

Phlogopite in jadeitite from the Osayama serpentinite mélange, Chugoku Mountains, Southwest Japan

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Accepted on October 31, 2002

Abstract Phlogopite was found as secondary mineral in a jadeitite clast (10 × 20 cm) from the Osayama serpentinite mélange of Chugoku Mountains. Phlogopites coexist with secondary analcime, and they are characterized by high-Si, low-Al and very low-Ti contents (i.e. low-eastonite and muscovite components), resembling some hydrothermal low-*T* phlogopites. The mineral paragenesis and compositional feature of phlogopite may suggest low-*T* (< 300°C) condition for hydro-alteration of jadeitite.

Keywords: phlogopite; hydrothermal alteration; jadeitite; Osayama serpentinite mélange

INTRODUCTION

Micas are rarely found as hydrous accessory minerals in jadeitites associated with serpentinite (e.g. Tazaki and Ishiuchi, 1976; Nishiyama, 1978; Harlow, 1994). Most of them are dioctahedral white micas such as paragonite and phengite. However, trioctahedral dark mica such as phlogopite and biotite is very rare in jadeitites. Phlogopite has been reported from Guatemalan jadeitite (Harlow, 1994). The author has discovered phlogopites in a jadeitite clast from the Osayama serpentinite mélange, Chugoku Mountains. This paper describes occurrence and chemistry of phlogopite in the Osayama jadeitite.

GEOLOGIC SETTING

The Osayama serpentinite mélange in the central Chugoku Mountains (Fig. 1) contains tectonic blocks of Paleozoic lawsonite-blueschist to epidote-blueschist facies rocks (Tsuji-mori 1998). The mélange unit is tectonically underlain by Late Triassic-Jurassic schists (pumpellyite-actinolite to greenschist facies) of the Suo metamorphic belt, and is overlain by serpentinitized massive peridotite unit of the Oeyama ophiolite (Fig. 2). The mélange unit is in contact with Jurassic molasses-type sediments on the north by a high-angle fault. Late Cretaceous granitic intrusions caused thermal recrystallization of serpentinite in the western part of the area (Nozaka and Shibata 1995; Tsujimori 1998). Phengite K-Ar ages from the Renge schists in this area gave 273 to 327 Ma (Tsuji-mori and Itaya, 1999). The matrix of the serpentinite mélange consists of schistose, friable,

fine-grained serpentinite with pebble to boulder-size fragments of serpentinitized peridotite. Chrysotile and lizardite are dominant serpentine minerals; rare winchitic to tremolitic amphiboles occur as metamorphic mineral in serpentinite. The protolith of serpentinite is moderately-depleted harzburgite and minor cumulus dunite. Relict olivine (Fo_{90.5-91.5}), orthopyroxene (2.4-3.0 wt.% Al₂O₃), clinopyroxene, and chromian spinel of Cr/(Cr+Al) ratio = 0.40-0.57 occur in serpentinitized harzburgite. Rare winchitic to tremolitic amphiboles and diopside occur as metamorphic minerals in the serpentinite. Several occurrences of jadeitite and omphacite have been identified (Kobayashi et al. 1987; Tsujimori 1997).

PETROGRAPHY

The phlogopite-bearing jadeitite is leucocratic massive rock. It consists mainly of jadeite with subordinate amounts of Ti-bearing (up to 3.6 wt.% TiO₂) grossular (< 1.0 mm) and secondary omphacite as well as with minor amounts of secondary analcime and phlogopite. Pectolite and the other Na-Ca hydrous silicates are not found in this sample. Phlogopite occurs as colorless tabular crystals (< 0.1 mm) and coexists with analcime (Fig. 3-a). Jadeite occurs as subhedral to euhedral prismatic crystals (0.3-5.0 mm in length). Some jadeite (Jd_{93.1-96.8}Di_{3.2-6.9}) is rimmed by secondary omphacite (Jd_{42.5-44.3}Di_{55.7-57.5}). Grossular (Gr_{94.5-100}Adr_{<3.4}, TiO₂ = 0.9-3.6 wt.%) commonly fills grain interstices between randomly oriented prismatic jadeite. Most of omphacite occurs as radial aggregates of needle-like crystals accompanied with secondary analcime in the

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Table 1. Representative electron-microprobe analyses of phlogopite in the Osayama jadeitites. Biotite in the Osayama omphacitite (Tsuji-mori, 1997) and phlogopite in jadeitite (Harlow, 1994), rodingites (Dubinska, 1995; Wares and Martin, 1980) and hydrothermal phlogopite (Belkin et al., 1988) are also listed.

