

# Omphacite-diopside vein in an omphacite block from the Osayama serpentinite melange, Sangun-Renge metamorphic belt, southwestern Japan

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## Abstract

Omphacite ( $Jd_{46.1-52.0}Ae_{0-8.4}Aug_{48.0-51.2}$ ) and diopside ( $Jd_{4.3-6.3}Ae_{0-0.4}Aug_{93.6-95.6}$ ) coexist in a vein cutting an omphacite block in a serpentinite melange of the Oeyama ophiolite, central Chugoku Mountains. The compositional gap between omphacite and diopside is significantly wider than for other omphacite-diopside pairs reported in the literature. The intergrowth texture of the omphacite-diopside vein suggests that the clinopyroxene pair was contemporaneously crystallized in the fracture from a Ca-bearing, alkali-rich fluid in a sub-solvus condition. Such a fluid may have been introduced from the surrounding serpentinitized clinopyroxene-bearing harzburgite. The stability of omphacite and Al-rich pumpellyite in the matrix and the omphacite-diopside vein indicate that the earlier matrix recrystallization and later fracture filling may have both taken place under high- $P$ - $T$  condition during the melange-forming stage.

**KEYWORDS:** omphacite, diopside, immiscibility, serpentinite melange, Oeyama ophiolite, Sangun-Renge metamorphic belt.

## Introduction

OMPHACITE is a common metamorphic mineral in blueschist and eclogite facies metamorphic rocks, and also in metasomatic sodic pyroxene-rich rocks such as jadeitite and omphacite. Coexisting omphacite-diopside pairs have been reported from high-pressure metamorphic rocks by several authors (e.g. Brown *et al.*, 1978; Carpenter, 1980a; Enami and Tokonami, 1984), and they provide some evidence of a miscibility gap in the omphacite-diopside system. Coexisting omphacite and diopside have now been found in a vein from an omphacite block in a serpentinite melange of the Oeyama ophiolite, central Chugoku Mountains. As described in this paper, the compositional gap is wider than the other omphacite-diopside pairs reported in the literature.

## Geology of the Osayama serpentinite melange

The Osayama serpentinite melange is located in the Sangun-Renge metamorphic belt of the Chugoku Mountains, in the Inner Zone of southwestern Japan. The Sangun-Renge metamorphic belt is the Carboniferous high- $P$ - $T$  type metamorphic belt,

which tectonically underlies the Early Palaeozoic Oeyama ophiolite (e.g. Ishiwatari, 1991), and may be a western counterpart of the Omi glaucophane schist (Banno, 1958) in the Hida Mountains.

The Osayama serpentinite melange is developed beneath the Osayama peridotite body of the Oeyama ophiolite in the central Chugoku Mountains, as a fragment of the Oeyama ophiolite (Fig. 1). It is a typical tectonic melange with a serpentinite matrix, which has probably been developed along a thrust fault between the Ordovician Oeyama ophiolite and Carboniferous Sangun-Renge blueschist nappe during the emplacement of the Oeyama ophiolite. The melange overlying the massive peridotite body is tectonically underlain by the 180 Ma basic schist of the Sangun-Chizu belt, which is in contact with the unmetamorphosed shallow marine sediments of the Jurassic Yamaoku Formation on the north because of a high-angle fault. All these rocks are unconformably overlain by the early Cretaceous Kyomiyama conglomerate. The massive peridotite body and the Sangun-Chizu schist are affected by contact metamorphism of the Cretaceous granitic intrusion on the west (Nozaka and Shibata, 1995). The protolith of the serpentinite matrix is harzburgite with minor dunite

