1 Revision 1

Adakite metasomatism in a back-arc mantle peridotite xenolith from the Japan Sea

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Abstract

11 Secondary orthopyroxenes occur as veinlets (<0.1 mm thick) cutting an olivine 12in a two-pyroxene peridotite xenolith frongsthine Shiribeshi Seamount in the Japan Sea. These orthopyroxenes are characterized by low Al₂O₃ (0.4–1.7 wt%), Cr₂O₃ (<0.1 wt%), 1314and CaO (0.3–0.4 wt%) contents, which are the same signatures of the secondary 15orthopyroxenes in peridotite xenoliths from island arcs. The trace-element patterns of 16 the melts in equilibrium with the secondary orthopyroxenes show enrichment in light 17rare earth element and Sr and depletion in heavy rare earth element, Nb and Ti. These 18 trace-element characteristics are highly consistent with those of slab-derived adakites. 19 The involvement of slab-derived melts in the mantle beneath the Japan Sea has been 20inferred from the geochemical characteristics of the volcanic rocks formed during 21opening of the Japan Sea. The source mantle of the enriched basalts in the Japan Sea is 22likely to have been metasomatized by adakitic melts in the same manner as the 23peridotite-hosted veinlet. The secondary orthopyroxenes in the peridotite xenolith from 24the Shiribeshi Seamount provide direct evidence for the metasomatic influx of adakitic 25melts into the back-arc mantle beneath the Japan Sea. Adakitic metasomatism, as 26documented in the Japan Sea, potentially plays an important role in mantle evolution 27and magma generation beneath global back-arc basins.

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29 Key-words: Mantle xenolith, Secondary orthopyroxene, Japan Sea, Adakite

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31 INTRODUCTION

The addition of H₂O drastically reduces the solidus temperatures of mantle peridotites (Kushiro, 1972); therefore, the influx of H₂O-rich substances released from the subducting slab into the overlying mantle wedge contributes significantly to the genesis of island arc magmatism (e.g., Tatsumi and Eggins, 1995). Recent active petrological investigations of peridotite xenoliths from island-arc volcanoes have

37 provided possible evidence for metasomatism caused by inputs of subduction 38components released from the subducting slab. In particular, secondary orthopyroxene 39 replacing olivine appears in island-arc peridotite xenoliths (e.g., Arai and Kida, 2000; 40 Arai and Ishimaru, 2008; Bali et al., 2007; Bénard and Ionov, 2012, 2013; Franz et al., 2002; Grégoire et al., 2001; Halama et al., 2009; Ishimaru et al., 2007; McInnes et al., 41422001, Shimizu et al., 2004). The presence of such secondary orthopyroxene implies that 43a possible metasomatic reaction between slab-derived Si-rich melts or fluids and 44peridotites in the mantle wedge takes place beneath island arcs. Ishimaru et al. (2007) 45and Shimizu et al. (2004) proposed that orthopyroxene replacing olivine between 46peridotite and felsic rock vein in island-arc peridotite xenoliths resulted from the 47reaction with slab-derived adakitic melts. Moreover, metasomatic melt inclusions with 48adakitic geochemical affinities have been also reported in island-arc peridotite xenoliths 49(Bali, 2008; Schiano et al., 1995).

50Several overviews of global back-arc magmatism and mantle processes from the 51viewpoint of petrology and geochemistry of back-arc basin basalts (BABB) have been 52carried out (e.g., Pearce and Stern, 2006; Taylor and Martinez, 2003), but studies using 53mantle peridotites have not yet been performed, due to their rarity. The Japan Sea is a 54Miocene back-arc basin formed by the detachment of the Japan arcs from the Asian 55continent (e.g., Kaneoka et al., 1992; Nohda, 2009; Otofuji et al., 1985; Tamaki et al., 1992). We have recently discovered mantle peridotite xenoliths enclosed in basaltic to 5657andesitic lavas from the Shiribeshi Seamount in the Japan Sea (Ichiyama et al., 2013, 582016; Fig. 1). Interestingly, veinlet-like secondary orthopyroxenes replacing olivine, as 59observed in island-arc peridotite xenoliths, are present in a sample of these peridotite 60 xenoliths. The secondary orthopyroxenes in the peridotite xenolith from the Japan Sea 61can provide important information on the mantle metasomatic processes during the 62 Japan Sea opening, and possibly global back-arc basins worldwide. In this study, we 63 present new geochemical data for the secondary orthopyroxenes in the mantle 64 peridotite from the Japan Sea, and discuss their petrogenesis and significance for Japan 65Sea magmatism.

