In-situ U-Pb zircon age dating deciphering the formation event of the omphacite growth over relict edenitic pargasite in omphacite-bearing jadeitite of the Itoigawa-Omi area of the Hida-Gaien belt, central Japan

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Omphacite replacing after relict edenitic pargasite has been found in an omphacite-bearing jadeitite block of the Itoigawa-Omi area in the Hida-Gaien belt. Omphacite occurs sporadically as fine-grained aggregate reaching a few cm in length in a jadeite-albite matrix, and sometimes contains edenitic pargasite as a core. The edenitic pargasite is chemically and optically homogeneous and does not show direct contact with jadeite and albite. An irregular shaped omphacite-diopside mixed area occurs near edenitic pargasite in a coarse omphacite aggregate. The texture suggests that the breakdown of edenitic pargasite was triggered by the addition of a hydrothermal fluid, from which jadeite and albite were precipitated later, passing through diopside and omphacite by the reaction:

At the periphery of pseudomorphic omphacite, a hydrothermal fluid removed the breakdown components of the reaction other than omphacite.

New in-situ LA-ICP-MS U-Pb dating revealed that zircons in edenitic pargasite yield apparent age up to ~590 Ma, with mean ages of 560 ± 16 Ma, interpreted as the minimum age of a precursor rock. A zircon age of 519 ± 21 Ma from jadeitite without omphacite corresponds to a timing of crystallization of omphacite, jadeite, and albite. The studied jadeitite is a typical R-type jadeitite, and the nearly total replacement from a precursor rock to the omphacite-bearing jadeitite has been attributed to hydrothermal activity at Middle Cambrian times.

Keywords: R-type jadeitite, Omphacite, Diopside, Jadeite, Subduction initiation

INTRODUCTION

Omphacite is a major constituent of eclogite and yields important information pertinent to understanding the pressure-temperature (*P-T*) trajectory of high- to ultra-

high-pressure metabasaltic rocks in subduction zones (e.g., Banno and Matsui, 1965; Nakamura and Banno, 1997). In relation to subduction zone metamorphism, omphacite coexisting with jadeite or augite has attracted a great deal of attention for studies of the nature of sodic clinopyroxene miscibility gaps (e.g., Carpenter, 1980; Yokoyama and Sameshima, 1982; Matsumoto and Hirajima, 2005).

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The origin of omphacite, especially in jadeitite and omphacitite, remains controversial. For examples, omphacite is simply considered a product of the miscibility gap between omphacite and jadeite (e.g., Yokoyama and Sameshima, 1982; Tsujimori et al., 2005) or omphacite and augite (e.g., Tsujimori, 1997; Tsujimori and Liou, 2004), irrespective of the origin of impure jadeite, either as the metasomatic product of jadeite + quartz or direct precipitation from a fluid phase (cf. Tsujimori and Harlow, 2012; Harlow et al., 2015; 2016). Direct precipitation of omphacite from a fluid phase has been also proposed as a formation mechanism of omphacitite in the Nishisonogi metamorphic rocks (Shigeno et al., 2012).

In the Itoigawa-Omi (Ohmi) area of the Hida-Gaien belt (the Hida marginal) belt, we found omphacite-bearing jadeitite (omphacite jadeitite) in which precursor amphiboles pseudomorphed by omphacite occurs in a jadeite and albite matrix. The amphibole has a composition of the edenitic pargasite of Leake et al. (1997). The replacement texture clearly shows that omphacite is a breakdown product of edenitic pargasite. Tsujimori and Harlow (2012) proposed two classes for the types of hydrothermal formation of jadeitite: one is precipitate type (P-type) jadeitite, and the other is metasomatic replacement type (R-type). The omphacite-bearing jadeitite of this study is R-type.

In this paper, we present petrology and mineralogy of the R-type omphacite-bearing jadeitite in the Itoigawa-Omi area and in-situ laser ablation inductively coupled plasma mass spectrometry (LA-ICP-MS) U-Pb dating of constituent zircons, and discuss the origin of omphacite and its petrogenetic significance.

GEOLOGY AND OCCURRENCE OF JADEITITE

Geological outline of the Hida-Gaien belt

The Hida-Gaien belt is a serpentinite mélangé belt in which Paleozoic accretionary complexes, high P/T metamorphic rocks, and Paleozoic sediments were tectonically disrupted and mixed (Komatsu, 1990; Kunugiza et al., 2004; Kunugiza and Maruyama, 2011), though each rock unit composes each geological unit as nappe in the western part of southwest Japan (Fig. 1). The major constituents of the Itoigawa-Omi area (Fig. 2) of the Hida-Gaien belt are: (a) Oeyama-Renge belt (Kunugiza and Goto, 2010)

Serpentinized peridotite, ca. 500 Ma jadeitite (Kunugiza et al., 2002; Tsutsumi et al., 2010), Ordovician metagabbro (e.g., Matsumoto et al., 1983), 370 Ma glaucophane-bearing eclogite (e.g., Tsujimori, 2002; Tsujimori, 2010), and the 300 Ma Renge metamorphic schist (Banno, 1958; Kunugiza et al., 2004; Matsumoto et al., 2011).

Figure 1. Jadeitite localities in Japan.

Figure 2. Geological map of the Itoigawa-Omi area of the Hida-Gaien belt and the locality of the omphacite-bearing jadeitite block.

(b) Akiyoshi belt (Nakamizu et al., 1989)

Permian accretionary complex with late Paleozoic reef limestone.

Occurrence of jadeitite of the Itoigawa-Omi area

Jadeitite occurs as boulders in the Omi (Hashidate) and Kotaki (Kotaki) riverbeds in the Itoigawa-Omi area (Fig.

