

STABILITY OF OSUMILITE IN GRANULITE FACIES
METAMORPHISM: KMAS SYSTEM AND
NATURAL OCCURRENCES

Makoto ARIMA¹, Yoichi MOTOYOSHI² and Kazuyuki SHIRAISHI²

¹Geological Institute, Yokohama National University, Hodogaya-ku, Yokohama 240

²National Institute of Polar Research, 9-10, Kaga 1-chome, Itabashi-ku, Tokyo, 173

Abstract: The stability relation of osumilite in granulite facies rocks is evaluated in the light of the recent study of osumilite-bearing granulites in the Paradise River area, Labrador, Canada (M. ARIMA and C. F. GOWER: Geol./Mineral. Assoc. Canada, Program with Abst., 10, 1987; to be publ. in J. Petrol., 1990). The mineral parageneses in the Paradise River granulites indicate that osumilite is stable at temperatures lower than those required for sapphirine-quartz stability. The *P-T* grid deduced from the Paradise River occurrence is in accordance with that proposed by E.S. GREW (Am. Mineral., 67, 762, 1982) for osumilite-sapphirine-quartz granulites in the Napier terrane, Antarctica. This conclusion is supported by high pressure experimental results in the KMAS system which suggest that osumilite is stable at temperatures lower than the univariant reaction $En + Sil = Sa + Q$.

1. Introduction

Osumilite $[K(Mg, Fe)_2(Al, Fe, Mg)_3(Sil, Al)_{12}O_{30}]$, first described by MIYASHIRO (1956) from volcanic rocks in Sakkabira, Osumi Peninsula, Japan, is a rare K-bearing ferromagnesian alumina silicate occurring in both igneous and metamorphic rocks. Since metamorphic osumilite was described by BERG and WHEELER (1976) from granulite facies metamorphic rocks in Nain, Labrador, Canada, osumilite-bearing granulites have been reported from four other localities worldwide such as: Napier, Antarctica (ELLIS, 1980; ELLIS *et al.*, 1980; GREW, 1982; SHERATON *et al.*, 1987), Vahugdzhur, Aldan Shield, Eastern Siberia (BOGDANOVA *et al.*, 1981), Rogaland, Norway (MAIJER *et al.*, 1977; JANSEN *et al.*, 1985; TOBI *et al.*, 1985), and Paradise River, Eastern Labrador, Canada (ARIMA and GOWER, 1987, 1990). Of these occurrences, Napier is the only regional granulite terrane where osumilites has been reported. Other four localities are granulite facies contact aureoles of anorthositic complexes. Osumilite, however, is probably a common and important mineral in regional granulite facies metamorphism, as its stable existence is suggested in numbers of granulite terranes by the presence of symplectic intergrowths diagnostic of osumilite breakdown products. These terranes are Wilson Lake, Canada (GREW, 1982), Labwor, Uganda (SANDIFORD *et al.*, 1987), Chogar, Aldan Shield, USSR (PERCHUK *et al.*, 1985), and Eastern Ghats, India (GREW, 1982; LAL *et al.*, 1987). It is noteworthy that a high temperature mineral assemblage of sapphirine-quartz occurs in all of the terranes.

In this paper the stability of osumilite in granulite facies pelitic rocks is examined in the light of the recent study of the mineral and chemographic relationships in osumilite-bearing granulites in the Paradise River area, Labrador, Canada (ARIMA and GOWER, 1987, 1990). We also report preliminary results of experimental works on the stability of osumilite at high temperatures and pressures in the K_2O - MgO - Al_2O_3 - SiO_2 (KMAS) system.

2. Previous Experimental Studies on the Stability of Osumilite

SCHREYER and SEIFERT (1967) synthesized osumilite at 1 kbar between 500–900°C and suggested that osumilite may be a metastable phase. The experiments by HENSEN (1977), however, showed that osumilite is probably stable with cordierite, orthopyroxene, feldspar and liquid at 1000–1100°C at 3.8–7.2 kbar. OLESCH and SEIFERT (1981) investigated the stability of osumilite in the KMAS- H_2O , suggesting that in the hydrous system the stability field of osumilite is limited to water pressure below 800 bar. These experimental studies collectively suggest that osumilite can be stable at P - T conditions comparable to those of granulite metamorphism under relatively low water fugacities, but no systematic experimental study has been done to establish the P - T stability relation of osumilite under the granulite facies metamorphic conditions.