	Osayama			1	2	3	4
	jadeitite		omphacitite	jadeitite	rodingite	rodingite	hydrothermal
	Phl	Phl	Bt	Phl	Phl	Phl	Phl
SiO ₂	41.72	41.33	38.46	37.55	42.24	39.96	39.58
TiO ₂	0.07	0.05	0.41	0.17	0.00	0.00	0.26
Al ₂ O ₃	16.76	17.05	17.14	23.32	15.35	13.93	14.61
Cr ₂ O ₃	0.07	0.05	0.01	0.00	0.20	0.11	-
FeO*	6.59	6.88	15.46	4.00	5.49	11.78	2.45
MnO	0.16	0.02	0.04	0.48	0.03	0.19	0.09
MgO	20.04	20.05	14.15	19.99	21.73	18.68	25.99
CaO	0.00	0.00	0.25	0.16	0.52	0.00	0.03
Na ₂ O	0.00	0.00	0.00	0.52	0.00	0.02	0.08
K ₂ O	10.18	10.05	9.63	9.17	9.45	9.97	10.25
Total	95.60	95.48	95.54	95.36	95.01	94.64	93.34
O=	22	22	22	22	22	22	22
Si	5.910	5.866	5.705	5.273	5.978	5.904	5.954
Ti	0.008	0.005	0.045	0.018	0.000	0.000	0.030
Al	2.799	2.852	2.997	3.860	2.560	2.425	2.590
Cr	0.008	0.006	0.001	0.000	0.022	0.013	0.000
Fe*	0.781	0.817	1.917	0.470	0.650	1.455	0.308
Mn	0.019	0.002	0.005	0.057	0.004	0.024	0.012
Mg	4.233	4.242	3.130	4.185	4.585	4.114	5.828
Ca	0.000	0.000	0.039	0.024	0.079	0.000	0.005
Na	0.000	0.000	0.000	0.142	0.000	0.006	0.023
K	1.839	1.819	1.822	1.643	1.706	1.879	1.967
Total	15.598	15.610	15.662	15.671	15.584	15.820	16.717
X _{Mg}	0.84	0.84	0.62	0.90	0.88	0.74	0.95

FeO* = total Fe as FeO. X_{Mg} = Mg/(Mg+Fe)

1. Guatemalan Jadeitite, Guatemala (Harlow, 1994)
2. Asbestos rodingite, Canada (Wares and Martin, 1980)
3. Lower Silesia rodingite, Poland (Dubinska, 1995)
4. Sabatini volcanic district, Italy (Belkin, 1988); average of 44 samples

interstices (Fig. 3-b). The omphacite aggregate occurs sometimes as irregular veins (< 3.0 mm in wide) cutting prismatic jadeite. The needle-like omphacite has significantly higher jadeite component (Jd_{50.6-53.6}Di_{46.4-49.4}) than that of omphacite rim gite may be formed at relatively low-*T*, because Ca-metasomatism to form rodingite occurs during low-*T* serpentinization (e.g. Schandl et al., 1989; O'Hanley et al., 1992). On the other hand, high-Si and low-Al phlogopites have been known in garnet peridotite (e.g. Arai, 1984), but phlogopites in mantle peridotite usu-

ally contain considerable amount of Ti. Moreover, high-Si and low-Al phlogopites were described from a geothermal region in Italy (Belkin et al., 1988). These hydrothermal phlogopites have significant amount of F instead of OH (Belkin et al., 1988). Belkin et al. (1988) implied that the crystallization of the hydrothermal phlogopite could occur at temperatures from 200 to 300°C and pressures from 14 to 22 MPa from volatile-rich geothermal fluid. The high-Si, low-Al and low-Ti features of phlogopites in the investigated sample may indicate low-*T* con

overgrowth on prismatic jadeite. Secondary analcime is directly in contact not only with omphacite but also with jadeite. The textural relationship indicates that the omphacite and phlogopite are not in equilibrium with jadeite.

