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Research Paper

Incipient charnockites from southern India: The role of brines

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ABSTRACT

Southern India and Sri-Lanka are the places where “incipient charnockites”, i.e. the local transformation of amphibolite-facies gneisses into orthopyroxene-bearing, igneous looking charnockites, have been discovered in the early sixties. The fact that some incipient charnockites occur along a network of brittle fractures, together with CO₂ remnants preserved in mineral inclusions, had called for the role of fluids during charnockite alteration. The present work presents new observations on fluid inclusions and microtextures of incipient charnockites from type localities in southern India. In addition to CO₂-rich fluid inclusions in quartz and feldspar, all of the occurrences have disrupted remnants of concentrated aqueous alkali chloride solutions. CO₂ inclusions are more abundant in paragneiss (Kerala) than in orthogneiss (Karnataka/Tamil Nadu). The finding of disrupted brine inclusions in the Kabbal charnockite is a key link between closely associated massive charnockites and Closepet Granite, both of which also share the brine remnants. All of the occurrences studied here have feldspar or feldspar-quartz microvein networks along grain boundaries of recrystallized quartz, feldspar and orthopyroxene. These metasomatic veins again indicate the action of alkali-exchanging fluids (i.e., saline solutions). Feldspar microveins, which have been found in most “massive” charnockites, along with the CO₂-rich fluid inclusions, suggest a commonality of incipient charnockite and massive charnockite, both types differing in intensity of interaction with metasomatizing pore fluids.

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1. Introduction

The average composition of the continental crust, which covers nearly one third of the Earth's surface, is widely believed to be andesitic or dacitic, with 57–66 wt.% SiO₂ (Rudnick and Gao, 2014). Despite years of studies, there are still discussions about the detailed composition, notably for its lower part, in contact with the Upper Mantle at the base of the continent. For a long time, the lower continental crust was considered to be dominantly mafic, but most recent estimates show that, except beneath collision zone, average SiO₂ content in the lower crust is between about 49 wt.% and 62 wt.%, partly overlapping with the SiO₂ content of the more acidic middle crust (53–70 wt.% SiO₂, Hacker et al., 2015). In fact, the greatest difference between middle and lower continental crust

is less the average composition than the metamorphic grade. Both are composed of metamorphic (migmatites) and magmatic (dominantly granitoids) rocks, showing a progressive replacement of hydrous minerals (biotite, amphibole) in the amphibolite-facies middle crust by H₂O-free mineral assemblages (garnet, orthopyroxene) in the granulite-facies lower crust (Rudnick and Fountain, 1995). Typical igneous (magmatic) intrusions in the metamorphic complexes are granites in the middle crust, orthopyroxene-bearing granitoids (charnockites) in the lower crust.

The transition zone between amphibolite- and granulite grade metamorphic rock, which can be taken as the limit between middle- and lower-crust, is marked by the formation of dark-greenish, pyroxene-bearing dehydration zones, intersecting the migmatite layering and occurring either in veins centimeters to meters thick or decimeter-size rounded patches (see illustrations in e.g. Rajesh and Santosh, 2012). These have first been named “charnockites in the making” (Pichamuthu, 1960), then “arrested charnockites” (Hansen et al., 1987), and finally “incipient charnockites”, a term now universally accepted. More than 50 years after its discovery, the

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phenomenon of “incipient” charnockite remains an enigma. First observations by C. S. Pichamuthu were done on vein-like alteration of granitic biotite-hornblende gneiss to an orthopyroxene-bearing rock in a quarry at Kabbal village in Karnataka State, southern India. The prograde nature of the granulite facies transition implied by the title of C. S. Pichamuthu’s 1960 paper in *Nature* remains one of the few deductions that are generally agreed upon by subsequent investigators (Ramiengar et al., 1978; Janardhan et al., 1982; Hansen et al., 1987; Stähle et al., 1987). Other aspects, including the relationship to the immediately adjacent Archean granulite facies terrain of southern India, the role of deformation control of vein emplacement, the relationship to the closely associated Closepet Granite, so evident in the Kabbal quarry (Friend, 1983), and the mechanism of dehydration, either fluid-absent melting (Thompson, 1990; Clemens, 1992; Battacharya and Sen, 2000) or migrating metasomatic fluids (Friend, 1985; Newton, 1992), all remain controversial.

Localized charnockitic alteration of the Kabbal type might be considered a mere petrologic curiosity but this form of patchy charnockitic alteration of amphibolite facies gneiss has later been found in most of the world’s high-grade terrains, including Siberia (Hopgood and Bowes, 1990), Greenland (McGregor and Friend, 1992), the Big Sur area of California (Hansen and Stuk, 1993), South Africa (Van Reenen et al., 1988), Finland (Perchuk and Gerya, 1993), Sweden (Harlov et al., 2006), Ukraine (Cinelu, 2008) and the extreme south of India and Sri Lanka (Holt and Wightman, 1983; Hansen et al., 1987). It is now evident that charnockitic alteration is a widespread phenomenon in space and time in the Earth’s crust in lithologies, either plutonic or supracrustal, that are capable of yielding orthopyroxene under metamorphic conditions of elevated temperature and pressure (Rajesh and Santosh, 2012).

McGregor and Friend (1992) gave a plausible reason why this widespread phenomenon was not reported much earlier. The dark mottling of gneiss that betrays the sporadic presence of orthopyroxene can only be observed on flat and fresh rock surfaces under conditions of inhibited weathering. These conditions are realized in active Indian and Sri Lankan rock quarries, where quarry workers utilize exfoliation surfaces to extract rock slabs, which they use for making fences or break into small equant fragments for road paving. Under conditions of tropical weathering, the distinctive charnockitic coloration vanishes in a matter of months if the quarry becomes inactive. A natural mode of preservation of the visual evidence of incipient charnockite is smooth surfaces in regions of low rate of chemical weathering, as on glacier-polished fiord-sides of southwest Greenland (McGregor and Friend, 1992).

The best-documented examples of charnockitic alteration lie in transitional granulite facies regional metamorphic terrains. This is true of the Kabbal type locality and nearby similar occurrences, which lie close to the regional orthopyroxene isograd (“Fermor Line”: Fig. 1). This feature raises what is probably the leading controversy concerning incipient charnockite: is it a harbinger of the granulite facies (i. e. a manifestation of the regional orthopyroxene isograd), a small-scale marginal reactivation of a pre-existing granulite facies terrain (Raith et al., 1989) or a misnamed rare occurrence of no regional significance (Frost and Frost, 2008)?

A second problem involves the possibility of localized partial melting. Certain textural features, especially the intersertal (orthopyroxene interstitial to plagioclase and quartz) texture seen in virtually all incipient charnockite samples (see e.g. Figs. 2B and 8B) has suggested to some authors that very localized melting may be an important mechanism of incipient charnockite formation (Burton and O’Nions, 1990; Perchuk et al., 2000). If partial melting of biotite gneiss is a significant factor, what relationship might be expected in association with major granite bodies such as the Closepet? Was partial melting provoked by an intergranular fluid phase (Friend, 1981) or could melting have taken place in the

absence of a free fluid, as advocated by the tenants of “fluid-absent melting”?

Because of their occurrence along a network of brittle fractures, most workers now regard the charnockitic veins at Kabbal, the discovery locality, and similar occurrences as marking pathways of metamorphic fluids through gneissic rocks. This idea was first put forward by Friend (1981) based on the nearly ubiquitous deformation features associated with alteration patches (Holt and Wightman, 1983; Hansen et al., 1987) which suggest access of exotic fluids following shear surfaces and dilatant structures. Further evidence of migrating fluids was provided by the discovery of pressurized CO₂ inclusions in quartz in the Kabbal charnockites and in other granulites from southern India (Hansen et al., 1984; Stähle et al., 1987; Santosh et al., 1991). More recently concentrated (Na, K)Cl brines have been identified in incipient charnockite in Sri Lanka (Perchuk et al., 2000), in massive charnockites immediately south of Kabbal in Karnataka (Srikantappa and Zarkar, 2009) and in granulites elsewhere (Touret, 1985; Crawford and Hollister, 1986). The latter type of fluid inclusion in granulites remains much less documented because of physicochemical factors unfavorable to preservation in uplift (high compressibility hence ease of disruption, high reactivity, etc.) (Touret, 2001; Touret and Huizenga, 2011). 1 Brine inclusions have not heretofore been reported in the Kabbal incipient charnockites.