3. Phase Relationships Involving Osumilite in Napier

The osumilite-bearing primary assemblages in the Napier terrane can be summarized as: $Os + Sap + Q \pm Opx \pm Gt \pm Sp$, $Os + Opx + Sil + Q \pm Gt$, $Os + Sap + Sil + Q \pm Gt$, $Os + Sap + Opx + Sil + Q$, $Os + Sap + Opx + Cd + Q$, and $Os + Opx + Cd + Q$ (ELLIS *et al.*, 1980; GREW, 1982, SHERATON *et al.*, 1987). ELLIS (1980) estimated peak metamorphism in the Tula Mountains at 8–10 kbar and 900–980°C. GREW (1980) estimated 900°+30°C and 7+1 kbar for the same area. In the western part of the Napier terrane, HARLEY (1983, 1985) estimated 900–950°C and 7–10 kbar. Such conditions would approximate the P - T stability of osumilite in pelitic compositions. GREW (1982) estimated the stability of osumilite with quartz in granulite facies pelitic rocks to be at temperatures above 750°C, lithostatic pressure not exceeding 8–9 kbar, and at water pressure not exceeding 1 kbar.

Based on the mineral and chemographic relationships in granulites in Napier, ELLIS *et al.* (1980) and GREW (1982) independently constructed P - T grids for the osumilite and sapphirine-quartz. In the grids formulated by GREW (1982), which involves osumilite, sapphirine, orthopyroxene, cordierite, garnet, phlogopite, K-feldspar, sillimanite, and quartz, an assemblage osumilite-quartz is stable at temperatures lower than those for sapphirine-quartz, unlike the grids proposed by ELLIS *et al.* (1980). A progressive P - T path has not been well defined in the Napier terrane (*cf.* SHERATON *et al.*, 1987). Without knowing the independently estimated temperature and pressure gradients for the area where osumilite- and sapphirine-quartz occur, it seems difficult to define the stability relationships between osumilite-bearing and sapphirine-quartz assemblages.

4. Osumilite in the Paradise River Area, Labrador

A progressive *P-T* path has been delineated for osumilite-bearing granulite facies metasedimentary gneisses in the Paradise River area, eastern Grenville Province, Labrador, Canada (ARIMA and GOWER, 1987, 1990). The osumilite-bearing gneiss occurs in the contact aureole of the Proterozoic gabbro-norite complex. The critical mineral assemblage in the pelitic gneiss varies, toward the intrusive contact, from assemblages containing muscovite + phlogopite + quartz, through those with phlogopite + sillimanite + quartz, to those with cordierite + K-feldspar + quartz, and finally to those with osumilite and sapphirine. An osumilite-in isograd is delineated at a distance of 1–3 km from the intrusive contact. The osumilite occurs in association with cordierite, orthopyroxene, sillimanite, sapphirine, spinel, K-feldspar, plagioclase, phlogopite, hematite, magnetite, corundum, and quartz. The stable osumilite-bearing assemblages deduced from petrographic features and from the phase relations in the KMAS system are $Os + Sa + Cd$, $Os + Sil + Cd + Q$, $Os + Cd + Kf + Q$, $Os + Cd + Opx + Q$, $Qs + Opx + Kf + Q$. Symplectic intergrowth of cordierite-orthopyroxene-K-feldspar-quartz is ubiquitously present which partly or completely replaced osumilite. The sapphirine-bearing assemblages are restricted to silica-deficient (quartz-absent) zones of the gneiss. This suggests that the assemblage Sa-Q is not stable under the peak metamorphic conditions in the Paradise River granulites.

