TRAPPED CARBON DIOXIDE IN EAST ANTARCTIC GRANULITES: EVIDENCE FROM FLUID INCLUSIONS

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Abstract: Charnockites from the Skallen area in the Lützow-Holm Bay region of East Antarctica show carbonic fluid inclusions trapped within garnet, feldspar and quartz. Microthermometric and micro-Raman laser analyses characterize these fluids to be pure CO_2 . With a maximum density of 1.05 g/cm³, these fluids are inferred to represent the traces of the ambient fluid which effected the reduction of water activities and dehyrated large segments of the Gondwanian deep crust. A sympathetic decrease in the densities of carbonic fluids trapped within early to late crystallizing minerals is recorded. This defines an uplift path which crossed higher to lower density isochores, in close correspondence with the T-convex metamorphic P-T trajectory computed for this terrane from mineral phase equilibria constraints. We provide the first direct evidence for the infiltration of CO_2 in the East Antarctic granulites, a well established mechanism believed to have been instrumental in generating charnockites in the other Gondwana continents.

1. Introduction

Granulite facies rocks are generated by a variety of mechanisms, including dehydration through melt extraction (WATERS and WHALES, 1985), vapor-absent metamorphism (LAMB and VALLEY, 1984), and dehydration through the influx of a CO₂-rich fluid (Newton et al., 1980). While considerable debate exists over the relative importance of each mechanism in stabilizing the anhydrous mineral assemblages which characterize granulites (e. g; TOURET and HARTEL, 1990), evidence from a number of granulite terranes indicates that the low water activity required to stabilize granulite mineral assemblages is generated by dilution or expulsion of the pore fluid through the influx of CO₂ (SANTOSH, 1991). This process is best demonstrated on a local scale in the case of veins and patches of incipient charnockites in intimate association with upper amphibolite facies gneisses in southern India (RAITH et al., 1989; SANTOSH et al., 1990) and Sri Lanka (HANSEN et al., 1987; SANTOSH et al., 1991a). CO₂ influx has also been demonstrated on a regional scale, in the case of massive charnockites (e.g.; SANTOSH, 1986, 1991; TOURET and HANSTEEN, 1988). The process of granulite genesis aided by CO₂ influx is popularly known as "carbonic metamorphism", first recognized by TOURET (1970), who coined the term "metamorphisme en milieu carbonique", and subsequently proposed as a global mechanism for the desiccation of the deep crust by NEWTON et al. (1980).

Evidence for CO_2 infiltration through faults and shears is provided by field structures associated with incipient charnockite formation, where orthopyroxene-bearing anhydrous granulites have developed as patches and veins within amphibolite facies gneisses along oriented zones of fluid pathways. The role of fluids in the petrogenesis of regional granulite masses is more difficult to demonstrate simply from field structures, and require a detailed characterization of the fluid phases trapped within the minerals during metamorphism. It has been demonstrated in a number of cases that peak metamorphic fluids are often preserved as high density carbonic inclusions in granulites (*e.g.*; SANTOSH, 1986; TOURET and HANSTEEN, 1988), signifying the importance of fluid inclusion study as a potential technique to understand fluid processes in the deep crust. In this regard, the granulite facies rocks of East Antarctica constitute a challenging problem for investigations related to trapped fluid phases. This paper presents the preliminary results of an ongoing detailed study on the mineral and fluid phase equilibria characteristics of the gneiss-charnockite terrane of the Lützow-Holm Bay (LHB) region in East Antarctica.

