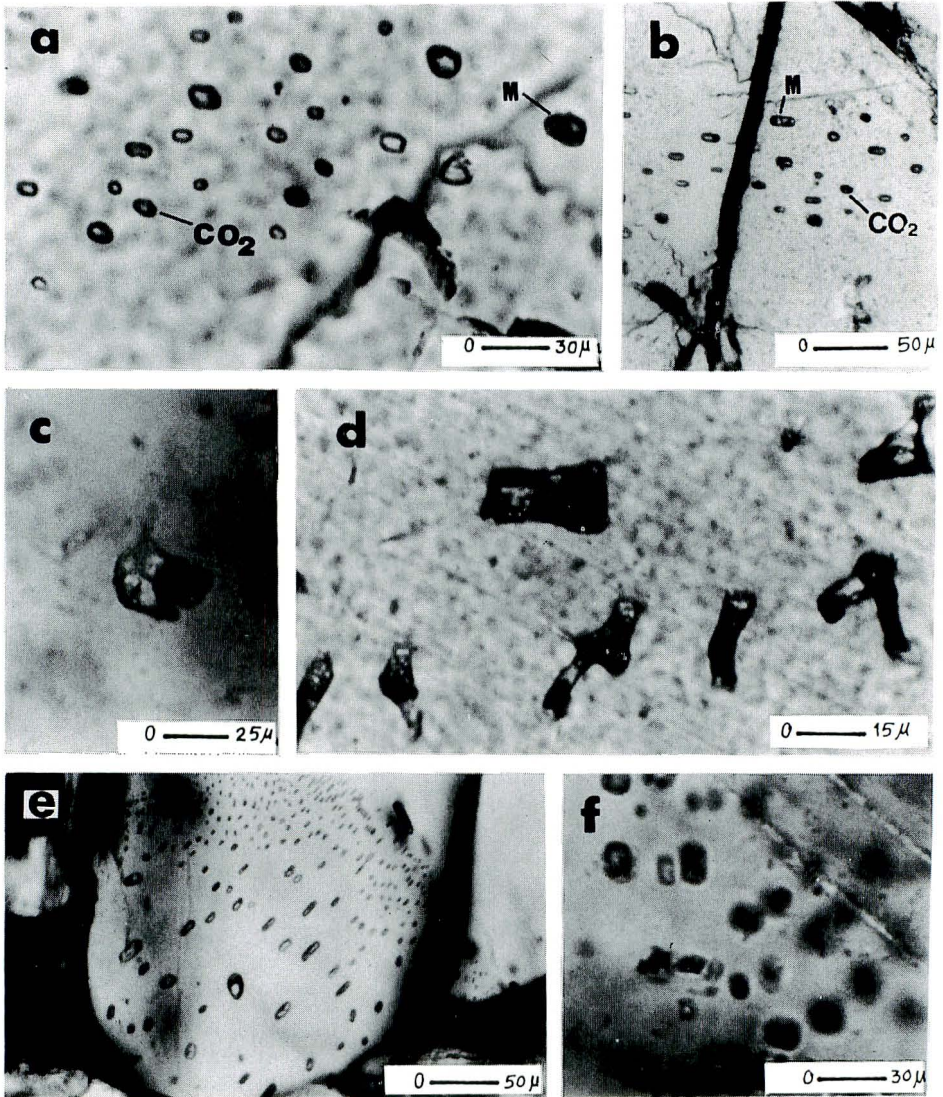


Fig. 17

temperatures (800–900°C) ascribe pressures of 4–5 Kbar for the south Indian and Sri Lankan samples.