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BRIEF OVERVIEW OF THE JAPAN SEA AND DESCRIPTION OF THE PERIDOTITE XENOLITHS FROM THE SHIRIBESHI SEAMOUNT

The Japan Sea has three main topographic basins: the Japan Basin, the Yamato Basin, and the Tsushima Basin (Fig. 1). The basement of the Japan Basin is composed of basaltic oceanic crust formed during Miocene back-arc spreading, whereas those of the Yamato Basin and Tsushima Basin are continental crust thinned or extended by back-arc rifting (Tamaki et al., 1992). Drill sampling performed during the Ocean
Drilling Program (ODP) Legs 127 and 128 obtained samples of basaltic lavas and sills
interbedded with Miocene sediments from three sites in the Japan Basin and Yamato
Basin (e.g., Allan and Gorton, 1992; Fig. 1). These volcanic rocks yield 17–20 Ma Ar–Ar
radiometric ages, indicating formation during the Miocene opening of the Japan Sea
(Kaneoka et al., 1992).

79The Shiribeshi Seamount is a Quaternary submarine stratovolcano formed on the 80 northeastern margin of the Japan Basin (Fig. 1). The Shiribeshi Seamount is a volcano 81 on back-arc side in the Northeast Japan Arc, and is composed mainly of high-K 82 calc-alkaline basaltic to andesitic lavas (Ichiyama et al., 2013; Shuto et al., 1991). Lava 83 samples were collected from dive sites on the flank and around the summit of the 84 seamount using the manned submersible SHINKAI 2000 during cruises performed by the Japan Agency for Marine-Earth Science and Technology (JAMSTEC). The detailed 85 86 metadata (collecting date, collecting point, ship name, etc.) and petrology of the host 87 volcanic rocks with the peridotite xenoliths were provided in the GANSEKI database¹ 88 (Ichiyama et al., 2011) and Ichiyama et al. (2013), respectively. The basaltic to andesitic lavas contain abundant small-sized peridotite xenoliths (<2 cm) of two-pyroxene 89 90 peridotites (lherzolites to harzburgite) and dunites. Ichiyama et al. (2016) suggested 91that the two-pyroxene peridotites represent residues after decompression melting 92during the opening of the Japan Sea, and that the dunites are products formed via the 93 reaction between the two-pyroxene peridotites and the pyroxene-undersaturated 94 magmas formed prior to the host magmas. The secondary orthopyroxenes occur as thin 95 mono-mineralogic veinlets (<0.2 mm thick) replacing olivine in a two-pyroxene 96 peridotite (Fig. 2). Importantly, all peridotite xenoliths from the Shiribeshi Seamount 97 have the fine-grained secondary reaction products of olivine + chromian spinel + glass 98 formed along the boundaries between the peridotites and their host lavas, but reaction 99 rims of orthopyroxene are not observed along any boundary.

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101 ANALYTICAL METHODS

102 The major-element concentrations of the secondary orthopyroxenes and olivines in 103 the veinlet-intruded peridotite were measured using electron microprobe analyzers 104 installed at Chiba University (JEOL JXA-8230) and JAMSTEC (JEOL JXA-8800). 105 Natural and synthetic minerals were used as standard samples. Acceleration voltage 106 and beam current in the analysis were set at 15 kV and 20 nA, respectively, with a probe 107 diameter of 3 μm. Trace-element concentrations (rare earth element [REE], Ti, Sr, Y, Zr,

¹ URL: http://www.godac.jamstec.go.jp/ganseki/search/e

Nb, and Ba) were determined using a laser-ablasion inductively coupled plasma mass
spectrometer (Agilent 7500s with 193 nm ArF excimer MicroLas GeoLas Q-plus laser)
at Kanazawa University with a laser spot of 110 μm. Details of the methods followed,
including analytical accuracy and precision, are described in Morishita et al. (2005).
Representative results are given in Table 1 for major element and Table 2 for trace
element.