A distinct type of charnockitic alteration was first identified in southernmost India by Ravindra Kumar et al. (1985). A rock quarry at Ponmudi, Kerala (Pt. P, Fig. 1 inset) exposes coarse-grained dark charnockite occupying meter-scale diffuse patches, kink-bands in foliation, and short shears, in an arrested state of replacement of light-colored biotite-garnet gneiss. Deformation control of charnockite emplacement is markedly similar to that at Kabbal, though involvement of granite intrusion is lacking at Ponmudi. The Ponmudi type of charnockitic alteration has been identified at many localities in Kerala (Ravindra Kumar and Chacko, 1986) and appears to be a characteristic feature of the Kerala Khondalite Belt (KKB, Fig. 1), a dominantly metasedimentary high-grade terrain (Chacko et al., 1987). Abundant CO₂-rich and some disrupted ultrasaline fluid inclusions in minerals have been identified in the KKB rocks (Klatt et al., 1988; Santosh et al., 1991).

An outstanding difference between the Kabbal charnockitic alteration (of hornblende-biotite orthogneiss) and that at Ponmudi (of garnet-biotite paragneiss) is the age of metamorphism: 2.51 Ga (Late Archean) at Kabbal (Friend and Nutman, 1991) and 0.62–0.48 Ga (Neoproterozoic/Eocambrian) for all of the Kerala occurrences (Ghose et al., 2004).

The present investigation is a reconnaissance of charnockitic alteration in some of the best-documented localities in southern India, searching for petrographic evidence of a discrete high-pressure metamorphic fluid, especially of the less-well-documented ultrasaline variety. Such evidence might include disrupted remnants of actual fluid inclusions, disseminated salt crystals, and grain-boundary veinlets of feldspar and myrmekite, which now appear to be common, if not ubiquitous, microtextures of granulites attributable to the action of migrating, alkali-exchanging fluids (Harlov and Wirth, 2000; Touret and Nijland, 2013). Samples selected are “duplex”: that is, they show, in a single hand-specimen, and sometimes in a single thin-section, the transition from host gneiss to charnockite.

2. The southern Karnataka-northern Tamil Nadu Archean charnockite association

2.1. Regional relations

Fig. 1 shows the generalized geology of the southern part of the Indian subcontinent and Sri Lanka, with emphasis on the

distribution of granulite facies outcrops. The northern part of the metamorphic/igneous terrain is the Dharwar Craton, which is mostly Late Archean in age, with a few smaller areas recording older ages (up to 3.4 Ga; [Beckinsale et al., 1980](#)). The dominant country rock is amphibolite facies tonalitic-trondhjemitic-granodioritic gneiss (TTG gneiss) of the sort that makes up the bulk of the Earth's crystalline shields. In the northern part of the Dharwar Craton, large, generally greenschist facies, sedimentary/volcanic terrains of Late Archean age called "greenstone belts" are discordant upon the older Peninsular Gneiss ([Chadwick et al., 2000](#)). A great linear expanse of batholithic rocks, the Closepet Granite of terminal Archean age (~2.5 Ga) divides the Dharwar Craton into Eastern and Western units. West of the Closepet Granite, the Peninsular Gneiss is dominantly about 2.9 Ga in age, whereas much of the Eastern Dharwar gneiss is 2.5–2.6 Ga ([Krogstad et al., 1991](#); [Jayananda et al., 2000](#)).

There is a distinct progression of metamorphic grade across the Peninsular Gneiss, from low amphibolite facies in the north to granulite facies in the south. The granulite facies is introduced by the appearance of orthopyroxene in quartzofeldspathic rocks along a generalized boundary known as the "Fermor Line" ([Radhakrishna et al., 2003](#)). The incipient charnockite in hornblende-biotite gneiss at the Kabbal locality lies in the

granulite facies transition zone. The progression of metamorphic grade records a paleodepth profile at the time of regional metamorphism, as was first recognized by [Pichamuthu \(1965\)](#). Geobarometric studies in the Western Dharwar Craton ([Raith et al., 1983](#)), the Closepet area ([Hansen et al., 1984a](#)) and the Eastern Dharwar Craton ([Hansen and Harlov, 2007](#)) show what seems to be an unbroken progression from about 0.4 GPa in the northern amphibolite facies to at least 0.8 GPa in the southern highest grade, dominantly "massive" charnockite.

The age of the Dharwar Craton granulite facies metamorphism is terminal Archean. In the southern Closepet Granite area ([Fig. 1](#)), zircon overgrowths yield emphatic recrystallization ages of 2.52 Ga for Closepet Granite, incipient charnockite and "massive" banded charnockite ([Friend and Nutman, 1991](#); [Mojzsis et al., 2003](#)). Importantly, unzoned zircon rims in charnockites have very low Th/U ratios, down to 0.001, much lower than if the zircons had crystallized from a melt of the present rock composition (~1.0), attesting to their metamorphic, as opposed to purely igneous nature. Whole-rock Rb–Sr isochrons of both incipient and massive charnockite in southern Karnataka are 2.5 ± 0.2 Ga ([Hansen et al., 1997](#)).

A curious distribution of Peninsular Gneiss, Closepet Granite and charnockitic gneiss occurs in the vicinity of $12^{\circ}30' \text{ N}$ ([Fig. 1](#)). Almost precisely where the mixed-facies outcrops like Kabbal are

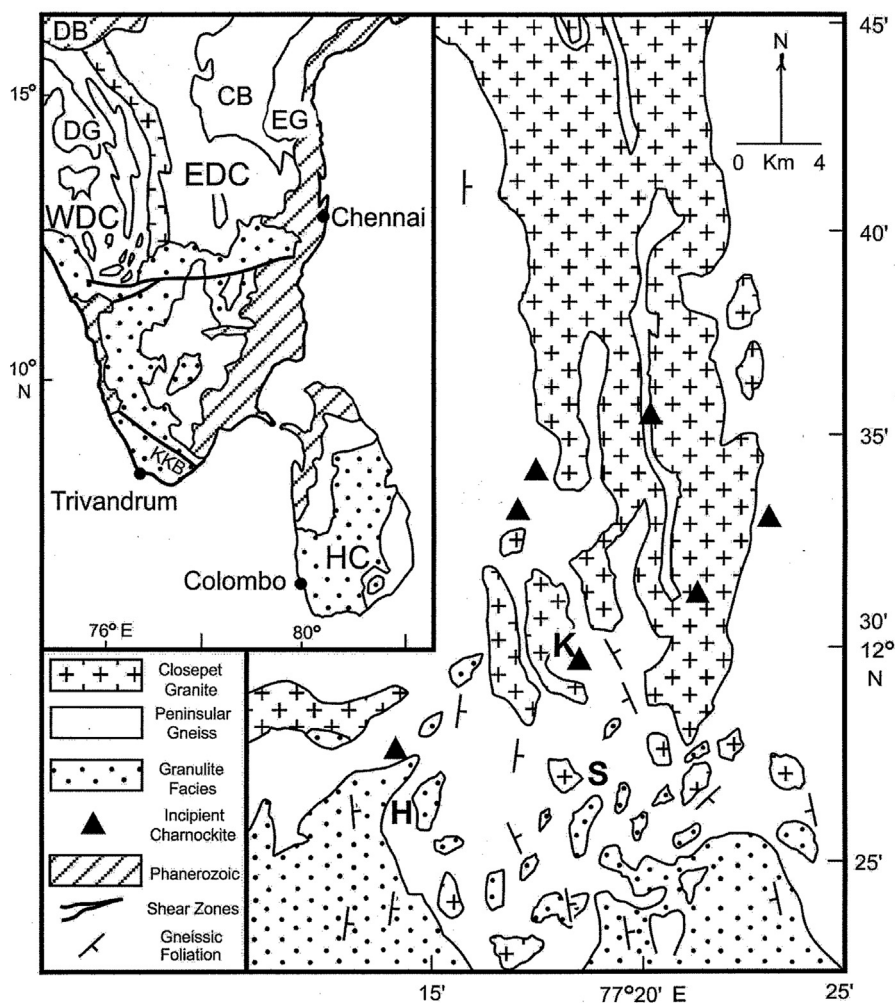


Figure 1. Generalized geology of the South India-Sri Lanka crystalline shield. The enlargement at 12° – 13°N was taken from [Suryanarayana \(1960\)](#) and [Devaraju and Sadashivaiah \(1969\)](#), with additions from [Mahabaleswar et al. \(1995\)](#). K: Kabbal, Karnataka; S: Satnur, Karnataka; H: Halaguru, Karnataka; DB: Deccan Basalts; DG: Dharwar Greenstones; CB: Cuddapah Basin; EG: Eastern Ghats; WDC: Western Dharwar Craton; EDC: Eastern Dharwar Craton; HC: Highland Complex; KKB: Kerala Khondalite Belt. Incipient charnockite locations from [Jayananda et al. \(1995\)](#). A: Arni, Tamil Nadu; P: Ponnudi, Kerala.