One of the most important implications of the occurrence of silicate melt-carbon dioxide inclusions is on the petrogenesis of massive-type charnockites. Although the incipient charnockites provide compelling field evidences of local dehydration controlled by

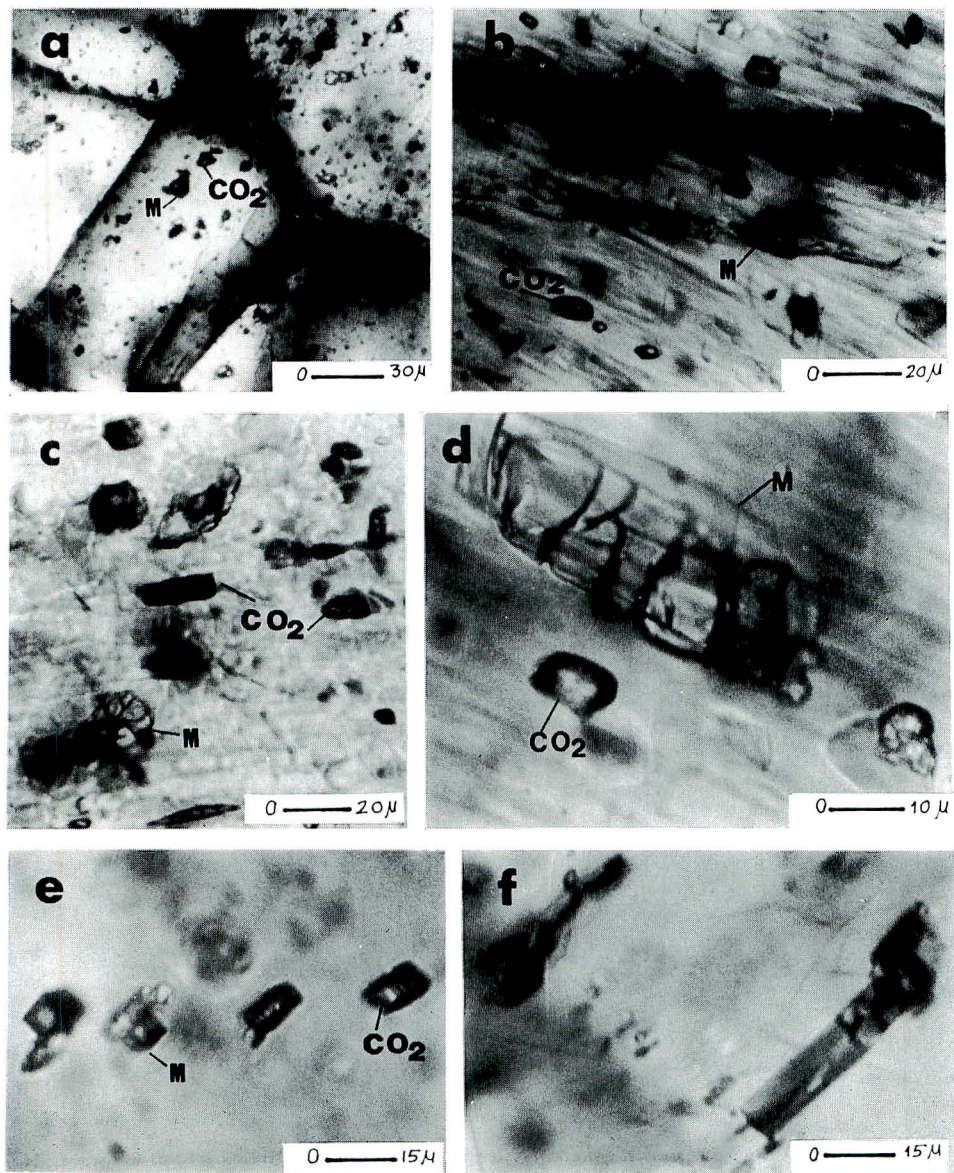


Fig. 18

the influx of CO_2 along structurally-controlled fluid pathways, there is no concrete evidence to prove that all granulites including massive charnockites formed by the flushing of fluids. Experimental studies have shown that in liquids of granitic composition, orthopyroxene is only a stable liquidus phase when low water contents are present. In order to stabilise orthopyroxene, CO_2 must be added to the system (GRANT, 1986). In the presence of pure CO_2 , complete crystallization of a granitic magma will occur above 1000°C with orthopyroxene as the only ferro-magnesium phase (WENDLANDT, 1981). If the liquid coexists with a mixed carbonic-aqueous vapour phase, the solidus will be considerably lowered to below 800°C and the water component will preferentially dissolve in the liquid