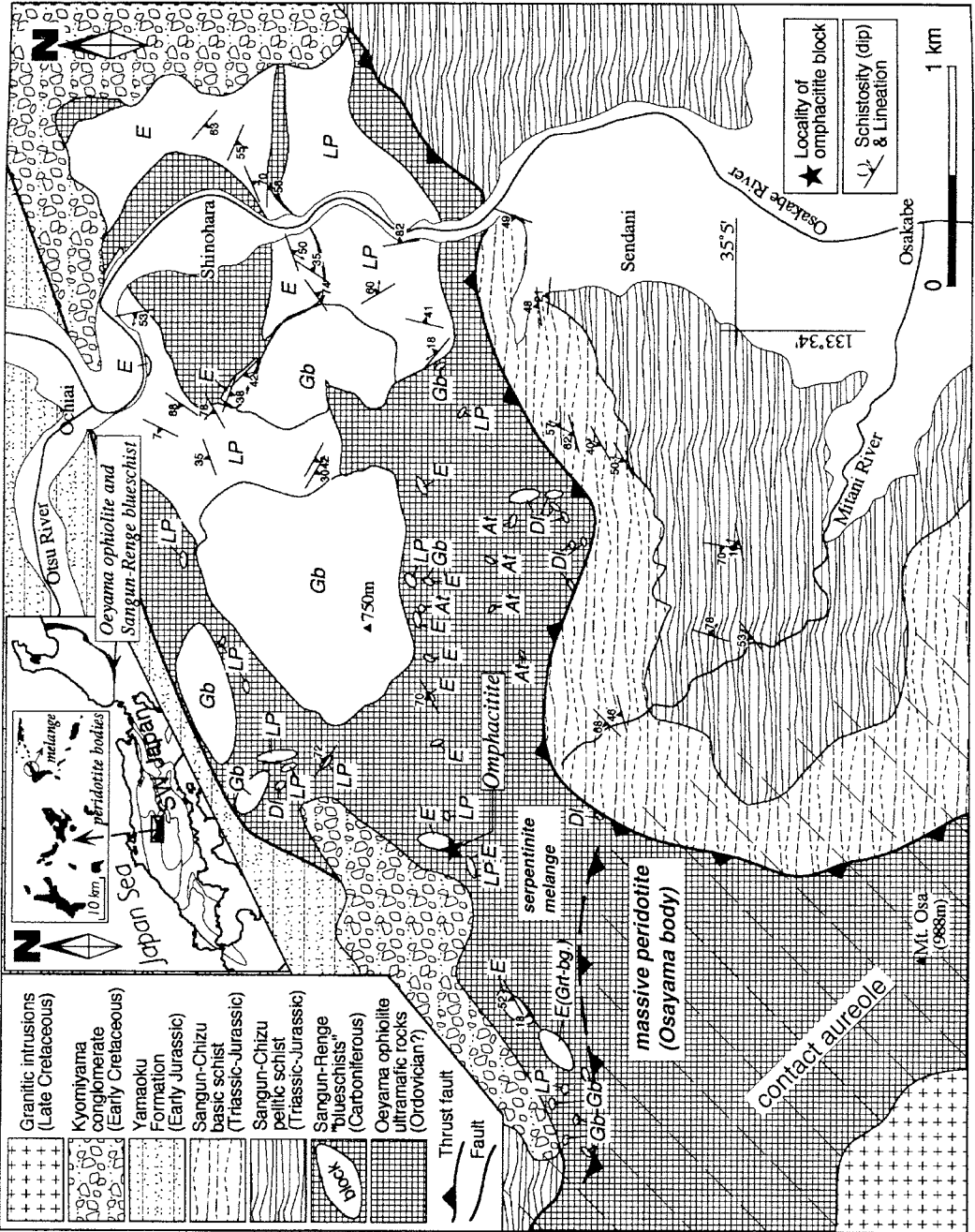


FIG. 1. Geological map of the Osayama serpentinite melange. [LP: schist of lawsonite-pumpellyite zone; E: schist of epidote zone; Gb: gabbro; Dl: dolerite; At: albitite]. The omphacitite block described in this paper occurs in the western part of the melange.

(Nozaka and Shibata, 1994; Tsujimori, in press), which may have been the residual peridotite of the Oeyama ophiolite.