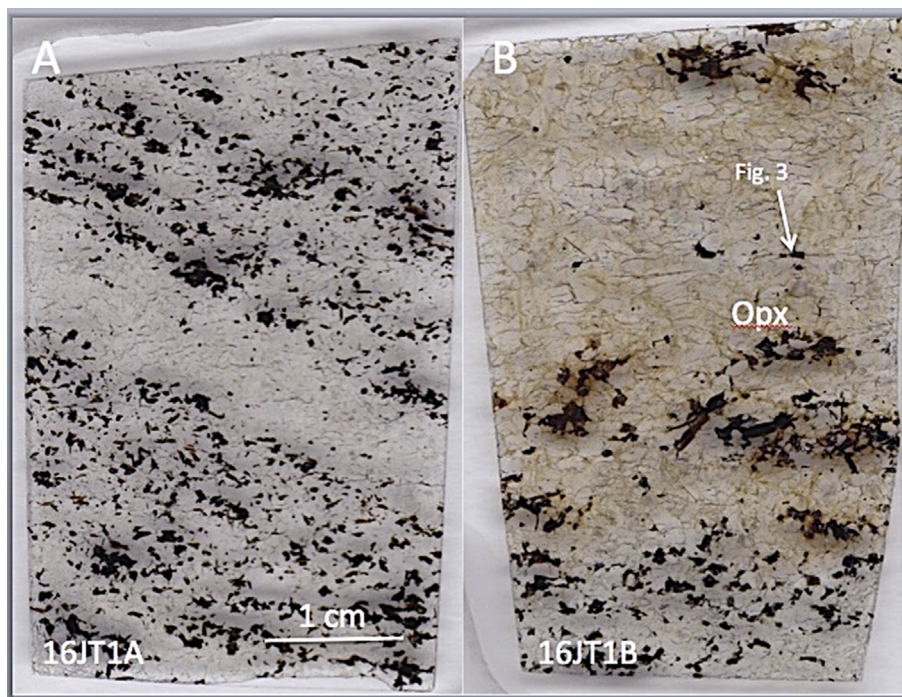


Figure 2. Duplex of double polished sections of Kabbal hornblende-biotite gneiss (A) and adjacent charnockitized equivalent (B) (same scale). B includes a small host gneiss layer in the lower part of the section. As the sections are about 2x thicker than normal thin sections (60 μm) Fe-bearing minerals are black: dominant hornblende and subordinate biotite in (A), Opx (orthopyroxene) aggregate in (B). The yellowish discoloration around orthopyroxene crystals results from myriad sub-micron ferrous chlorite veinlets and causes the dark color of charnockite in hand specimen. Arrow: Fracture-surrounded crystal shown in Fig. 3.

encountered, the Closepet Granite phases out and disappears into a terrain of charnockitic gneisses. Because of the pronounced N–S paleopressure gradient, the metamorphic grade progression must, to some extent, record a Late Archean profile of the middle to lower crust. In the critical paleodepth range, mixed-facies outcrops show enigmatic relations of hornblende-biotite gneiss, granite and incipient charnockite (Friend, 1981).

2.2. Kabbal, Karnataka

2.2.1. Microtextures

Fig. 2A shows sections of the sample 3-1A,B of Janardhan et al. (1982) and Newton and Tsunogae (2014). The enlargement is about twice actual size. Duplex samples of broad (2.5 cm \times 3.5 cm) doubly polished thick sections (80–100 μm) were made of either

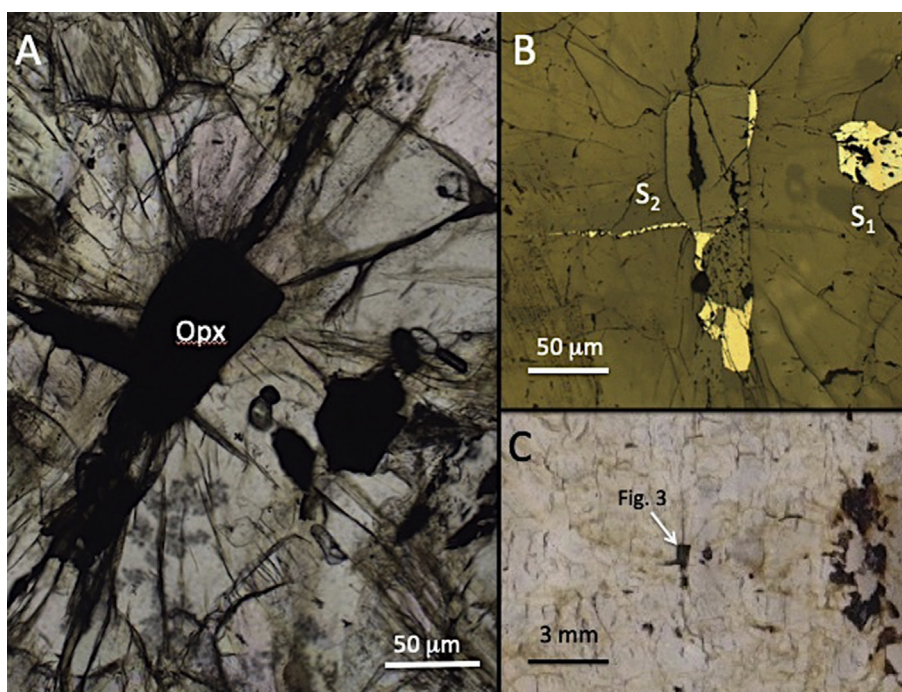


Figure 3. Kabbal charnockite, Sample 16JT1B, cf. Fig. 2B. (A) Subidiomorphic orthopyroxene crystal, surrounded by a radial set of microfractures. (B) The same in reflected light. S_1 = sulfide (pyrite), S_2 = pyrrhotite. (C) Crystal shown in A and neighboring intersertal orthopyroxene aggregate (cf. Fig. 2B).

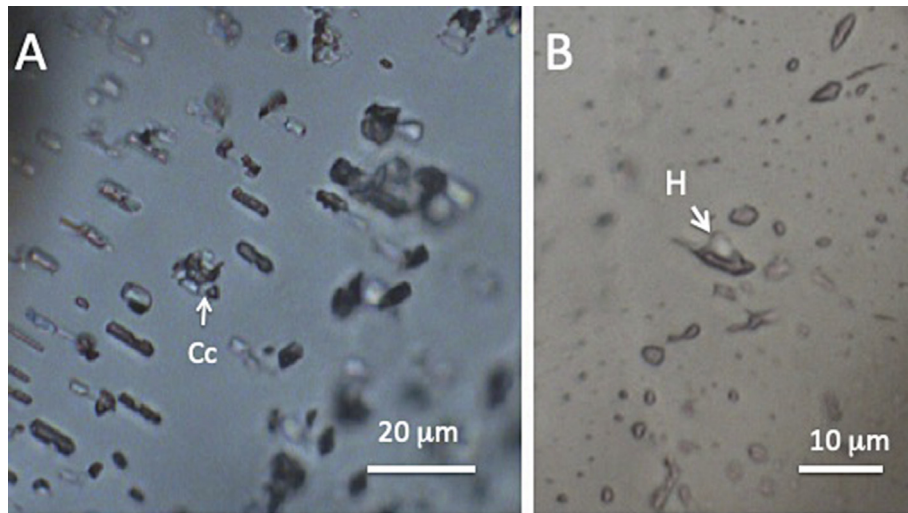


Figure 4. Types of fluid remnants in Kabbal charnockite (sample 16JT1B, cf. Fig. 2B). (A) Trail of supercritical tubular CO₂ inclusions (dark gray), some of them containing carbonates (Cc). Slightly out of focus, carbonates progressively replace CO₂ inclusions at the right of the section. (B) Cluster of irregular gaseous inclusions, presumably low-density H₂O, the largest (with traces of necking-down) containing an isotropic halite cube (H). Larger inclusions are surrounded by tiny inclusions, derived from transposition (decrepitation) of the former larger inclusions.