2. Geologic Background

The LHB region (Fig. 1) is mostly a Proterozoic terrane which has undergone amphibolite to granulite grade metamorphism (YOSHIDA et al., 1983; MOTOYOSHI et al., 1989). The geology and tectono-metamorphic history of this terrane were studied in detail by YOSHIDA (1978, 1979) and are summarised in YOSHIDA et al. (1983), HIROI et al. (1983) and SHIRAISHI et al. (1987). The region is dominantly underlain by pelitic to psammitic gneisses, intermediate to acid charnockites, metabasites, gneissose granites and subordinate amounts of calc-silicates and ultramafic rocks. As one of the important members of the Gondwana supercontinent of which the Antarctic continent was a part, these lithologic units have serveral features in comon with the leptynite-khondalite and charnockite suites of rocks in the Gondwana segments of southern India and Sri Lanka. Charnockites in the LHB region, as in the other terranes, show a range of mineral assemblages from garnet, biotite and orthopyroxene paracharnockites with or without graphite to hornblende, biotite and orthopyroxene orthocharnockites in the absence of garnet and graphite. The field relations in the LHB region as studied in Skallen by YOSHIDA (1978) clearly indicate that the charnockites have resulted by the prograde transformation of the gneisses. Here, coarse charnockitic rocks occur cutting the gneissic fabric with a metabasite layer in the gneiss traceable into the charnockite without any obliteration of its structural fabric. This has been cited as evidence for the *in situ* transformation of gneisses to charnockites. Mineral phase equilibria studies (YOSHIDA, 1979; SUZUKI, 1983; MOTOYOSHI et al., 1989; SANTOSH and YOSHIDA, in prep.) ascribe metamorphic P-T conditions of ca. 650-850°C and 6-8 kbar to the charnockites in the Skallen area. The gneisses and charnockites show comparable pressuretemperature ranges, with no major detectable gradients, precluding the possibility of local increase in P-T conditions as the cause for amphibolite facies gneiss to charnockite transformation. The other strong possibility is a fluid-controlled transformation process. This study examined garnet and orthopyroxene-bearing charnockites and upper amphibolite facies gneisses from the Skallen area for tracing the role of fluids.







3. Fluid Inclusions

Fluid inclusions were studied in garnet, K-feldspar and quartz from charnockite (specimen 0309A), metabasite (0615) and garnet-biotite gneiss (614) samples (sample details and localities as documented in YOSHIDA, 1978). The charnockite sample comprises an assemblage of quartz-K-feldspar-plagioclase-garnet-orthopyroxene with accessory biotite, hornblende and opaques. The metabasite has a similar assemblage, except for the subordinate abundance of quartz and K-feldspar, and the presence of clinopyroxene as an additional phase. The gneiss assemblage is characterized by quartz-K-feldspar-plagioclase-garnet-biotite. Doubly polished thin wafers of unaltered whole rocks were prepared, with a thickness of 0.5–1.0 mm. Inclusion petrography was carried out using a normal petrological microscope in transmitted light. Microthermometric measurements were carried out using a temperature-calibrated U.S.G.S. adapted gas flow-type heating-freezing system housed at the Geological Survey of Japan Laboratory at Tsukuba. Preliminary laser-excited Raman spectral analyses of representative inclusions were done at the Physics and Chemistry Research Institute (Tokyo)

using Jobin-Vyon RAMANOR U-1000 Raman microprobe. A detailed discussion of the inclusion petrography, together with size and phase-type populations and their implications, are presented elsewhere (SANTOSH and YOSHIDA, 1991). The following is a brief account of the salient fluid characteristics.

In charnockite, the dominant inclusion category is a monophase carbonic type (Fig. 2a), completely filled at room temperature with dense carbon dioxide (see below). In feldspar and garnet, they occur mostly in a scattered and azonal pattern; in quartz they are either scattered or distributed along narrow trails which pinch out within individual grains. Quartz contains more than one generation of inclusions, as indicated by the cross-cutting trails. This might reflect the capture of fluids at various stages during metamorphic uplift, even under low pressure-temperature conditions. Carbonic inclusions occur in both the earlier arrays and late arrays. In the latter case, they coexist with mixed carbonic-aqueous inclusions along sub-linear arrays that cut across several grains. Quartz from the gneisses also contains monophase carbonic inclusions of the type described from charnockites above, but the dominant category comprises two visible phases at room temperature, nucleating a third phase upon slight cooling, consistent with a mixed carbonic-aqueous composition (Fig. 2b). From statistical approach, SANTOSH and YOSHIDA (1991) recorded an increase in the abundance of carbonic inclusions as much as five times while passing from gneiss to charnockite in the Skallen area, which they considered as a reflection of the decrease in the partial pressure of water in the charnockite. They traced the chronology of fluid entrapment from early high density carbonic, through intermediate mixed carbonic-aqueous, to late aqueous regimes, vis-a-vis the uplift history.

A. charnockite

B. gneiss



Fig. 2. Photomicrographs of fluid inclusions in charnockite (A) and gneiss (B) from the Skallen area. The host mineral is quartz in both cases. Inclusions in charnockite are dominantly monophase as seen from the photograph, completely filled with CO₂ at room temperature. The dominant category in the gneisses comprises inclusions which are bi-phase at room temperature (as seen in the photograph), but develop a third phase upon cooling, consistent with a mixed carbonicaqueous composition.