2). Though no outcrop of jadeitite has been found in Hashidate and Kotaki, jadeitite is considered to occur as blocks in serpentinite because jadeitite boulders are distributed only in the lower part of a small stream cross-cutting serpentinite. The samples of this study were collected as debris from alluvial deposits of a newly constructed dam outside of the registered area as a natural monument by the Ministry of Education, Culture, Sports, Science, and Technology of Japan.

The mineral assemblages of jadeitite of this area are mainly jadeite–albite with no quartz (Iwao, 1953; Chihara, 1971; Oba et al., 1992). Natrolite jadeitite (Miyajima et al., 1999) and analcime jadeitite (Ishida et al., 2005; Morishita et al., 2007) are not uncommon in this area. Euhedral jadeite grains were found in albite, natrolite, and analcime matrices in each jadeitite sample (e.g., Miyajima et al., 1999). New minerals, such as itoigawaite with an ideal formula of $\text{SrAl}_2\text{Si}_2\text{O}_7(\text{OH})_2 \cdot \text{H}_2\text{O}$ (Miyajima et al., 1999), renegeite with an ideal formula of $\text{Sr}_4\text{ZrTi}_4\text{Si}_4\text{O}_{22}$ (Miyajima et al., 2001), and matsubaraite with an ideal formula of $\text{Sr}_4\text{Ti}_5(\text{Si}_2\text{O}_7)_2\text{O}_8$ (Miyajima et al., 2002), have been found in jadeitite, and will be called new Sr–Ti–Zr minerals in this paper. Thus, a hydrothermal origin has been proposed for jadeitite in this area (Miyajima et al., 1999; Kunugiza et al., 2000; Morishita et al., 2007).

Omphacite sporadically occurs as irregular aggregates in jadeitite (Yokoyama and Sameshima, 1982; Ishida et al., 2005; Morishita et al., 2007). It shows various colors in hand specimen depending on Fe and Ti contents. Omphacite in this study is magnesian and is dark-greenish in color.

PETROGRAPHY OF OMPHACITE-BEARING JADEITITE

‘Jadeitite’ block including omphacite-bearing jadeitite was found at the Hashidate dam site of the Omi River (Fig. 2), reaching about 3 m in diameter. The block consists of three different parts (Fig. 3a): jadeitite with pseudomorphous omphacite after edenitic pargasite (OE part), jadeitite without pseudomorphous omphacite (JD part), and a biotite vein with a maximum thickness of several tens of cm (BV part). The JD part is found between the OE and BV parts.

Jadeitite with pseudomorphous omphacite after edenitic pargasite (OE part)

In the OE part (Fig. 3b), jadeitite consists of jadeite and albite with small amounts of omphacite, edenitic pargasite, and zircon. Jadeite is fine-grained, occurring as in-

dividual grains and aggregates in an albite-rich matrix. Fine-grained jadeite also occurs as aggregate around pseudomorphous omphacite. The needle-like aggregate of jadeite in an albite matrix is similar in texture to Figure 3 in Ishida et al. (2005), which they called ‘dendritic texture’ consisting of fine-grained jadeite and omphacite. Albite is coarse to medium-grained, sometimes showing albite twinning. The modal proportion of jadeite/albite varies within a thin section.

Omphacite occurs sporadically as fine-grained aggregate reaching a few cm in length in a jadeite–albite matrix, and sometimes contains edenitic pargasite as a core. The omphacite aggregate sometimes shows prismatic shape, inheriting the morphology of the pegmatitic amphibole (Figs. 3b and 4a). The extinction angle of omphacite is essentially the same within a grain (Fig. 4a), but birefringence color and back-scattered electron image (BEI) show the presence of fine-scale inhomogeneity (Figs. 4a and 4b). Note that a fine-grained, irregular shaped omphacite–diopside mixed area occurs near edenitic pargasite in a coarse omphacite aggregate (Figs. 4c and 4d).

The edenitic pargasite core is homogeneous under a microscope and the contact between the edenitic pargasite core and the pseudomorphous omphacite rim is irregular but sharp (Fig. 4a). There is no edenitic pargasite in direct contact with jadeite and albite.

Zircon occurs as euhedral to subhedral grains in a jadeite–albite matrix and as inclusions in omphacite and edenitic pargasite (cf. Fig. 7a).

Jadeitite without pseudomorphous omphacite (JD part)

Jadeitite of the JD part consists mostly of fine-grained jadeite with small amounts of fine-grained omphacite, pargasite, and albite (Fig. 3c). It lacks the pseudomorphous omphacite after edenitic pargasite. Euhedral zircons occur, showing faint alignment in a jadeitite matrix (Fig. 3c).

Biotite vein (BV part) crosscutting jadeitite

The biotite vein occurs with maximum thickness of several tens of cm and sometimes penetrates along a crack in the omphacite-bearing jadeitite (Fig. 3a). The BV is greenish in color and consists mainly of phlogopite and chlorite. At the boundary between the JD and BV parts, a dark green pargasite zone about 1 cm thick occurs (Fig. 3d). Under a microscope, acicular pargasite crystals occur perpendicular to the jadeitite wall and are intergrown with fine-grained jadeite aggregate (Fig. 3d). At the opposite side, pargasite crystals are interfingered with acicular phlogopite and chlorite crystals.

Figure 3. Photograph of omphacite-bearing jadeitite block at Hashidate of the Omi River in the Itoigawa–Omi area. (a) The block consists of three different parts, OE, JD, and BV parts (see text for explanation), (b) Photomicrograph of OE part (open nicol). Note pseudomorphic omphacite after edenitic pargasite (Ed-Prg). (c) Photomicrograph of JD part (crossed nicols). Zircons were dated by using LA-ICP-MS (see Fig. 8), and (d) Photomicrograph of the boundary between JD and BV parts (open nicol).

