

Thermal and chemical evolution of the subarc mantle revealed by spinel-hosted melt inclusions in boninite from the Ogasawara (bonin) Archipelago, Japan

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3 (Bonin) Archipelago

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10 **ABSTRACT**

11 Primitive melt inclusions in chrome spinel from the Ogasawara Archipelago comprise
12 two discrete groups of high-SiO₂, MgO (high-Si) and low-SiO₂, MgO (low-Si) boninitic suites
13 with ultra-depleted dish- and V-shaped, and less depleted flat rare earth element (REE) patterns.
14 The most magnesian melt inclusions of each geochemical type were used to estimate the T-P
15 conditions for primary boninites, that range from 1345°C-0.56 GPa to 1421°C-0.85 GPa for the
16 48-46 Ma high-Si and low-Si boninites, and 1381°C-0.85 GPa for the 45 Ma low-Si boninite.
17 The onset of the Pacific Slab subduction at 52 Ma forced upwelling of depleted mid-ocean ridge
18 basalt mantle (DMM) to yield proto-arc basalt (PAB). With the rise of DMM, refractory
19 harzburgite ascended without melting. At 48-46 Ma, introduction of slab fluids induced melting
20 of the PAB residue and high-T harzburgite, resulted in the low-Si and high-Si boninites,
21 respectively. Meanwhile, convection within the mantle wedge brought the less depleted residue

22 of PAB and DMM into the region fluxed by slab fluids, which melted to yield the less depleted
23 low-Si boninite at 45 Ma, and fertile arc basalts, respectively.

24 **INTRODUCTION**

25 The Izu-Ogasawara (Bonin)-Mariana (or IBM) Arc has been a locus of intense study on
26 oceanic arc evolution because it provides complete records of the volcanic arc development since
27 the onset of the Pacific Plate subduction beneath the Philippine Sea Plate up to present (e.g.,
28 Ishizuka et al., 2006, 2011). Along the entire IBM forearc MORB-like proto-arc basalt (PAB;
29 Reagan et al., 2010) extruded at 52-48 Ma, followed by arc tholeiitic lavas on the Ogasawara
30 Archipelago (Umino et al., 2009). The dissected tholeiitic rocks are overlain by 48-46 Ma highly
31 depleted, high-silica boninite with minor low-silica boninite (Maruberiwan Formation), which
32 became covered with less depleted, low-silica boninite associated with calc-alkalic magmas at 45
33 Ma (Mikazukiyama Formation). By 44 Ma the volcanic front retreated westward and fertile arc
34 magmas (Hahajima Island Group basalt, or HGB) superseded boninites (Kanayama et al., 2012;
35 2014). This variation in magma geochemistry represents the thermo-chemical evolution of the
36 mantle wedge during the establishment of the IBM arc.

37 Boninites potentially provide significant constraints on the P-T-X conditions of the
38 varying mantle wedge because of their primitive compositions compatible with refractory upper
39 mantle minerals (Umino, 1986; Crawford et al., 1989; Pearce et al., 1992; Kanayama et al.,
40 2012). Specifically, melt inclusions in forsteritic olivine in boninite have been used for
41 estimating primary magma compositions, based on which Tongan low-Si boninite was proposed
42 to derive from hot mantle plumes with a mantle potential T (MPT) >1500°C (e.g., Sobolev &
43 Danyushevsky, 1994; Sobolev and Chaussidon, 1996). However, most olivine-hosted melt
44 inclusions suffered post-entrapment crystallization and diffusive reequilibration with the hosts,

45 which are irreversible by heating experiments (Kamenetsky et al., 2002; Kent, 2008). Especially
46 Fe-loss is critical in estimating T-P conditions of primary boninite. Therefore, the original melt
47 compositions upon entrapment were restored by raising FeO to the liquid line of descent, which
48 was assumed to be equal to bulk compositions. Unfortunately, bulk compositions of porphyritic
49 arc magmas like boninites mostly differ from the liquid compositions due to processes such as
50 crystal accumulation and imperfect fractionation, and mixing of magmas.

51 In contrast, chrome spinel is an excellent container of primitive magmas because the
52 trapped liquid compositions are little modified by crystallization of the host spinel due to limited
53 Cr contents in melts (e.g., Shimizu et al., 2001). Furthermore, chrome spinel is resistant to
54 deterioration compared to silicate minerals such as olivine, and chemically stable against low-
55 grade metamorphic alteration. Chrome spinel is also eminently pressure-resistant as it may
56 preserve ultra-high-pressure minerals (Yamamoto et al., 2009), so it is expected that chrome
57 spinel may preserve primitive compositions of inclusions trapped under high temperatures and
58 pressures.

59 This paper focuses on primitive melts preserved in chrome spinel, the earliest crystallized
60 phase from boninite magmas in the Ogasawara Archipelago (Umino, 1986), to estimate the
61 genetic conditions for primary magmas, and discusses the thermo-chemical evolution of the
62 mantle wedge during the early stages of the IBM subduction zone development.

63 **SPINEL-HOSTED MELT INCLUSIONS**

64 **Mode of Occurrence of Melt Inclusions**

65 Boninites are widely exposed on the Mukojima and Chichijima islands, Ogasawara
66 Archipelago, where chrome spinel samples were taken from the beach sand “Uguisu-zuna”,
67 natural mineral concentrates of orthopyroxene and minor amounts of olivine, spinel +/-

68 clinopyroxene (Umino et al., 2009). Poorly developed river systems and generally consistent
69 geochemistry and mineralogy of Uguisu-zuna and the surrounding geology support the view that
70 those mineral concentrates were eroded from the nearby exposures and wave-sorted (Fig. DR-1).
71 Melt inclusions are randomly distributed within spinel and have mainly oval or rounded
72 triangular shapes constrained by crystallographic orientation of the host spinel. This indicates a
73 primary origin for these inclusions trapped during the growth of the host spinel (Schiano, 2003).
74 All melt inclusions in a single spinel crystal have the same texture and geochemical
75 characteristics, indicating that these inclusions were trapped at the same time and experienced
76 the same cooling history during ascent and quenching upon eruption.

