



Streaming of saline fluids through Archean crust: Another view of charnockite–granite relations in southern India

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ABSTRACT

The complementary roles of granites and rocks of the granulite facies have long been a key issue in models of the evolution of the continental crust. “Dehydration melting”, or fluid-absent melting of a lower crust containing H₂O only in the small amounts present in biotite and amphibole, has raised problems of excessively high temperatures and restricted amounts of granite production, factors seemingly incapable of explaining voluminous bodies of granite like the Archean Closepet Granite of South India. The existence of incipient granulite-facies metamorphism (charnockite formation) and closely associated migmatization (melting) in 2.5 Ga-old gneisses in a quarry exposure in southern India and elsewhere, with structural, chemical and mineral-inclusion evidence of fluid action, has encouraged a wetter approach, in consideration of aqueous fluids for rock melting which maintain sufficiently low H₂O activity for granulite-facies metamorphism.

Existing experimental data at elevated T and P are sufficient to demonstrate that, at mid-crust pressures of 0.5–0.6 GPa and metamorphic temperatures above 700 °C, ascending immiscible CO₂-rich and concentrated alkali chloride aqueous fluids in equilibrium with charnockitic (orthopyroxene-bearing) gneiss will inevitably begin to melt granitic rocks. The experimental data show that H₂O activity is much higher (0.5–0.6) than previously portrayed for beginning granulite facies metamorphism (0.15–0.3). Possibilities for metasomatism of the deep crust are greatly enhanced over the ultra-dry models traditionally espoused. Streaming of ultrasaline fluids through continental crust could be a mechanism for the generation of the discrete mid-crust layer of migmatites suggested to characterize younger tectonometamorphic regions. The action of CO₂-rich and hypersaline fluids in Late Archean metamorphism and magmatism could record the beginning of large-scale subduction of volatile-rich surficial materials.

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1. Introduction

The orthopyroxene-bearing granitic rock called charnockite was first described in southern India and named by Holland (1900). Charnockitic rocks are the dominant component of the Archean granulite-facies terrane of northern Tamil Nadu and southern Karnataka States. Felsic rocks bearing orthopyroxene have been identified as major units in Precambrian crystalline shields worldwide (Field et al., 1980; Hubbard and Whitley, 1979; Ridley, 1992).

The origin of charnockite has been a subject of controversy. A key aspect of the charnockite problem is whether charnockite is most fundamentally an igneous or a metamorphic rock. Evidence for an igneous origin exists in orthopyroxene-bearing phases of stratified granitic intrusions that have been described in southern Norway (Petersen, 1980) and South Africa (Bohlender et al., 1992). In the crystallization

sequences of these bodies, an early orthopyroxene-bearing phase was followed by non-charnockitic biotite granite. Fluid inclusions in minerals of the Norway occurrence are CO₂-rich in the charnockitic portion and H₂O-rich in the non-charnockitic portion (Madsen, 1977), suggesting some form of control by the volatile components in charnockite–granite relations. In contrast to these unambiguously igneous examples of a charnockite–granite relationship, the Indian charnockitic rocks appear to be granulite facies metamorphic variants of the felsic gneisses that make up the bulk of Archean cratons (Martin, 1993). They are complexly deformed polyphase rocks, some of whose protoliths are much older than the granulite facies metamorphism that generated the orthopyroxene (Hansen et al., 1997; Mojzsis et al., 2003). In their major element chemistry, structures and metasedimentary and metabasic enclaves, the Indian charnockites are little different from the lower-grade gneisses of the Dharwar Craton (Cooray, 1969).

A particularly enigmatic example of charnockite–granite relations is found in southern Karnataka State. The outcrop of the Closepet Granite, a linear 250-km-long complex of intrusive granites, crosses the

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lower-grade craton from north to south and impinges upon the charnockitic terrane (Fig. 1). At this petrologic juncture, a charnockitic equivalent of Closepet Granite might be expected, but is not in evidence; instead, a coarsely porphyritic (commonly megacrystic) K-feldspar granite is succeeded abruptly by granulite facies gneisses. Although radiometric ages of whole rocks and zircons show that granulite facies metamorphism and granite emplacement were coeval at 2.52 ± 0.01 Ga (Friend and Nutman, 1991; Hansen et al., 1997; Jayananda et al., 1995; Mojzsis et al., 2003), and therefore presumably co-genetic, definitive petrogenetic relations of charnockite and granite have so far not been elucidated. An outstanding problem exists in the very low H_2O activity inferred by some workers to be necessary for orthopyroxene stability relative to biotite and amphibole in felsic rocks (Burton and O'Nions, 1990; Sen and Bhattacharya, 1990), whereas megacrystic granite is considered to implicate fluid-present, possibly even post-consolidation recrystallization (Glazner and Johnson, 2013).

The present paper seeks to define possible relationships of charnockite and granite in the Dharwar Craton of southern Karnataka, with particular reference to a possible role of active fluid phases, as is suggested by recently published descriptions of fluid inclusions in granite and charnockite. We make use of available experimental and

thermodynamic data on melting of quartzofeldspathic rocks and on the stability of orthopyroxene in such rocks. Particular emphases are whether the relatively low H_2O activity that could sustain orthopyroxene stability could be high enough to promote melting, what kinds of fluids (saline, carbonic, or other) could have been active, and whether possible fluids correspond to the observed fluid inclusions. Definition of such fluids might have implications for the general importance of fluids in evolution of the deep continental crust.

2. Regional and local petrologic relations

There exists a marked north-to-south increase in metamorphic pressure discernable in gneisses and associated metasedimentary units in the western (Raith et al., 1983), Closepet area (Hansen et al., 1984), and eastern (Hansen et al., 1995) parts of the Craton. Paleopressures of ≈ 0.4 GPa in the northern amphibolite-facies Peninsular Gneiss, corresponding to about 15 km depth, give way progressively southward to paleopressures up to 0.8 GPa in the granulite facies terrane, corresponding to paleodepths of 30 km. The N-S traverse is therefore an excavated mid-to-lower crust profile (Pichamuthu, 1965).