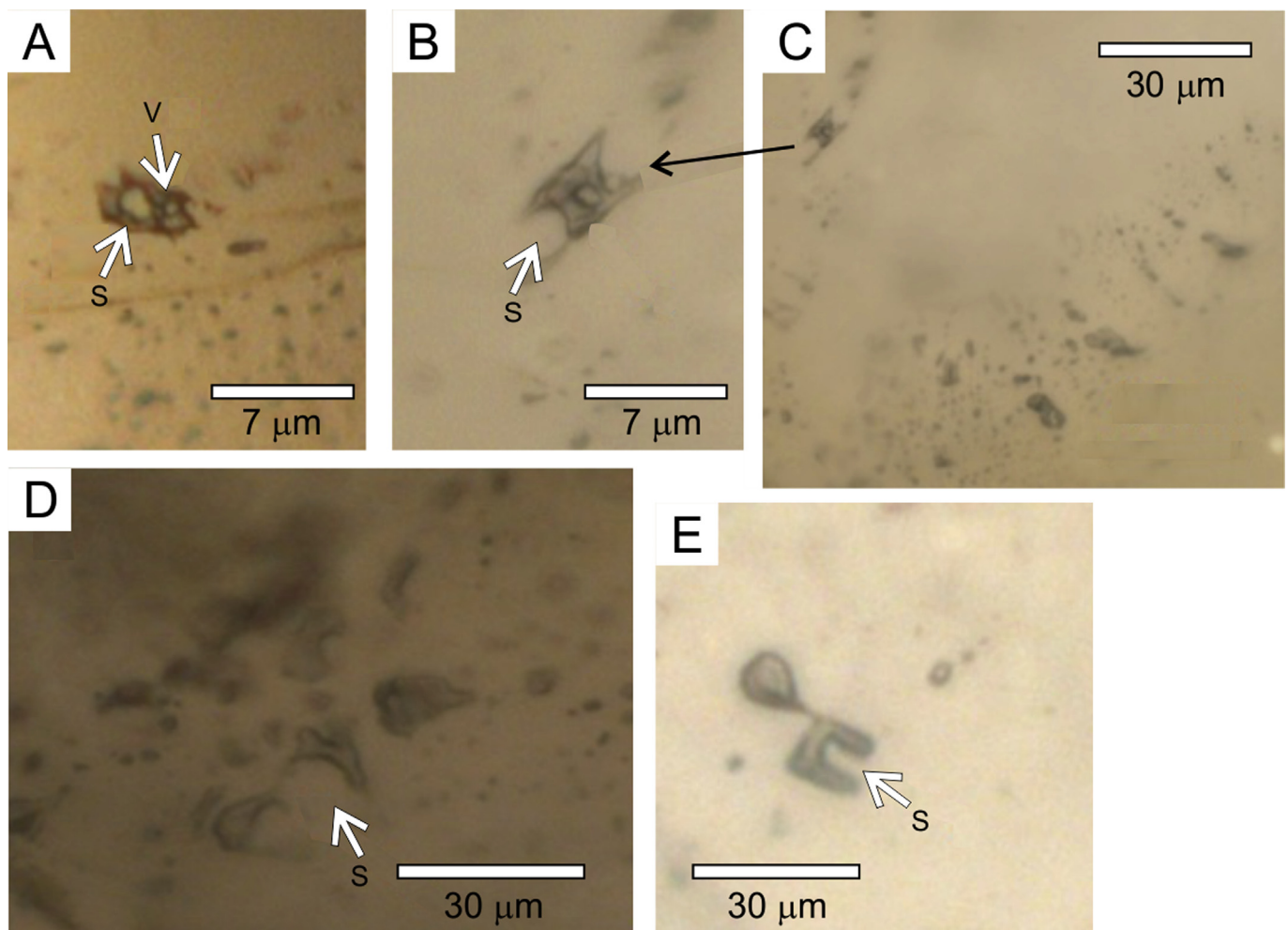


Figure 5. Primary brine inclusions in Kabbal charnockite (sample 16JT1B). (A and C) Isolated, primary brine inclusions composed of a small irregular cavity squeezed around one or several crystals, most of them isotropic (S). V = Vapor bubble (Collapsed inclusions; Touret, 2001). (B) Detail of the primary inclusion shown in C. Below this inclusion, trail of secondary inclusions, all elongated in the same direction and surrounded by micro-inclusions, also aligned along this direction (Stress-induced transposition; Roedder, 1984). (D) Cluster of brine inclusions. The arcuate shape (S) is attributed to evolution by underpressing or implosion (Vityk and Bodnar, 1995). (E) Isolated brine inclusion showing a “necking down” structure (Roedder, 1984). Arrow (S): NaCl crystal without visible boundary against the quartz host, diagnostic for NaCl because of nearly identical refringence.

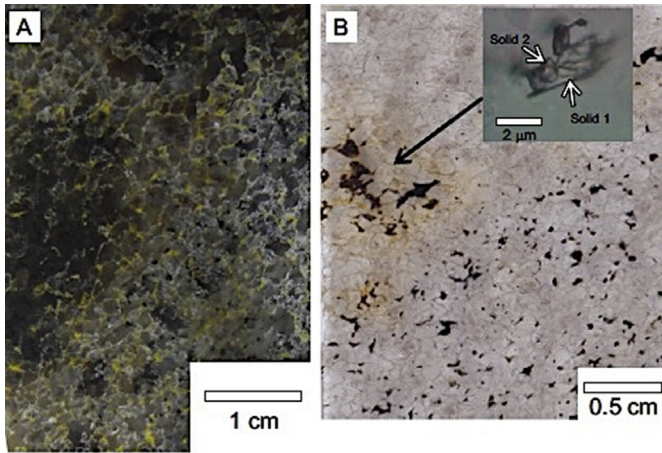


Figure 6. Arni rock (sample T-6-30 of Hansen et al., 1987). (A) Polished slab of the hand specimen with stained K-feldspar. Charnockite = dark ovoids in the left-hand side of the slab. Note network of yellow K-feldspar microveins along intergrain boundaries. (B) Detail of the contact of charnockite (upper left) and gneiss (lower right) in thin section. The orange-yellow aureole around orthopyroxene is the usual chloritic alteration responsible for the dark color of charnockite Howie (1967). Inset: Collapsed brine inclusion in quartz within the orthopyroxene aggregate: solid 1 = NaCl, solid 2 is slightly more colored and refringent mineral, possibly Fe-bearing chloride.

gneiss-charnockite “close-pairs” (retrieved < 20 cm apart in the field) or with the transition zone visible in a single section. Fig. 2A is the host rock, well-banded hornblende-biotite granitic gneiss. Sample 2B (Fig. 2B) is the immediately adjacent charnockitic portion showing typical features: (1) coarse orthopyroxene crystals often concentrated at the margins of a charnockitic patch. They have marked interstitial habit, already mentioned by Hansen et al. (1987); (2) increased crystal sizes of quartz and feldspar. Increase of

modal K-feldspar is evident in this sample; (3) variable retrogression of orthopyroxene to secondary biotite and turgid mixtures of chlorite and oxide minerals; (4) brownish haloes around orthopyroxene formed by myriad sub-micron veinlets in quartz and plagioclase crystals of a yellowish Fe-chlorite. The chlorite microveins impart the characteristic dark color of charnockite to hand specimens and relatively unweathered outcrops (Howie, 1967). In addition, occasional grain-boundary linings of pure K-feldspar and quartz-albite intergrowths (myrmekite) were observed in the charnockitic portion of Fig. 2B.

Fig. 3A shows a relatively common additional feature. Quartz and feldspar crystals surrounding an altered subidiomorphic orthopyroxene show intense radial microfracturing. Fig. 3B shows the same orthopyroxene grain and environs in reflected light, revealing sulfide minerals as part of the alteration. Fig. 3C shows the location of the cracked crystal with respect to Fig. 2B. The fracture pattern near the orthopyroxene suggests that the microfracturing may be due to volume increase of the orthopyroxene in alteration or, equally possibly, that hydraulic fracturing by fluid overpressuring occurred near the end of orthopyroxene crystallization, as observed during the formation of graphite veins in Sri Lanka (Touret et al., 2018).

2.2.2. Fluid inclusions

Preparation of the Kabbal sample for fluid inclusion study followed standard procedures used for all of the samples of this study, on the sections used for looking at microtextures. Sections are approximately 3 times thicker than standard petrographical thin sections. It explains the color change observed for some minerals under the microscope, e.g. dark green or black for orthopyroxene. The emphasis was on reconnaissance search for evidence of disrupted remnants of former ultrasaline fluid inclusions.