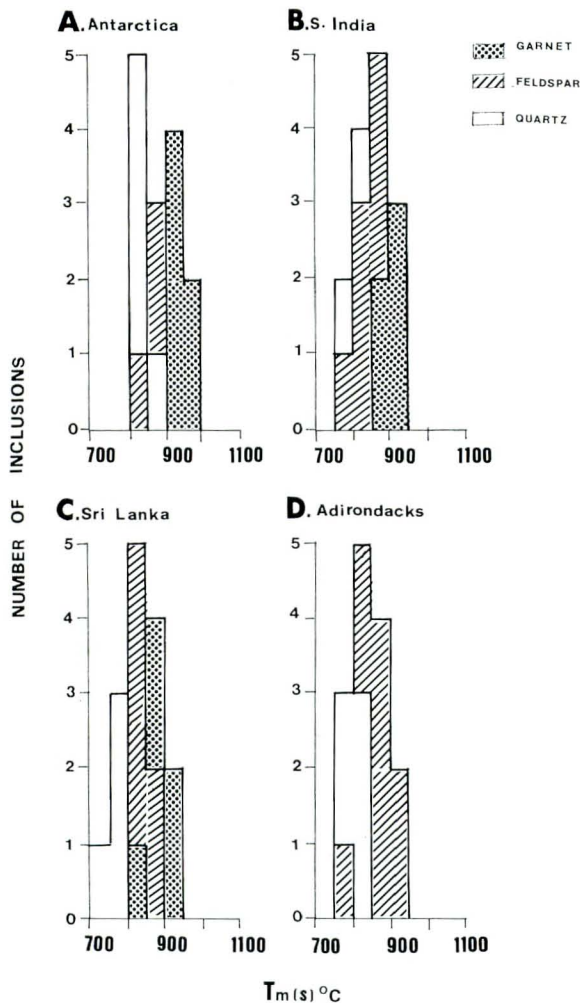


Fig. 19 Histograms showing melting temperatures of solid phases within silicate-melt inclusions in different minerals in massive charnockites from four granulite terrains. See text for details.

resulting in the crystallization of orthopyroxene and biotite (GRANT, 1986). Recently, PETERSON and NEWTON (1989) made experimental determination of the enstatite and phlogopite melting curves in the KMASH-CO₂ system for pure H₂O and CO₂ endmembers at 720°C. The results show that CO₂ induced melting can occur at granulite facies temperatures. A combination of data from GRANT (1986) and PETERSON and NEWTON (1989) imply that for identical melt compositions, high water activities will result in the formation of a biotite granite, whereas low water activities may give a charnockite assemblage. One of the potential mechanisms of lowering the water activity is by the addition of a CO₂-rich fluid phase.

The silicate-melt inclusions now identified in massive charnockites from a number of terrains in this study indicate the presence of a melt phase during the early crystallization history of these granulites. In all cases, they are accompanied by carbonic inclusions. From the foregoing discussion, it follows that at least in some granulite facies rocks, CO₂ could have induced melting. From PETERSON and NEWTON's (1989) experimental study, melt inclusions could be consistent with infiltrated liquids attendant upon migmatization, or liquids formed *in situ* in a CO₂-induced migmatization. Conversely, they could be residual CO₂-enriched melts from the crystallization of charnockitic plutons (R.C. NEWTON, pers. comm., 1990). Further studies are required to resolve these aspects. However, the present observations undoubtedly place CO₂ as a critical variable which determines the stability of granulite facies assemblages.