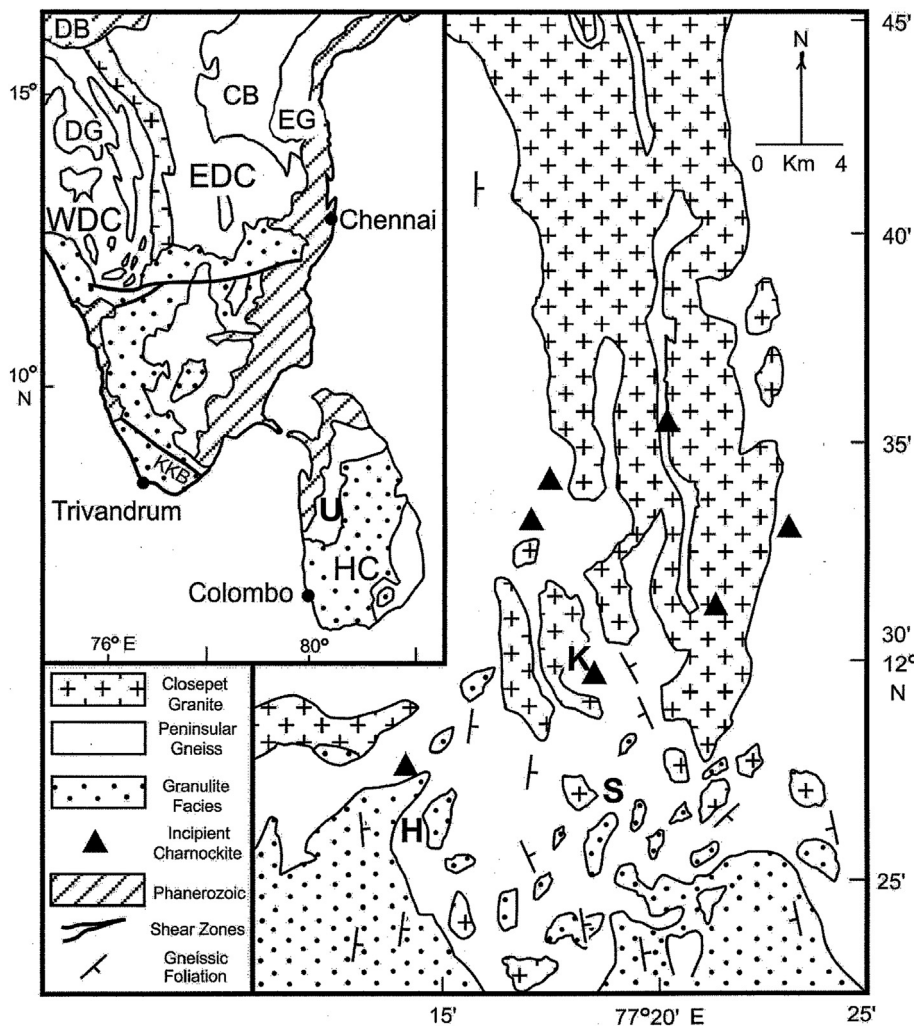


Fig. 1. Granite-gneiss-charnockite relations in the South India Archean high grade terrane, compiled from the maps of Suryanarayana (1960) and Devaraju and Sadashivaiah (1969), with some input from Mahabaleswar et al. (1995). The main map shows that Closepet Granite and massive charnockite outcrops have almost no overlap. Small outcrops of charnockitic gneiss occur within the southern margin of Closepet Granite; these include the incipient charnockites at Kabbaldurga and several other localities. The inset map shows relations of the southern India and Sri Lanka high grade terranes. Despite a 2-billion year age difference between the Kabbaldurga partially charnockitized hornblende-biotite gneiss and the closely similar occurrence at Udadigana, Sri Lanka, the mechanism of charnockite formation seems to have been essentially the same: by infiltration of CO_2 -rich and concentrated alkali chloride solutions at granulite facies P-T conditions, with partial melting in both cases. Symbols: DB: Deccan Basalt; EG: Eastern Ghats; CB: Cuddapah Basin; DG: Dharwar Greenstones; WDC: Western Dharwar Craton; EDC: Eastern Dharwar Craton; KKB: Kerala Khondalite Belt; HC: Sri Lanka High Grade Terrane; K: Kabbaldurga; H: Halaguru; S: Satnur; U: Udadigana.

A remarkable aspect of the areal relations (Fig. 1) is that charnockitic gneiss and Closepet Granite have only minor overlap; the granite outcrops become sporadic southward and phase out at about 12°30'N, almost exactly where massive charnockite outcrops appear. Petrologic studies of this critical area have revealed a new aspect of the charnockite problem, “incipient charnockite”: vein-like or patchy orthopyroxene-forming alteration of granitic gneiss. This phenomenon was first identified in a rock quarry at Kabbaldurga (Fig. 1) by Pichamuthu (1960). Incipient charnockite received little attention for 20 years until several similar occurrences were reported from the same area (Ramiengar et al., 1978). Patchy orthopyroxene-forming alteration of biotite gneiss has since been identified in high-grade terranes worldwide (Harlov et al., 2006; Hopgood and Bowes, 1990; McGregor and Friend, 1992; Van Reenen et al., 1988); this phenomenon now appears to be characteristic of transitional amphibolite-facies to granulite-facies terranes that traverse paleodepth intervals from upper to lower continental crust.

The Kabbaldurga quarry and nearby localities expose amphibole-biotite gneiss with localized charnockitic alteration, and closely associated intrusion of granite in the forms of discrete dikes and nebulous K-feldspar emplacement. The important issue immediately raised by the lithologic relations and their location in the regional transition to the granulite-facies is the nature of a petrogenetic relationship of granite with the first appearance of orthopyroxene.

3. Charnockite-granite relations: the fluid connection

Spatial relations of charnockite and granite in the Kabbaldurga quarry and similar localities show, even in a casual inspection, that both igneous and metasomatic processes are intimately involved (Friend, 1983). Charnockite and granite occupy deformational features

in host gneiss (Fig. 2a). The deformation involves a new foliation superposed on older gneissic structure (Fig. 2b). Chemical analyses of charnockitic lenses and immediately adjacent hornblende-biotite gneiss show that K-metasomatism is a characteristic aspect of this kind of charnockite formation (Stähle et al., 1987). Other slight chemical changes in the charnockitization process (loss of Ca, Mg, Fe and Ti, and gain of SiO₂) also indicate an open-system alteration (Hansen et al., 1987). The location of alteration in shears, foliation warps and dilatant structures suggests deformational conduits for metasomatizing fluids (Friend, 1985).

Further evidence of fluid-induced metamorphism exists in CO₂-rich fluid inclusions in quartz of incipient charnockite; this feature is also common to many or most granulites. Recently, inclusions of highly saline fluid and disseminated halite crystals have been identified in closely associated massive charnockite, incipient charnockite and megacrystic granite near the Kabbaldurga locality (Newton et al., 2014; Srikantappa and Malathi, 2008; Srikantappa and Zarkar, 2009; Touret et al., 2019). The saline fluids and the carbonic fluids could have coexisted as immiscible phases at high-grade temperatures and pressures (Gibert et al., 1998; Johnson, 1991). The presence of fluid inclusions of both types in massive charnockite, incipient charnockite and Closepet Granite suggests that there may be a fluid-mediated petrogenetic link among the three facies.