The Kabbal charnockite contains recognizable fluid inclusions of at least two types (Fig. 4), in contrast to the correlative Kabbal gneiss, which is almost entirely lacking in fluid inclusions. The

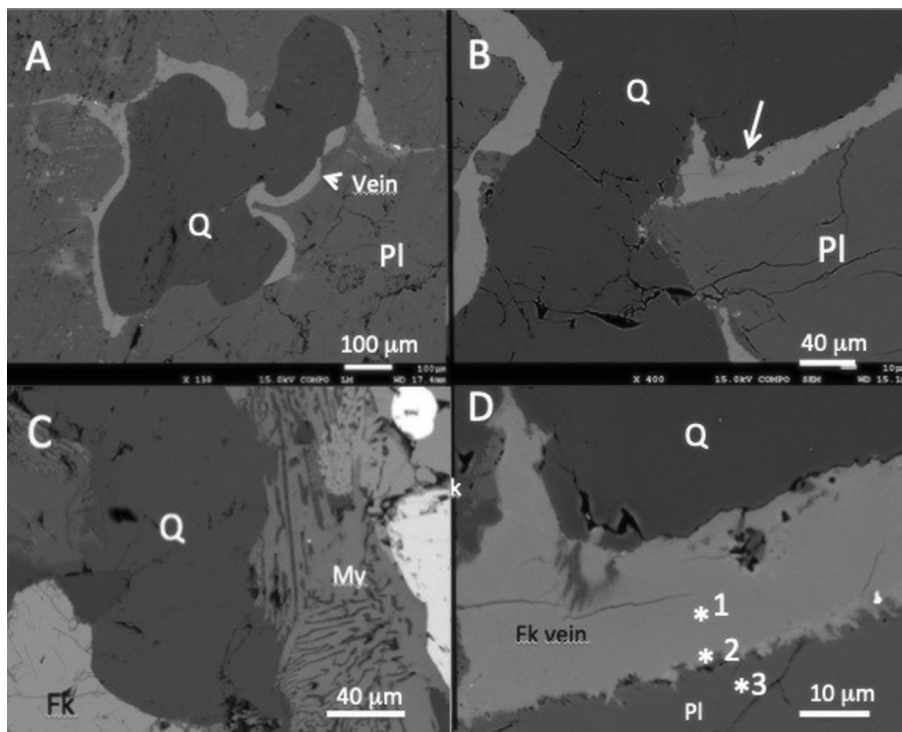


Figure 7. SEM (Jeol JSM-7600F) of grain boundary veins in Arni charnockite. (A and B) Nearly continuous microvein (white arrow, light gray) along quartz-feldspar (Pl = plagioclase) grain boundary. (C) Quartz-feldspar intergrowth (My = myrmekite) at the contact between large quartz and K-feldspar crystals. (D) Detail of the K-feldspar vein shown in B. 1,2,3: Position of the spot analyses (see text).

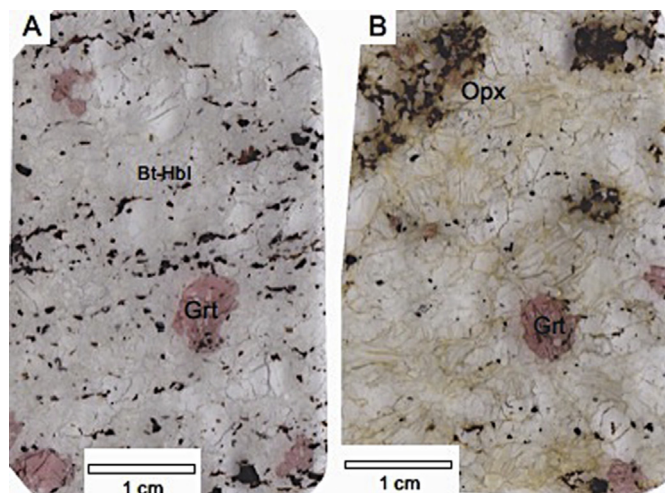


Figure 8. Duplex polished sections of the Ponnudi charnockite sample 16JT4. (A) Garnet-biotite gneiss. Gneiss foliation is marked by biotite (dominant) and some hornblende. (B) Charnockite. Progressive replacement of garnet (Grt) by orthopyroxene (Opx): isolated garnet crystal in the center of the section (cf. A), smaller crystal surrounded by Opx (black) above, Opx aggregates (interstitial texture) including small garnet remnants on top of the section.

majority of the fluids enclosed in quartz are small dark CO₂ bubbles of low density, as shown by the high homogenization temperatures, in the range 25–30 °C as also reported by Newton and Tsunogae (2014). These occur most commonly in trails of secondary inclusions, individuals nearly identical in orientation and shape, often elongated or tube-like, occurring along healed cracks (Fig. 4). A microtexture not reported previously, is that the CO₂ trails often grade into more diffuse zones containing many small carbonate crystals. In the first author's experience, dispersed minute carbonate crystals are very common in all granulites. They can possibly be interpreted by a late reaction between CO₂ remnants and more mobile brine fluids at low temperature.

Hansen et al. (1984b) and Stähle et al. (1987) found rare large primary CO₂ inclusions in the Kabbal charnockite quartz with homogenization temperatures down to –25 °C (density ≈ 1.1 gm/cm³). CO₂ isochores indicate a pressure of about 0.6 GPa for = 700 °C, corresponding to P–T conditions of incipient charnockite formation (Hansen et al., 1987).

We found in our sample a number of brine inclusion remnants. Brine inclusions have not previously been reported in the Kabbal incipient charnockite. The remnants consist of clusters of very small aqueous inclusions frequently containing a halite cube and occasionally a small gas bubble (Fig. 4B). The clusters are surrounded by swarms of tiny inclusions too small to identify with certainty. This texture has been replicated experimentally by Vityk and Bodnar (1995) and is attributable to decrepitation and transposition of high-pressure brine inclusions in unloading. The disrupted state of the brine inclusions illustrate why this type of fluid has been mostly overlooked in previous studies of granulites.

The secondary inclusions described above were relatively easy to identify in this sample. We were able to find a few remnants of primary brine inclusions in quartz, shown in Fig. 5. They are small, a few microns in diameter, isolated or in clusters of a few individuals, surrounded, like the secondary inclusions, by a “cloud” of tiny cavities. The contours of primary brine inclusion remnants are very irregular, squeezed around an aggregate of solids, most of them isotropic (Fig. 5A and B). This type of disruption (transposition) leading to collapsed inclusions (Touret, 2001) (also called “implosion-decrepitation by Van den Kerkhof and Hein (2001)) is very common in high-grade metamorphic rocks which have followed

some nearly isobaric cooling path after peak metamorphism. The disruption results from the steep dP/dT isochores of aqueous fluids compared to CO₂. Another indication that inclusion disruption occurred through implosion is given by arcuate inclusion shapes, consistent with some textures produced experimentally (Vityk and Bodnar, 1995) (Fig. 5D). Other inclusions (Fig. 5E) show spectacular “necking down” textures (Roedder, 1984), indicating that the disruption has been progressive, occurring over a relatively wide temperature range. It is impossible to determine microscopically the original salinity of the primary brine fluids except to state that they were very concentrated, halite-saturated at peak metamorphic temperature (if they were halite saturated at peak metamorphic temperature, say 700 °C, they would correspond to a salinity of at least 55 wt.% NaCl, data from Sourirajan and Kennedy (1962)).

2.3. Arni, Tamil Nadu

Hansen et al. (1987) found a second type of charnockitic alteration in the northern margin of the Archean high grade terrain in northern Tamil Nadu (Point A, Fig. 1 inset). This occurrence is in biotite gneisses, similar to those at Kabbal but lacking in hornblende. The incipient charnockite introduces the granulite facies in a region where there appears to be an unbroken north-to-south depth-zone succession of amphibolite facies quartzofeldspathic gneisses to high-grade garnetiferous charnockite (Hansen and Harlov, 2007). Scattered orthopyroxene in the biotite gneiss exposed in the quarry at Arni, Tamil Nadu, is revealed by dark ovoids, 1–3 cm across in isolated spots, in contrast to the more pervasive vein-like alteration at Kabbal. The deformation control of the distribution of ovoids is subtle but definite in their alignment, marking a secondary foliation at an acute angle to the earlier fabric (Hansen et al., 1987).

2.3.1. Microtextures

Fig. 6A, is a polished slab of Arni rock T-6-30 of Hansen et al. (1987) stained with sodium cobaltinitrite, which reagent colors exposed K-feldspar bright yellow. The preparatory etching with HF fumes leaves plagioclase with bleached surfaces. The unaltered host gneiss shows scattered blebs of interstitial K-feldspar. This contrasts with a trace-work of K-feldspar enveloping large recrystallized quartz and plagioclase grains in the charnockitic spots. As in the Kabbal charnockite, the orthopyroxene is interstitial relative to quartz and feldspar; the light-colored minerals form a moat around the pyroxene (Fig. 6B). Quartz and plagioclase are laced with innumerable tiny yellowish-orange veinlets (not to be confused with the yellow-stained K-feldspar grain boundary veins of Fig. 6A). Orthopyroxene in the charnockitic part of the rock (Fig. 6B) shows the typical granulite intersertal texture, subidiomorphic orthopyroxene crystals crystallizing along quartz and feldspar intergrain boundaries.

Rare CO₂ inclusions with densities too low to be primary fluids trapped during high-grade metamorphism were found in previous work on this locality (Hansen et al., 1984b), but none were observed in the present study. We found a few relatively large, irregular primary brine inclusions in coarsely recrystallized quartz between intersertal orthopyroxene (Fig. 6B, inset). Some brine inclusions contain only one isotropic halite crystal and an empty space, while others, like the one shown in Fig. 6B, have an aggregate of irregular isotropic minerals, some of them transparent-white (halite), others slightly colored (greenish), possibly Fe-bearing halides. These objects are undoubtedly the remnants of collapsed brine inclusions. They constitute an important link with the incipient charnockite at Kabbal.