CHEMICAL COMPOSITION OF PHLOGOPITE

Electron microprobe analysis was carried out with an AKASHI ALPHA-30A scanning electron microscope equipped with a Philips EDAX-9100 energy-dispersive analytical system at Kanazawa University. Natural and synthetic minerals such as periclase, hematite, wollastonite, albite and chromite are used as standards, and the program based on Bence and Albee's (1968) method written by A. Ishiwatari is used for data processing. The analyses were done at 20 kV accelerating voltage, 1 nA sample current on MgO and 5 μ m probe diameter. Representative chemical analyses of phlogopite are listed in Table 1 and illustrated in Fig. 4.

Phlogopite has a composition of $Al^{VI} = 0.70-0.72$ ($Al_2O_3 = 16.3-17.4$ wt.%) and Si p.f.u. = 5.82-5.94 (O=22) with $X_{Mg} = Mg/(Mg+Fe) = 0.84-0.85$ and $TiO_2 < 0.1$ wt.%. Sodium is not detected. It is characterized by high-phlogopite [$K_2R^{2+}_6Si_6Al_2O_{20}(OH)_4$] and low-eastonite [$K_2R^{2+}_4Al_2Si_2Al_4O_{20}(OH)_4$] component and low titanium con-

tent. The phlogopite of the Osayama jade resembles biotite from omphacitite in the Osayama serpentinite mélange, except for X_{Mg} . The biotite in omphacitite has a composition of $Al^{VI} = 0.72-0.75$ ($Al_2O_3 = 17.1-17.5$ wt.%) and Si p.f.u. = 5.71 (O=22) with $X_{Mg} = 0.62$ and $TiO_2 = 0.4$ wt.% (Tsujimori, 1997).

DISCUSSION AND CONCLUSION

Phlogopites occur in some metasomatic rock associated with serpentinite and also in geothermal region. Metasomatic phlogopite was described in the Guatemalan jadeitite (Harlow, 1994). It is characterized by high Al^{IV} (= 1.13) ($Al_2O_3 = 23.3$ wt.%) and low Si p.f.u. (= 5.27). Metasomatic phlogopites have also been reported from rodingites associated with serpentinite. For instance, phlogopite occurs as halos around euhedral chromite from the rodingitized serpentinite in the Jeffrey mine, Canada (Wares and Martin, 1980). It has a composition with high-Si (Si = 5.98) and low Al ($Al^{VI} = 0.54$, $Al_2O_3 = 13.9$ wt.%). Phlogopite was described as inclusions in vesuvianite from rodingitized mafic dike in the Jardonow-Gogolow serpentinite massif, Poland (Dubinska, 1995). It shows a composition with high-Si (Si = 5.78) and low-Al ($Al^{VI} = 0.33$, $Al_2O_3 = 13.9$ wt.%). The high-Si and low-Al features of phlogopites in rodingite resemble those in the investigated sample. Phlogopites in rodingite