Most comprehensive studies of the Closepet Granite emphasize classical igneous processes of origin (Condie et al., 1986; Jayananda et al., 1995; Moyen et al., 2001, 2003), with almost no discussion of possible involvement by fluids. The massive charnockite at the southern terminus of the Closepet Granite was considered by Moyen et al. (2003) to be the root zone from whence the granite was extracted. The cause of crustal melting was considered to be heat from invading mantle-derived basaltic magmas; these magmas were almost entirely blended

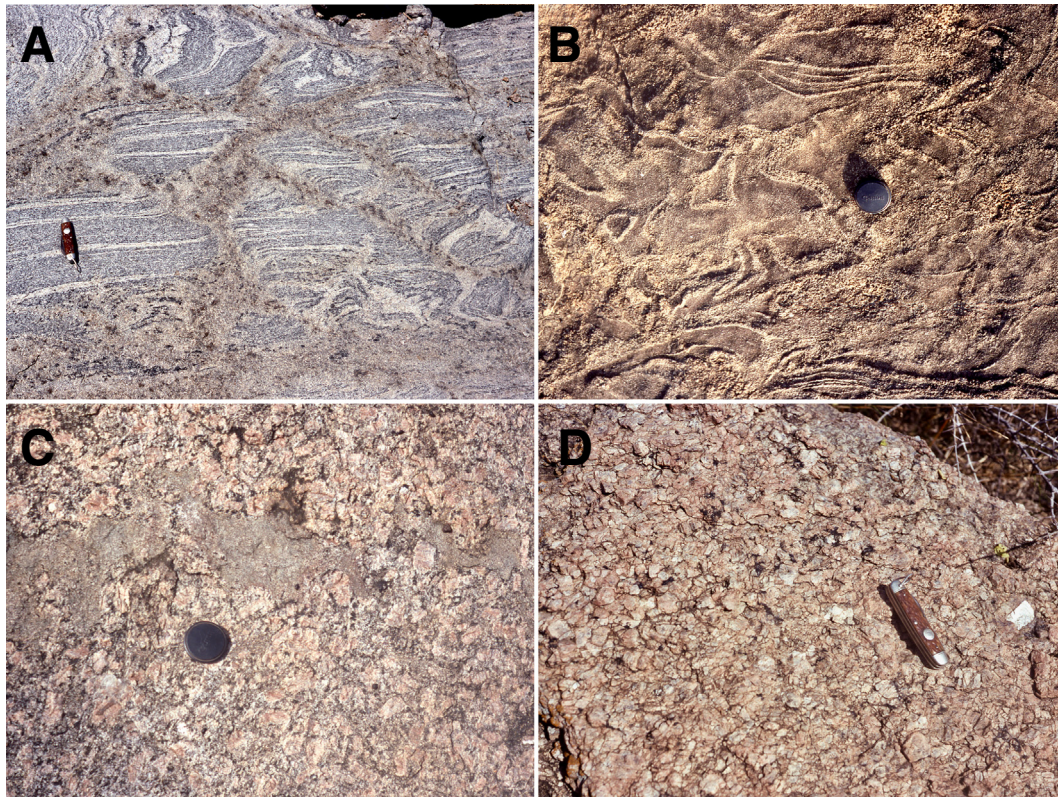


Fig. 2. Charnockitic (orthopyroxene-bearing) veins in granitic gneiss in rock quarries, southern Karnataka. **A:** Orthopyroxene development is attended by diffuse feldspathization, Belikatnur. **B:** Charnockitization and feldspathization are attended by development a new foliation coinciding with the N-S regional linear grain in the Closepet area. The cross-foliation gives rise to fold-interference patterns visible on weathered surfaces. Belikatnur. **C:** Coarsely porphyritic and mylonitic granite in a quarry surface near Ramnagaram, Karnataka State (just north of main map area at 77.3°E). Faint vestiges of former gneissic structure can be seen at the bottom of the view, dipping gently to the right. **D:** Megacrystic K-feldspar rock, near Ramnagaram.

with anatectic granitic liquids to form the K-rich porphyritic monzogranite. These hypotheses do not specifically address the discrete geographic distribution of massive charnockite and granite by any physical mechanism of emplacement.

Friend (1983) advanced an ingenious fluid-based hypothesis for the relationship among amphibolite-facies gneiss, massive charnockite, incipient charnockite and Closepet Granite. He envisioned that fluids rising through a deformation zone in the Late Archean crust, which fluids he believed to be dominantly CO_2 , formed charnockite by scavenging H_2O and K_2O from hornblende-biotite gneiss in the lower crust, and granite by implanting these components in the upper crust, thus melting it when H_2O rose to a critical level. This hypothesis neatly explains the depth-zone arrangement of charnockite, hornblende gneiss and granite, but invokes the passage of an improbably large amount of CO_2 -rich fluid. In order to effect charnockitic metamorphism, calculations made with the Holland and Powell (1998) thermodynamic data set on the reaction of biotite plus quartz to orthopyroxene plus K-feldspar plus fluid show that the H_2O content of CO_2 -rich fluids must be quite low. The lower crustal reservoir from which the H_2O supposedly was extracted would yield at most 0.6–0.8 wt. percent H_2O (Rutter and Wyllie, 1988). However, the granite liquid generated would have at least 8 wt. percent H_2O at the P-T conditions inferred for the transition zone (Makhluf et al., 2017). Thus, even complete dehydration of the lowermost one-half of the crust below Kabbaldurga could have yielded only very limited quantities of granite.

An outstanding problem which neither the classical igneous approach or the fluid-based hypothesis addresses is the evidence for large-scale potassium metasomatism in the Closepet Granite. Rocks of the southern Closepet vary from K-rich porphyritic granite to altered gneiss with vestigial foliation (Fig. 2c), to megacrystic orthoclases (Fig. 2d) to the nearly pure K-feldspar “Brick-red Rock” (Jayananda and Mahabaleswar, 1989; Radhakrishna, 1958). Seemingly, some unique H_2O -mediated action is indicated (Putnis et al., 2007).

The recent discovery of concentrated alkali chloride fluid inclusions in massive charnockite from Halaguru and Satnur (“H” and “S” in Fig. 1), in the Closepet Granite north of Kabbaldurga (“K”) and in incipient charnockite at Kabbaldurga, puts a different light on the H_2O budget and metasomatism concerns. The H_2O contents of saline fluids that could be at once charnockitizing and anatectic might be much higher than for CO_2 -rich fluids of the same H_2O activity; H_2O activities attending melting and charnockite formation might be within the ranges of observed saline inclusions in high-grade rocks.

4. Saline fluid inclusions and micro-veins: evidence of alkali metasomatism

Evidence of fluid-induced metamorphism and metasomatism exists in the form of fluid remnants preserved as mineral inclusions. Two types of fluid are ubiquitous in all granulites worldwide, highly saline aqueous fluids and nearly pure CO_2 (Touret and Huizenga, 2012). There are significant differences in amount and fluid density in the various occurrences. The maximum amount and density of CO_2 occur in the highest-temperature (UHT) granulites, up to and exceeding 1.1 g/cm^3 (Touret and Huizenga, 2012). In incipient charnockites, CO_2 inclusions are present, but in smaller amounts than in UHT or massive charnockites (Touret et al., 2019). Where the gneiss protolith contains graphitic material, as in incipient granulite paragneisses of Kerala, southern India, the amount of included CO_2 is greater than in incipient charnockite of the Kabbaldurga type (Touret et al., 2019).