5. *P-T* Grid for Osumilite-Bearing Assemblages in the KMAS System

A *P-T* grid involving sapphirine, enstatite, cordierite, sillimanite, spinel, corundum, and quartz in the $MgO-Al_2O_3-SiO_2$ (MAS) system is depicted in Fig. 1. The grid is constructed on the basis of the experimental data by SCHREYER and SEIFERT (1969), ACKERMAND (1970), CHATTERJEE and SCHREYER (1972), NEWTON (1972), NEWTON *et al.* (1974), and ARIMA and ONUMA (1977). An assemblage with sapphirine + quartz is constrained to fields of higher temperatures than univariant reactions $En + Sil = Sa + Q$ and $Cd + Sil = Sa + Q$. The *P-T* position of the invariant point I(1) is a subject of debate. NEWTON *et al.* (1974) located the invariant point I(1) at 950°C and 7.5 kbar for the synthetic system and at 825°C and 6.7 kbar for the natural system. HENSEN (1987), however, estimated the *P-T* position of the invariant point I(1) at 950° + 50°C and 10 + 1 kbar, based on the experimental data by ACKERMAND (1970), NEWTON (1972), and SEIFERT (1974). Recent experimental data by BERTRAND *et al.* (1989) suggest that Mg-cordierite can be stable as high as 13 kbar at 1000°C, supporting the *P-T* estimation of I(1) at 1000°C and 10 kbar by HENSEN (1987).

In the Paradise River area, sapphirine does not coexist with quartz in the osumilite-bearing gneiss, suggesting that the equilibrium conditions of the Paradise osumilite-bearing assemblages were located at temperatures lower than those required for sapphirine + quartz stability. This is in accordance with the *P-T* grid in the KMAS system proposed by GREW (1982) but not consistent with that of ELLIS *et al.* (1980). Based on the mineral assemblages in the Paradise River area, ARIMA and GOWER (1990) extended the grid proposed by GREW (1982) to quartz-absent assemblages (Fig. 2). In the quartz-absent system, the univariant reaction $En + Sil + Kf = Os + Sa$

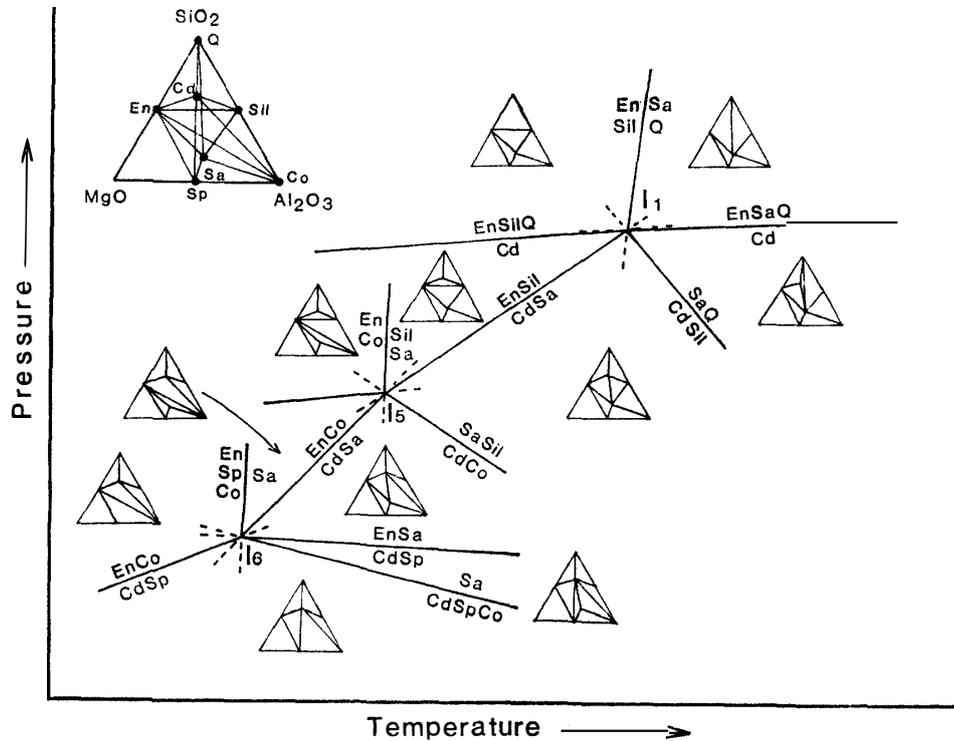


Fig. 1. *P-T* grid showing phase relations in MAS system. The univariant reactions $En + Sil = Sa + Q$ and $Cd + Sil = Sa + Q$ define the lower temperature stability limit of the assemblage sapphirine + quartz. HENSEN (1987) estimated *P-T* position of the invariant point I(1) at about 900–1000°C and 9–10 kbar.

intersects the univariant reaction $En + Sil = Sa + Cd$ at the invariant point I(4) (Fig. 2). This suggests that osumilite can coexist with sapphirine in quartz absent compositions at higher temperatures than univariant lines $En + Sil + Kf = Os + Sa$ and/or $En + Cd + Kf = Os + Sa$.