4. Role of Fluids in Granulite Formation: Concluding Remarks

4.1. Field Evidence

Field evidence for fluid-controlled granulite formation mostly comes from localities where gneisses and granulites occur in intimate association in hand-specimen scale, precluding the possibility of any significant temperature gradients or removal of a melt phase. Compelling evidence for deep crustal fluid pathways are documented from two terrains in this study, namely South India and Sri Lanka. The probable existence of similar features are predicated for a third terrain, Antarctica. The simplest field truth is the structural control of charnockite formation in the "arrested" incipient charnockite localities, where granulites start forming as clots and patches, spread along the pathways of fluid infiltration into veins and branching out "trees", and finally to anastomosing network, overprinting all penetrative deformational structures. Abrupt coarsening of grain size, complete obliteration of gneissic fabric and warping of foliation in the precursor rocks about the coarse charnockitic patches all point to the *in situ* transformation of upper amphibolite facies gneisses into dry granulitic charnockites along the course of fluid influx. Increased reaction kinetics along zones of high fluid flow, brittle-dominated rheologic properties and decompression-related genetic history are also suggested from the field characteristics.

On the other hand, the gneiss to granulite progression on a regional scale as observed in the massive charnockite terrains of South India, Sri Lanka, Antarctica and Adirondacks

are different from the locally generated incipient charnockites and no doubt, are exposures of the exhumed sections of the deep crust. Since they are monotonous granulite exposures in the absence of associated gneisses, granulitization by pervasive fluid flow at the base of the crust remains an attractive option.

4.2. Mineralogic Evidence

The transformation from gneiss to granulite involves the breakdown of hydrous minerals like biotite and/or hornblende to orthopyroxene (common for both incipient and massive charnockites). In the paracharnockite localities of South India, biotite transforms to pyroxene and in the orthocharnockite localities, hornblende is the major mafic constituent which is broken down. Both examples are present in Sri Lanka also. Mineral phase equilibria considerations require that water activities should be considerably low to break down mica or amphibole so as to produce the pyroxene. According to experimental constraints, the conversion of phlogopite+quartz to enstate+K-feldspar+vapor at the temperatures at which these rocks equilibrated require the $X_{\text{H}_2\text{O}}$ to lie below a value of 0.35. A detailed computation of the water activity in one typical locality (Mannantala) shows that the charnockite equilibrated at $X_{\text{H}_2\text{O}}=0.2$. Calculations also reveal that in localities of gneiss-charnockite reaction fronts, only a slight decrease in water activity (0.05–0.1) in the gneisses is required to drive the dehydration reaction and generate a charnockite at the observed temperatures of ca. 700°C. The removal of water and lowering of water activity can be achieved by (1) the vapor-absent metamorphism of previously dry lithologies, (2) removal of water in a melt phase through partial melting and melt extraction or (3) diluting or driving out the water by flushing with non-aqueous fluids. Combined field and analytical data presented in this work show that mechanism (1) and (2) did not operate or were not critical in generating the charnockites in the different terrains investigated, which leaves option (3) of mineral transformation effected by CO_2 buffering.

In addition to the mechanism of dehydration, considerable information on the tectonic evolution of granulites could be tapped from mineralogy. In the paracharnockite localities, charnockite formation involves the consumption of garnet and quartz along with biotite in the gneisses. This reaction is essentially pressure-dependent and proceeds by isothermal decompression. In areas where cordierite is associated, a number of reaction textures are observed, including cordierite corona around garnet, cordierite-hypershene symplectites etc. The reaction, garnet+quartz=cordierite+hypersthene has been noted to be the key reaction in the formation of cordierite-bearing charnockites along the Achankovil shear zone in southern India, which is also pressure-dependent. These mineral reactions and microstructures indicate that incipient charnockite formation was slightly post-peak metamorphic and occurred through isothermal decrease of pressure under an extensional tectonic regime. Except in the Adirondack Mountains, isothermal uplift histories have been traced for the other granulite terrains of present study.