77 Melt inclusions consist of glass with or without crystals. 21% of the inclusions examined
78 are of entirely homogeneous glass (Figs. 1A, B, DR-2A, B) with or without shrinkage bubbles on
79 the wall. The rest 79% carry daughter or captured crystals in addition to glass. Rare melt
80 inclusions develop an outer zone of rapidly grown daughter pyroxene crystals surrounding a
81 homogeneous glass pool in the core (Figs. 1C, DR-2C). However, both the outer zone and the
82 core have identical bulk major element compositions (Fig. DR-3). About 27% of inclusions are
83 “variolitic”, composed entirely of variolitic pyroxene crystals, interstitial glass and vesicles (Figs.
84 1D, DR-2D). Both the “melt (glass)” dominant inclusions and the variolitic inclusions lack any
85 compositional gradient through the inclusions and in the adjacent host spinel (Figs. 1, DR-2).
86 The textural relationships and chemical homogeneity of these melt inclusions strongly suggest
87 that the primary melt compositions at the time of entrapment are well preserved, in spite of the
88 postentrapment crystallization in some inclusions. We therefore take the bulk compositions of
89 these melt-dominant and variolitic inclusions as representative of quenched pristine melt
90 compositions.

91

92 **Major and Trace Element Geochemistry**

93 The melt inclusions have 52-63 wt% SiO₂, 7-23 wt% MgO and 0.01-0.25 wt% TiO₂ on
94 an anhydrous basis (Fig. 2; Table DR-1). With few exceptions, almost all fit the category of
95 boninite defined by the IUGS (SiO₂ >52 wt%, MgO >8 wt% and TiO₂ <0.5 wt%; Le Maitre,
96 2002). Most inclusions form two roughly subparallel groups showing an inverse correlation of
97 MgO and SiO₂, which are generally consistent with fractionation paths of olivine followed by
98 orthopyroxene (based on alphaMELTS by Smith and Asimow, 2005), combined with mixing of
99 melts on the paths (Figs. 2A, DR-5). One group with higher MgO and SiO₂ falls within the field
100 of high-silica boninite of Kanayama et al. (2012), but are more Mg-rich than the whole rocks at a
101 given SiO₂ (“high-Si (boninitic) inclusions”, hereafter). The other group is somewhat scattered
102 but plot between the high-Si inclusions and the bulk low-Si boninites, and are hence called “low-
103 silica (low-Si) (boninitic) inclusions”. Almost all high-Si inclusions have low CaO (<8 wt%) and
104 CaO/Al₂O₃ ratios <0.8, characteristics of low-Ca boninite (Crawford, 1989). On the other hand,
105 the low-Si inclusions have 6.5-12 wt% CaO and spread over the fields of both low-Ca and high-
106 Ca boninites.

107 High-Si boninitic inclusions are severely depleted in REEs with chondrite-normalized
108 patterns, which can be divided into dish-shaped (high-Si-D) and V-shaped (high-Si-V) at a
109 Gd/Yb ratio of 0.7 (Fig. 2B). Dish-shaped REE patterns are identical to those of the bulk
110 boninites in overall shape and abundance (Fig. DR-6). On the contrary, V-shaped REE patterns
111 have never been found among the bulk rocks. High-Si-D and high-Si-V inclusions are enclosed
112 in spinel with Cr/ (Cr + Al) of 0.85-0.91 and TiO₂ <0.12 wt% derived from the Maruberiwan

113 Formation. Although individual spinel grains have only a single type of inclusions, both high-Si-
114 D and high-Si-V inclusions coexist in spinel samples from the same locality.

115 The low-Si boninitic inclusions are of two distinct types. One has less depleted, flat REE
116 patterns with low $Zr/Ti < 0.02$ (low-Si-F), and is only found in spinel derived from the
117 Mikazukiyama Formation (Fig. 2C). Most of them possess a high-Ca boninitic affinity with
118 CaO/Al_2O_3 ratios > 0.8 . The other type, low-Si-D inclusions, is found in spinel from the
119 Maruberiwan Formation and shows depleted dish-shaped REE patterns with variable Zr/Ti ratios
120 (0.01-0.03), CaO (6-11 wt%) and CaO/Al_2O_3 ratios (0.5-1). The host spinel of the low-Si
121 inclusions has low $Cr/(Cr + Al)$ of 0.72-0.87 and 0.04-0.24 wt% TiO_2 .

122 The variolitic melt inclusions (Fig. 1D) tend to show lower H_2O contents (< 2.4 wt%) and
123 H_2O/Ce ratios than the other inclusions (Fig. DR-4), suggesting water loss due to degassing. We
124 therefore corrected water contents of variolitic samples by assuming their original H_2O/Ce ratios
125 to be equal to the average H_2O/Ce ratios of undegassed samples of the same geochemical type.
126 Because all low-Si-F inclusions exhibit a variolitic texture, the H_2O contents were restored based
127 on the maximum H_2O/Ce ratio of the type, which gives the minimum H_2O estimates. The
128 restored H_2O and H_2O/Ce ratios mostly range from 2 to 5 wt% and 9800-43200, respectively.

129 **Genetic Conditions of Primary Boninites**

130 If magma retains its primary composition, it should be multiply saturated with olivine and
131 orthopyroxene at the temperature and pressure of segregation, which can be estimated by the
132 combination of appropriate geothermobarometers. We applied the method of Putirka (2008)
133 based on olivine-liquid equilibration (Putirka et al., 2007) and olivine-orthopyroxene-liquid
134 equilibration (Putirka, 2008). Olivine-spinel equilibria suggests oxygen fugacity of boninite
135 magma to be 0 to +1 in log unit above fayalite-magnetite-quartz (FMQ) buffer (Evans et al.,

136 2012), whereas $\text{Fe}^{3+}/\text{Fe}^{2+}$ ratios of pillow margin glass estimated by XANES (Ishibashi et al.,
137 2012) and sulfur speciation of melt inclusions in spinel determined by soft X-ray photoelectron
138 spectroscopy (Shimizu et al., 2014) indicate f_{O_2} of $\Delta\log(\text{FMQ}) = +1 \sim +2$ for the Ogasawara
139 boninite. We therefore assumed f_{O_2} of $\Delta\log(\text{FMQ}) = 1 \pm 1$ for the T-P estimates. CO_2
140 concentrations of boninitic melt inclusions in chrome spinel from the Ogasawara Archipelago are
141 very low (<50 ppm; Shimizu and Shimizu, 2013) and hence H_2O is considered as the only
142 volatile species. The compositions of primary boninite magmas are assumed to be those of the
143 most magnesian melt inclusions among the four geochemical types described above, i.e. high-Si-
144 D, high-Si-V, low-Si-D and low-Si-F melt inclusions (Table DR-1). The estimated equilibration
145 T and P are 1343-47°C at 0.54-0.59 GPa for the low-Si-D boninite, and 1415-28°C at 0.74-0.96
146 GPa for the high-Si-D and high-Si-V boninites (Fig. 3). The genetic T-P of 1378-1384°C and
147 0.81-0.88 GPa for the low-Si-F boninite are considered to be the maximum estimates, because its
148 H_2O content is the minimum estimate.

