極東ネパールヒマラヤ苦鉄質グラニュライトの低い流体 H₂O 活動度(予報) Low H₂O activity of fluid in mafic granulite in far-eastern Nepal (forecast)

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Abstract

The P-T-X_{H2O} condition of mafic granulite in far-eastern Nepal has been investigated using multi-equilibrium thermobarometry. Based on retrograde zoning in amphiboles and the amphibole + plagioclase symplectite surrounding garnet and clinopyroxene, the mafic granulite has been overprinted by amphibolite facies metamorphism. The thermobarometric result indicates the presence of low H₂O activity fluids with high CO₂ under granulite facies metamorphic condition. The inferred peak P-T condition is P= c. 8.4 ± 0.2 kbar and T= c. 750 ± 10 °C at $X_{H2O}=$ 0.3. This study suggests that it is necessary to know about the precise fluid activity of water when inferring the P-T condition of metabasites.

Keywords: Nepal Himalaya; low H₂O activity in fluid; mafic granulite; retrograde reaction

1. Introduction

Dynamics of fluids is attracting much attention, because fluids affect a fundamental control on crustal processes as well as metamorphic P-T condition (e.g. Spear, 1993; Yardley, 2009; Thompson, 2010). For example, the addition of fluids decreases in the temperatures of dehydration and melting reactions of rocks, and facilitates growth of hydrous phases at the upper-middle crustal levels (e.g. Ferry and Gerdes, 1998; Thompson, 2010). Their effects are not well known quantitatively, and so constitute a major uncertainty in estimating of metamorphic P-T conditions at prograde or peak-T stages and interpreting the effect on retrograde reaction on cooling.

The Himalayan orogen formed along the northern margin of the Indian continent as a result of the India–Asia collision since c. 45-55 Ma (e.g. Rowley, 1996; Yin and Harrison, 2000; Zhu et al., 2005). The High Himalayan Crystalline Sequence (HHCS) has partly experienced eclogite facies metamorphism, and the subduction-related ultra-high pressure (UHP) and high pressure (HP) rocks have been found in the western and eastern Himalayas (e.g. Liu and Zhang, 1997; de Sigoyer et al., 2000; Kaneko et al., 2003; Parrish et al., 2006; Zhang et al., 2010). Retrogressed eclogites overprinted by amphibolite-granulite facies metamorphism, which are mostly recorded by amphibole zoning and omphacite replaced by clinopyroxene + plagioclase symplectite, have been reported in the Arun Valley of eastern Nepal (e.g. Lombardo and Rolfo, 2000; Groppo et al., 2007) and in Bhutan (e.g. Chakungal et al., 2010). Ferrando et al. (2007) has indicated that the fluid compositions in the granulized eclogite change from salinity aqueous fluid with CO₂ during

early eclogite metamorphism, through CO₂-rich fluid during granulite metamorphism, to aqueous fluid with minor CO₂ during amphibolite facies metamorphism. Hence, the P-T- X_{H2O} evolution of metabasites in the HHCS is a key to understand the exhumation process of deep crustal rocks in the Himalaya orogen.

In this study, the amphibole zonings of retrogressed mafic granulite in the far-eastern Nepal Himalaya, where spatial distributions of P-T conditions (Imayama et al., 2010) and metamorphic ages (Imayama et al., 2012) have been well documented, were investigated. The $P-T-X_{H2O}$ condition of mafic granulite is estimated by multi-equilibrium thermobarometry using TWQ program (Berman, 1991), which is inversely calculated from the compositions of equilibrium mineral assemblages in nature. And then, it is discussed about the origin of low H₂O fluid activities and the effect on retrograde reaction.



Fig. 1. Geological map along the Tamor-Ghunsa transect of far-eastern Nepal, showing mineral isograds and location of mafic granulite used for chemical and P-T analyses. Modified after Schelling (1992), Goscombe et al. (2006), and Imayama et al. (2010, 2012).