A series of high temperature and pressure experiments have been done with a piston cylinder apparatus at Yokohama National University to define *P-T* positions of the univariant reaction $Os = En + Sil + Kf + Q$ in the KMAS system. A crystal mixture ($Os + En + Kf + Sil + Q$) was used as a starting material. Run products were examined by XRD, and intensities of X-ray reflections of osumilite and other phases in the run products were compared with those of the starting mixture. The results suggest that osumilite is stable at 1100°C and 12 kbar, 900°C and 11 kbar, but breaks down to the assemblage $En + Sil + Kf + Q$ at 1100°C 13 kbar, 900°C and 12 kbar. The univariant reaction $En + Sil = Sa + Q$ (Figs. 1 and 2) is located at 1000°C and 10 kbar and 1100°C and 14 kbar (CHATTERJEE and SCHREYER, 1972; ARIMA and ONUMA, 1977). The present results suggest that osumilite is probably stable at temperatures lower than the univariant reaction $En + Sil = Sa + Q$, being in accordance with the *P-T* grid depicted in Fig. 2.

- and temperatures in a model pelitic system. IGCP-235 Metamorphism and Geodynamics, Granulite Metamorphism, Program and Abstract, 9.
- BOGDANOVA, N. G., TRONEVA, N. Z., ZABOROVSKAYA, N. B., SUKHANOV, M. K. and BERKIN, S. I. (1981): The first find of metamorphic osumilite in the USSR. *Dokl. Akad. Nauk SSSR*, **250**, 690–693.
- CHATTERJEE, N. D. and SCHREYER, W. (1972): The reaction $\text{enstatite}_{88} + \text{sillimanite} \rightleftharpoons \text{sapphirine}_{88} + \text{quartz}$ in the system $\text{MgO-Al}_2\text{O}_3\text{-SiO}_2$. *Contrib. Mineral. Petrol.*, **36**, 49–62.
- ELLIS, D. J. (1980): Osumilite-sapphirine-quartz granulites from Enderby Land, Antarctica; P-T conditions of metamorphism, implications for garnet-cordierite equilibria and the evolution of the deep crust. *Contrib. Mineral. Petrol.*, **74**, 201–210.
- ELLIS, D. J., SHERATON, J. W., ENGLAND, R. N. and DALLWITZ, W. B. (1980): Osumilite-sapphirine-quartz granulites from Enderby Land, Antarctica—Mineral assemblages and reactions. *Contrib. Mineral. Petrol.*, **72**, 123–143.
- GREW, E. S. (1980): Sapphirine-quartz association from Archean rocks in Enderby Land, Antarctica. *Am. Mineral.*, **65**, 821–836.
- GREW, E. S. (1982): Osumilite in the sapphirine-quartz terrane of Enderby Land, Antarctica; Implications for osumilite petrogenesis in the granulite facies. *Am. Mineral.*, **67**, 726–787.
- HARLEY, S. L. (1983): Regional geobarometry-geothermometry and metamorphic evolution of Enderby Land, Antarctica. *Antarctic Earth Science*, ed. by R. L. OLIVER *et al.* Canberra, Aust. Acad. Sci., 25–30.
- HARLEY, S. L. (1985): Garnet-orthopyroxene bearing granulites from Enderby Land, Antarctica; Metamorphic pressure-temperature-time evolution of the Archean Napier Complex. *J. Petrol.*, **26**, 819–856.
- HENSEN, B. J. (1977): The stability of osumilite in high grade metamorphic rocks. *Contrib. Mineral. Petrol.*, **64**, 197–204.
- HENSEN, B. J. (1987): P-T grids for silica-undersaturated granulites in the system $\text{MAS}(n+4)$ and $\text{FNAS}(n+3)$ -tools for the derivation of *P-T* paths of metamorphism. *J. Metamorph. Geol.*, **5**, 255–271.