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115 **Results**

The secondary orthopyroxenes are characterized by much lower CaO (0.3-0.4 wt), 116 117 Al_2O_3 (0.4–1.7 wt%), and Cr_2O_3 (mostly <0.1 wt%) contents than those in the two-pyroxene peridotites (Fig. 3). The Mg# (= Mg/[Mg + Fe]) values are between 0.88 118 119 and 0.89. The trace-element contents of the secondary orthopyroxenes are markedly low, 120especially for middle REE (MREE) such as Eu, Gd, and Tb, which were below the 121detection limit (Fig. 4a). The primitive mantle-normalized trace element patterns of the 122secondary orthopyroxenes exhibit slight enrichment in light REE (LREE) without any 123significant troughs and spikes. The secondary orthopyroxenes differ from those in the 124two-pyroxene peridotites in possessing a higher LREE content (Fig. 4a).

125 Olivines in the veinlet-intruded two-pyroxene peridotite have values of NiO = 0.36-0.39 wt.% and MnO = 0.25-0.32 wt.% in Mg# = 0.87, and are lower in Mg# than 127 those in the other two-pyroxene peridotites at the same NiO content (Fig. 5).

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129 **DISCUSSION**

130 Origin of the secondary orthopyroxenes: Evidence for reaction with adaktic melts

131 The lack of orthopyroxene reaction rims along the boundaries between the peridotite 132xenoliths and the host lavas from the Shiribeshi Seamount indicates that the secondary 133orthopyroxenes in the peridotite xenolith cannot be attributed to the reaction with the 134host magmas during transportation to the surface. The secondary orthopyroxenes are 135characterized by lower contents of CaO, Al₂O₃, and Cr₂O₃ than those in the residual 136two-pyroxene peridotites and host lavas (Fig. 3). The trace-element patterns of the secondary orthopyroxenes show slight LREE enrichment, which is different from the 137138highly depleted patterns expected for residual peridotites after simple decompression 139melting caused by the back-arc spreading that formed the Japan Basin (Ichiyama et al., 1402016; Fig. 4a).

141 The reaction of olivine with Si-rich melt can generate orthopyroxene products (e.g., 142 Kelemen et al., 1998). The petrological and geochemical evidence described above 143 suggests that the secondary orthopyroxenes can be explained by the reaction between

144the mantle peridotites and SiO₂-saturated melts. Arai and Ishimaru (2008) reviewed 145island-arc peridotite xenoliths and suggested that secondary orthopyroxenes in 146island-arc peridotites can be formed by the reaction of olivines with Si-rich liquids 147derived from the subducted slab. Moreover, secondary orthopyroxenes replacing olivines are generally lower in CaO, Al₂O₃, and Cr₂O₃ contents than primary residual 148149orthopyroxene. For instance, the secondary orthopyroxenes in peridotite xenoliths from 150the Avacha Volcano in Kamchatka (Ishimaru et al., 2007) and from the Tallante area, 151Spain (Shimizu et al., 2004), are very similar to those from the Shiribeshi Seamount (Fig. 3). 152