149 **DISCUSSION**

150 There is a general consensus that boninitic magmas derive from severely depleted source
151 mantle which experiences flux melting by introduction of hydrous fluids (or melts) liberated
152 from a descending slab (e.g. Pearce et al. 1992; Bédard, 1999; Kanayama et al., 2012). These
153 studies ascribed high Zr/Ti ratios of high-Si boninites to contribution of slab melt under
154 amphibolite facies. The degree of depletion of the source mantle can be assessed by
155 concentrations of heavy REEs such as Yb, which are predominantly immobile through slab
156 dehydration and sediment partial melting. Figure 4 shows Yb-Zr/Ti variations modeled by 5%
157 and 20% batch partial melts of residual DMM (Workman and Hart, 2005) after extraction of 0%
158 to 20% fractional melts, mixed with 5% batch partial melt of amphibolitic slab (based on altered

159 oceanic basalt at Site 1149; Kelley et al., 2003). The model indicates that Yb concentrations are
160 more sensitive to the degree of source depletion than that of batch melting. Both the PAB and the
161 HGB have the least depleted source mantle and little contribution from slab melts. Irrespective to
162 the degree of melting of the residual DMM, the highest Yb and the least Zr/Ti for the 45 Ma low-
163 Si-F inclusions indicate less than 11-16% source depletion and contribution of 8-15% slab melt,
164 smallest among all melt inclusions. The 48-46 Ma high-Si-D and high-Si-V inclusions have the
165 least Yb and the highest Zr/Ti, indicating the largest source depletion up to 19-24% and mixing
166 of 10-20% slab melt. Although the 48-46 Ma low-Si-D inclusions are similarly low in Yb, the
167 model suggests slightly lower depletion of 17-20%.

168 Subsidence of the old, dense Pacific Plate caused forearc spreading and decompression
169 melting of DMM-like asthenosphere to form MORB-like PAB magma for the first few million
170 years after the subduction initiated (Ishizuka et al., 2011; Kanayama et al., 2012). If the PAB
171 source had a MPT of $\sim 1410^{\circ}\text{C}$, which is slightly above the present suboceanic ambient mantle
172 (Herzberg et al., 2007), the residue would become $\sim 1345^{\circ}\text{C}$ at 0.5 GPa, coincident with the T-P
173 conditions for the 48-46 Ma low-Si-D boninite magma, supporting the remelting of the PAB
174 residue (Fig. 3). However, it is incompatible with the coexistence of extremely hot ($>1400^{\circ}\text{C}$ at
175 0.8 GPa), severely depleted source mantle of the high-Si boninite magmas. If, as the plume-
176 origin model of boninite suggests, the pre-boninite decompression melting of the DMM was
177 solely responsible for the extreme depletion of the boninite source, it should have ascended from
178 the depth of 3.5 GPa with a MPT of 1500°C . The ascent of such high-T peridotite to <1 GPa
179 should cause extensive decompression melting to produce picritic magmas, which have never
180 been found among the pre-boninite PAB. This indicates that the depleted proto-boninite source
181 already existed below the DMM-like PAB source before the subduction began. This is supported

182 by the unradiogenic Os isotopic composition of harzburgite drilled in the Izu Forearc, which
183 experienced melt extraction in Proterozoic age and became the source for the IBM boninite
184 magmas (Parkinson et al., 1998; Suzuki et al., 2011). With the DMM asthenosphere, the
185 refractory harzburgitic source ascended along the adiabatic decompression path without melting,
186 resulted in the high temperatures of the high-Si boninites (Fig. 3). Considering 1380°C and 0.8
187 GPa are the maximum estimates for the low-Si-F boninite genesis, it would be close to the
188 adiabatic melting path of the PAB source. Compared to the low-Si-D boninite, the low-Si-F
189 boninite has the higher T-P and less depleted source, suggesting its derivation from the periphery
190 of the asthenospheric upwelling, which suffered lower degrees of melting and ascended off the
191 forearc spreading axis. By 45 Ma, convection in the wedge mantle driven by drag force of the
192 subducting slab brought the less depleted low-Si-F boninite source and DMM-like asthenosphere
193 into the region fluxed by slab fluids, which melted to generate the low-Si-F boninite magma, and
194 then arc tholeiitic and calc-alkalic magmas on the Hahajima Island Group and the western scarps
195 of the Ogasawara Ridge.

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312 **FIGURE CAPTIONS**

313 Figure 1. Representative COMPO images (left) and electron microprobe analyzer (EMPA)
314 elemental maps showing textures, and Fe and Mg distributions respectively of melt inclusions

315 and the host chrome spinel. Note the absence of Fe-rich and Mg-poor halos in the spinel host
316 adjacent to the melt inclusions, confirming the absence of diffusive Fe-loss from the melt.
317 Figure 2. Boninitic melt inclusions (circles and squares) compared with bulk boninites (crosses)
318 of the 48-46 Ma Maruberiwan (Mb) and 45 Ma Mikazukiyama (Mk) Formations from
319 Ogasawara. Examples of fractionation paths for primary boninites (solid and broken lines) are
320 shown at 0.2 GPa and fO_2 of 0.5 log unit above fayalite-magnetite-quartz (FMQ) buffer using
321 alphaMELTS (A), chondrite-normalized REE patterns of high-Si-D and high-Si-V (B) and low-
322 Si-D and low-Si-F (C) inclusions. Broken lines encircle the bulk boninites. See Fig. DR-5 for
323 further explanation.

324 Figure 3. T-P estimates of multiple saturation for the primary boninite magmas. The ranges of T-
325 P estimates denote 1σ of H_2O and fO_2 of $\Delta\log(FMQ) = 1\pm 1$. Note that varying fO_2 by 2 in log
326 unit brings a little difference in T-P estimates, but increase in H_2O content by 1 wt% reduces the
327 estimated T and P by $34^\circ C$ and 0.17 GPa, respectively. Adiabatic melting paths are after
328 Herzberg and Gazel (2009). The broken arrow is that for the DMM asthenosphere with $MPT =$
329 $1410^\circ C$. The solid arrow is an adiabatic decompression path of the harzburgite source for the
330 high-Si boninite, calculated using $dT/dP = 13.3^\circ C/GPa$ (Putirka et al., 2007). The harzburgite
331 solidus is based on the residual DMM after 20% fractional melting by alphaMELTS.