The grain-boundary veins are starkly revealed in SEM images (Fig. 7A–D). They are identical in aspect to those now known to be present in many or most high-grade charnockitic rocks (Coolen,

1980; Perchuk and Gerya, 1993; Hansen et al., 1995; Harlov et al., 2006). SEM-EDS analyses were done on a microvein located along the boundary between quartz (Qtz) and plagioclase (Pl), showing the microvein to be nearly pure K-feldspar, with some increase of the albite content near the contact with plagioclase (Fig. 7D): (1) (Center of the microvein) $\text{Si}_{3.04}\text{Al}_{0.96}\text{O}_8\text{K}_{0.96}\text{Na}_{0.04}$. (2) (Close to the plagioclase boundary): $\text{Si}_{2.99}\text{O}_8\text{Al}_{1.03}\text{K}_{0.87}\text{Na}_{0.13}$. (3) (Plagioclase): $\text{Si}_{2.79}\text{O}_8\text{Al}_{1.20}\text{Na}_{0.69}\text{Ca}_{0.26}\text{K}_{0.01}$ (Analyses done at GeoRessources, Nancy, Andreï Lecomte, analyst). K-feldspar grain-boundary veins are invariably accompanied by diffuse K-feldspar alteration of the interiors of the plagioclase (“replacement anti-perthite”) common in high-grade gneiss (Griffin, 1969).

3. Southern Neoproterozoic/Eocambrian province

3.1. Regional relations

The Archean portion of the Southern Indian Shield is terminated by a series of prominent E–W shear zones (Fig. 1). A zone of igneous and metamorphic rocks of mixed Proterozoic ages, some demonstrating retrogression after granulites, is Late Proterozoic to Eocambrian (Ghose et al., 2004). Biotite-garnet paragneiss, some graphite-bearing, is a prominent country rock in the southwest tip of India. Garnet-biotite-sillimanite gneiss (khondalite) and calc-silicate gneiss are widespread also in the Kerala Khondalite Belt.

Incipient charnockite is a characteristic feature of the Late Precambrian metamorphism of southern India and the Highland Complex of central Sri Lanka (Fig. 1). This mixed facies rock type has been described many times as exposed in quarries (Holt and Wightman, 1983; Ravindra Kumar et al., 1985; Ravindra Kumar and Chacko, 1986; Hansen et al., 1987; Milisenda et al., 1991; Raith and Srikantappa, 1993; Perchuk et al., 2000; Endo et al., 2012, 2013). Despite the fact that incipient charnockite in southernmost India and Sri Lanka is nearly 2 Ga younger than the southern Karnataka/

northern Tamil Nadu variety, and that it affects garnet-biotite paragneiss as well as hornblende-biotite orthogneiss, the distinctive darkly mottled outcrop appearance is very similar in both types. The associated characteristic shears and warping of foliation in orthopyroxene-bearing patches are again ubiquitous.

3.2. Ponmudi, Kerala

3.2.1. Microtextures

This sample (Fig. 8) is equivalent to K-18-6A of Hansen et al. (1987). Modal analyses show that the charnockitic alteration is essentially the biotite-quartz dehydration reaction to orthopyroxene plus K-feldspar (Newton and Tsunogae, 2014). There was a substantial gain of modal K-feldspar, requiring that some K_2O was introduced in the open-system reaction, since the amount of biotite destroyed was only a few percent. Garnet involvement in the reaction was small but definite, according to mass balance calculations. This conclusion is supported by the distribution of orthopyroxene in the charnockitic portion, conspicuously clustered around garnet crystals (Fig. 9). Large orthopyroxene crystals contain garnet remnants. Thin quartz-K-feldspar veinlets cross-cut some grain-boundaries. Fe-Ti oxide minerals are always present in garnet-orthopyroxene clusters—these are readily explainable by liberation of Ti from biotite and the different Mg/Fe ratios of orthopyroxene and garnet. Other features in common with the Kabbal-type charnockite are microfracturing of large recrystallized quartz or feldspar and the ubiquitous halo of ultrathin planar greenish veins permeating new quartz and feldspar around orthopyroxene clusters.

3.2.2. Fluid inclusions

As a whole, fluid inclusions in Ponmudi samples, especially CO_2 -bearing, are significantly more abundant than in those from Karnataka, but the main types (CO_2 and disrupted brines) are the same. Primary CO_2 inclusions were observed in plagioclase and quartz

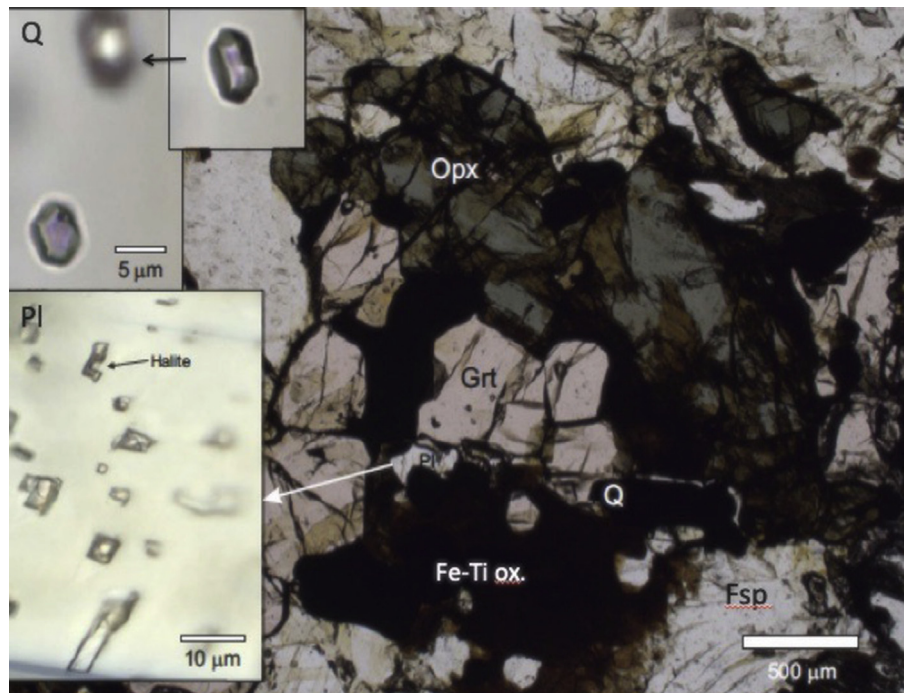


Figure 9. Replacement of garnet (Grt) by orthopyroxene (Opx) in Ponmudi Sample 16JT6 (Fig. 8B). Near the contact with garnet are large crystals of Fe-Ti oxide, due to the much higher Fe-content of garnet relative to orthopyroxene. Other minerals involved in the replacement are plagioclase (Pl) and quartz (Q), the latter mineral grading in a microvein intersecting Fe-Ti oxide and Opx. In the lower right corner of the section, a feldspar phenocryst (Fsp) shows curved microfractures converging towards Opx = hydraulic fracturing, cf. Fig. 3A. Inset (Left) Pl: Primary low-density CO_2 inclusions in plagioclase. A halite cube in one inclusion suggests immiscibility between CO_2 and NaCl-saturated aqueous solutions at the time of trapping. Q: Primary high-density CO_2 inclusions in quartz, negative crystal shape (sample 16JT4).

within the replacement reaction zone between garnet and orthopyroxene, clearly visible in double-polished thin section. In feldspar, these characteristically inhabit clusters of square cavities (Fig. 9, inset). A few inclusions contain a small isotropic cube (H in Fig. 9), suggesting a possible immiscibility between CO₂ and halide-bearing brine at the time of the inclusions' formation. CO₂ homogenization temperatures are close to critical (between +26 and +30 °C) with homogenization into the liquid. It indicates a relatively low CO₂ density, much too low to correspond to peak metamorphic conditions despite the primary character of the inclusions. But this is a typical pattern for CO₂ inclusions in feldspar because of its good cleavage and consequent weakness. Alternatively, few conspicuously isolated CO₂ inclusions were found in quartz, with spectacular negative crystal shapes (Fig. 9, inset). It was not possible to make microthermometry measurements in the present study, but these inclusions are strikingly similar to inclusions found in Tanzania high-pressure granulites, either Furua (Coolen, 1980) or Ulaguru Mountains (Herms and Schenk, 1988). It can safely be assumed that these inclusions contain the high-density CO₂ fluid found in the KBB rocks by Klatt et al. (1988), or Santosh et al. (1991) (T_H below -20 °C, compatible with the crystallization conditions of the charnockite at ~0.6 GPa and 700 °C).