MINERAL CHEMISTRY

Analytical procedure

Pyroxene, amphibole, plagioclase, and biotite were analyzed by the JEOL JXA8230 electron probe micro analyzer with 5 WDS detectors at the University of Toyama. Operating conditions were 5 μm diameter beam, 15 kV, 20 nA probe current, and an analytical procedure after Shukuno (2003). Ferric iron contents of pyroxene and amphibole were calculated based on 6 oxygens and 4 cations, and 23 oxygens and 13 cations excluding Na, K, and Ca cations, respectively. The representative analyses of these minerals are presented in Table 1.

Pyroxene

Pyroxene compositions of the omphacite-bearing jade-

itite block are shown in Figures 5a and 5b on the basis of the classification of Morimoto et al. (1988). Note that the compositional ranges of jadeite and omphacite overlap between the OE and JD parts (Figs. 5a and 5b). Jadeite and omphacite are depleted in aegirine (Ae) components. They are highly magnesian at around $\text{Mg}/(\text{Mg} + \text{Fe})$ atomic ratio $[X_{\text{Mg}}] = 0.9$ (Table 1). Though kosmochlor (Sakamoto and Takasu, 1997) has been described in a jadeitite sold in the Itoigawa–Omi area, Cr_2O_3 content of jadeite in the omphacite-bearing jadeitite is very low (Table 1).

Diopside occurs only in a coarse omphacite aggregate as a fine-grained diopside–omphacite mixed area near the edenitic pargasite core (Figs. 4c and 4d). It is almost pure diopside, and its compositional variation toward omphacite in Fig. 5a is not only due to the miscibility gap between diopside and omphacite, but also to analytical error due to the fine-grain size of the minerals.

Figure 4. Photomicrograph and back-scattered electron image of pseudomorphic omphacite after edenitic pargasite (Ed-Prg) of jadeitite in the OE part. In a coarse grain, fine-grained, irregular shaped mixture of diopside and omphacite (Di-Omp mix) occur near relic edenitic pargasite.

Amphibole

Amphiboles of the omphacite-bearing jadeitite are edenite to pargasite of Leake et al. (1997), and show separated compositional groups in each part (Fig. 6). The amphibole core in the omphacite of the OE part is edenitic in composition, being around the boundary between edenite and pargasite. Si ranges from 6.35 to 6.58 pfu, with average of 6.49 and X_{Mg} goes from 0.85 to 0.96, with an average of 0.89 for 36 analyses. For descriptive purposes in this paper, the amphibole of the OE part is called edenitic pargasite (Ed-Prg), because the amphibole of the JD and BV parts is pargasite as described below.

Amphibole of the JD part is pargasite, with Si ranging 5.95–6.23 pfu, with an average of 6.1, and X_{Mg} of 0.87–0.96, with an average of 0.91 for 14 analyses (Fig. 6). Acicular amphibole of the BV part is also pargasite

with Si of 6.12–6.28 pfu, with an average of 6.2, but is slightly Mg-poor, with X_{Mg} of 0.80–0.86, with an average of 0.83 for 10 analyses (Fig. 6).

Sodic plagioclase

Plagioclase of the omphacite-bearing jadeitite is albite with X_{An} less than 0.01 and K_2O content less than 0.1 wt% (Table 1). No relict plagioclase or Ba-rich feldspar described by Morishita (2005) was found in this omphacite-bearing jadeitite.

Biotite

Biotite of the BV is phlogopite, with X_{Mg} of 0.87–0.89 and TiO_2 less than 0.04 wt% (Table 1).

Table 1. Representative chemical compositions of jadeite, omphacite, diopside, amphibole, plagioclase and biotite of omphacite-bearing jadeitite of the Itoigawa-Omi area