The blueschist-facies schists, fragments of the Oeyama ophiolite (serpentinized peridotite, gabbro, dolerite) and metasomatic rocks (albitite, jadeitite, omphacite, tremolite schist, etc.) are enclosed as tectonic blocks of various size (10 cm to 1.5 km in length) in a serpentinite matrix (Tsujimori, in press). The blueschist-facies schists are divided into a lawsonite-pumpellyite zone (230–270°C at 6–8 kbar) and an epidote zone by the mineral assemblages of the mafic blueschists. The epidote zone blocks contain two varieties i.e. a garnet-free, lower grade block (>300°C at 6–8 kbar) and a garnet-bearing higher-grade block (garnet-glaucophane schist) (400–530°C at 11–13 kbar). The gabbro and dolerite blocks also contain blueschist-facies mineral assemblages corresponding with the lawsonite-pumpellyite zone. Tsujimori (1995a) found relics of the eclogite-facies mineral assemblage (620°C at 15 kbar) in a garnet-glaucophane schist block. Tsujimori (1995b) reported a chromian omphacite-bearing tremolite schist of peridotite origin, suggesting that a part of the Oeyama ophiolite has experienced a high-*P*–*T* metamorphism with the Sangun-Renge metamorphic rocks. The blueschist blocks give phengite K-Ar ages of around 320 Ma (Tsujimori and Itaya, submitted).

### Petrography

The omphacite block, 1 × 1 m in size, occurs at a small outcrop in the central part of the melange. This is a well foliated, dark-green to black rock. It is surrounded by sheared serpentinite.

The omphacite-diopside veins (< 0.4 mm wide) consist mainly of omphacite and diopside with minor amounts of pumpellyite and biotite. The veins cut the foliation at high angles (Fig. 2a). Omphacite in the vein occurs as radiating anhedral crystals which are intergrown with fan-shaped diopside crystal (< 0.3 mm) (Fig. 2b). Omphacite shows weak pleochroism between *X'* = colourless and *Z'* = faint green, and exhibits lower birefringence than the coexisting diopside. Diopside in the vein has a radiating habit and tends to be oriented perpendicular to the vein wall. The extinction positions of the two adjacent clinopyroxenes are different from each other. The two-pyroxene pair may have grown from the vein wall, and the texture suggests that the omphacite and diopside have contemporaneously crystallized in the vein.

The omphacite matrix consists mainly of omphacite with small amounts of chlorite, pumpellyite and titanite. Albite, apatite and biotite occur as accessories in the matrix, and quartz is absent.

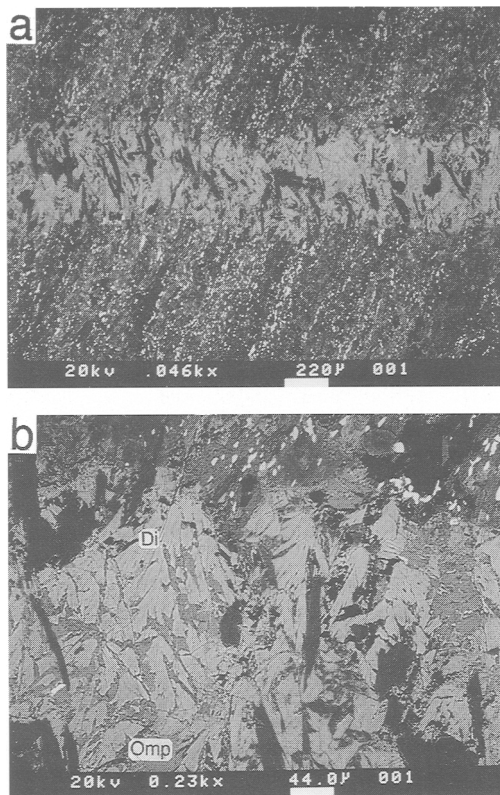


FIG. 2. (a) Back-scattered electron image of the occurrence of the omphacite-diopside vein cutting a foliated omphacite matrix. (b) Back-scattered electron image of coexisting omphacite (Omp) and diopside (Di) of the omphacite-diopside vein.

Titanite and apatite are not found in the omphacite-diopside vein. The albite-rich layer (< 3 mm wide) is rarely observed in the matrix. The foliation is defined by preferred orientation of elongated omphacite crystals. Matrix omphacite occurs as prismatic crystals (0.05–0.2 mm in length), and shows weak pleochroism between *X'* = colourless and *Z'* = pale green. A small number of crystals have a thin green rim. Chlorite occurs as oriented, tabular, colourless crystals, and shows a grey or brown interference colour. The elongation is positive. Pumpellyite is colourless and occurs as prismatic crystals (< 0.05 mm in length). Apatite occurs as anhedral crystals (< 0.1 mm).