It is interesting to note that these wholly (quartz) or partly (feldspar) preserved CO₂ inclusions have only been found so far in reacting minerals enclosed within garnet and orthopyroxene, which have protected them despite the relative weakness of the feldspar mineral. This zone is also remarkably fresh, devoid of microfractures, which are very abundant in the rest of the rock, showing often a curved pattern similar to that found in Karnataka (see above). Quartz in the groundmass contains a number of empty cavities (decrepitated inclusions), as well as very small brine inclusions, too small to be characterized precisely but usually containing very small NaCl cubes. Even smaller (less than one micron) trails of inclusions are parallel to or in continuity with microfractures. As in the Northern Province (and in many other granulites elsewhere, see e.g. Touret et al., 2018), it can then be assumed that these microfractures are created by the decrepitation of inclusions during uplift. It may be significant that all of the fluid types found in Ponnudi have their counterparts in the sample from Kabbal, where charnockite formation operated on a different lithology (orthogneiss versus paragneiss) and in a much older event (Late Archean as opposed to Neoproterozoic).

3.3. Kottavattam, Kerala

One of the most classical occurrences of incipient charnockite is exposed in a large quarry in the Kerala Khondalite Belt (Raith and Srikantappa, 1993). A striking nearly rectilinear array of charnockite lenses gives the impression of brittle cross-fracturing with dark charnockite occupying the fractures. Santosh et al. (1991) and Raith and Srikantappa (1993) found abundant medium-density CO₂ inclusions in quartz, typical of other similar occurrences in the Kerala Khondalite Belt. Raith and Srikantappa (1993) interpreted the fracture system as resulting from fluid over-pressure, with large-scale rock bursting as a consequence (Srikantappa et al., 1985).

We examined a large polished slab from the Kottavattam quarry supplied by M. Raith. We did not examine the fluid inclusions in this specimen, but noted a couple of features supporting the idea of fluid overpressure. One unmistakable evidence of a geode, or open fluid cavity in which orthopyroxene could crystallize, is shown in Fig. 10, right-hand side. Details of such a geode are shown in Fig. 11. The concentric array of coarse quartz and feldspar crystals, as well as a single quartz crystal much larger than the others, are typical features of geodal cavities in Alpine veins. The width of the large quartz crystal decreases progressively towards the top, recalling the "Tessiner

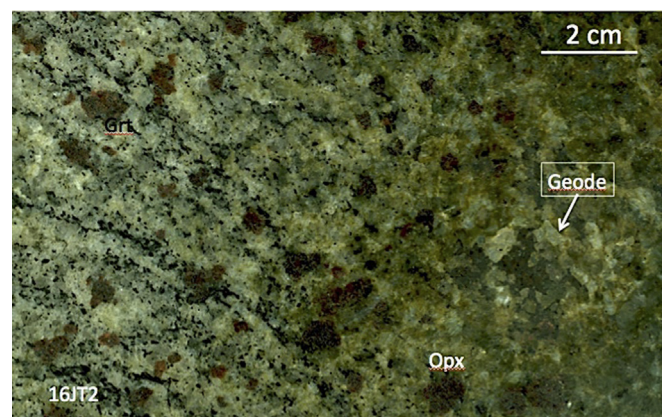


Figure 10. Kottavattam: large polished rock slab (about 10 cm × 20 cm) of the contact between garnet-bearing paragneiss (left) and charnockite (right), sample 16JT2. Grt = garnet, Opx = orthopyroxene aggregates, including garnet, feldspar and quartz crystals (cf. Fig. 9). Note the darker color and the sudden disappearance of the gneiss foliation in the charnockite. A former geode (open cavity mantled by idiomorphic quartz crystals) can be seen on the right-hand side of the slab (cf. Fig. 11).

habitus" (Rykart, 1995) which, in the Alps, is typical for quartz grown in the presence of CO₂-rich fluids (Mullis et al., 1994). Evidence that some of the fluids could also have been concentrated brines is the abundance of myrmekite (intergrown sodic plagioclase and quartz) in K-feldspar microveins forming linings between large feldspar and quartz crystals, as revealed by Na cobaltinitrite staining (Fig. 12).

4. Petrologic interpretations

4.1. Importance of ultrasaline fluids

A principal finding of the present work is ultrasaline inclusions in recrystallized quartz of incipient charnockites of southern India. This kind of fluid inclusion is well-documented in the study of charnockitic veins in Sri Lanka by Perchuk et al. (2000) but had not previously been identified at either the Kabbal or Ponnudi incipient charnockite type localities. Migrating brines exchanging alkalis and

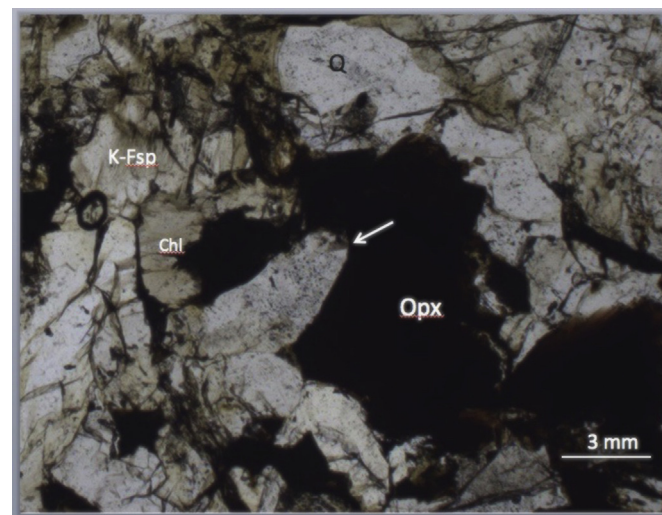


Figure 11. Detail in polished thin section of a geode within Kottavattam sample 16JT2, smaller but very similar to that shown in Fig. 10. Q = quartz, K-sp = microperthite, Chl = chlorite, Opx = altered orthopyroxene. Arrow: larger quartz crystal showing the typical "Tessiner habitus". Black dots in quartz = fluid inclusions, most of them decrepitated, some still containing traces of CO₂.

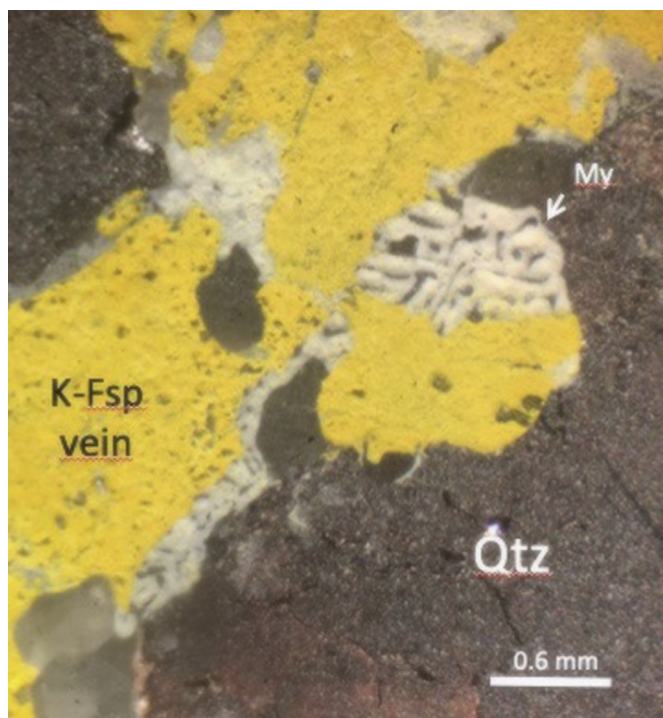


Figure 12. Binocular microscope view on polished surface of Kottavattam charnockite sample. Quartz-plagioclase intergrowth (myrmekite = My) develops at the contact between K-feldspar microvein (stained bright yellow by cobaltinitrite) and quartz (Qtz). The large quartz crystal (speckled) is etched with HF fumes.

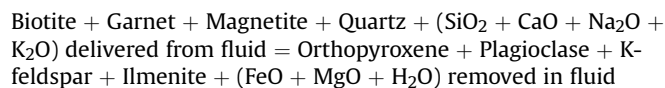
alkaline earth elements with wall rocks provides an appealing explanation for the open-system alteration documented at both localities by detailed chemical studies (Hansen et al., 1987; Ståhle et al., 1987; Newton and Tsunogae, 2014). Especially prominent is the K-metasomatism involved in charnockite formation, as well as K-feldspar and myrmekite grain-boundary veinlets. These textures implicate the passage of base-exchanging fluids, most probably concentrated chloride solutions. Coeval CO₂ fluid inclusions at both localities could have been immiscible with the saline fluids at the probable metamorphic conditions of about 0.6 GPa and 700 °C (Johnson, 1991; Gibert et al., 1998). The CO₂ would have been a more inert fluid, less capable of alkali metasomatism. At the difference of saline fluids, it is also unevenly distributed among the different samples, very scarce or lacking at Kabbal. Its greater amount in Ponmudi is an indication of local derivation by reaction of organic matter contained in the sedimentary protolith with H₂O liberated during prograde metamorphism. Isotope data indicate however that, like for other granulites, some contribution from the mantle cannot be excluded (Santosh et al., 1991).