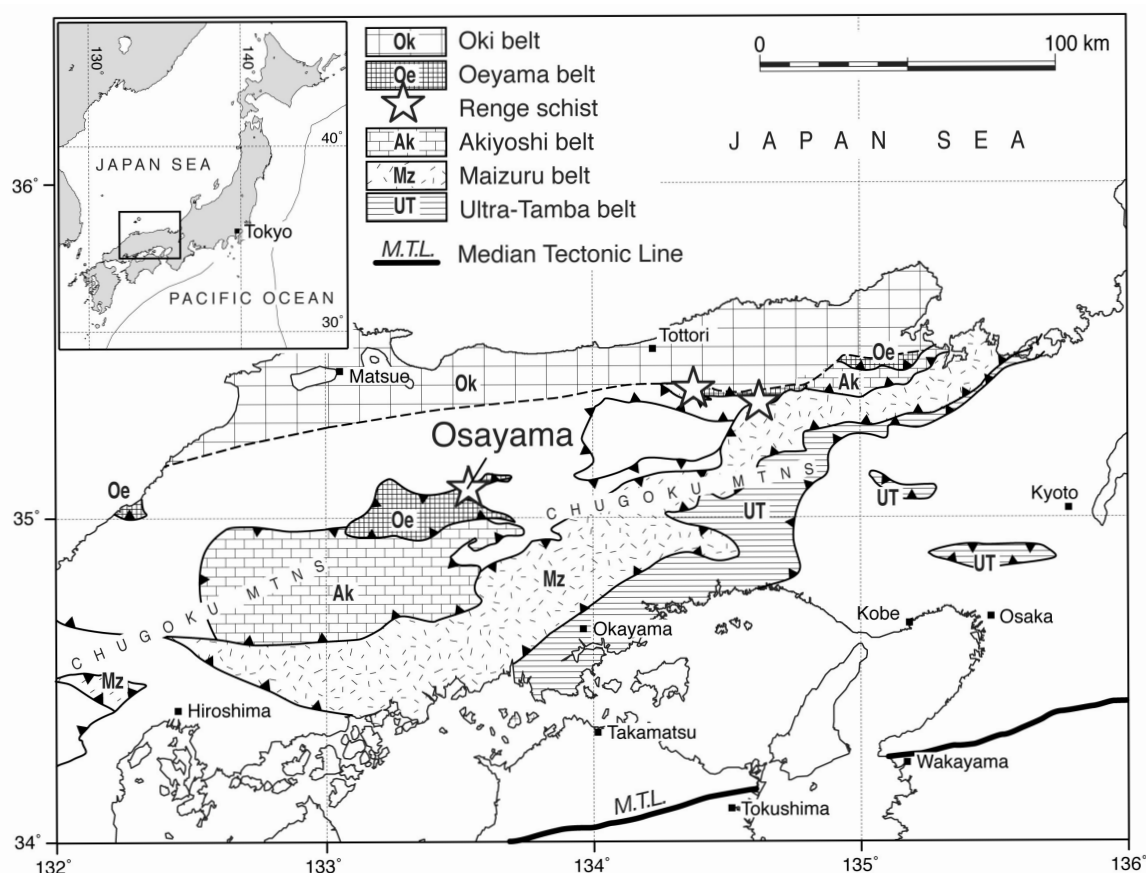


Fig. 1. Simplified map of the Chugoku Mountains, showing various petroTECTONIC units and the locality of the Osayama Mtn.