### Mineral chemistry

Chemical analyses of minerals were carried out with an AKASHI ALPHA-30A scanning electron micro-

TABLE 1. Representative microprobe analyses of clinopyroxenes in the omphacitite

wt. %	Omphacite-diopside vein						Matrix			Green rim
	Omphacite			Diopside			Omphacite			
SiO <sub>2</sub>	56.79	56.50	55.86	52.72	53.66	53.05	56.76	55.58	55.98	51.48
TiO <sub>2</sub>	0.45	0.20	0.29	0.15	0.06	0.01	0.19	0.23	0.11	0.14
Al <sub>2</sub> O <sub>3</sub>	10.51	11.54	11.00	2.12	1.43	1.20	9.73	9.64	8.15	2.53
Cr <sub>2</sub> O <sub>3</sub>	0.03	0.15	0.16	0.12	0.17	0.15	0.20	0.22	0.12	0.11
FeO*	7.13	7.06	7.51	10.57	10.88	10.33	6.09	5.77	7.34	19.53
MnO	0.13	0.16	0.19	0.33	0.40	0.34	0.11	0.04	0.22	0.41
MgO	6.12	5.39	7.00	11.13	10.07	11.06	7.06	7.79	7.38	4.11
CaO	11.55	11.71	11.38	21.79	22.96	23.06	13.76	14.16	14.60	18.15
Na <sub>2</sub> O	7.46	7.31	7.04	1.34	0.79	0.79	6.35	6.74	6.21	3.91
K <sub>2</sub> O	0.03	0.03	0.03	0.09	0.00	0.03	0.00	0.06	0.00	0.00
Total	100.20	100.05	100.46	100.34	100.42	100.02	100.25	100.23	100.11	100.37
Number of cations on the basis of 6 oxygens										
Si	2.032	2.022	1.998	1.977	2.013	1.998	2.029	1.997	2.028	2.008
Ti	0.012	0.005	0.008	0.004	0.002	0.000	0.005	0.006	0.003	0.004
Al	0.443	0.487	0.464	0.094	0.063	0.053	0.410	0.408	0.348	0.116
Cr	0.001	0.004	0.004	0.004	0.005	0.004	0.006	0.006	0.004	0.003
Fe*	0.213	0.211	0.225	0.331	0.341	0.325	0.182	0.173	0.222	0.637
Mn	0.004	0.005	0.006	0.010	0.013	0.011	0.003	0.001	0.007	0.014
Mg	0.326	0.288	0.374	0.622	0.563	0.621	0.376	0.417	0.399	0.239
Ca	0.443	0.449	0.436	0.876	0.923	0.930	0.527	0.545	0.567	0.759
Na	0.517	0.507	0.488	0.097	0.057	0.058	0.440	0.470	0.436	0.296
K	0.001	0.001	0.001	0.004	0.000	0.001	0.000	0.003	0.000	0.000
Total	3.993	3.981	4.004	4.020	3.980	4.003	3.978	4.026	4.012	4.076

\* Total Fe.

scope 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 were used as standards, and a program based on Bence and Albees (1968) method and written by A. Ishiwatari was used for data processing. The analyses were done at 20 kV accelerating voltage, 1 nA sample current on MgO and <10  $\mu$ m beam diameter. Representative chemical analyses of clinopyroxenes in omphacitite are listed in Table 1.

#### Clinopyroxenes

The nomenclature of sodic pyroxene is based on the classification of Morimoto *et al.* (1988). The estimation of Fe<sup>2+</sup> and Fe<sup>3+</sup> was based on the assumption of four total cations and six oxygens. Jadeite, aegirine and augite (diopside) components of the metamorphic pyroxene were calculated as follows: (1) Jd = Al<sup>VI</sup>; (2) Ae = Fe<sup>3+</sup>; (3) Di(Aug)

= Ca, (4) and these components were normalized to 100. Analysed clinopyroxenes are plotted in a jadeite-aegirine-diopside ternary diagram (Fig. 3).