Sample	OE	OE	OE	OE	OE	OE	OE	OE	OE	OE	OE	OE	OE	OE	OE
Mineral	Jd	Jd	Jd	Omp	Omp	Omp	Omp	Di	Di	Ed-Prg	Ed-Prg	Ed-Prg	Ed-Prg	Ed-Prg	Pl
SiO ₂	59.55	58.81	58.49	57.37	57.19	57.22	57.40	55.42	55.57	46.34	46.33	46.23	45.61	45.87	68.86
TiO ₂	0.00	0.00	0.02	0.36	0.58	0.33	0.41	0.05	0.00	0.44	0.44	0.39	0.42	0.44	0.00
Al ₂ O ₃	24.65	22.53	22.09	12.34	12.11	13.52	12.88	1.35	0.90	13.49	13.41	13.5	13.4	13.66	19.39
Cr ₂ O ₃	0.07	0.00	0.00	0.00	0.01	0.07	0.01	0.00	0.02	0.03	0.03	0.01	0.00	0.03	0.00
FeO	0.38	0.56	0.64	1.17	1.19	0.77	1.12	1.64	1.60	5.21	5.21	4.26	5.28	5.52	0.00
MnO	0.00	0.01	0.01	0.03	0.00	0.03	0.04	0.04	0.13	0.08	0.08	0.07	0.07	0.07	0.04
MgO	0.23	1.54	2.44	9.02	8.99	7.99	8.23	17.15	17.54	16.79	16.8	17.47	16.99	16.78	0.00
CaO	0.43	2.41	2.91	11.83	11.96	11.14	11.77	23.41	24.08	10.76	10.76	10.31	10.29	10.12	0.02
Na ₂ O	14.51	13.5	12.39	8.08	8.08	8.60	8.31	0.95	0.64	4.64	4.64	4.70	4.80	4.74	12.03
K ₂ O	0.01	0.00	0.37	0.00	0.00	0.01	0.02	0.00	0.01	0.27	0.27	0.26	0.24	0.29	0.01
Total	99.81	99.36	99.36	100.20	100.1	99.67	100.2	99.99	100.5	98.03	97.94	97.20	97.09	97.52	100.34
Si	2.02	2.01	2.01	2.00	1.99	2.00	2.00	2.00	2.00	6.54	6.55	6.52	6.48	6.48	3.00
Ti	0.00	0.00	0.00	0.01	0.02	0.01	0.01	0.00	0.00	0.05	0.05	0.04	0.05	0.05	0.00
Al	0.99	0.91	0.90	0.51	0.50	0.56	0.53	0.06	0.04	2.24	2.23	2.24	2.24	2.27	0.99
Cr	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Fe ³⁺	0.00	0.00	0.00	0.03	0.03	0.01	0.01	0.01	0.01	0.00	0.00	0.18	0.22	0.25	0.00
Fe ²⁺	0.01	0.02	0.02	0.01	0.00	0.01	0.02	0.04	0.04	0.61	0.62	0.33	0.41	0.40	0.00
Mn	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.01	0.01	0.01	0.01	0.01	0.00
Mg	0.01	0.08	0.13	0.47	0.47	0.42	0.43	0.92	0.94	3.54	3.54	3.67	3.60	3.53	0.00
Ca	0.02	0.09	0.11	0.44	0.45	0.42	0.44	0.90	0.93	1.63	1.63	1.56	1.56	1.53	0.00
Na	0.95	0.90	0.83	0.54	0.55	0.58	0.56	0.07	0.04	1.27	1.27	1.29	1.32	1.30	1.02
K	0.00	0.00	0.02	0.00	0.00	0.00	0.00	0.00	0.00	0.05	0.05	0.05	0.04	0.05	0.00
O=	6	6	6	6	6	6	6	6	6	23	23	23	23	23	8

Sample	JD	JD	JD	JD	JD	JD	JD	JD	JD	JD	JD	BV	BV	BV	BV
Mineral	Jd	Jd	Jd	Jd	Omp	Omp	Omp	Prg	Prg	Prg	Pl	Prg	Prg	Bt	Bt
SiO ₂	58.89	59.02	58.65	59.03	56.32	56.71	56.28	42.30	42.60	43.41	69.75	43.12	43.14	37.91	37.70
TiO ₂	0.00	0.03	0.01	0.04	0.07	0.02	0.00	0.16	0.15	0.16	0.00	0.00	0.00	0.04	0.04
Al ₂ O ₃	25.43	24.69	24.67	24.50	14.10	11.82	10.69	18.18	18.37	19.07	20.14	16.91	16.70	18.45	18.50
Cr ₂ O ₃	0.00	0.00	0.00	0.00	0.08	0.00	0.02	0.02	0.00	0.07	0.01	0.00	0.00	0.01	0.00
FeO	0.07	0.20	0.13	0.22	1.90	2.15	2.28	5.57	5.74	5.72	0.00	6.95	6.51	5.03	5.04
MnO	0.03	0.04	0.01	0.01	0.06	0.04	0.08	0.13	0.13	0.08	0.03	0.12	0.12	0.01	0.01
MgO	0.09	0.21	0.29	0.37	7.08	8.50	9.28	15.23	15.11	15.62	0.00	14.59	14.86	22.36	22.03
CaO	0.37	0.53	0.56	0.93	11.11	12.82	13.66	10.63	10.7	10.34	0.01	10.54	10.75	0.00	0.02
Na ₂ O	14.93	14.89	14.63	14.74	8.57	7.70	6.99	4.30	4.00	4.72	11.97	4.68	4.60	0.49	0.48
K ₂ O	0.02	0.01	0.01	0.00	0.03	0.00	0.03	0.43	0.37	0.35	0.02	0.38	0.32	10.38	10.32
Total	99.83	99.61	98.96	99.83	99.31	99.74	99.30	96.95	97.16	99.55	101.93	97.28	97.01	94.69	94.14
Si	1.99	2.00	2.00	2.00	1.98	1.99	1.99	6.02	6.03	5.99	2.99	6.18	6.20	5.45	5.45
Ti	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.02	0.02	0.02	0.00	0.00	0.00	0.00	0.00
Al	1.01	0.99	0.99	0.98	0.58	0.49	0.45	3.05	3.07	3.10	1.02	2.86	2.83	3.12	3.15
Cr	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.01	0.00	0.00	0.00	0.00	0.00
Fe ³⁺	0.00	0.00	0.00	0.00	0.04	0.05	0.05	0.37	0.42	0.49	0.00	0.18	0.14	0.00	0.00
Fe ²⁺	0.00	0.01	0.00	0.01	0.02	0.02	0.02	0.30	0.25	0.17	0.00	0.66	0.65	0.60	0.61
Mn	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.02	0.02	0.01	0.00	0.01	0.01	0.00	0.00
Mg	0.00	0.01	0.01	0.02	0.37	0.45	0.49	3.23	3.19	3.21	0.00	3.12	3.18	4.79	4.74
Ca	0.01	0.02	0.02	0.03	0.42	0.48	0.52	1.62	1.62	1.53	0.00	1.62	1.65	0.00	0.00
Na	0.98	0.98	0.97	0.97	0.58	0.52	0.48	1.19	1.10	1.26	0.99	1.30	1.28	0.14	0.13
K	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.08	0.07	0.06	0.00	0.07	0.06	1.90	1.90
O=	6	6	6	6	6	6	6	23	23	23	8	23	23	22	22

Jd, jadeite; Omp, omphacite; Di, diopside; Ed-Prg, edenitic pargasite; Prg, pargasite; Pl, plagioclase; Bt, biotite.