Unlike CO_2 inclusions, concentrated saline inclusions in granulites are highly transformed during retrograde evolution, largely because the steep dP/dT slopes of the isochoric curves cause the fluids to become relatively decompressed with respect to the mineral hosts during post-metamorphic cooling. If an inclusion was lacking in daughter crystals, most or all of the fluid would be lost. If it contained a solid phase, a small irregular cavity would remain, squeezed around the crystal(s).

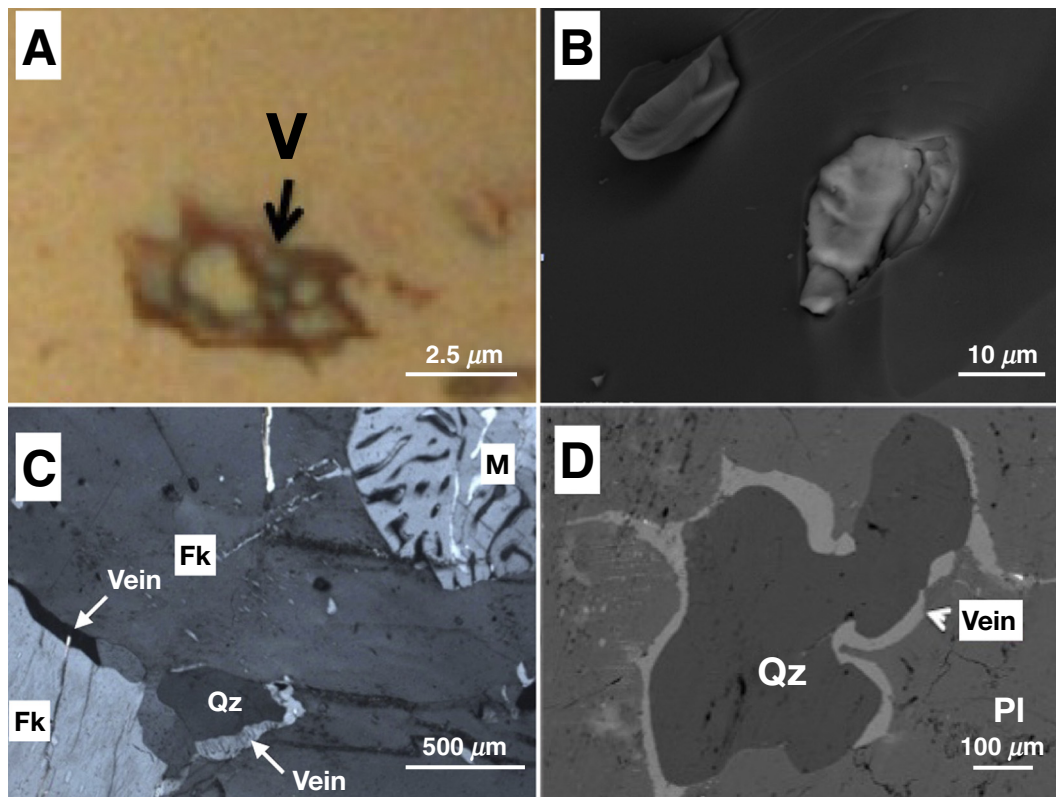


Fig. 3. Microtextures of incipient charnockite. A: Collapsed hypersaline inclusion in incipient charnockite, Kabbaldurga. B: Trapped halite in charnockitic vein, Satnur (Fig. 1). C: Grain-boundary myrmekite in an incipient charnockite from Sri Lanka. D: Grain-boundary K-feldspar micro-veins in incipient charnockite at Arni, Tamil Nadu (Touret et al., 2019).

This condition is termed “collapsed inclusion” (Touret, 2001). Fig. 3a is a typical example of a collapsed saline inclusion in the Kabbaldurga incipient charnockite. In some cases, the liquid phase has completely vanished, leaving a halite crystal trapped within a cavity in the host crystal. Fig. 3b shows such a trapped halite crystal in quartz from an orthopyroxene-bearing vein in the Archean granulite transition zone at Satnur (Fig. 1).

Because of ubiquitous disruption, the only information obtainable about the original salt concentration of a fluid inclusion is that it must have been very high (at least 30 mol% NaCl equivalent) to have coexisted with an immiscible CO₂-rich fluid (Gibert et al., 1998; Shmulovich and Graham, 1999). Such fluids are very mobile and reactive and would leave few vestiges of their original nature, once liberated.

Fluid inclusions in quartz of the southern Closepet Granite are not much different from those of the incipient charnockites. The texturally earliest inclusions are high-salinity brines and immiscible CO₂ (Srikantappa and Malathi, 2008); the latter type of inclusion is found mainly in quartz segregations (Bhattacharya et al., 2014). The main differences from the incipient charnockites are the somewhat lower concentration of salt (maximum 35 wt% NaCl) and lower density. These types of fluids are found in many alkali-rich granites, such as the granite ejecta from Ascension Island (Harris and Sheppard, 1987) and intrusions of the Ranchi area of eastern India (Mishra et al., 2007).

The second important line of evidence for brine infiltration in granulites is in the form of thin K-feldspar linings or micro-veins on the margins of plagioclase, quartz and mafic minerals. These microstructures were first identified in granulites from central Finland and the Lake Baikal region of Siberia (Perchuk and Gerya, 1993) and subsequently from the Dharwar Craton (Hansen et al., 1995), the Ivrea Zone of northern Italy (Harlov and Wirth, 2000) and many other occurrences worldwide (Aranovich and Safonov, 2018; Safonov and Aranovich, 2014), and appear to be a characteristic feature of most granulites. Similar grain-boundary feldspar linings have also been interpreted as the frozen remains of migrating interstitial melts (Sawyer, 1999; Hasalová et al., 1997), but other authors believe that the veinlets formed by alkali exchange of plagioclase with migrating concentrated chloride solutions. The plagioclase is nearly always changed to a slightly more calcic composition at the grain boundary adjacent to a K-feldspar vein (Perchuk and Gerya, 1993; Hansen et al., 1995; Harlov and Wirth, 2000; Touret and Huizinga, 2012; Rajesh et al., 2014). This is the opposite of what would be expected if the grain-boundary linings were a frozen granitic liquid: normal zoning of the adjacent plagioclase should be present in that case. Harlov and Förster (2002) noted that K-feldspar micro-veins in granulites from Alaska and northern Italy are found only in quartz-bearing rocks; this suggests that plagioclase has reacted with quartz and a K-bearing fluid to produce the micro-veins; a K-rich fluid low in silicate components would plausibly be a chloride solution. Based on two-feldspar thermometry, the veins formed from about 740 °C, near peak metamorphism, to less than 600 °C, generally followed by myrmekite (interstitial plagioclase-quartz intergrowths) below 600 °C (Perchuk et al., 2000). An example from the high-grade terrane of Sri Lanka is shown in Fig. 3c. K-feldspar micro-vein networks occur in textural equilibrium with orthopyroxene in incipient charnockite in the Archean granulite transition zone of northern Tamil Nadu, and appear to be part of the fluid-induced recrystallization that produced the orthopyroxene (Touret et al., 2019). An example is shown in Fig. 3d.