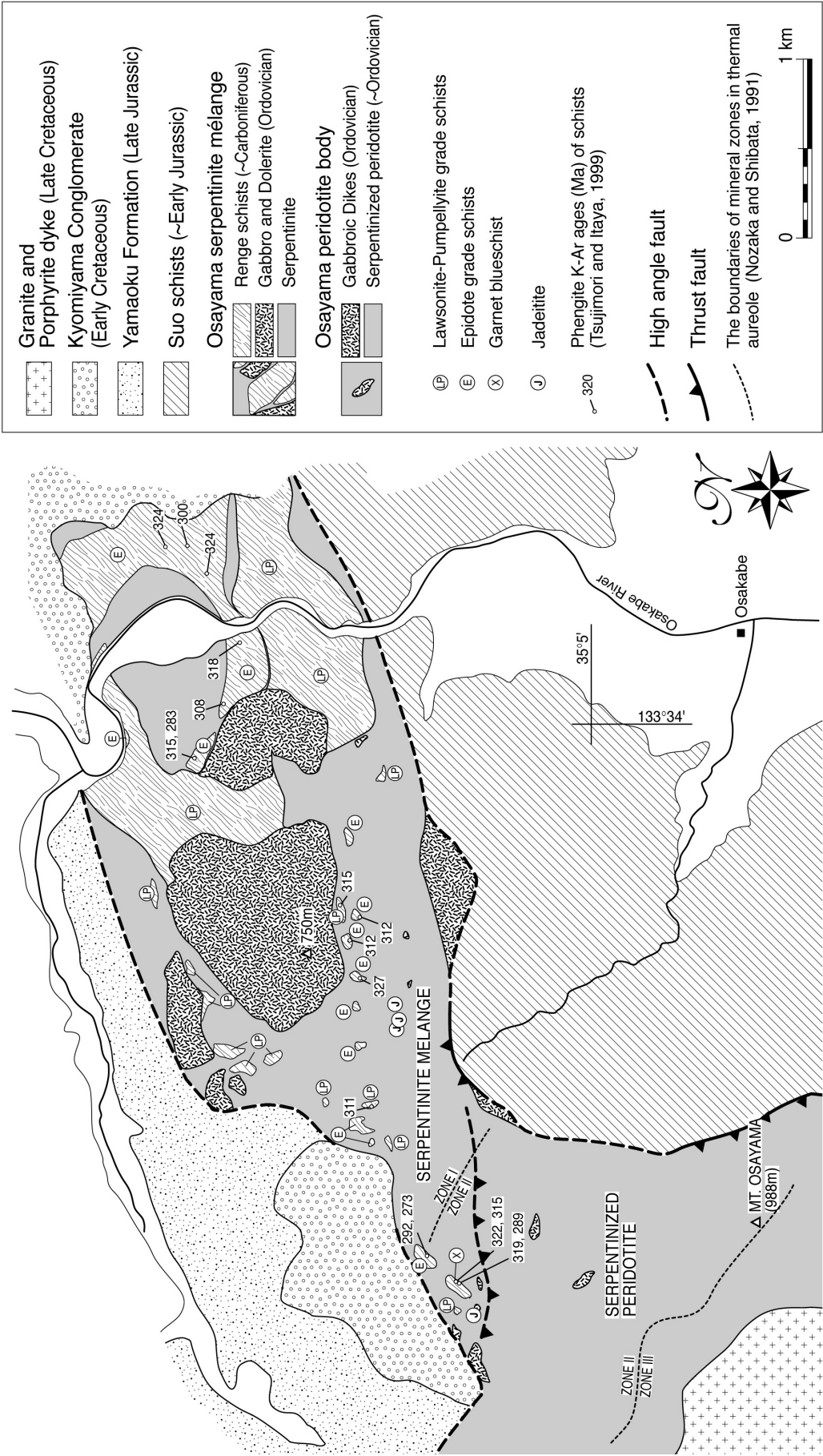


Fig. 2. Geologic map of the Osayama serpentinite mélangé (after Tsujimori, 1998).

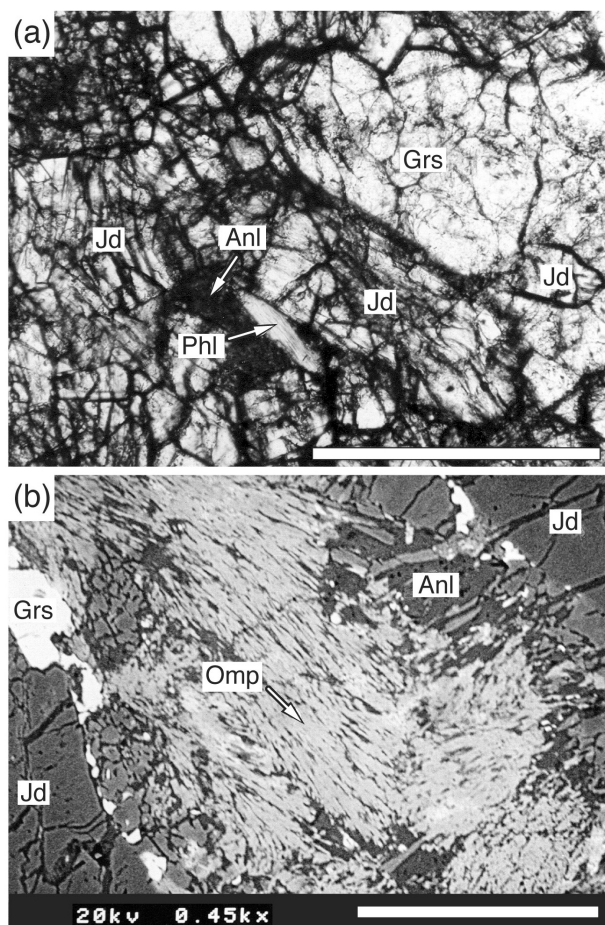


Fig. 3. Photomicrograph showing microtextures of jadeitite. (a) Open-polarizer view showing the occurrence of phlogopite. Phl = phlogopite; Jd = jadeite; Grs = grossular; Anl = analcime. Scale bar represents 0.5 mm. (b) Back-scattered electron image of needle-like omphacite and analcime. Scale bar represents 0.1 mm. Omp = omphacite.

dition for their formation.