2. Metamorphism in the Himalaya

The subduction-related UHP and HP metamorphisms are mainly recognized at the leading margin of the Indian plate. In the western Himalaya syntaxis, the UHP metamorphism from coesite-bearing eclogite has been dated at c. 50-46 Ma, based on U-Pb zircon and allanite ages in the Kaghan Valley of Pakistan (Kaneko et al., 2003; Parrish et al., 2006), and at c. 54 Ma, based on a Lu-Hf and Sm-Nd garnet isochron ages in the Tso Morari dome in the Ladakh region of India (de Sigoyer et al., 2000). In the eastern Himalayan syntaxis, based on zircon U-Pb ages, Liu and Zhang (1997) and Zhang et al. (2010) reported the high-pressure granulite-facies metamorphic event in the HHCS occurred at T=c. 750–850 °C and P=c. 14–18 kbar at c. 37-32 Ma, which had been related to the northward subduction of the Indian continent beneath south Tibet. The granulized eclogites that were produced during Early Miocene occur in the Arun Valley of eastern Nepal

(e.g. Cottle et al., 2009; Corrie et al., 2010) and in Bhutan (e.g. Chakungal et al., 2010; Grujic et al., 2011). Although far-eastern Nepal (study area) is located only at approximately 50 km east of the Arun Valley area, both the UHP and HP rocks are previously unreported.

In contrast to the UHP and HP rocks, the intermediate P/T-type metamorphism (kyanite zone) in the HHCS has regionally been recognized in the Himalaya as the Eohimalayan stage in the Late Eocene–Oligocene (c. 38–32 Ma: Vannay and Hodges, 1996; Simpson et al., 2000; Godin et al., 2001). After this Barrovian-type metamorphism, high-T (mainly sillimanite zone) at medium- to low-pressure metamorphism caused the widespread anatexis in the Early–Middle Miocene, Neohimalayan stage at c. 26–18 Ma (e.g. Simpson et al., 2000; Daniel et al., 2003; Viskupic et al., 2005). However, Imayama et al. (2012) recently indicated that the Eohimalayan event during the Early Oligocene was not only characterized by kyanite grade metamorphism in the HHCS, but also by Early Oligocene anatectic event that was recorded in the middle HHCS



Fig. 2. Photomicrographs of mafic granulite H2608 along the Tamor-Ghunsa section in far-eastern Nepal. (a) Garnet porphyroblasts with inclusions of clinopyroxene, amphibole and ilmenite. (b) Symplectite of amphibole + plagioclase surrounding garnet porphyroblast.

3. Petrography

Mafic granulite H2608 along the Tamor-Ghunsa section in far-eastern Nepal was collected for chemical and P-T analyses (Fig. 1). Mafic granulite H2608 constitutes the middle part of the HHCS, and was intercalated within the sillimanite migmatites in kyanite/sillimanite-K-feldspar zone above muscovite-out isograd. The host sillimanite migmatites have the retrograde path from P=c. 7–10 kbar at T=c. 730–780 °C at c. 33-28 Ma to P=c. 4-7 kbar at T=c. 650-725 °C at c. 27-23 Ma determined by P-T pseudosection and zircon U-Pb ages (Imayama et al., 2012). Mafic granulite is intruded by muscovite-biotite leucogranite dyke. Zircons from the muscovite-biotite leucogranite are two successively overgrown rims with ages of 18.3 ± 0.3 Ma associated with the anatectic melting, and 16.3 ± 0.2 Ma with melt crystallization on cooling (Imayama et al., 2012). Sample H2608 mainly contains garnet, amphibole, clinopyroxene, ilmenite, plagioclase, and quartz. Biotite and calcite are present in minor amount. On the basis of microstructural observations, the metamorphic history is divided into three stages. The peak-T granulite-facies assemblage (garnet + clinopyroxene + amphibole + biotite + plagioclase + quartz + ilmenite) is composed of garnet and clinopyroxene porphyroblasts and matrix minerals. A weak foliation is defined by plagioclase and amphibole. Garnets are large porphyroblast up to 1 cm in length, and include clinopyroxene, amphibole, plagioclase, and ilmenite (Fig. 2a), indicating that their minerals are stable during garnet growth. Second metamorphic stage is characterized by symplectic amphibole + plagioclase surrounding garnet porphyroblasts (Fig. 2b), representing the later amphibolite facies metamorphism. Clinopyroxene is partly replaced by symplectic corona of amphibole + plagioclase around its margin. These retrograde microstructures from granulite to amphibolite facies metamorphism are probably developed according to the following reaction: garnet + clinopyroxene + fluid \rightarrow amphibole + plagioclase symplectite (or amphibole + plagioclase in matrix rims). Amphibole + ilmenite symplectite has also formed at the rim of garnet, and is associated with the growth of retrograde amphibole in the matrix rims. The production of calcite along the fracture of plagioclase is interpreted to reflect the latest metamorphic event at low temperature conditions.