Figure 5. Compositional diagram of Morimoto et al. (1988) for clinopyroxenes of omphacite-bearing jadeite: (a) OE part with pseudomorphous omphacite and (b) JD part without pseudomorphous omphacite.

LA-ICP-MS U-Pb AGE OF ZIRCON

Analytical procedure

In-situ zircon U-Pb age dating for ordinary petrographic rock thin section was carried out using an iCAP-Q (Thermo Fisher Scientific) ICP-MS with an Analyte G2 excimer laser ablation system (193-nm wavelength, Teledyne CETAC Technologies) at the Pheasant Memorial Laboratory for Geochemistry and Cosmochemistry, Institute for Planetary Materials, Okayama University at Misasa. The fixed laser power was 4.0 mJ and the pulse repetition rate was 5 Hz for all analyses. The beam size was controlled using a 25 μm circular aperture and the ablation duration was 40 seconds, as a sequel to yield a $\sim 20 \mu\text{m}$ pit depth. During the ablation, $^{202}\text{Hg}^+$, $^{204}\text{Pb}^+$, $^{206}\text{Pb}^+$, $^{207}\text{Pb}^+$, $^{208}\text{Pb}^+$, $^{232}\text{Th}^+$, $^{235}\text{U}^+$, and $^{238}\text{U}^+$ were sequentially collected during each ~ 0.14 second cycle. The total instrumental mass and elemental discriminations were corrected using

Figure 6. Compositional diagram of Leake et al. (1997) for amphiboles from the OE, JD, and BC parts of omphacite-bearing jadeite.

the discrimination factor obtained through repeated analyses of the reference zircon (apparent age of 561 ± 2 Ma; Usui et al., 2002). The reference zircon was examined every 5–10 ablations of unknown analyses. All ages and errors of analyzed zircon were calculated using ISOPLOT (version 4.15) by K. Ludwig. The 91500 standard zircon used for quality control of the analyses yielded 1063.2 ± 3.3 Ma ($n = 5$) (Ref. Age: 1065 Ma; Wiedenbeck et al., 1995). The estimated uranium concentration of zircon was typically within the range of 20–50 $\mu\text{g/g}$ and the total counts of ^{206}Pb for each spot was less than ~ 10000 . In this case, the effect of common lead contamination could be significant. However, it was difficult to apply reliable common lead correction to the obtained data because of statistically large errors of the Pb signals. Therefore, in this experiment we selected the data, which were plotted on a U-Pb concordia, and calculated the age without the common lead correction. As a result, we obtained a total of 22 concordia ages from two samples (203A from the OE part and NA03 from the JD part) (Table 2).

Results

A total of 22 analyses obtained from the two samples of omphacite-bearing jadeite show age groups of 560 ± 16 Ma and 519 ± 21 Ma (Figs. 9a and 9b). The older ages are from the OE part with pseudomorphous omphacite (sample 203A) and the younger ages are from the JD part without pseudomorphous omphacite (sample NA03). Occurrence of dated zircon grains is shown in Figures 7 and 8, respectively.

Age determination of the OE part focused on zircons in and/or around edenitic pargasites. Within a thin section, ages of more than 590 Ma (analyses 203A X1–5 and 203A X2–1 in Table 2) were obtained from zircons occurring fully or partly in edenitic pargasite grain (Fig. 8a), although other zircons in the same thin section show

Table 2. In-situ LA-ICP MS U-Pb zircon ages of omphacite-bearing jadeitite of the Itoigawa-Omi area

Spot#	$^{207}\text{Pb}/^{206}\text{Pb}$	2 σ	$^{207}\text{Pb}/^{235}\text{U}$	2 σ	$^{206}\text{Pb}/^{238}\text{U}$	2 σ	$^{206}\text{Pb}/^{204}\text{Pb}$	ρ	Conc. Age (Ma)	2 σ^*	Weighted mean Age (Ma)	2 σ
203A X1-5	0.0596	0.0013	0.790	0.031	0.0963	0.0021	1026	0.33	592.0	11.0	560	16
203A X2-1	0.0601	0.0017	0.795	0.041	0.0959	0.0022	989	0.16	591.0	12.0		
203A Y1-1	0.0579	0.0014	0.746	0.031	0.0934	0.0021	2050	0.13	573.0	10.6		
203A Y1-2	0.0608	0.0015	0.761	0.034	0.0909	0.0020	1718	0.26	563.0	11.2		
203A Y1-3	0.0589	0.0015	0.763	0.035	0.0940	0.0021	1210	0.04	578.0	10.6		
203A Y1-4	0.0575	0.0012	0.730	0.026	0.0922	0.0020	2672	0.16	565.0	10.0		
203A Y1-5	0.0601	0.0023	0.715	0.051	0.0863	0.0021	1032	0.08	536.0	12.0		
203A Y2-2	0.0601	0.0014	0.719	0.030	0.0869	0.0019	1395	0.20	540.0	10.0		
203A Y2-3	0.0584	0.0013	0.681	0.027	0.0846	0.0019	1010	0.23	524.0	10.0		
203A Y2-4	0.0595	0.0016	0.733	0.036	0.0893	0.0020	1639	0.03	553.0	10.6		
NA03 X1-1	0.0595	0.0013	0.631	0.023	0.0769	0.0016	3836	0.07	483.2	8.4	519	21
NA03 X1-3	0.0590	0.0011	0.614	0.019	0.0755	0.0016	2148	0.11	475.8	7.8		
NA03 X2-2	0.0595	0.0010	0.695	0.018	0.0847	0.0018	2807	0.20	530.0	8.2		
NA03 X2-4	0.0579	0.0012	0.676	0.023	0.0847	0.0018	3169	0.09	524.0	9.0		
NA03 X3-3	0.0613	0.0011	0.788	0.023	0.0933	0.0020	3873	0.21	582.0	9.6		
NA03 X3-4	0.0606	0.0011	0.717	0.020	0.0859	0.0019	5134	0.11	539.1	8.6		
NA03 X3-5	0.0606	0.0011	0.709	0.021	0.0850	0.0019	1660	0.12	533.9	8.7		
NA03 X4-1	0.0589	0.0014	0.640	0.025	0.0789	0.0019	1181	0.33	493.0	11.0		
NA03 X4-4	0.0577	0.0020	0.737	0.048	0.0927	0.0025	1066	0.18	570.0	14.0		
NA03 X5-1	0.0580	0.0015	0.671	0.029	0.0839	0.0023	1037	0.20	520.0	12.0		
NA03 X5-3	0.0600	0.0017	0.734	0.037	0.0889	0.0024	1375	0.26	551.0	13.0		
NA03 X5-4	0.0581	0.0019	0.674	0.039	0.0843	0.0024	1010	0.08	522.0	12.0		