4.3. Evidence from Geochemistry

Although gneiss-granulite conversion in the incipient charnockite zones has been

earlier though to be an isochemical phenomenon, geochemical studies of close-pair samples indicate that granulite formation, even if on a localised scale, is attended by distinct patterns of elemental abundance and mobility. However, these patterns are contrastingly different for incipient charnockites as compared to regional granulites (including massive charnockites). While the incipient charnockites have undepleted chemistry (which preclude the model of granulite genesis involving the removal of a melt phase) and very low K/Rb values, the massive charnockites are characterised by large ion lithophile-element depletion and high K/Rb values. Among the incipient charnockite localities, those characterized by the consumption of garnet, biotite and quartz to produce orthopyroxene (paracharnockite) exhibit a marked enrichment in incompatible elements like Ti and P, with simultaneous loss of Rb and Ba. In contrast, the orthocharnockite localities show a pronounced LILE-enrichment, with gain of K, Rb and Ba, with loss of CaO, suggesting extensive replacement of plagioclase in the gneisses by K-feldspar in the charnockite through K-Na-Ca exchange reactions with influxing carbonic fluids. The depletion in Fe, Ti and P in these rocks correlate with progressive dissolution of hornblende, biotite, magnetite and accessory minerals like apatite. These distinct chemical mobilities are finger-prints of a fluid-controlled transformation process.

4.4. Evidence from Fluid Inclusions

In all the terrains of present study, in most of the granulite minerals, trapped fluid phase occurs commonly and several lines of evidence presented in this work point to their capture during the peak P-T conditions of charnockite formation. The occurrence of such inclusions itself testifies to a fluid-present metamorphic history. From microthermometry and laser-induced Raman spectrometry, it is found that the composition of the dominant fluid species corresponds to pure carbon dioxide. Dilutants like methane or nitrogen, if present, occurs in insignificant amounts of less than 10 mol per cent. Visually, charnockitic minerals contain many more arrays of carbonic inclusions than the gneisses, the latter having predominantly mixed carbonic-aqueous inclusions. Optical decrepitations indicate almost double the number of explosions in charnockites than the gneisses. Extraction of fluids by stepped thermal explosion and quantitative measurements of carbon dioxide yield the final proof of contrastingly higher abundance of CO₂ in the charnockite, requiring external addition of carbon dioxide during charnockite formation and its capture by various minerals during their growth. No such contrast between gneiss and charnockite can be expected had the fluids pervaded the rocks after peak metamorphism. A traverse across the gneiss-charnockite reaction front in the Mannantala incipient charnockite quarry reveals a pronounced increase in the abundance of CO₂ and shift towards heavier carbon in the carbon stable isotope value of the fluid as the charnockite is approached. Graphite in the charnockite is also enriched with heavier carbon than that in the gneiss. These features indicate that externally derived carbon dioxide was flushed along shears and fractures, which advected from the centre of the charnockite lens into the adjacent gneisses over short distances. These carbonic fluids were critical in effecting dehydration and granulite formation.

One of the important findings of this study is the side-by-side occurrence of silicate-melt CO₂ inclusions in massive charnockites from all the four terrains investigated. The silicate melt inclusions point to the existence of a melt-phase during charnockite formation and the coexisting carbonic inclusions signify that melting was induced by carbon dioxide-rich fluids. Experimental phase equilibria considerations indicate that at the temperature ranges of equilibration of these rocks, it is possible to induce melting by CO₂-rich fluids at the base of the crust and generate orthopyroxene-rich granulitic assemblages. This opens up a radically revised thinking about the mechanism of deep crustal metamorphism and generation of large segments of dry granulites. It thus follows that even if different mechanisms of granulite genesis operated in different terrains, CO₂ played a critical role in stabilizing their dry mineral assemblages.

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