332 Figure 4. Yb vs Zr/Ti of boninitic melt inclusions compared with pre-boninite PAB, bulk
333 boninites and post-boninite HGB. Symbols are the same as in Fig. 2. The Yb-Zr/Ti relationships
334 is modeled by mixing of 5% (gray) and 20% (black) batch partial melts of residual DMM after
335 extraction of 0-20% fractional melts (denoted such as "10%"), and 5% batch partial melt of
336 amphibolitic slab (based on altered oceanic basalt at Site 1149; Kelley et al., 2003) with a
337 proportion of clinopyroxene: amphibole:plagioclase = 7:88:5 (Taylor et al., 1994). Numbers on

338 the broken lines (“70:30”, etc.) are the proportions of peridotite melt:slab melt. Increase in slab
339 melting up to 15% does not change the model results.

340 ¹GSA Data Repository item 2014xxx, Boninitic melt inclusions in chrome spinel from the
341 Ogasawara Archipelago, is available online at www.geosociety.org/pubs/ft2014.htm, or on
342 request from editing@geosociety.org or Documents Secretary, GSA, P.O. Box 9140, Boulder,
343 CO 80301, USA.

Fig. 1

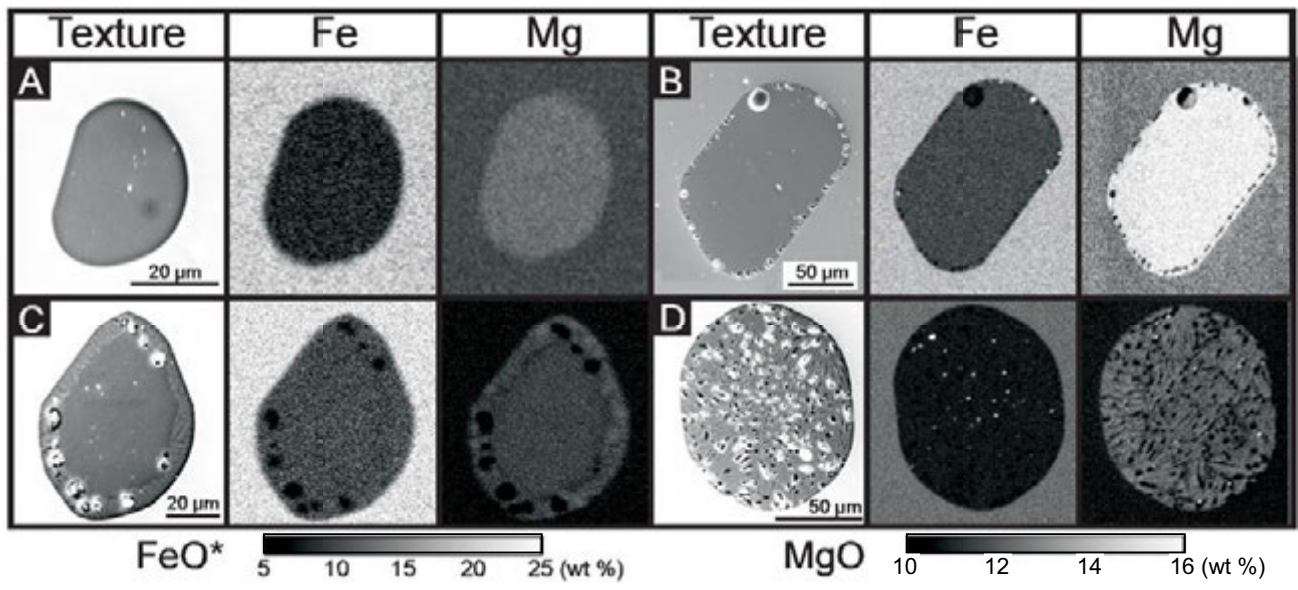


Fig. 2

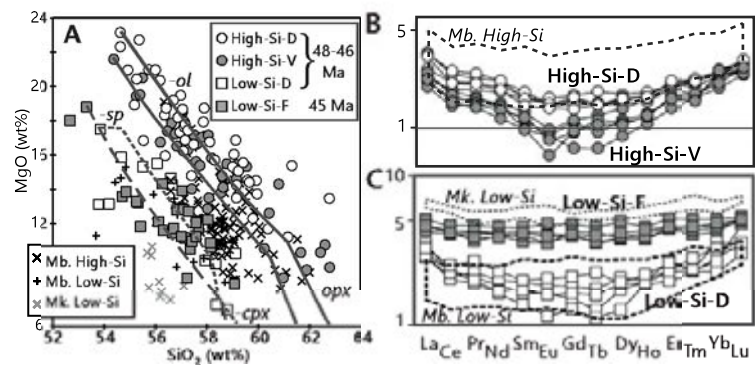


Fig. 3

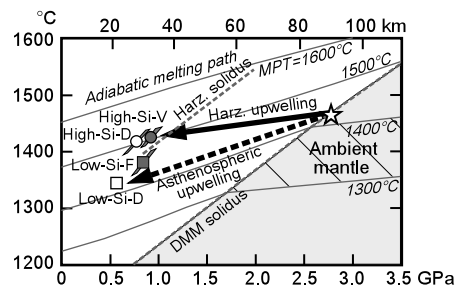
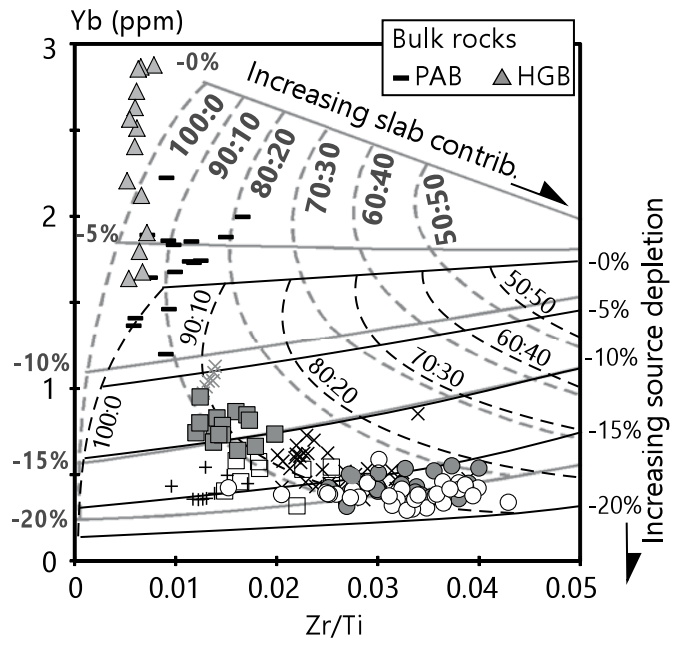


Fig. 4



**BONINITIC MELT INCLUSIONS IN CHROME SPINEL FROM THE OGASAWARA
ARCHIPELAGO**

DATA REPOSITORY for “Thermal and chemical evolution of the subarc mantle revealed by spinel-hosted melt inclusions in boninite from the Ogasawara (Bonin) Archipelago” by Susumu Umino, Keitaro Kitamura, Kyoko Kanayama, Akihiro Tamura, Naoya Sakamoto, Osamu Ishizuka, and Shoji Arai.