Identification of brine remnants in the Kabbal incipient charnockite demonstrates a genetic link with the closely associated Closepet Granite, also containing disseminated halite crystals and brine remnants (Srikantappa and Malathi, 2008) and the massive charnockites (Srikantappa and Zarkar, 2009).

Despite the disparity of ages (2.5 Ga vs. 0.5 Ga) and in the lithologies affected (hornblende-biotite orthogneiss at Kabbal versus garnet-biotite paragneiss at Ponmudi), the outcrop structures, microtextures and fluid inclusion types are similar for the two occurrences, suggesting a commonality of causal factors. Hansen et al. (1987) and Newton and Tsunogae (2014) presented chemographic evidence that the major difference between Kabbal and Ponmudi was somewhat higher temperature and/or lower H₂O activity during the latter metamorphism.

Some workers (Raith et al., 1989) have resisted the idea that the incipient charnockite occurrences along the Fermor Line in southern Karnataka and northern Tamil Nadu represent the first regional appearance of orthopyroxene (i. e. the Late Archean orthopyroxene isograd), in spite of the conspicuous association of incipient charnockite localities with the granulite facies transition zone (Fig. 1). The present finding of remnants of concentrated chloride brine inclusions in Kabbal and Arni charnockites, as well as grain-boundary feldspar (-quartz) microveins in both rocks, and in massive charnockites farther south, further support a more direct link between incipient charnockites and massive charnockites, which types may differ more in degree than in kind.

Brines remnants in all types of incipient charnockites are strong indications that these fluids played a major role during charnockitisation. Rock analyses of “close pairs” (alteration veins and immediately unaltered host gneiss) from Ponmudi (Hansen et al., 1987; Newton and Tsunogae, 2014) show open system alteration with orthopyroxene formation, according to the reaction, determined from mass balance calculations:



Except for SiO₂, the solubility of all involved elements is high in concentrated chlorine-bearing aqueous solutions, increasing linearly with NaCl equivalent-molality (Newton and Manning, 2002). While quartz solubility in the system SiO₂–H₂O–NaCl–CO₂ is maximal at lower crustal pressures and temperatures if the solvent is pure H₂O, the decline in quartz solubility with NaCl content (salting-out effect) is less severe than in CO₂–H₂O fluids at comparable H₂O activities (Newton and Manning, 2010). Moving easily through the rocks along intergrain boundaries, brines are the best fluids which can promote the metasomatic transformation leading to the formation of incipient charnockite. The origin of these fluids is still a mystery. High salinity brines are very common in metamorphosed pelites, inherited from saline pore waters or dissolved evaporites, e.g. in the Bamble province of Southern Norway (Touret and Nijland, 2013). But the fact that they occur also in Karnataka orthogneiss calls for a different origin. It has been shown that UHT-granulites, found at a distance of few kilometers of most incipient granulites occurrences in Southern India and Sri-Lanka (Kelsey and Hand, 2015), are massive fluid reservoirs at peak metamorphic conditions (Omori and Santosh, 2008). Such a distance is not an obstacle for the very mobile brine solutions, as further indicated by their regional influence during the last stage of the retrogression (Touret and Nijland, 2013). In short, brine fluids issued from neighboring HT- and especially UHT-granulites are the best candidates for promoting the formation of incipient charnockites.

4.2. Temperature-pressure-H₂O activity regimes

Most authors agree that both the Kabbal-type and the Ponmudi-type charnockitization pressure regimes were in the range of 0.5–0.6 GPa, corresponding to a paleo-depth range of 15–18 km (Hansen et al., 1984a; Raith and Srikantappa, 1993; Perchuk et al., 2000). Although most workers put associated temperatures near 700 °C, considerable uncertainty exists for mineralogical thermometry of parageneses lacking garnet, which mineral anchors the most successful thermometers (garnet-biotite: Ferry and Spear, 1978; garnet-orthopyroxene: Lee and Ganguly, 1988; Aranovich and Berman, 1997).

Two thermometric calibrations applicable to the alteration at Kabbal and Arni are the Na distribution between microcline and plagioclase (Whitney and Stormer, 1977), which may be applicable

to feldspar grain-boundary veins, and the orthopyroxene-biotite Fe-Mg distribution thermometer of Sengupta et al. (1990). Applying the former thermometer to our analyses of a K-feldspar vein in Arni T-6-30 (Fig. 7) and immediately adjacent plagioclase, with input of $\text{NaAlSi}_3\text{O}_8$ activity 0.63 for plagioclase Ab_{69} from Newton et al. (1980) gives 610 °C at 0.55 GPa. For Kabbal charnockite 3-1 (Hansen et al., 1987), the Sengupta et al. (1990) formulation gives 690 °C, and for Arni T-6-30, 663 °C. These estimates are comparable to the 690 °C yielded by the Ferry and Spear (1978) thermometer for Pomudi K-18-6. The preliminary indication of the thermometry is that the K-feldspar grain-boundary veins are an integral part of the orthopyroxene-forming reaction. Charnockite formation took place in such a low-temperature range that, considering the low H_2O activity in equilibrium with orthopyroxene and K-feldspar at 0.55 GPa and 700 °C (<0.5: Aranovich and Newton, 1998; Newton et al., 2014), partial melting would not have taken place as part of charnockite formation. Rather, the indication is that orthopyroxene formation was a late, low-temperature subsolidus event, following upon granite emplacement in the case of Kabbal. The somewhat higher temperature (~750 °C) and H_2O activity (>0.6) found for the Kabbal-type charnockite at Udadigana, Sri Lanka (Perchuk et al., 2000) would be feasible for the beginning of melting, according to the brine-granite melting experiments of Aranovich et al. (2013). The mutually exclusive relationship between massive charnockite and Closepet Granite in southern Karnataka (Fig. 1) could represent an arrested critical horizon of increasing H_2O activity in solutions moving upwards in the Late Archean crust, as envisioned by Friend (1983).

The salinity of an aqueous (K, Na)Cl solution can be characterized by the apparent H_2O activity, based on orthopyroxene-K-feldspar coexistence (Aranovich and Newton, 1997, 1998). For the H_2O activity of this assemblage including also hornblende, estimated to be 0.5–0.6 at conditions of incipient charnockite formation (Perchuk et al., 2000), the salinity of a pore-fluid brine would be 0.71–0.78 wt.%, or 46–60 wt.% salt components. For the assemblage lacking hornblende, the H_2O activity would be 0.4–0.5, and the salinity would be nearly 70 wt.% salt. This concentration would be near chloride saturation at 700 °C and 0.55 GPa (Aranovich and Newton, 1997). These estimates indicate that the H_2O activity was higher (0.5–0.6) than most previous estimates (0.1–0.3, e.g. Santosh et al., 1991). Melting is then possible in some cases of localized orthopyroxene generation, as has been advocated by tenants of fluid-absent melting (Clemens, 1992). This possibility could explain the enigmatic close field associations of incipient charnockite and granite found together in the type locality at Kabbal. But melting, if any, is not the prime cause of incipient charnockite formation. It occurred when adequate composition was produced metasomatically by infiltration of high salinity aqueous solutions.

5. Conclusions

(1) The present work enhances the interpretation that incipient charnockite may be a more wide-spread and important factor in progressive metamorphism of the deep crust than previously recognized, and that concentrated saline solutions may have played a major role in Precambrian high grade metamorphism. Possible sources of highly saline fluids are deeply buried metasediments, disseminated halite in altered sea-floor basalt and peridotite (Sharp and Barnes, 2004), or orthomagmatic fluids (Ryabchikov and Hamilton, 1971) from deep crustal intrusions. These fluids are temporarily stored in lower crustal UHT-granulite domains before migrating towards the sites of formation of the incipient charnockites.

- (2) The presence of K-feldspar grain-boundary veins in the Arni incipient charnockite, which kind of veins are ubiquitous in massive charnockites immediately to the south (Hansen and Harlov, 2007) is evidence for continuity of the metamorphic progression as a coeval depth-zone profile subjected to through-crustal fluid-streaming processes. This hypothesis is more consistent with geochronology than the idea that the incipient charnockite is a reactivation of underlying older granulites, as postulated by Raith et al. (1989).
- (3) Ultrasaline fluid inclusions in Kabbal and Arni charnockites reinforce the concept of a fluid-mitigated petrogenetic link between massive charnockite, incipient charnockite and Closepet Granite, so closely associated in the Archean granulite transition zone of India.

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