In the investigated sample, jadeite and grossular may be primary minerals. The formation of our jadeitite requires relatively high- P and low- T condition, because jadeite is stable only at HP conditions and grossular is stable at relatively low- T under low X_{CO_2} conditions (Kerrick, 1974). All analcime in the jadeitite is interpreted to be secondary mineral related to the breakdown of jadeite. Omphacite is also regarded as a retro-grade phase. Using the thermodynamic calculation by the THERMOCALC program (ver. 3.1) (Holland and Powell, 1998; Powell et al., 1998) for the equilibrium $\text{Jd} + \text{H}_2\text{O} = \text{Anl}$, an approximate P of the breakdown of jadeite is estimated from the jadeite component assuming ideal jadeite-diopside solid solution. The omphacite (maximum Jd = 54%) - analcime assemblage gives $P = 300\text{--}550$ MPa at $T = 160\text{--}300^\circ\text{C}$ for the hydro-alteration of jadeitite. The phlogopite + analcime assemblage suggests that K^+ ions were added to form phlogopite during low- T ($< 300^\circ\text{C}$) hydro-alteration of the jadeitite.

ACKNOWLEDGEMENTS

This research was supported financially in part by JSPS Research Fellowship for Young Scientists, and Grant-in-Aid for JSPS Fellows. The author thanks to T. Itaya who provided the opportunity for publication in this bulletin. The author also extends his thanks to A. Ishiwatari for many discussions during his stay at Kanazawa University (1994-2000).

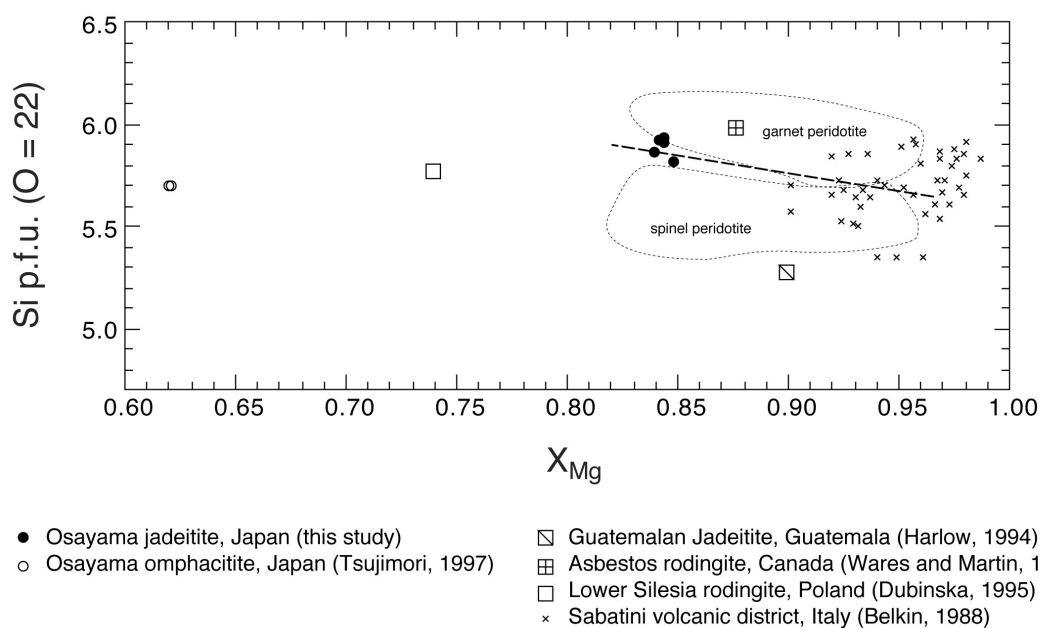


Fig. 4. The X_{Mg} versus Si p.f.u. (O=22) diagrams for phlogopites in the Osayama jadeitite. Phlogopites from Guatemalan jadeitite (Harlow, 1994), rodingite of Lower Silesia, Poland (Dubinska, 1995), rodingite of the Jeffrey mine, Canada (Wares and Martin, 1980) and hydrothermal volcanic region (Belkin et al., 1988) are also plotted. Broken line represents the boundary between garnet peridotite (upper) and spinel peridotite (lower) phlogopite fields after Arai (1984).

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