younger ages, ranging from 578 to 524 Ma (Table 2, and Figs. 8e and 8f). As cathodoluminescence (CL) images of these zircons show clear oscillatory zoning (Figs. 8c and 8d), it is considered that these old ages are those of precursor rock.

Zircons of the JD part are coarse-grained (Fig. 9a) and damaged to some extent as shown by CL images (Figs. 9b – 9f). The age of ~ 519 Ma overlaps with the ion-microprobe age of ~ 516 Ma for zircons from jadeitite in this area (Kunugiza et al., 2002; Kunugiza and Goto, 2010). Within the thin section, age of 582 Ma (NA03 X3-3) is found in the central clear portion of a zircon grain (Fig. 9d), and ages of less than 500 Ma were obtained from the edges and broken portion of zircon grains (Figs. 9b and 9e).

DISCUSSION

Formation of omphacite

The presence of pegmatitic edenite pseudomorph in the omphacite-bearing jadeitite of this study suggests that the

omphacite grew over edenitic pargasite (Figs. 4a and 4b). A diopside-omphacite mixed area near edenitic pargasite (Figs. 4c and 4d) would have recorded the breakdown processes from edenitic pargasite to omphacite. Because edenitic pargasite is rich in Ca compared to omphacite, assuming NaAl content to be constant, the breakdown reaction of ideal edenite to form omphacite and diopside can be written as follows.

The breakdown reaction suggests the temporal presence of diopside to incorporate excess Ca and Mg components at the breakdown site of edenitic pargasite crystal. In addition to pseudomorphous morphology, the extinction angle of individual omphacite grains being essentially the same within omphacite aggregate suggests that the single chain structure of omphacite was inherited from the dou-

Table 2. (Continued)

Spot#	Th (ppm)	U (ppm)	Th/U
203A X1-5	10	70	0.14
203A X2-1	7	46	0.16
203A Y1-1	11	64	0.17
203A Y1-2	9	46	0.20
203A Y1-3	11	50	0.22
203A Y1-4	41	99	0.41
203A Y1-5	6	29	0.20
203A Y2-2	11	63	0.17
203A Y2-3	17	76	0.22
203A Y2-4	6	36	0.18
NA03 X1-1	8	39	0.21
NA03 X1-3	19	67	0.28
NA03 X2-2	47	149	0.31
NA03 X2-4	9	46	0.20
NA03 X3-3	34	91	0.37
NA03 X3-4	18	79	0.23
NA03 X3-5	12	69	0.17
NA03 X4-1	21	105	0.20
NA03 X4-4	4	27	0.15
NA03 X5-1	7	90	0.07
NA03 X5-3	10	53	0.19
NA03 X5-4	7	41	0.17

ble chain structure of edenitic pargasite.

The trigger of the breakdown reaction of edenitic pargasite should be the hydration of a precursor rock. The edenitic pargasite core is clear and chemically homogeneous and might have been stable in the absence of fluid. Instead of dry conditions, infiltrating fluid capable of precipitating jadeite and albite changed the chemical composition of a precursor rock and reacted with edenitic pargasite to form omphacite. At the periphery of the omphacite aggregate, there is no diopside and Mg_3SiO_5 component of the breakdown reaction, and whereas in the core of a coarse omphacite aggregate, the diopside-omphacite mixed area remains intact. This contrast can be explained if newly formed omphacite has prevented a further reaction from consuming edenitic pargasite.

A solid-solid reaction between edenitic pargasite, for instance, jadeite and albite upon the change of temperature and/or pressure cannot explain the presence of diopside being richer in Ca than as edenitic pargasite, and the absence of excess component minerals other than omphacite both at the periphery of the omphacite aggregate

and at jadeite and albite matrix. Thus, these mineralogical conditions imply the hydration reaction of edenitic pargasite.

The hydration reaction of edenitic pargasite to form dry omphacite in this study may not be a peculiar case for clinopyroxene. The stable occurrence of diopside is well known in serpentinized peridotite, below the stability field including tremolite (cf. Evans, 1977). The hydrothermal origin of jadeitite is often related to a silica-deficient fluid from serpentinized mantle-wedge peridotite (e.g., Tsujimori and Harlow, 2012; Harlow et al., 2015). Such a fluid is considered to have the potential to dehydrate amphibole to form omphacite. Morishita et al. (2007) represented the positive anomalies of Sr and Eu, and a strong positive high field strength element anomaly of omphacite from the green-colored omphacite-bearing jadeitite of this area, and attributed these compositional characteristics to a hydrothermal fluid related to serpentinization in subduction zones. The breakdown of edenitic pargasite was thus due to the stability of omphacite in the presence of a silica-deficient fluid from serpentinized peridotite.