153The vein-like secondary orthopyroxenes in the Avacha peridotite xenoliths show a 154trace-element pattern similar to those in the Shiribeshi peridotite, with the exception of 155significant positive Zr and Hf anomalies in the former (Fig. 4a). Ishimaru et al. (2007) 156interpreted that the secondary orthopyroxenes in the Avacha peridotites were produced 157by the infiltration of slab-derived adakitic melts into the mantle wedge. In contrast, Bénard and Ionov (2013) argued that these secondary orthopyroxenes resulted from the 158159fractionation-reactive percolation of boninite melts into the mantle wedge, which could 160cause the percolating melts to become enriched in LREE and Zr-Hf via assimilation and 161re-equilibration with peridotite wall rocks. Although the origin of the secondary 162orthopyroxene veins in the Avacha peridotite xenoliths is still controversial, there is no 163 such Zr anomaly in the trace-element patterns of the secondary orthopyroxenes in the 164 Shiribeshi peridotite; therefore, it is unlikely that fractionation-reactive percolation 165took place effectively in the orthopyroxene veinlets. The patterns of these veinlets are 166 closer to those of an orthopyroxene phenocryst in a magnesian adakite from the 167 Izu-Bonin-Mariana (IBM) arc (Li et al., 2013) than those in a typical calc-alkali 168island-arc andesite in the Northeast Japan arc (Kobayashi and Nakamura, 2001; Fig. 1694a). We calculated the trace-element compositions of the melts in equilibrium with the 170secondary orthopyroxenes using two datasets of partition coefficients between 171orthopyroxene and melt: (1) hydrous basaltic melt and orthopyroxene experimentally 172determined by McDade et al. (2003); and (2) whole-rock and orthopyroxene phenocryst 173calculated from an IBM magnesian adakite of Li et al. (2013; Table 2). The melt 174compositions calculated using these two datasets are characterized by enrichment in 175LREE and depletion in Nb and heavy REE (Fig. 4b). Additionally, the patterns 176calculated using the latter partition coefficients show distinct Sr spikes and possible Ti 177troughs. These chemical characteristics of the calculated melt compositions are in good 178agreement with those of an average adakite and adakites from the IBM arc and Japan 179Sea (Fig. 4b). Therefore, these results suggest that the causal agent for the secondary orthopyroxenes in the Shiribeshi Seamount was adakitic melts, which are likely
generated by melting of the subducting slab (e.g., Drummond et al., 1996; Martin et al.,
2005).

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184 IMPLICATIONS FOR ADAKITIC MELT METASOMATISM BENEATH THE JAPAN SEA

185Basalts drilled from the floor of the Japan Sea can be subdivided into two types on the basis of their geochemical characteristics: depleted (D-type) and enriched (E-type) 186basalts. D-type basalts have geochemical signatures similar to those of mid-ocean ridge 187188 basalt, whereas E-type basalts are characterized by the enrichment of large-ion 189 lithophile elements and LREE and by radiogenic Pb isotopic composition compared with 190 the D-type basalts (Allan and Goton, 1992; Cousens et al., 1994; Hirahara et al., 2015). 191Hirahara et al. (2015) suggested that the source mantle of the E-type basalts was 192metasomatized by the influx of melts derived from subducting slab sediments, while 193 that of the D-type basalts was more depleted and higher mantle potential temperature than that of the E-type basalts. Tamura (2003) estimated the primary magma 194195compositions for the D-type and E-type basalts (Fig. 5), and revealed that the E-type primary magma (and source mantle in equilibrium with it) was Fe-rich in composition 196 197 relative to the D-type. Tamura (2003) proposed that slab-melt from subducted sediment 198possibly added to Fe-rich E-type mantle source. The presence of such slab melts is 199 supported by the formation of Miocene adakites in back-arc margins during the opening 200 of the Japan Sea: these adakites were derived from the melting of altered basaltic crust 201and sediment caused by the upwelling of the asthenospheric mantle into the mantle 202wedge (Fig. 4b; Sato et al., 2012). Ichiyama et al. (2016) proposed that the peridotite 203xenoliths from the Japan Sea were the residues after the extraction of the D-type and 204E-type basaltic melts, on the basis of the clinopyroxene REE and chromian spinel 205compositions, and that the E-type residues underwent higher degrees of partial melting 206than the D-type, due to the reduction of the solidus temperature possibly by H₂O supply 207via infiltration of the slab-derived melts.