- JANSEN, J. B. H., BLOK, R. J. P., BOS, A. and SCHEELINGS, M. (1985): Geothermometry and geobarometry in Rogaland and preliminary results from the Bamble area, S. Norway. *The Deep Proterozoic Crust in the North Atlantic Provinces*, ed. by A. C. TOBI and J. L. R. TOURET. Dordrecht, D. Reidel, 499–516 (NATO Advanced Science Institutes, Ser. C, 158).
- LAL, R. K., ACKERMAN, D. and UPADHYAY, H. (1987): P-T-X relationships deduced from corona textures in sapphirine-spinel-quartz assemblages from Paderu, southern India. *J. Petrol.*, **28**, 1139–1168.
- MAIJER, C., JANSEN, J. B., WEEVERS, J. and POORTER, R. P. E. (1977): Osumilite, a mineral new to Norway. *Nor. Geol. Tidsskr.*, **57**, 187–188.
- MIYASHIRO, A. (1956): Osumilite, a new mineral and its crystal structure. *Am. Mineral.*, **41**, 104–116.
- NEWTON, R. C. (1972): An experimental determination of the high-pressure stability limits of magnesium cordierite under wet and dry conditions. *J. Geol.*, **80**, 398–420.
- NEWTON, T. C., CHARLU, T. V. and KLEPPA, O. J. (1974): A calorimetric investigation of the stability of anhydrous cordierite with application to granulite facies metamorphism. *Contrib. Mineral. Petrol.*, **44**, 295–311.
- OLESCH, M. and SEIFERT, F. (1981): The restricted stability of osumilite under hydrous conditions in the system $\text{K}_2\text{O-MgO-Al}_2\text{O}_3\text{-SiO}_2\text{-H}_2\text{O}$. *Contrib. Mineral. Petrol.*, **76**, 362–367.
- PERCHUK, L. L., ARANOVICH, L. Ya., PODLESSKII, K. K., LAVRANT'eva, I. V., GERASIMOV, V. Yu. and FED'KIN, V. V. (1985): Precambrian granulites of the Aldan shield, eastern Siberia, USSR. *J. Metamorph. Geol.*, **3**, 265–310.
- SANDIFORD, M., NEALL, F. B. and POWELL, R. (1987): Metamorphic evolution of aluminous granulites from Labwor Hills, Uganda. *Contrib. Mineral. Petrol.*, **95**, 217–225.
- SCHREYER, W. and SEIFERT, F. (1967): Metastability of an osumilite end member in the system $\text{K}_2\text{O-MgO-Al}_2\text{O}_3\text{-SiO}_2\text{-H}_2\text{O}$ and its possible bearing on the rarity of natural osumilites. *Contrib. Mineral. Petrol.*, **14**, 242–58.
- SCHREYER, W. and SEIFERT, F. (1969): Compatibility relations of the aluminous silicates in the system $\text{MgO-Al}_2\text{O}_3\text{-SiO}_2\text{-H}_2\text{O}$ and $\text{K}_2\text{O-MgO-Al}_2\text{O}_3\text{-SiO}_2\text{-H}_2\text{O}$ at high pressure. *Am. J. Sci.*, **267**, 371–388.

- SEIFERT, F. (1974): Stability of sapphirine; A study of the aluminous part of the system $MgO-Al_2O_3-SiO_2-H_2O$. *J. Geol.*, **83**, 173–204.
- SHERATON, J. W., TINGEY, R. J., BLACK, L. P., OFFE, L. A. and ELLIS, D. J. (1987): Geology of Enderby Land and western Kemp Land, Antarctica. *BMR Bull.*, **223**, 51.
- TOBI, A. C., HERMANS, G. A. E. M., MAIJER, C. and JANSEN, J. B. H. (1985): Metamorphic zoning in the high-grade Proterozoic of Rogaland-Vest Agder SW Norway. *The Deep Proterozoic Crust in the North Atlantic Provinces*, ed by A. C. TOBI and L. R. TOURET. Dordrecht, D. Reidel, 477–497 (NATO Advanced Science Institutes, Ser. C, 158).

(Received February 28, 1990; Revised manuscript received May 8, 1990)

•