Analytical methods and procedures

Chrome spinels were handpicked under binocular after cleaning and sieving the sand samples with pure water. Spinel crystals embedded in epoxy resin on a slide glass were ground and polished to expose the core of crystals with melt inclusions, which were utilized for major and trace element analyses, including H₂O (Table DR-1).

Major elements of glass and minerals were analyzed by a JEOL JXA-8800 electron probe microanalyzer (EPMA) at Kanazawa University. The accelerating voltage was 15 kV and the specimen current was 12 nA. Analyses of glass followed the procedures of Noguchi *et al.* (2004) using broad beam diameters <30 μm . The corrections were made according to ZAF method. Relative errors (1σ) of melt inclusion analyses are better than 0.4% for SiO₂, 11.3% for TiO₂, 1.3% for Al₂O₃, 3.0% for FeO, 23.3% for MnO, 1.2% for MgO, 1.7% for CaO, 7.8% for Na₂O, 7.9% for K₂O and 6.7% for Cr₂O₃.

Trace element compositions (REEs, V, Cr, Co, Ni, Li, B, Sc, Rb, Sr, Y, Zr, Nb, Cs, Ba, Hf, Ta, Pb, Th and U) of glass were analyzed by laser ablation (193 nm ArF excimer: MicroLas GeoLas Q-plus) inductively coupled plasma mass spectrometry (Agilent 7500s) (LA-ICP-MS) at Kanazawa University (Morishita *et al.*, 2005a, b). Each analysis was performed by ablating spots of 30 or 40 μm in diameter at 5 Hz with energy density of 8 J/cm² per pulse. Signal integration times were 50 seconds for a gas background interval and 50 seconds for an ablation interval. BCR-2G (USGS glass reference material) was used as the primary calibration standard and its element concentration values are selected from the GeoReM database (see Jochum and Nohl., 2008). Repeated analyses of NIST 610, NIST612 and BIR show that reproducibility is better than $\pm 11\%$ (1σ)

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except B, Cs, Ta and U (Table DR-2). Data reduction was facilitated using ^{29}Si as internal standards for glass, based on SiO_2 contents obtained by EPMA analysis, and followed a protocol essentially identical to that outlined by Longerich et al. (1996). Details of the analytical method and data quality for the LA-ICP-MS system at Kanazawa University are described in Morishita et al. (2005a, b) and Ichiyama et al. (2013).

H_2O concentrations in glass of melt inclusions were determined by using a Secondary Ion Mass Spectrometry (SIMS) Cameca IMS-6F of the Creative Research Institution, Hokkaido University. Analytical procedures followed Miyagi and Yurimoto (1995) using natural hornblende with 1.66 wt% H_2O as a standard. Water contents (in wt%) were determined by comparing $^1\text{H}/^{30}\text{Si}$ ratios of unknown samples with averages of those of the standard hornblende measured on the same day.

Water content

H_2O contents vary from 1 to 5 wt% and $\text{H}_2\text{O}/\text{Ce}$ ratios from 2000 to 43200. Interstitial glass in melt inclusions consisting mainly of variolitic pyroxene crystals and vesicles (Fig. 1D) tends to show lower H_2O concentrations (<2.4 wt%). These glass inclusions have consistently lower $\text{H}_2\text{O}/\text{Ce}$ ratios compared to glass in melt inclusions comprising pure glass and a few shrinkage vesicles of the same geochemical type (Fig. DR-4). H_2O and Ce do not fractionate during partial melting of the source mantle and fractional crystallization of magmas due to their similar partition coefficients between melt and relevant mineral phases (Michael, 1995; Hauri et al., 2006), and hence melt inclusions of the same geochemical type are expected to have similar $\text{H}_2\text{O}/\text{Ce}$ ratios. Thus, the coincidence of the variolitic texture and the lower H_2O contents and $\text{H}_2\text{O}/\text{Ce}$ ratios compared to those of pure glass inclusions of the same geochemical type suggests that the variolitic melt inclusions experienced degassing and partially lost their original water during ascent to the surface. We therefore have corrected water contents of degassed samples and estimated the original water contents based on Ce contents, by assuming the original $\text{H}_2\text{O}/\text{Ce}$ ratios of the degassed samples to be equal to the average $\text{H}_2\text{O}/\text{Ce}$ ratio of undegassed samples of the same geochemical type. Because all low-Si-F (flat chondrite-normalized REE pattern) inclusions show a variolitic texture with low $\text{H}_2\text{O}/\text{Ce}$ ratios, the original $\text{H}_2\text{O}/\text{Ce}$ ratio of the type was assumed to be 9800, the maximum ratio among Type F inclusions observed. This gives the minimum

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estimates of the original H₂O contents for low-Si-F inclusions. The restored H₂O contents range from 1 to 6 wt% with restored H₂O/Ce ratios varying from 9800 to 43200.

Procedure of T-P estimate of olivine-orthopyroxene multiple saturation

T-P estimates for the primary boninitic magmas to be multiply saturated with olivine and orthopyroxene were determined by solving simultaneously the olivine-liquid geothermobarometry (equation (4) of Putirka et al., 2007) and olivine-orthopyroxene-liquid geothermobarometry (equation (42) of Putirka, 2008), following the procedure described in Putirka (2008). Major element compositions including Fe₂O₃, and H₂O contents are used as input. First, (1) The Fe³⁺/Fe²⁺ ratio at prescribed f_{O2} (here $\Delta\log(\text{FMQ}) = 0$ to +2) and the liquidus temperature is determined by using alphaMELTS (Smith and Asimow, 2005) at initial guess of pressure. (2) T and P obtained from the geothermobarometry are then used to calculate Fe³⁺/Fe²⁺ ratio at prescribed f_{O2} by alphaMELTS. (1) and (2) are repeated iteratively until T, P, and Fe³⁺/Fe²⁺ ratio become stable within 0.5%. Varying f_{O2} by 2 in log unit brings a little difference in T-P estimates compared to the uncertainty inherited from the geothermobarometry, while increase in H₂O content by 1 wt% reduces the estimated T and P by 34°C and 0.17 GPa, respectively.