As discussed above, the veinlet-forming secondary orthopyroxenes in the 208two-pyroxene peridotite xenolith from the Shiribeshi Seamount should have been 209210produced by the infiltration of slab-derived adakitic melts. The olivines in the 211veinlet-intruded peridotite xenolith are plotted in a FeO-rich direction relative to those 212in other common two-pyroxene peridotites, while maintaining the same NiO content 213(Fig. 5). This should have been caused by Si-rich adakitic metasomatism because high 214compatibility of Ni for olivine can maintain the Ni budget for olivine during olivine 215consumption (Kelemen et al., 1998) despite the increase in Fe/Mg ratios. Interestingly,

216the olivine composition in the veinlet-intruded peridotite is highly consistent with the 217calculated olivine composition in equilibrium with the primary E-type magmas 218estimated by Tamura (2003; Fig. 5). Metasomatism caused by adakitic melt infiltration 219into the mantle beneath the Japan Sea can also account for the Fe-enrichment and 220trace-element characteristics of the E-type basalts (Fig. 6). The secondary 221orthopyroxenes probably provide direct evidence for the metasomatic influx of 222slab-derived melts into the mantle beneath a back-arc basin in the Japan Sea. Pearce 223and Stern (2006) proposed, considering BABB geochemistry, that slab-derived melts (or 224supercritical fluid) were input into the source mantle of global BABB as a deep 225subduction component. Although adakitic volcanism on back-arcs is rare, adakitic 226metasomatism, as proposed in this study, potentially plays an important role in mantle 227 metasomatism and magma generation beneath global back-arc basins.

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229 **1. Conclusions**

- (1) Secondary orthopyroxenes occur as veinlets replacing olivine in two-pyroxene peridotite from the Shiribeshi Seamount in the Japan Sea. The orthopyroxenes are characterized by low Al₂O₃, Cr₂O₃, and CaO contents, and the trace-element patterns of the melt in equilibrium with them are very similar to those of adakites. The veinlet-forming secondary orthopyroxenes in the peridotite were produced by the reaction between mantle peridotites and slab-derived adakitic melts beneath the Japan Sea.
- 237(2) The involvement of adakitic melts in the mantle beneath the Japan Sea consistently 238accounts for the geochemical characteristics of the enriched basaltic magmas formed 239during the opening of the Japan Sea, and the secondary orthopyroxenes in the 240peridotite from the Shiribeshi Seamount provide direct evidence for the 241metasomatic influx of slab-derived melts into the mantle beneath the Japan 242Sea. Adakitic metasomatism, as documented in the Japan Sea, also potentially 243plays an important role in mantle evolution and magma generation beneath 244global back-arc basins.
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385 Figure Captions

386 Fig. 1

- Distribution of the basement geology in the northern Japan Sea (Tamaki et al., 1992)
 and location of the Shiribeshi Seamount. Drill sites during ODP (black dots) are also
 shown.
- 390
- 391 Fig. 2
- 392
- 393 The microphotographs were taken with cross-polarized light.
- 394
- 395 Fig. 3

Plots of (a) CaO and (b) Cr₂O₃ contents against Al₂O₃ content for the secondary orthopyroxenes in the Shiribeshi peridotite xenoliths. The primary orthopyroxenes in the two-pyroxene peridotites and orthopyroxene phenocrysts in the host lavas are from Ichiyama et al. (2016) and Ichiyama et al. (2012), respectively. The regression lines for the two-pyroxene peridotites are illustrated as thick gray lines. The secondary orthopyroxenes from the Avacha and Tallante peridotites are from Ishimaru et al. (2007) and Shimizu et al. (2004), respectively.

403

404 **Fig. 4**

405Primitive mantle-normalized trace element patterns of (a) the secondary 406 orthopyroxenes and (b) the melts in equilibrium with the secondary orthopyroxenes. For 407comparison, trace-element patterns of primary orthopyroxenes in the Shiribeshi 408 two-pyroxene peridotites, a secondary orthopyroxene in an Avacha peridotite (Ishimaru 409 et al., 2007) and an orthopyroxene phenocryst in a magnesian adakite from the IBM arc (Li et al., 2013) and in a calc-alkali andesite from the Northeast Japan arc (Akagi 410Volcano; Kobayashi and Nakamura, 2001) are illustrated in (a). Also, whole rock trace 411412element patterns of the Shiribeshi basaltic to andesitic host lavas, a boninite, and an 413adakite from the IBM arc (Li et al., 2013), an average adakite from the Japan Sea area 414(from Iwaine Formation) (Sato et al. 2012), and an average adakite (Drummond et al., 4151996) are illustrated in (b). McDounogh and Sun (1995) was used for normalization 416values.