In essence of the formation of omphacite-bearing jadeitite, a hydrothermal fluid capable of crystallizing jadeite and albite reacted with relict edenitic pargasite of a precursor rock to form omphacite. In the omphacite-bearing jadeitite of this study, omphacite, jadeite, and albite are hence in equilibrium. This inference is contradicted by the notion that omphacite is usually late in jadeitite (Harlow et al., 2016).

Edenitic pargasite as relict mineral of a precursor rock

Amphiboles of jadeitite are usually considered as secondary or retrogressive products after omphacite and jadeite. For instance, Shigeno et al. (2012) found two types of omphacitite in Nishisonogi metamorphic rocks in western Kyushu, (1) omphacitite layer in a metabasite block and (2) omphacitite lens in an albitite block, and described minor amounts of winchite in the former and magnesiohornblende in the latter. Though their description of amphibole is limited, they considered that magnesiohornblende postdated omphacite because the former crosscuts the latter.

Our study provides an opposite view for amphibole in jadeitite. It is apparent, as discussed above, that edenitic pargasite reacted with a hydrothermal fluid to form omphacite. Edenitic pargasite is thus a relict mineral of a precursor rock and remains stable as a core of coarse-grained omphacite where fluid was absent. However, the origin of edenitic pargasite remains unclear because

Figure 7. Occurrence and cathodoluminescence (CL) images of dated zircon grains of omphacite-bearing jadeitite (sample 203A) from OE part. (a) photomicrograph (open nicol): A coarse grained pseudomorph omphacite contains diopside-omphacite mixed area (see Figs. 4c and 4b and text for explanation), (b) photomicrograph (crossed nicols), and (c) to (f) CL images.

of the nature of its composition. Pegmatitic morphology and the mode of edenitic pargasite being small suggest a precursor rock of pegmatitic portions of plagiogranite,

but the nature of edenitic pargasite requires hydrothermal recrystallization after igneous activity.

Figure 8. Occurrence and cathodoluminescence (CL) images of dated zircon grains of omphacite-bearing jadeitite (sample NA03) from JD part. (a) photomicrograph (crossed nicols: see Fig. 3c), and (b) to (f) CL images.

Pargasite as a reaction product between jadeitite and evolved hydrothermal fluid

In the omphacite-bearing jadeitite, pargasite occurs in the

JD and BV parts. These amphiboles show separated compositional groups from edenitic pargasite of the OE part (Fig. 6) and hence suggest different origins for them.

In BV part, pargasite forms the outermost wall zone

Figure 9. In-situ LA-ICP MS U-Pb zircon ages of omphacite-bearing jadeitite: (a) sample 203A with pseudomorphous omphacite from the OE part and (b) sample NA03 from the JD part.

of the biotite vein in which acicular grains occur perpendicular to the host jadeitite of JD part (Fig 3d). Acicular pargasite coexists with jadeite in the JD part side and with phlogopite in the other side of the BV part. The fluid phase to form jadeitite should have been rich in Na and the fluid phase to form BV was rich in K. This difference can correspond to the changes in chemical composition of the fluid phase. It is thus considered that the acicular pargasite wall is the reaction product between jadeitite and an evolved K-rich fluid.

Pargasite in the JD part is similar in composition to that in BV part except for a slightly higher X_{Mg} (Fig. 6). Because of this, it is not a relict amphibole of a precursor rock, but a reaction product probably due to infiltrating fluid at the time of the formation of the BV. As the JD part sample is collected near the BV, it is suggested that the mineralogy and petrology of this jadeitite can be modified to some extent by infiltrating fluid.

Correspondence between petrogenetic sequence and in-situ zircon dating

We note three petrological stages from the study of the omphacite-bearing jadeitite. The first stage is the formation of relic edenitic pargasite, the second stage is the formation of pseudomorphous omphacite after pargasite, synchronized with the precipitation of jadeite and albite, and the third stage is represented by the formation of biotite vein. However, from *in-situ* zircon age dating, we obtained two age groups of zircons, 560 ± 16 Ma from the OE part with pseudomorphous omphacite and 519 ± 21 Ma from the JD part without it. Thus, the correspondence between the three petrological stages and the two age groups of in-situ zircon dating are of much interest.

Note that the younger age is harmonious with the ion-probe zircon ages of 519 ± 17 Ma and 512.3 ± 6.9 Ma from albite jadeitite (Kunugiza et al., 2002) and 497 ± 23 Ma from lavender jadeitite consisting mostly of jadeite, with small amounts of natrolite, titanite, and rengerite (Tsutsumi et al., 2010). The coincidence among zircon ages from the jadeitites of varying mineralogy in this area suggests a major hydrothermal event forming jadeitite to have taken place during the earliest Paleozoic ranging from 519 to 497 Ma. Fu et al. (2010) pointed out that most zircons in jadeitite originating from a hydrothermal fluid are relic igneous crystals inherited from a protolith. However, in this area, Kunugiza et al. (2002) proposed that these ages represent hydrothermal events to form jadeitite because of the absence of negative Eu anomalies in zircons.

The third stage of the BV formation apparently postdated the second stage of major jadeitite formation. Despite this, it is considered that the age of the third stage is within the range of the second stage, because the dated JD sample, NA03, was collected near the boundary with the biotite vein and the composition of pargasite is essentially the same in both the JD and BV parts. This inference provides a view that the composition of hydrothermal fluid in this area changed from early Na-rich fluid forming omphacite, jadeite and albite, passing through an evolved fluid precipitating Sr-Ti-Zr minerals, and finally to a K-rich fluid to form the BV.