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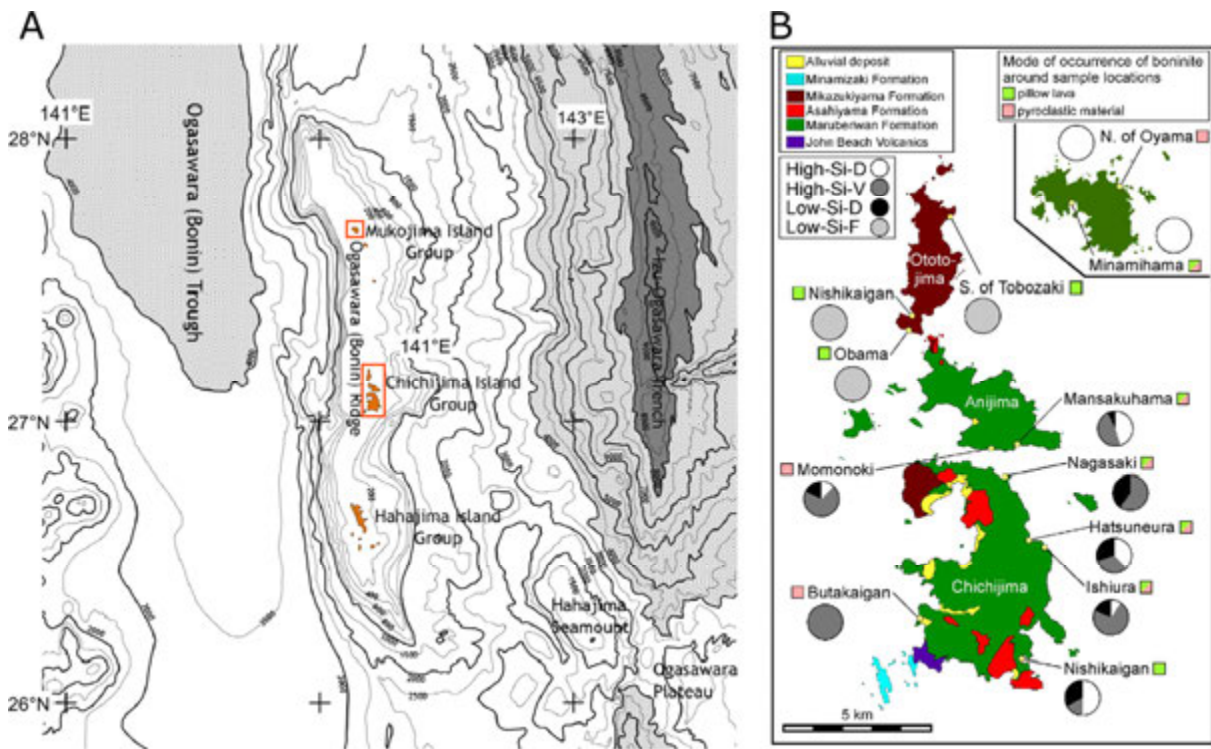


Figure DR-1. Locations of the Ogasawara Archipelago (A) and samples of chrome spinel together with the mode of occurrence of the host rocks and geochemical types (B).

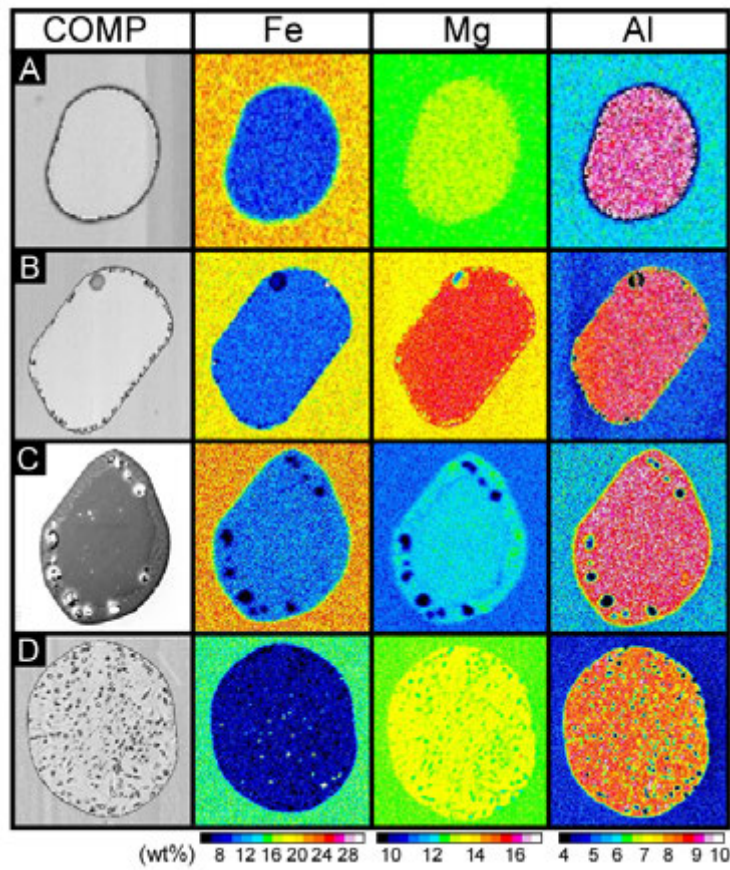


Figure DR-2. SEM (COMPO) images (left) and pseudo-colored EMPA maps (right) of Fe, Mg, and Al of melt inclusions and the host chrome spinel shown in Figure 1. Note the absence of Fe-rich and Mg-poor halos in the spinel host adjacent to the melt inclusions, confirming the absence of diffusive Fe-loss from the melt.