In Fig. 3 is presented the analytical results to characterize the composition of the entrapped fluid within the carbonic inclusions. The histograms in the inset compile the melting temperatures observed during freezing experiments of these inclusions. The peak melting temperatures lie near -56.6° C, which is the triple point for pure CO₂. This indicates that the trapped fluid phase has a near-pure CO₂ composition. Depression in the melting temperature of pure CO₂ is generally brought about by the presence of impurities, like nitrogen or methane. In the absence of graphite, methane is an unlikely candidate. Nitrogen is sometimes present in traces associated with the carbonic inclusions in paracharnockites of the Kerala Khondalite Belt in southern India (*e.g.*; KLATT *et al.*, 1988). We examined representative inclusions from the Skallen charnockites under a laser Raman probe and obtained characteristic spectral peaks for CO₂ (Fig. 3). Although the CH₄ and N₂ regions were also scanned for the same inclusions, no pulses were recorded. This indicates that the dominant fluid system in the Skallen charnockite is pure carbon dioxide. Studies in other regional charnockite terranes have also shown pure carbon dioxide trapped within inclusions (*e.g.*; TOURET and HANSTEEN, 1988).

Homogenization experiments of the carbonic inclusions in the different minerals



RAMAN SHIFT cm¹

Fig. 3. Composition of the trapped fluid phase as measured by the melting temperatures during microthermometric experiments (histograms in the inset) and laser-excited Raman spectroscopy (showing characteristic CO₂ spectra).



Fig. 4. Histograms showing the homogenization temperatures of carbonic inclusions in garnet, feldspar and quartz from charnockites in the Skallen area. The lowest homogenization temperatures are yielded by garnet-bound inclusions, which have the highest densities.

yielded interesting results (Fig. 4). In all the cases, the phase transformation was characterized by homogenization into the liquid phase. Inclusions in garnet homogenized first, followed by those in feldspar, and finally in quartz. The late array-bound inclusions in quartz showed homogenization at an entirely different higher temperature range. The lowest homogenization temperature, recorded in garnet, is translated into a density of *ca*. 1.05 g/cm³. From garnet through feldspar to quartz, the earliest generation inclusions show a density variation from 1.05 to 0.90 g/cm³. The late arraybound inclusions in quartz stand apart, with the lowest density at *ca*. 0.83 g/cm³. Carbonic inclusions in the gneisses show density variation mostly in the lower range, *cd*. 0.80– 0.88 g/cm³, and appear to have trapped only the late fluids which infiltrated through the charnockite and penetrated the adjacent gneisses.

4. Discussion

Charnockite formation in East Antarctica involved the transformation of upper amphibolite facies gneisses to anhydrous granulites on a regional scale in fairly constant P-T conditions. Observed mineral transformations, namely the consumption of garnet and biotite to produce orthopyroxene may be represented by the reaction:

 $Garnet + Biotite + Quartz = Orthopyroxene + Alkali feldspar + Plagioclase + H_2O.$

At granulite facies temperatures of $650-800^{\circ}$ C, the transformation of biotite to orthopyroxene through the reaction, biotite + quartz (gneiss) = orthopyroxene + K-feldspar + vapor (charnockite) requires that the water activity was considerably reduced, to a value of possibly near 0.20. The fluid inclusion characteristics documented in this study suggest that the reduction in water activity to promote dehydration reactions was achieved by the influx of CO_2 -rich fluids. The timing of entrapment of fluids in granulite minerals has been considered in detail elsewhere (SANTOSH *et al.*, 1991b), where it is shown that symmetamorphic fluid capture within inclusions occurs during charnockite formation.

Charnockite formation in Skallen also involved the consumption of garnet, similar to that recorded from the charnockites of southern India (SANTOSH, 1987; SANTOSH *et al.*, 1990) and may imply a decompression reaction, *i.e.*, a drastic reduction in pressure under relatively constant temperature, through isothermal uplift. This can be further verified with the present data set on fluid densities from the different minerals. Figure 5 shows the corresponding CO_2 isochores for the various densities recorded from garnet, feldspar and quartz in the Skallen charnockites. A systematic decrease in the densities of trapped fluids can be observed from early to late crystallizing minerals. Such a situation implies the crossing of higher to lower density isochores during metamorphic



Fig. 5. P-T diagram showing isochores (iso-density lines) corresponding to the various densities of carbonic fluids in garnet, feldspar and quartz from charnockites in Skallen. Regional metamorphic conditions are shown as a P-T box as computed from mineral phase equilibria. The fluid characteristics are consistent with a P-T path which crosses higher to lower density isochores (broad arrow). This isothermal uplift path is consistent with the P-T trajectory derived by MOTOYOSHI et al. (1989) for the Lützow-Holm Bay granulites based on combined data from mineral phase equilibria and geochronology (trace of which is shown as a curved, dotted line).