Omphacite in the omphacite-diopside vein has a significantly higher jadeite component (Jd<sub>46.1-52.0</sub>Ae<sub>0-8.4</sub>Aug<sub>48.0-51.2</sub>) than the omphacite in the matrix (Jd<sub>37.1-46.3</sub>Ae<sub>0-5.3</sub>Aug<sub>53.7-60.4</sub>). The FeO (total iron) content (av. 7.4 wt.%) of the vein omphacite is slightly higher than that of the matrix omphacite (av. 6.6 wt.%). Diopside coexisting with omphacite in the vein is poor in the jadeite component (Jd<sub>4.3-6.3</sub>Ae<sub>0-0.4</sub>Aug<sub>93.6-95.6</sub>). The green rim on matrix omphacite is also poor in jadeite, but has a slightly higher aegirine component than the vein diopside (Jd<sub>4.2-11.3</sub>Ae<sub>11.0-16.1</sub>Aug<sub>73.9-82.0</sub>).

Some jadeitites in the Osayama melange also contain omphacite. However, the omphacite in the jadeitite shows quite different occurrences. The omphacite (Jd<sub>42.5-71.3</sub>Ae<sub>0-5.1</sub>Aug<sub>28.7-57.5</sub>) occurs as overgrowths on pure (Jd<sub>>98</sub>) or impure (Jd<sub>84-96</sub>) jadeite, and as veinlet clots (2-5 mm wide), consisting of radial aggregates of needle-like crystals.

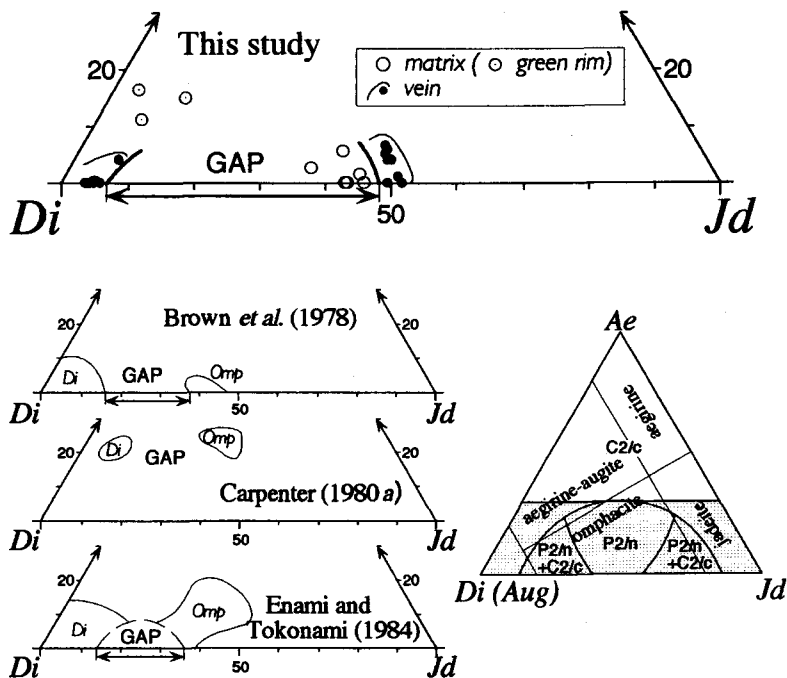


FIG. 3. Chemical compositions of the clinopyroxenes in the omphacitite block from the Osayama serpentinite melange plotted on a jadeite (Jd)-aegirine (Ae)-diopside (Di) ternary diagram. The compositional ranges reported by Brown *et al.* (1978), Carpenter (1980a) and Enami and Tokonami (1984) are also shown in small diagrams.

The omphacite in jadeitite may have been retrogressively formed in jadeitite.

#### Other minerals

Pumpellyite in omphacitite has an Al-rich composition with  $Al/(Al+Fe^*+Mg) = 0.78-0.81$ . The  $Fe^*/(Fe^*+Mg)$  ratio (0.28–0.39) is slightly lower than that of pumpellyite from other blueschist blocks in the Osayama serpentinite melange (0.41–0.69). Chlorite has a composition of  $Fe^*/(Fe^*+Mg) = 0.40$  and  $Al/(Al+Fe^*+Mg) = 0.33$ . Biotite in omphacitite has a composition of  $Mg/(Mg+Fe^{2+}) = 0.62$ ,  $TiO_2 = 0.4$  wt.% and  $Si = 5.8$  (p.f.u. for  $O = 22$ ).