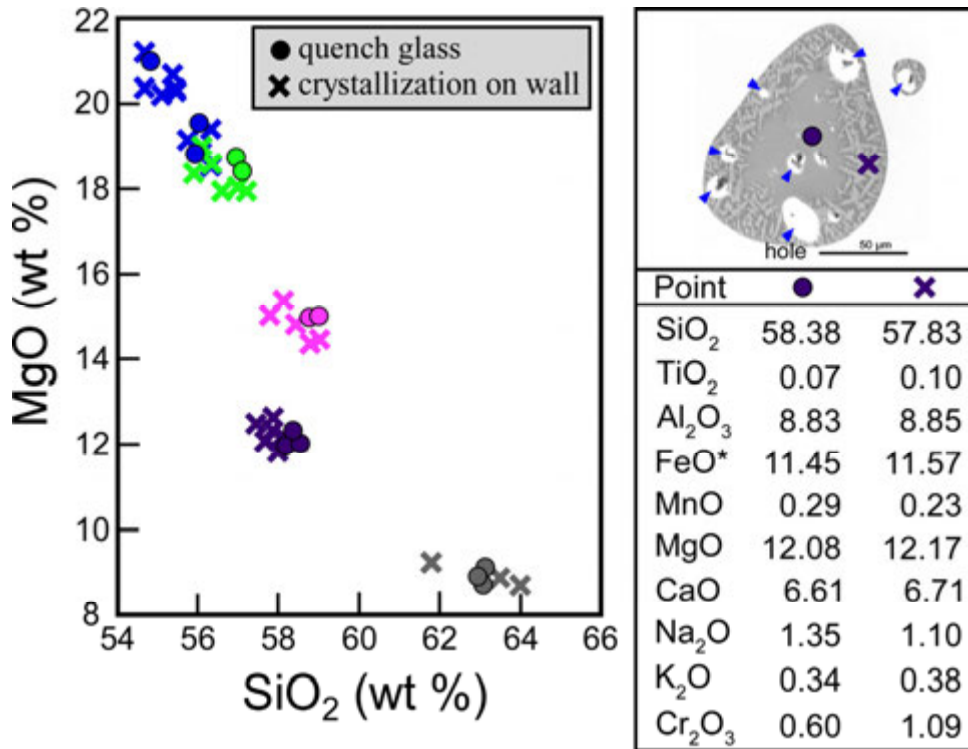


Figure DR-3. Analyses of melt inclusions (left) composed of the central glass pool (dots) surrounded by an outer zone with quench crystals and interstitial glass (crosses). A SEM image (upper right) shows dark grey glass rimmed by a zone with light grey quench crystals. Bright spots are holes (blue arrows) due to edge effect. Symbols with the same colors are analyses of the same inclusions. Note that both outer zones and central glass pools have identical bulk compositions, indicating that crystallization of the outer zone did not affect the composition of the remaining melt in the core.

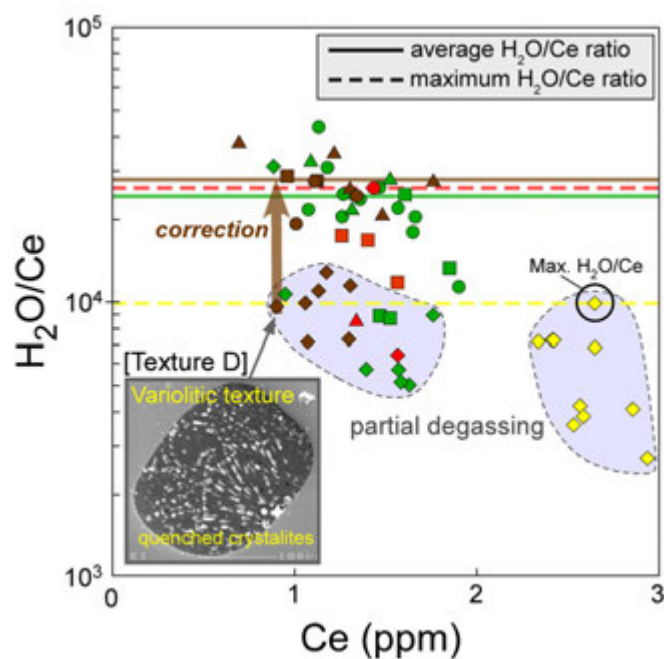


Figure DR-4. H_2O/Ce plotted against Ce of melt inclusions of high-Si-D (green) and high-Si-V (brown), and low-Si-D (red) and low-Si-F (yellow). Variolitic inclusions (solid diamonds) (Figs. 1D, DR-2D) have consistently lower H_2O/Ce ratios compared to glass in melt inclusions comprising pure glass and a few shrinkage vesicles (circles, squares and triangles) (Figs. 1A, 1B, 1C, DR-2A, 2B, 2C) of the same geochemical type. These variolitic inclusions are considered to have partly lost their water upon entrapment due to degassing. For these inclusions, the original water contents were restored based on Ce contents, by assuming the original H_2O/Ce ratios of the variolitic samples to be equal to the average H_2O/Ce ratio of undegassed samples of the same geochemical type.

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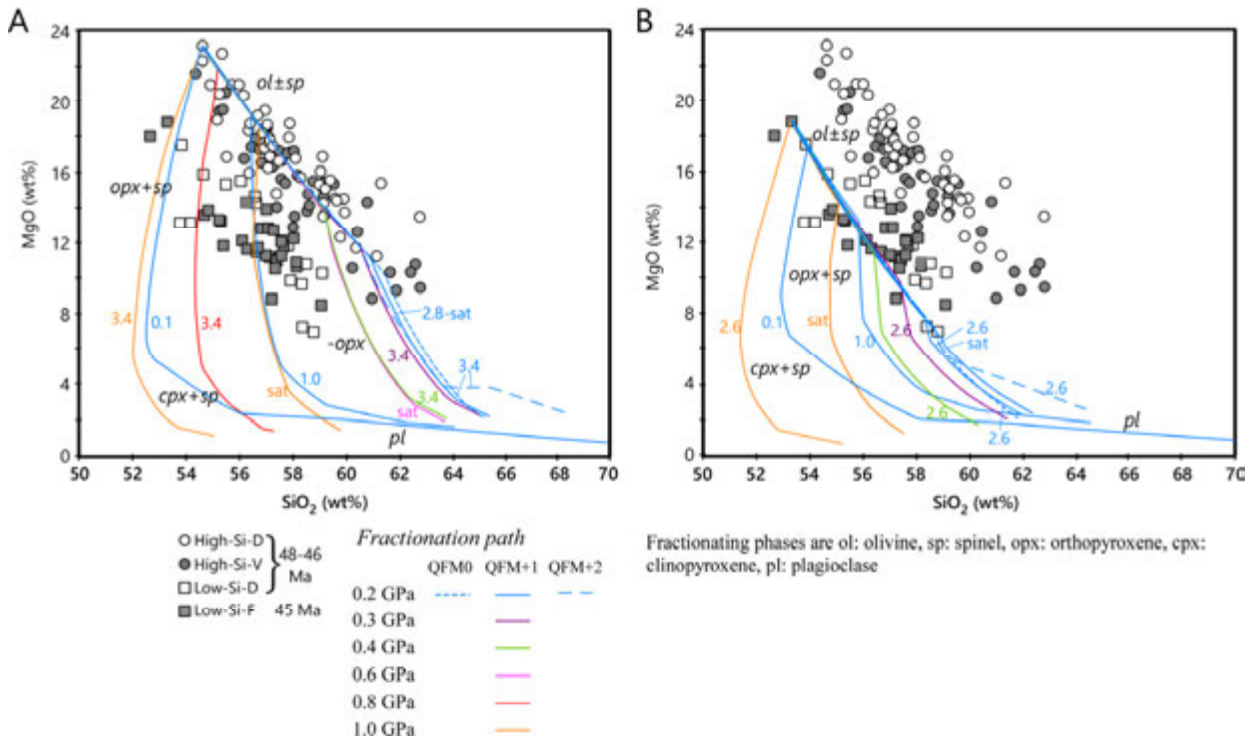