We conclude that the accumulated evidence for the important involvement of concentrated saline solutions in charnockite-gneiss-granite relations warrants an analysis of the physical chemistry of the granite-alkali-chloride-(CO₂) system as it may bear upon, and contribute to an understanding of, the southern India Archean high-grade terrane and Closepet Granite.

5. Phase equilibrium of fluid-rock systems

Sufficient experimental phase equilibrium data now exist to calculate with some confidence possible petrogenetic relationships among hornblende-biotite gneiss, analogous gneiss with orthopyroxene, possible pore-space fluids having an appropriate activity of H₂O, and the pressure-temperature conditions under which such fluids could react to melt the rocks. The calculation makes use of several experimental sources:

- 1) The temperatures and H₂O contents of a simple KAlSi₃O₈-NaAlSi₃O₈-SiO₂ granitic melt in the presence of concentrated (Na, K)Cl aqueous solutions of the Na/(Na + K) ratio necessary to coexist with both alkali feldspar and plagioclase (about 0.75) have been determined experimentally at deep-crust pressures (0.6–1.4 GPa: Aranovich et al., 2013);
- 2) The minimum melting temperatures of a simple granite in the presence of CO₂-H₂O fluids have been determined experimentally at pressures up to 1 GPa (Ebadi and Johannes, 1991). This makes possible the determination of a critically important quantity, the H₂O activity (a_{H₂O}), at the melting point, based on experimentally determined activity-concentration relations of CO₂-H₂O fluids (Aranovich and Newton, 1999);
- 3) Experimental phase-equilibrium data on the dehydration reaction of biotite plus quartz to orthopyroxene plus K-feldspar in simple systems (Aranovich and Newton, 1998), together with measured thermodynamic data for alkali feldspar (Thompson and Hovis, 1979), plagioclase (Newton et al., 1980) and (Na,K)Cl solutions at elevated P and T (Aranovich and Newton, 1997), serve to construct adequately the P-T-a_{H₂O} relations of the charnockite-forming reaction in granitic hornblende-biotite gneiss.

The first step is to chart the P-T-a_{H₂O} relations of first (minimum temperature) melting of a simple granite. The above sources of experimental data allow conversion of the CO₂-H₂O-granitic melt data to comparable values at the same activities of H₂O in saline solutions. The assumption is made that Cl and CO₂ in the experimental liquids are negligibly small (Papale et al., 2006; Webster, 1997). Fig. 4 shows the available granite melting observations on the plane of temperature and H₂O

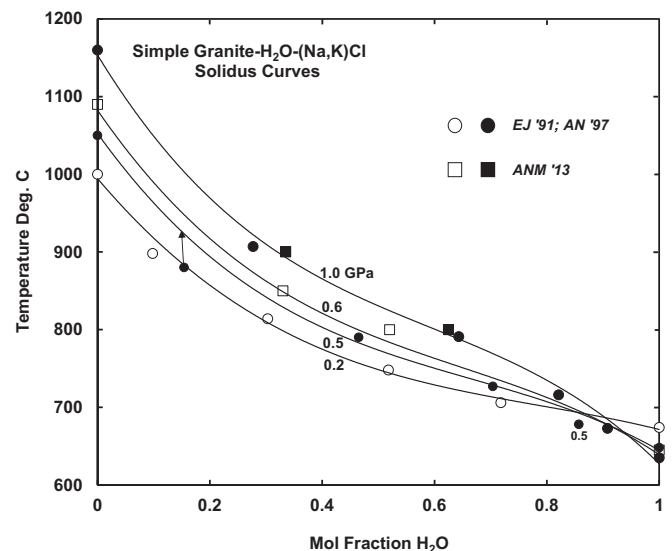


Fig. 4. First-melting (lowest-temperature) melting curves of simple (albite-K-feldspar-quartz) granite in the presence of (Na₇₅K₂₅)Cl aqueous solutions. Experimental points of CO₂-H₂O-granite from Ebadi and Johannes (1991: EJ '91) converted to Saline fluid concentrations of equivalent H₂O activity (Aranovich and Newton, 1997: AN '97) and melting point determinations of Aranovich et al. (2013: ANM '13). Dry melting points from Holtz et al. (2001).

Table 1

Pressure coefficients of first-melting temperatures T_m of simple granite saturated with (Na,K)Cl solutions, as functions of H_2O mol fraction X_{H_2O} .

	P^3	P^2	P^1	P^0
A	−1354.3	2756.2	−983.6	671.3
B	−1281.2	2800.6	86.25	1417.7
C	0	177.0	1322.0	1368.0
D	0	−86.15	679.8	866.5
E	−637.3	1097.8	−323.4	1024.5

$T_m = AX_{H_2O}^4 + BX_{H_2O}^3 + CX_{H_2O}^2 + DX_{H_2O} + E$ T_m in Deg. C.

$A = -1354.3P^3 + 2756.2P^2 - 983.6P + 671.3$ etc. P in GPa.

concentration in (Na_{0.75}K_{0.25})Cl solutions. The fluid-saturated melting temperatures T_m may be retrieved in the pressure range 0.2–1.0 GPa from a quartic function in X_{H_2O} , the H_2O mol fraction:

$$T_m = AX_{H_2O}^4 + BX_{H_2O}^3 + CX_{H_2O}^2 + DX_{H_2O} + E \quad (1)$$

whose coefficients are given in Table 1. The overall fit of the regression equations to the experimental points is better than $R^2 = 0.99$.

Fig. 5 shows calculated solidus curves of a saline fluid-saturated simple granite at 700–800 °C as functions of pressure. The unique effect of alkali chloride solutions on melting is apparent in the remarkable back-bending of the solidi. This behavior is quite in contrast to that of melting in the presence of CO_2 - H_2O solutions of the same H_2O concentrations. The solidus back-bend results from the strong decrease of H_2O activity in alkali chloride solutions as pressure rises above 0.3 GPa (Aranovich and Newton, 1997). This effect in turn stems from pressure-induced ionic dissociation of the solute salts (Aranovich and Newton, 1996). Consequently, an ascending alkali chloride solution of

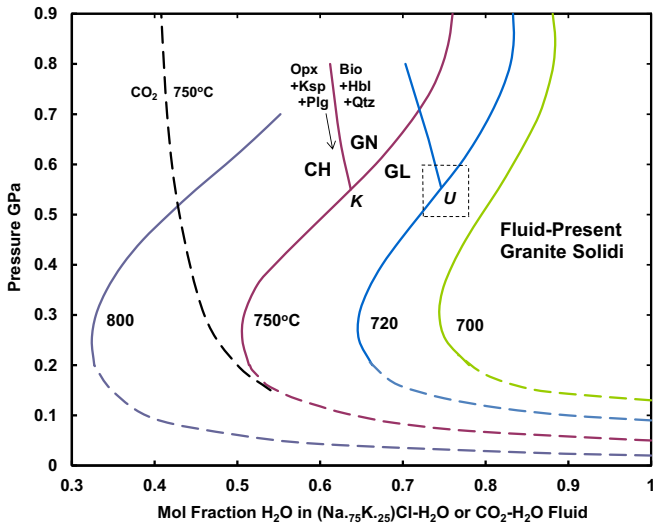
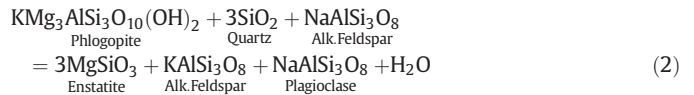