#### Discussion

##### Comparison with the other omphacite-diopside pairs

The occurrence of coexisting omphacite and diopside pyroxene pairs has previously been reported from high-*P* metamorphic rocks. The coexistence is often ascribed to the presence of a miscibility gap between omphacite and diopside

pyroxene. For example, omphacite and diopside coexist in a blueschist specimens from Piemont, Italy ( $Jd_{35-50}$  and  $Jd_{3-12}$ ; Brown *et al.*, 1978) and from the Russian River, Franciscan Complex, U.S.A. ( $Jd_{28-41}$  and  $Jd_{8-15}$ ; Carpenter, 1980a). The paired clinopyroxenes reported by Brown *et al.* (1978) occur in an unusual manganese-rich, metamorphic rock containing manganese minerals such as braunite, hollandite, piemontite, manganian phengite and manganian phlogopite. The omphacite coexisting with diopside contains up to 2.5 wt.% MnO and less than about 5 mole% aegirine component. Peak metamorphic condition was estimated as  $8 \pm 1$  kbar and  $300 \pm 50^\circ C$ . The paired clinopyroxenes reported by Carpenter (1980a) form sector-zoned composite grains constituting emerald-green veins (up to 1 cm wide) and cutting an epidote-blueschist. The veins rarely contain quartz and white mica. The omphacite has about 25 mole% aegirine component.

Several studies on the microstructure of sodic pyroxene (e.g. Matsumoto *et al.*, 1975) have revealed that cation ordering can occur in omphacite with compositions close to  $Jd_{50}Di_{50}$ , causing a symmetry change from *C*-face-centred (*C2/c*) to primitive (*P2/n*). Carpenter (1979) determined the composi-

tional limits of the ordered  $P2/n$  omphacite relative to disordered  $C2/c$  pyroxenes in blueschist-facies rocks. Recently, Carpenter *et al.* (1990) thermodynamically described the cation ordering in omphacite using Landau theory.

Two different possible equilibrium phase relations between omphacite and diopside were presented. Carpenter (1980*b*) suggested the phase relations in the jadeite-diopside system shown in Fig. 4*a*. The apparent consolute temperature for the solvus between  $P2/n$  omphacite and  $C2/c$  diopside is about 500°C. He proposed a second-order transformation mechanism for the  $C2/c \rightleftharpoons P2/n$  transition. On the other hand, Enami and Tokonami (1984) found two different coexisting clinopyroxene pairs,  $C2/c$  omphacite ( $Jd_{35}$ )– $C2/c$  diopside ( $Jd_{15}$ ) in the epidote-amphibolite facies stage (above 500°C), and a  $P2/n$  omphacite ( $Jd_{44}$ )– $C2/c$  diopside ( $Jd_{10}$ ) in later epidote-blueschist facies stage (below 500°C) from the Iratsu eclogitic amphibolite mass, Sambagawa belt, Japan. They proposed a first-order transformation model for the transformation  $C2/c$  to  $P2/n$  to explain the persistence of the  $C2/c$  omphacite structure. Although the two interpretations for

omphacite transformation from  $P2/n$  to  $C2/c$  are quite different, the solvus between  $P2/n$  omphacite and  $C2/c$  diopside becomes wider with decreasing temperature in both cases. Although the space group of omphacite coexisting with diopside in the vein from the Osayama is not yet determined, it is probably ordered ( $P2/n$ ), judged from its high jadeite component and low temperature of formation.

The compositional gap between the omphacite and diopside in the Osayama vein is significantly wider than for other studies (Fig. 3). This may be due to the lower temperature of formation. The presence of Al-rich pumpellyite and the absence of epidote in the Osayama omphacitite constrains the metamorphic temperature condition to <350°C at 6–8 kbar. This temperature is lower than for the other omphacite-diopside pairs. As mentioned above, the coexisting omphacite and diopside in the Osayama omphacitite have a radiating habit. This texture suggests that the omphacite and diopside underwent a relatively rapid rate of growth. The rapid growth would probably have resulted in omphacite with a degree of cation order, less than the case of equilibrium growth at that temperature. In general, the miscibility gap is highly sensitive to cation ordering. Although the omphacite-diopside pair in the Osayama serpentinite melange may not have achieved strict thermodynamic equilibrium, the simultaneous growth of the two clinopyroxenes with a wide compositional gap suggests the lowest temperature (< 350°C) ever reported. The low temperature has undoubtedly promoted cation ordering.