Fig. 3. Amphibole compositions plotted in (a) $Mg/(Mg + Fe^{2+})$ -Si diagram (b) Ti-Al^{IV} diagrams for mafic granulite in far-eastern Nepal. Classification of Ca-amphiboles is based on Leake et al. (1997). Data of different amphibole domains are plotted as shown in legends.

4. Mineral chemistry

Mineral compositions were analyzed using Japanese Electron Optics Laboratory (JEOL) Superprobe 733 electronprobe microanalyser (EPMA) at Hokkaido University. The operating conditions are 15 kV accelerating voltage, and 12 nA beam current. Natural and synthetic silicates and oxides are used as standards. Conventional ZAF method is employed for matrix correction. Amphibole analyses were calculated on the basis of 23 oxygens assuming all Fe as Fe^{2+} , and then Fe^{3+} was recalculated assuming total cation = 13 excluding Ca, Na, and K (Triboulet, 1992: 13 eCNK method).

Amphiboles in mafic granulite H2608 are brown to dark green in color. Amphiboles are calcic hornblends with (Na+K) in A site ranging between 0.27-0.46 pfu. Compositional zoning in amphibole is observed, and tschermakitic hornblende core is rimmed by magnesio-hornblend (Fig. 3a). Amphiboles included in garnet show the similar composition as in the matrix core (Fig. 3a). From core to rim, Ti and Al^{IV} values decrease (Fig. 3b). Because high Ti and Al^{IV} values in calcic amphibole are generally considered as reliable indicator of high temperatures (e.g. Raase, 1974; Zhenk and Schlz, 2004), the compositional zoning of amphiboles in

mafic granulite is interpreted to be formed at retrograde stage. The changes from core to rim in Na (M4 site) and Al^{VI} values, as indicator of pressure (e.g. Brown, 1977), are much less obvious. Garnets are enriched in almandine and grossular ($X_{Alm} = 0.53-0.54$, $X_{Gls} = 0.27-0.28$) with lesser amounts of pyrope ($X_{Pyp} = 0.15-0.17$). Chemical zoning patterns in Mn, Ca, Mg, and Fe of garnets are rarely observed from core to rim, and the crystals reflect growth zonation modified to some extent by diffusion. Calcic clinopyroxene occur in garnet and matrix. Both clinopyroxenes are augite with Mg/(Mg + Fe) ratios of 0.65-0.66 in matrix and 0.61-0.63 in garnet. Na₂O (0.09-0.29 wt. %) contents of their clinopyroxenes are generally low. Plagioclase is bytownite, and the anorthite content increases $X_{An} = 0.72-0.73$ in cores to $X_{An} = 0.76-0.78$ in rims.



Fig. 4. TWQ results for mafic granulite H2608 calculated (a) at X_{H20} = 1.0 and (b) at X_{H20} = 0.3. Numbered equilibrium are listed in lower parts of figures. The following solid solution models are used to calculate multi-equilibrium thermobarometry; garnet (Berman, 1991), amphibole (Mader et al., 1994), plagioclase (Furman and Lindsley, 1988), clinopyroxene and mixed H₂O-CO₂ fluid (Berman, 1988). Alm; almandine, Py; pyrope, Gr; grossular, Di; diopside, Hd; hedenbergite, Tsc; tschermakite, An; anorthite.