Fig. 5. Saline fluid-saturated solidus curves of simple granite as functions of pressure measured by Aranovich et al. (2013), compared with melting in the presence of CO_2 - H_2O solutions at 750 °C by Ebadi and Johannes (1991). The extreme difference in melting behavior of the two fluid media is apparent, and arises from a unique property of alkali chloride solutions, pressure-induced decrease of H_2O activity, which reverses the pronounced negative dP/dT slopes of hydrous melting curves. Curves for the formation of orthopyroxene in hornblende-biotite gneiss in the presence of such solutions, consistent with experimental data and field observations, intersect the melting curves at high angles in the range 700–750 °C. This property apparently militates that concentrated alkali chloride solutions rising through the crust at granulite facies conditions will inevitably melt quartzofeldspathic rocks. Incipient charnockite with incipient melting at Kabbaldurga, Karnataka, (Janardhan et al., 1979) and Udadigana, Sri Lanka (Perchuk et al., 2000), points “K” and “U”, respectively, are explained as intersections of the melting curves and charnockite-forming curves, “buffered” equilibria that will arise spontaneously from upward streaming of saline solutions through the crust.

a given concentration will undergo strong increase of H_2O activity in the mid-crust pressure range.

Calculation of the pressure-temperature conditions for incipient charnockite equilibrium in the presence of alkali chloride solutions encounters the difficult problem of complex mixing properties of amphibole and biotite solid solutions. This problem may be circumvented by use of an empirical or “geo-experimental” method (Newton, 2011, 2018). In this way, thermodynamic parameters may be extracted from the mineral chemistry of natural parageneses where pressures and temperatures of crystallization are sufficiently well known by conventional geothermometry and geobarometry. This method was used by Perchuk et al. (1981) to derive thermodynamic properties of minerals from their compositions in natural granulites. For the Kabbaldurga and similar occurrences, temperatures of 700–750 °C and pressures of 0.5–0.6 GPa appear to be well established (Hansen et al., 1984; Perchuk et al., 2000; Stähle et al., 1987). Assuming with Friend (1983), Burton and O’Nions (1990) and Perchuk et al. (2000) that the incipient formation of charnockite took place at the melting point of granitic hornblende-biotite gneiss, reference to Fig. 5 defines alkali chloride brine compositions for Kabbaldurga (point “K”, assumed to be 750 °C) and point “U”, accepting the estimate of Perchuk et al. (2000) of 720 °C for the Udadigana occurrence.

A simple-system reaction forms the basis for calculation of the dehydration of biotite gneiss to charnockite:



The condition for thermodynamic equilibrium of this reaction at a given temperature and pressure is:

$$\begin{aligned} 0 = & \Delta G_0^{\circ} + RT \ln(a_{or}) + RT \ln(f_{H_2O}) + RT \ln(a_{H_2O}) + RT \ln(a_{en}^3/a_{phl}) \\ & + RT \ln(a_{ab}^{Ksp}/a_{ab}^{Plg}) \end{aligned} \quad (3)$$

where ΔG_0° is the standard Gibbs free energy of the reaction of Expression 2, f_{H_2O} is the fugacity of H_2O at T and P , a_{H_2O} is the activity of H_2O in a saline or carbonic fluid having H_2O mol fraction X_{H_2O} , a_{en} and a_{phl} are the activities of the components $MgSiO_3$ and $KAlMg_3Si_3O_{10}(OH)_2$ in orthopyroxene and biotite, respectively, a_{or} is the activity of $KAlSi_3O_8$ in K-feldspar, and a_{ab} is the activity of $NaAlSi_3O_8$ in alkali feldspar (Ksp) and plagioclase (Plg). The last term of Expression 3 vanishes for equilibrated feldspars.

The “geo-experimental” estimates for the Kabbaldurga incipient charnockite define the quantities f_{H_2O} and a_{H_2O} . These quantities, together with the compositions of the Kabbaldurga feldspars, are compatible with the presence of hornblende as a virtual reactant in Expression 2. Also consistent is the fact that the feldspar compositions given by Janardhan et al. (1979) and Perchuk et al. (2000), yield, within limits of the reported compositions, the same ab activities in plagioclase and alkali feldspar at the inferred temperatures and pressures. This is a necessary condition for equilibrated feldspars. The values used for the parameters of Expression 3 are given in Table 2.

The high value of H_2O activity at 750 °C is in accord with that given by Perchuk et al. (2000), which apparent agreement lends credence to the complex thermodynamic mixing properties of biotite and hornblende used in their analysis. Their values for $a_{en}(opx)$ and $a_{phl}(bio)$ are also close to the simple ideal solid solution model for enstatite-ferrosilite and phlogopite-annite solid solutions, respectively:

$$a_{en} = Mg / (Mg + Fe^{2+}) \quad \text{and} \quad a_{phl} = [Mg / (Mg + Fe^{2+})]^3 \quad (4)$$

This agreement shows that the simple system reaction of Expression 2 is a valid indicator of H_2O activity in granulites. The lower thermal stability of the annite (Fe^{2+}) component of biotite is apparently

Table 2

Parameters of the reaction: Biotite (+ Hornblende) + Quartz = Orthopyroxene + Plagioclase + K-feldspar + Fluid (Expression 2, text).

Site	T °C	$\Delta G^\circ(2)$ kJ/mol	$\frac{RT \ln f_{H_2O}}{2}$ kJ/mol	X_{H_2O}	a_{H_2O}	X_{an}^{pl}	X_{or}^{ks}	a_{or}	X_{Mg}^{opx}	X_{Mg}^{bio}	$\frac{a_{en}^3/a_{phl}}{calc}$	$\frac{a_{en}^3/a_{phl}}{model}$
K	750	-61,023	72,020	0.637	0.537	0.31	0.74	0.781	0.45	0.53	0.552	0.633
U	720	-56,900	69,470	0.746	0.646	0.27	0.75	0.797	0.40	0.50	0.424	0.512

Mineral data: Janardhan et al. (1979); Kabbaldurga; Perchuk et al. (2000); Udadigana.

 a_{en}^3/a_{phl} (calc) from Eq. (2). a_{en}^3/a_{phl} (model) = $X_{Mg}^3(opx)/X_{Mg}^3(bio)$ $X_{Mg} = Mg/(Mg + Fe^{2+})$

compensated by the stabilizing factors Ti, F and additional Al in natural biotite (Berman et al., 2007). Our estimate of transitional granulite-facies H_2O activity (0.5–0.6) is much higher than that given by Sen and Bhattacharya (1990) and Burton and O’Nions (1990): 0.15–0.3.