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uplift, possible only when the uplift trajectory is characterized by a T-convex isothermal path. In the case of an isobaric uplift path, the early to late crystallizing minerals would show trapped fluids with progressively increasing densities, where the resultant T-concave path would cross lower to higher density isochores in a P-T grid. Example for the latter case has been demonstrated by TOURET and HANSTEEN (1988) from the high pressure granulites of the Nilgiri Hills in southern India. The T-concave uplift path obtained from the present study is in close correspondence with that delineated by MOTOYOSHI *et al.* (1989) for the LHB region, based on regional mineral phase equilibria characteristics *vis-a-vis* geochronological data-base.

This study supports the notion that the low P_{H_2O} required to generate granulites in the deep crust is largely achieved by the infiltration of CO₂-rich fluids, a process popularly known as carbonic metamorphism (TOURET, 1970; NEWTON et al., 1980; SANTOSH, 1986). The entrapment pressures of the highest density fluid inclusions in the Skallen charnockites correlate with the minimum pressure estimates from mineral phase equilibria, and their isochores penetrate the P-T box of regional metamorphism. This is a suggestive, although not conclusive, criterion which proves that these carbonic inclusions were entrapped under peak metamorphic conditions, and that they preserve the remnants of the metamorphic fluids. Of late, the mechanism of generating low water activity to stabilize granulite mineral assemblages has been the subject of heated debates. From detailed phase equilibria, fluid inclusion and carbon stable isotope traverse across a gneiss-charnockite reaction front in southern India, it was recently demonstrated that CO₂ advection in the deep crust serves as a fundamental mechanism for the evolution of dehydrated crust in Precambrian terranes (SANTOSH et al., 1990). From the incipient charnockite quarry in the Kurunegala district of Sri Lanka, BURTON and O'NIONS (1990) argued that charnockite formation was controlled by dehydration melting with limited melt seggregation, in the absence of externally flushed fluids. Their study, however, shows a lack of comprehension, as they fail to take into consideration the field structures at Kurunegala, with spectacular fluid-pathways along which charnockites have developed. Recent experimental data of PETERSON and NEWTON (1990) indicate that in the P-T conditions attained commonly during granulite facies metamorphism, introduction of CO_2 into a biotite gneiss would result in melting. Hence, it is important to be able to distinguish between dehydration melting in the absence of CO₂, and melting induced by the influx of CO_2 . Clearly, this is a topic on which future research in charnockite terranes should concentrate. Decompression melting induced by external CO₂ influx is more probable, because the general occurrence of high density carbonic inclusions in these rocks and their genetic setting in a decompression environment. Models for granulite formation are not unequivocal, but evidence from charnockitic terranes in the Gondwanian lower crustal domains has converged to indicate that CO₂ was indeed a potential variable which dictated granulite genesis, regardless of the mechanism which operated (for further details on this topic, the reader is referred to a recent review by SANTOSH, 1991, and references therein). An evaluation of the source of CO_2 for the Skallen rocks requires detailed stable isotope studies. The common association of calcareous rocks with charnockites in this area would suggest that CO₂ given-off by metamorphic decarbonation reactions could have operated at least on outcrop level. Charnockites also occur in the absence of associated calcareous rocks in the vast granulite terrane from Ongul through Skallen to Botnnest. However, not all localities associate calcareous rocks, requiring an external source for CO_2 . Our ongoing studies will further resolve some of these aspects.

Acknowledgments

MS thanks INOUE Foundation for Science and the Japan Society for Promotion of Science for supporting his research in Japan, and the Director, Centre for Earth Science Studies for encouragement. Laboratory facilities for the fluid inclusion microthermometric work were kindly provided by Dr. M. SASADA of the Geological Survey of Japan. Prof. A. MASUDA and Dr. K. TAKAHASHI (RIKEN) provided all encouragement for Raman microprobe work, and Dr. H. KAGI (University of Tokyo) rendered valuable analytical assistance. It is indeed a pleasure to thank Prof. Y. YOSHIDA, Dr. Y. MOTO-YOSHI and Dr. M. FUNAKI of the National Institute of Polar Research for organizing a wonderful Symposium and for the opportunity to present our results. Useful comments from two referees aided in improving the manuscript.

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(Received March 15, 1991; Revised manuscript received May 23, 1991)