417

418 **Fig. 5**

419 Plots of olivine Fo (= $100 \times Mg / [Mg + Fe]$) versus NiO contents of the two-pyroxene 420 peridotites. The olivine compositions in equilibrium with the primary D- and E-type 421 basalts are based on the calculation of Tamura (2003). The mantle olivine array is from 422 Takahashi et al. (1987).

423

424 **Fig. 6**

425 Schematic illustration of adakite metasomatism in the Japan Sea. Please see the text426 for details.













	Secondary Or	Primary				
SiO ₂	57.04	57.15	56.76	56.78	56.29	56.35
TiO ₂	0.02	0.00	0.04	0.00	0.08	0.02
Al_2O_3	1.04	0.83	1.45	1.26	2.41	2.24
Cr_2O_3	0.00	0.00	0.04	0.06	0.30	0.29
FeO*	7.81	8.18	8.18	7.89	6.23	6.31
MnO	0.34	0.32	0.30	0.30	0.18	0.16
MgO	33.64	33.74	33.83	33.71	33.99	34.61
CaO	0.35	0.36	0.34	0.29	0.60	0.43
Na ₂ O	0.00	0.00	0.01	0.02	0.03	0.00
K2O	0.01	0.00	0.01	0.00	0.01	0.01
Total	100.24	100.58	100.95	100.32	100.13	100.42
0	6	6	6	6	6	6
Si	1.974	1.975	1.956	1.965	1.941	1.937
Ti	0.001	0.000	0.001	0.000	0.002	0.001
Al	0.042	0.034	0.059	0.051	0.098	0.091
Cr	0.000	0.000	0.001	0.002	0.008	0.008
Fe*	0.226	0.236	0.236	0.228	0.180	0.181
Mn	0.010	0.009	0.009	0.009	0.005	0.005
Mg	1.736	1.738	1.737	1.739	1.747	1.774
Ca	0.013	0.013	0.012	0.011	0.022	0.016
Na	0.000	0.000	0.001	0.001	0.002	0.000
K	0.000	0.000	0.000	0.000	0.001	0.000
Total	4.002	4.006	4.012	4.006	4.005	4.013
Mø#	0 885	0.880	0 881	0 884	0 907	0 907

Table 1 Representative chemical composition of orthopyroxenes in the peridotite xenoliths from the Shiribeshi Seamount

*Total Fe as Fe²⁺

Table 2 Trace element and rare earth element contentrations (ppm) of sedondary orthopyroxenes in peridotite xenoliths from the Shiribeshi Seamount and partition coefficients used for the estimation of the melt compositions.

	Orthopyroxe	Partition coefficients			
Spot No.	#701	#702	#703	D^{*}	D^{**}
Ti	676	591	351	0.141	0.321
Rb	-	-	-	-	0.019
Sr	0.813	1.76	1.30	0.0044	0.003
Y	0.860	0.651	0.751	0.101	0.158
Zr	2.10	1.60	1.53	0.027	0.020
Nb	0.059	0.072	-	0.0028	0.070
Ba	0.387	2.12	2.22	-	0.004
La	0.095	-	0.049	0.003	0.008
Ce	0.382	0.560	0.158	0.005	0.011
Pr	0.048	0.063	0.023	-	0.015
Nd	0.173	0.254	0.103	0.009	0.018
Sm	-	0.050	-	0.021	0.032
Eu	-	-	-	0.031	0.049
Gd	-	-	-	-	0.088
Tb	-	-	-	-	0.104
Dy	0.112	0.080	0.077	-	0.155
Но	0.033	0.022	0.025	-	0.217
Er	0.129	0.096	0.105	0.121	0.250
Tm	0.027	0.022	0.025	-	0.305
Yb	0.253	0.199	0.261	0.164	0.291
Lu	0.043	0.038	0.055	0.186	0.288

*: McDate et al. (2003)

**: Calculated from orthopyroxene / host rock of the IBM adakite (sample: D22-53) from Li et al. (2013)