The hornblende-biotite gneiss versus charnockite P-T boundary may be extended to higher pressures based on the assumption that the ratio a_{en}^3/a_{phl} is independent of pressure over a range of a few thousand bars at constant temperature. This is justifiable because the densities and Mg coordinations are closely similar in the two phases. The calculated charnockite-amphibolite facies gneiss boundaries are shown in Fig. 5.

6. Applications to petrogenesis

The pronounced back-bend in brine-saturated granite melting curves (Fig. 5) betokens melting of quartzofeldspathic rocks in the presence of alkali chloride solutions that ascend at 700–750 °C to mid-crust levels. The K-metasomatism shown by transitional granulite facies rocks in southern Karnataka would be an additional melt-producing factor for tonalitic and trondhjemitic gneisses (Newton, 1990). These factors combine with the charnockite-forming subsolidus equilibrium, Expression 2, to provide a plausible explanation for the enigmatic outcrop relations of biotite-hornblende gneiss, charnockite and granite in the Kabbaldurga quarry.

The sudden appearance of porphyritic granite at Latitude 12°30’N (or about 0.55 GPa paleopressure) is interpreted as the result of large-scale melting of gneiss at a critical depth by increase of H_2O activity in ascending alkali chloride solutions in equilibrium with charnockitic hornblende-biotite gneiss. The arrow in Fig. 5 suggests the path of H_2O concentration in ascending solutions. Over a pressure range approaching 0.55 GPa at 750 °C, solutions in equilibrium with both biotite

and orthopyroxene would accumulate a small amount of H_2O from dehydration reactions, but would lose some H_2O to migmatitic melts (Touret and Dietvorst, 1983), perhaps fluxed by local excesses of potassium. Large-scale melting occurs in rocks of granitic composition where the H_2O mol fraction in the saline solution increases to 0.63 (point “K” in Fig. 5); it is possible that this composition was conditioned by influx of K into trondhjemitic or tonalitic gneisses (Newton, 1990). Fig. 5 also shows the calculated temperature–pressure conditions (720 °C and 0.5–0.6 GPa) for the incipient charnockite in hornblende-biotite gneiss at Udadigana (Perchuk et al., 2000). These authors reported halite-bearing fluid inclusions in quartz of the incipient charnockite, and textures indicative of partial melting of the gneiss. Their calculated a_{H_2O} interval in Fig. 5 has been converted to mol fractions in a $Na_{.75}K_{.25}Cl$ aqueous solution. The high H_2O activity inferred for the Udadigana occurrence confirms the present assessment for incipient charnockite, since the calculations of Perchuk et al. (2000) were based in part on activity models for hornblende and biotite solid solutions, in contrast to our “geo-experimental” determination. A calculated phase-equilibrium diagram (Fig. 6) for fluids in the system H_2O - CO_2 - $NaCl$ at 750 °C and 0.55 GPa (Aranovich et al., 2010) shows that the minimum H_2O activity of 0.5 for Kabbaldurga is quite far from salt saturation at the time of melting and incipient charnockite formation. Salt saturation was reached in residual fluids in post-metamorphic cooling.

The present study may bear upon the debated relationship between granulites and granites. According to classical theory, the deep continental crust was converted to the granulite facies (garnet-pyroxene gneiss) over time by removal of H_2O , alkalis and incompatible elements in anatectic granitic liquids (Fyfe, 1973). Melting has most commonly been construed as fluid-deficient or fluid-absent (Clemens, 1990). Fluid-absent melting (dehydration melting) was proposed by Bhattacharya and Sen (2000) to explain the granite-granulite relations at Kabbaldurga.

Experimental data on hornblende stability in granitic gneiss provides evidence in favor of fluid-present metamorphism and melting. Modal analyses of host gneiss and incipient charnockite at Kabbaldurga (Hansen et al., 1987; Stähle et al., 1987) agree that hornblende breakdown is the main contributor to the generation of the orthopyroxene; biotite also is a contributor, but survives, and is a ubiquitous associate of orthopyroxene in incipient charnockite. This behavior contrasts strongly with the much higher thermal stability of hornblende than biotite in experimental fluid-absent melting of granodiorite at 0.5 GPa (Naney, 1983) and of tonalite at 1.0 GPa (Rutter and Wyllie, 1988). Apparently, hornblende is more vulnerable than biotite to a fluid of low H_2O activity. The persistence of biotite at decreased a_{H_2O} is largely due to the stabilizing influence of increased Ti content (Harlov et al., 2006; Harlov and Förster, 2002). Small amounts of remanent amphibole are likewise conditioned by high Ti content.

Jayananda et al. (1995) suggested, on the basis of a thorough chemical survey of the porphyritic granite of the southern Closepet, that it could have been generated by melting of upper mantle greatly enriched in alkalis and large-ion lithophile elements (LILE: rare earth elements, Ba, etc). In a subsequent series of papers, Moyen et al. (2001, 2003) modified this hypothesis by invoking mantle-derived basaltic magmas as the primary vector of the trace elements, with subsequent nearly complete homogenization of the invading magmas with crustal anatectic melts.

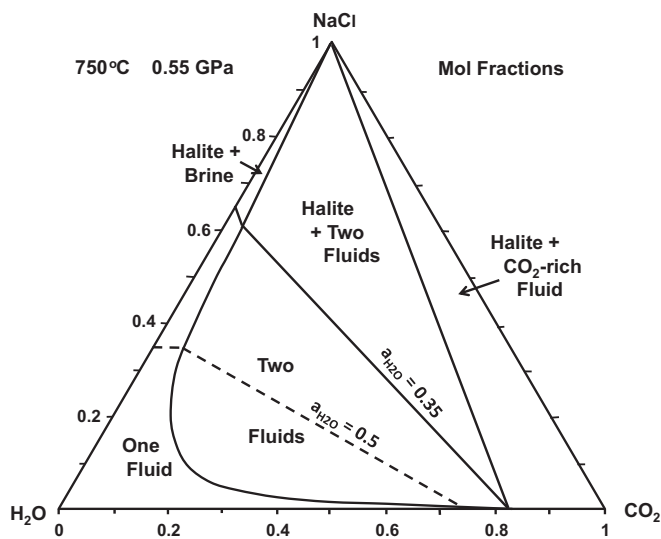


Fig. 6. Activity–concentration relations in H_2O - $NaCl$ - CO_2 solutions from Aranovich et al. (2010) at 0.55 GPa and 750 °C. The diagram shows that saline solutions corresponding to those inferred for incipient melting and incipient charnockite formation at Kabbaldurga and comparable transitional granulite facies occurrences can coexist with immiscible CO_2 -rich fluids and are salt-undersaturated in the inferred metamorphic P-T range.