The zircon age of 560 ± 16 Ma of this study is the oldest among zircon ages from Japanese accretionary complexes (e.g., Kunugiza and Goto, 2010, and Tsujimori, 2017; Tsujimori and Harlow, 2017) and those from jadeitite in the world (cf. Harlow et al., 2015). Zircons older than 590 Ma as inclusions in edenitic pargasite from the OE part sample 203A (Figs. 8c and 8d) may represent a previous event in a precursor rock. Nevertheless, the geological meaning of the 40 m.y. time gap be-

tween the older 560 ± 16 Ma and younger 519 ± 17 Ma ages remains unsolved, as exact age data overlap considerably between them. Possibly, even relict zircons of the OE sample experienced recrystallization due to huge hydrothermal activity to form omphacite, jadeitite, and albite.

Omphacite-bearing jadeitite as R-type

The omphacite-bearing jadeitite of the Itoigawa-Omi area has been classified as the R-type of Tsujimori and Harlow (2012), because of the presence of relict amphiboles in jadeitite (cf. Kunugiza and Goto, 2010). According to Harlow et al. (2015), by definition, R-type jadeitite partially preserves textural, mineralogical, or geochemical evidence of a pre-existing protolith. However, the R-type is rare, and only two localities from the western Italian Alps (Compagnoni et al., 2012) and the Nishisonogi area of Japan (Shigeno et al., 2012) have been cited as established examples. In the Monviso metaophiolite of the western Italian Alps, relics of igneous pyroxene containing oriented titanite needles occur as dusty cores of jadeite grains in a jadeitite block (Compagnoni et al., 2012). For the omphacite-bearing jadeitite of the Itoigawa-Omi area, the relict mineralogy of a precursor rock is edenitic pargasite and zircon with an age of ~ 560 Ma, and thus becomes an additional and rare example of R-type jadeitite.

Hydrothermal activity to form R-type jadeitite at time of subduction initiation

The rarity of R-type jadeitite could be explained by the rarity of huge hydrothermal activity in the vicinity of peridotite. As proposed by Tsujimori and Harlow (2012), P-type jadeitite requires a hydrothermal fluid capable of precipitating jadeite in veins and channels in relation to serpentinization of peridotite. In addition to such a hydrothermal fluid, R-type jadeitite requires the replacement and recrystallization of constituent minerals of a precursor rock to a considerable extent, and thus the huge hydrothermal activity keeping a chemically open system.

Huge hydrothermal activity near peridotite can be envisaged at subduction initiation. Subduction initiation begins to serpentinize the still hot peridotite of a newly formed mantle wedge, thereby the huge amount of hydrothermal fluid flow forms jadeitite, especially R-type jadeitite. After the subduction initiation, the subduction zone becomes cool and the proceeding of serpentinization of hanging wall peridotite loses the potential to provide silica-deficient fluid to precipitate jadeite. Hence, P-type jadeitite can be precipitated, but R-type jadeitite is hardly

formed in high a P/T type subduction zone.

The earliest Paleozoic age of ~ 520 Ma of jadeitite in this area is accepted as the timing of subduction initiation to form the accretionary complex of Japan (e.g., Kunugiza and Goto, 2010). Maruyama et al. (1997) examined the paleogeographic maps of the Japanese Islands and proposed a tentative age of ca. 500 Ma for the change from a passive margin to an active margin of the Proto-Pacific Plate. After their study, zircon ages of around 500 Ma have been accumulated from jadeite rock, meta-granite, and serpentinite. In the Kochi area of the Kurosegawa tectonic belt, an outer layer of the Hida-Gaien belt (Isozaki and Maruyama, 1991) yielded a zircon age of 501 ± 5 Ma from jadeite-quartz-glaucophane rock (Tsutsumi et al., 2010), and considered it as a protolith age of felsic igneous rock origin. The zircon ages of 467 ± 3 Ma and 488 ± 3 Ma were found from serpentinite of the Kochi area and interpreted as protolith age, the zircon of which was formed by melt infiltration during serpentinization and rodingitization (Hu et al., 2017). The meta-granite of the Hitachi metamorphic belt contains inherited zircon of ~ 500 Ma (Sakashima et al., 2003; Tagiri et al., 2011). The detrital zircons ranging 430–540 Ma in ion-probe age have been described in the Renge metamorphic rocks (Tsutsumi et al., 2003, 2010). Therefore, the hydrothermal event of ~ 520 Ma appears to have a huge scale and potential to form R-type jadeitite.

CONCLUDING REMARKS

Omphacite of the omphacite-bearing jadeitite of the Itoigawa-Omi area is a pseudomorph after a relict edenitic pargasite of a precursor rock formed by hydrothermal activity to precipitate jadeite and albite. The omphacite-bearing jadeitite is thus one of typical R-type jadeitite, and omphacite is not a postdated mineral after jadeitite minerals. In-situ LA-ICP-MS dating of zircons included in edenitic pargasite and omphacite of jadeitite with pseudomorphic omphacite yielded more than 590 Ma, and supports the presence of a precursor rock including edenitic pargasite. Zircons from jadeitite without pseudomorphic omphacite yielded ~ 519 Ma, being similar to the previous ion-microprobe ages of this area, and strengthens the notion that ~ 520 Ma is the age of subduction initiation to form the Japanese Islands. Subduction initiation begins to serpentinize the still hot peridotite of a newly formed mantle wedge; therefore, the huge amount of hydrothermal fluid flow formed jadeitite, especially R-type jadeitite.

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