4. TWQ program

In order to estimate P-T conditions at peak-T, multi-equilibrium is computed using the TWQ program (Berman, 1991) with internally consistent themodynamic data (Berman, 1988). Only P-T estimates for mafic granulite (H2608) with low-variance assemblages can be calculated by the method. The average compositions of cores in matrix minerals (garnet, clinopyroxene, amphibole, and plagioclase) are used for

the calculation. If the sample is well equilibrated and used compositional data are good, all curves for mineral equilibria should intersect. Equilibria for sample H2608 is given in Fig. 4, which are calculated in the SiO₂-FeO-MgO-CaO-Al₂O₃-H₂O-(CO₂) system with the following end members: almandine, pyrope, grossular, diopside, hedenbergite, tschermakite, quartz, anorthite, and mixed H₂O-CO₂ fluid. Only three equilibria are independent of all 14 reactions in the diagram. When the activity of water = 1.0 in mixed H₂O-CO₂ fluid is assumed (i.e. X_{H2O} = 1.0), equilibrium curves (2) and (4) for traditional garnet-clinopyroxene-plagioclase-quartz barometry and garnet-clinopyroxene thermometry are not well converged with the other 12 curves at one *P*-*T* point (Fig. 7a). In contrast, when the proportion of H₂O and CO₂ in fluid changes to 3:7 (i.e. X_{H2O} = 0.3), all 14 reaction curves are *P*=*c*. 8.4 ±0.2 kbar and *T*=*c*. 750 ±10 °C at X_{H2O} = 0.3, in consistent with those (*P*=*c*. 7–10 kbar and *T*=*c*. 730–780 °C) at peak-*T* of the host sillimanite migmatite (Imayama et al., 2010, 2012).

5. Discussion: Source and evolution of the mixed H2O-CO2 fluid in mafic granulite

Based on fluid inclusion analyses, CO_2 -rich fluids were previously recognized in the granulized eclogite in eastern Nepal (e.g. Ferrando et al., 2007) and in the high pressure granulite in eastern Himalaya syntaxis (e.g. Shen et al., 2010). Whereas fluid inclusion analyses may have been subjected to post-trapping modifications and re-equilibration (e.g. Ferry and Gerdes, 1998), the fluid composition inferred from the natural mineral compositions using TWQ analyses is an independent evidence of CO_2 -rich fluid during granulite facies metamorphism in mafic granulite in the HHCS.

Ferrando et al. (2007) suggests that the CO_2 -rich fluids in the granulized eclogite are probably attributed to the preferential incorporation of H_2O into the melts produced during anatexis of hosting augen gneiss. However, the solubility of CO_2 in the silicate melt is strongly reduced at low pressures, in contrast to that of H_2O (e.g. Lowenstern, 2001), and the degree of partial melting in mafic granulite H2608 itself is minor. Marbles or calc-silicate rocks, which could be the source of CO_2 influx from metamorphic reactions, are also very minor in the middle part of the HHCS along the Tamor-Ghunsa section (Goscombe et al., 2006; Imayama et al., 2010). Therefore, the incorporation of H_2O in anatectic melts and CO_2 infiltration through decarbonation reactions are not likely as the process to produce CO_2 -rich fluid in mafic granulite in far-castern Nepal.

According to Chakungal et al. (2010), mafic rocks in the Bhutan Himalaya were most likely derived from a subcontinental lithospheric mantle source and were later modified by crustal contamination, based on negative Nb and Ti anomalies and an enrichment of Th and light REE. Carbon dioxides are common in basaltic magmas derived from mantle (e.g. Goldberg et al., 2008), and so we interpreted the carbon dioxides component in fluids in mafic granulite is the remnant of original magmatic fluid. In the middle crust, the activity of water in an aqueous fluid could be internally buffered to various values by mixing during metamorphism between the original CO_2 -rich magmatic and the later H_2O -rich metamorphic fluids.

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including pseudosection modeling, will be published in the Mineralogy and Petrology or one of the other international journals in near future.

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