Invasion of mantle-derived basaltic magmas into the lower crust is appealing as a source of heat for granulite-facies metamorphism (Bohlen, 1987) and generation of granitic magmas (Huppert and Sparks, 1988). If the basaltic magmas emanate from an enriched subcontinental mantle, they could impart alkalis, volatiles and LILE components to the anatectic products. This idea leaves open the question of why the Moyen et al. (2003) root zone of the Closepet Granite coincides almost exactly with the regional orthopyroxene isograd, and how the basaltic magmas were apparently able to homogenize so completely with the anatectic products to yield the voluminous porphyritic granite.

A contrasting hypothesis, one that specifically involves the action of the volatile components, is that of Litvinovsky and Podladchikov (1993). In this scenario, mantle-derived and volatile-rich basaltic magmas invade the lower crust, where they generate anatectic granitic melts. The volatiles, dominantly H₂O, are transmitted directly by diffusion into granitic magma without intervention of a separate fluid phase. The volatile transfer imparts H₂O-compatible components (non-H₂O volatiles, alkalis, halogens, trace elements) to the granites with negligible hybridization of the basaltic and granitic magmas. The basaltic magmas must freeze upon loss of their volatiles; granitic magmas then pursue their own agenda in ascending to the mid- and upper crust. This hypothesis has the merit that the involvement of H₂O moderates the possible temperature regimes of metamorphism and anatexis.

By invoking a specific role of volatile-mediated transmission, the Litvinovsky and Podladchikov (1993) hypothesis potentially solves an important problem of the origin of within-plate or anorogenic granites, the apparent decoupling of major and trace element budgets. Granites are, in their major element compositions, almost entirely products of crustal melting (Leake, 1990; Wyllie et al., 1976), whereas, in some regards, as in Sr and Nd isotopes, the granites may appear to have a mantle source (De Paolo, 1981; Soesoo, 2000).

We propose a more direct role of volatile components in genesis of the Closepet Granite, one that involves a discrete fluid phase or phases. Alkali chloride and CO₂ are much more soluble in basaltic magmas than in granitic magmas, and solubility is pressure-dependent (Webster et al., 1999). Upon entering the crust, basaltic magmas of enriched mantle origin may, in addition to losing H₂O to anatectic magmas, emit carbonic and ultrasaline fluids of low H₂O activity. The CO₂ and Cl components promote outgassing and freezing of magmas (Holloway, 1976; Webster et al., 1999). Because of low H₂O activity, these fluids may travel upward in the crust, carrying heat, fluxing granulite-facies metamorphism, and perhaps leaching K and LILE, but provoking only limited anatexis, until, at mid-crust levels, H₂O activity increases dramatically by virtue of the effect of decreasing pressure on reassociation of alkali and chloride ions into neutral molecules. This hypothesis could explain why, at mid-crust levels, generation of granitic melt succeeded granulite-facies metamorphism in the Late Archean crust of southern India, and emphasizes an important role of metasomatism in the origin of the granite.

Some authors have argued that megacrystic K-rich granites and granodiorites record fluid-present, relatively low-temperature, possibly post-consolidation processes (Glazner and Johnson, 2013). Three-dimensional modeling of various spectroscopic data on K-feldspar megacrysts from the southern Closepet revealed alkali and volatile element distributions that betoken interaction at magmatic temperatures with a “halogen-rich” fluid (Slaby et al., 2012).

Jayananda and Mahabaleswar (1991) noted the apparently syn-deformational emplacement of the southern Closepet Granite. According to them, many of the granites are mylonitic. The strain pattern in the rocks is especially evident in deformation of K-feldspar megacrysts (Fig. 2c), and as shown elegantly by remanent magnetic anisotropy of oriented samples from the Closepet (Moyen et al., 2003). The strain-induced fabric is consistent with Late Archean transcurrent faulting in the Dharwar Craton, which motion created the dominant regional lineation and elongate outcrop pattern of the Closepet (Chadwick et al.,

2000). In the present interpretation, a vertically oriented transcrustal fault zone served as a conduit for saline and carbonic fluids of deep-seated origin. The Closepet Granite may share this mode of origin with other swarms of alkaline granites related to transcurrent fault zones, such as the Paleozoic granites of the Arabian-Nubian Shield. Sylvester (1989) suggested that the source rocks of these granites were tonalitic gneisses, but could not explain the seemingly high temperatures required to melt them. Concentrated alkali chloride aqueous solutions could provide both H₂O and alkalis to provoke melting at high grade metamorphic conditions.

A mid-crust layer of partial melting has been invoked as an important zone of dislocation in formation of gneiss domes and metamorphic core complexes (Vanderhaeghe and Teyssier, 2001). Uplift is considered to be gravity-driven and occurs along low-angle normal faults lubricated by interstitial silicate melt (Teyssier and Whitney, 2002). The positive dP/dT slope of saline-aqueous rock melting may be a critical condition in exhumation of metamorphic/igneous terranes, as it enables melting of crustal rocks by ascending fluids. Several authors have presented evidence of fluid-present anatexis in generation of migmatite complexes (Höller and Hoinkes, 1996; Smit and Van Reenen, 1997). Fluid-inclusion studies of some migmatites reveal the presence of concentrated saline solutions (McLelland et al., 2002; Olsen and Touret, 1985).

Stähle et al. (1987) showed that oxygen and carbon isotopes extracted from fluid inclusions of the Kabbaldurga charnockite are consistent with a mantle origin, but argued that the CO₂-rich fluids might have been released from inclusions in underlying older granulites, a conceivable origin if the granulites in question were themselves of more or less direct mantle origin. However, whole-rock Rb-Sr dates of both massive and incipient charnockites in southern Karnataka gave identical ages of 2.50 ± 0.02 Ga (Hansen et al., 1997) and zircon U-Pb ages of Closepet Granite and closely associated massive charnockites are synchronous at 2.51 ± 0.02 Ga (Friend and Nutman, 1991; Mojzsis et al., 2003). Also, the Stähle et al. (1987) hypothesis, restated as a general principle by Raith et al. (1989), does not explain the coeval saline inclusions and disseminated halite common to all members of the granite-charnockite association.

The great expanse of Late Archean granulite facies terranes of the Superior Province of Canada (Percival et al., 1992) may imply a commensurate depth scale (i.e. upper mantle involvement). High oxidation states of granulites of the southern India Archean high-grade terranes, as recorded in oxide and sulfide mineralogy, are not readily explainable by closed-system processes, but more likely record infiltration of oxidizing fluids (Harlov et al., 1997; Samuel et al., 2019). The latter study invoked high sulfate content of the saline solutions as an oxidant. An underlying mantle rich in volatiles, halogens and sulfur, in communication with the middle and upper crust through shear zones, may have been the source of the saline and carbonic fluids. Conceivably, the advent of large-scale fluid-involved high-grade metamorphism records the beginning of major flow of surficial materials (limestone, altered sea-floor sediments) into the upper mantle through subduction.

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