#### Geological significance of the omphacite-diopside vein

Omphacite veins have been found at many localities in the Franciscan metamorphic rocks, such as in blueschist, amphibolite, greenstone and metagreywacke. Essene and Fyfe (1967) described some omphacite veins cutting blueschists from the Franciscan Complex. The vein omphacite occurs as prismatic crystals normal to the vein wall, or as radial aggregates, and Essene and Fyfe interpreted these textures as having been formed in a hydrostatic environment during omphacite crystallization. Coleman (1961) reported jadeite veins cutting blueschist in the Franciscan Complex, New Idria, U.S.A., and he interpreted the jadeite vein as having been produced by mobilization of a liquid rich in the jadeite component and low in  $H_2O$  during tectonic movement of the host blueschist. Philippot and Kienast (1989) described an omphacite-rich vein that cut mylonite zones in an eclogite-facies metagabbro, Monviso, Western Alps, Italy, and they found sector-zoned omphacite-chloromelanite

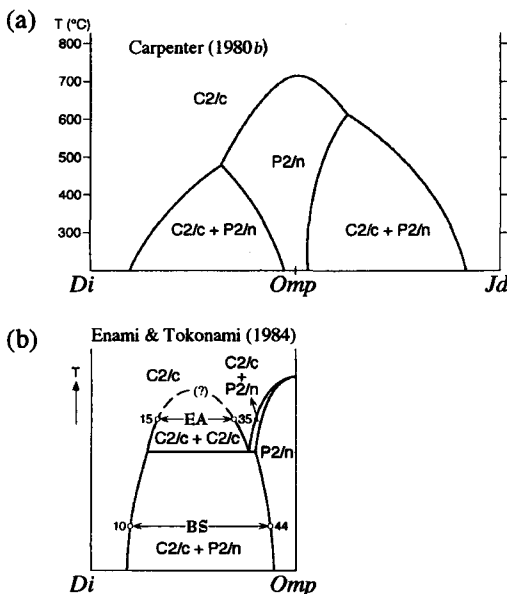


FIG. 4. (a) Phase relations in the  $Fe^{3+}$ -free diopside-jadeite system proposed by Carpenter (1980*b*). See Fig. 3 for abbreviations. (b) Phase relations between  $Fe^{3+}$ -free diopside and omphacite proposed by Enami and Tokonami (1984), which were partly based on Carpenter's diagram. BS: blueschist-facies, EA: epidote-amphibolite facies.

(they used Essene and Fyfe's (1967) nomenclature) in the veins. It was inferred that the vein pyroxenes of the eclogite-facies metagabbro were precipitated from aqueous solution during successive periods of fracturing and healing.

The texture of our omphacite-diopside vein suggests that the clinopyroxene pair was formed contemporaneously in the fracturing stage following the metasomatism from a Ca- and alkali-rich fluid at a lower temperature than the solvus between omphacite and diopside. Such a fluid might be introduced from the surrounding serpentinite, because CaO may have been released from clinopyroxene-bearing harzburgite during its serpentinization. The stability of omphacite and Al-rich pumpellyite, both in the matrix and in the vein, indicates that the matrix-forming stage and the later fracturing stage may have been under relatively high-pressure conditions of 6–8 kbar at < 350°C. The omphacite-diopside vein may have formed during the melange-forming stage at relatively high-*P–T*.

### Conclusion

The compositional gap between the coexisting omphacite (Jd<sub>46–52</sub>) - diopside (Jd<sub>4–6</sub>) in the Osayama omphacite vein is significantly wider than that for other omphacite-diopside pairs reported in the literature. This may be due to the lower temperature of their formation. They may have grown simultaneously from a vein-filling fluid in the omphacite block at relatively high pressure during the melange-forming stage.

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