Figure DR-5. Examples of fractional crystallization paths for primary boninite magmas of the high-Si-D (A) and the low-Si-F (B) by alphaMELTS (Smith and Asimow, 2005), compared with boninitic melt inclusions (circles and squares). The numbers beside the paths are H₂O contents (wt%) in the primary magmas. Calculations were done under fO₂ of Δlog(FMQ)=+1 at pressures 0.2-1.0 GPa, and also under fO₂ of Δlog(FMQ)=0 and +2 at 0.2 GPa. Change in fO₂ by 2 log units only slightly shifts the fractionation paths. The high-Si boninitic inclusions are mostly plot around low-P (<0.4 GPa), high-H₂O (>2.8 wt%) paths. Higher pressure or lower H₂O content leads to earlier crystallization of orthopyroxene and rapid decreases of MgO, away from the clusters of melt inclusion compositions. The variation of the low-Si inclusions also consistent with low-P (<0.3 GPa), high-H₂O (>2.6 wt%) conditions, suggesting minimal H₂O loss during fractionation. Those plotted at higher MgO than the olivine-fractionation paths suggest mixing of relatively primitive and differentiated melts.

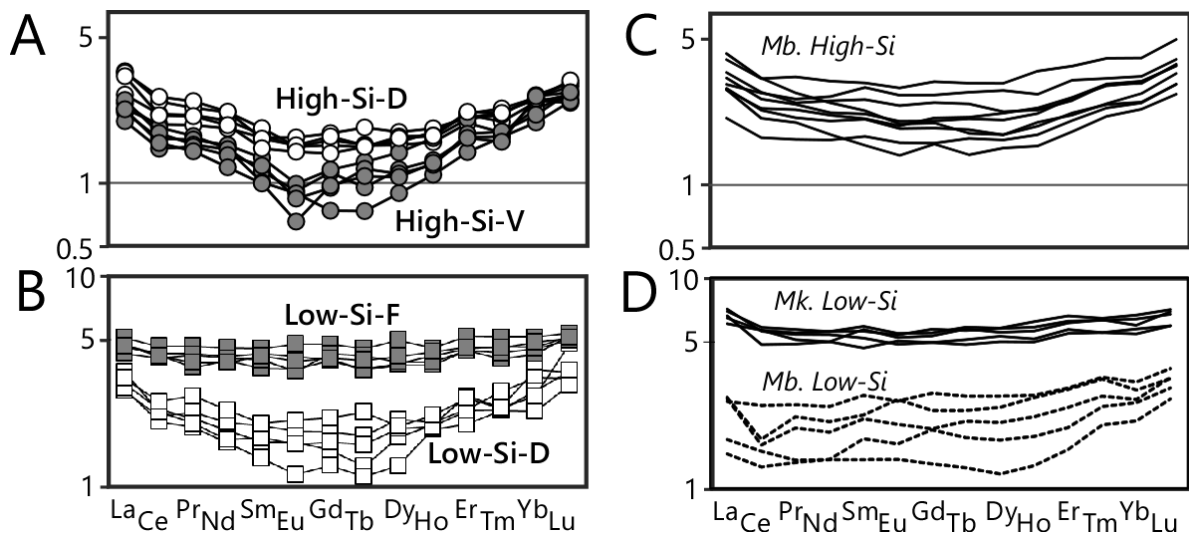


Figure DR-6. Chondrite-normalized REE patterns for the high-Si-D and high-Si-V (A) and the low-Si-D and low-Si-F (B) melt inclusions, compared with the whole-rock compositions (after Kanayama et al., 2012) of high-Si boninite from the 48-46 Ma Maruberiwan Formation (Mb.) (C), and those of low-Si boninites from the 45-Ma Mikazukiya (Mk.) and 48-46 Ma Maruberiwan (Mb.) Formation (D).

TABLE DR-1. REPRESENTATIVE ANALYSES OF MELT INCLUSIONS IN CHROME SPINEL AND THE ESTIMATED T-P CONDITIONS FOR THE PRIMARY BONINITES

Formation Geochem.Type No. of analyses	Maruberiwan												Mikazukiya			
	High-Si-V* 3	High-Si-V 3	High-Si-V 2	High-Si-V 3	High-Si-D* 3	High-Si-D 3	High-Si-D 3	High-Si-D 2	Low-Si-D* 1	Low-Si-D 2	Low-Si-D 3	Low-Si-D 3	Low-Si-F* 5	Low-Si-F 3	Low-Si-F 5	Low-Si-F 2
SiO ₂	52.63 ± 0.27	55.89 ± 0.41	58.38 ± 0.51	58.36 ± 0.08	52.20 ± 0.54	56.09 ± 0.42	56.43 ± 0.16	54.95 ± 0.06	53.53 ± 0.22	54.91 ± 0.68	56.12 ± 0.31	57.34 ± 0.17	51.38 ± 0.89	57.75 ± 0.45	53.48 ± 0.31	54.30 ± 0.88
TiO ₂	0.09 ± 0.03	0.08 ± 0.05	0.06 ± 0.00	0.07 ± 0.04	0.06 ± 0.02	0.09 ± 0.03	0.08 ± 0.03	0.05 ± 0.04	0.14 ± 0.01	0.10 ± 0.03	0.10 ± 0.04	0.14 ± 0.03	0.12 ± 0.04	0.16 ± 0.04	0.19 ± 0.04	0.13 ± 0.03
Al ₂ O ₃	7.70 ± 0.09	8.16 ± 0.06	10.77 ± 0.14	8.62 ± 0.04	6.42 ± 0.11	7.37 ± 0.15	6.87 ± 0.04	7.10 ± 0.07	10.73 ± 0.10	9.03 ± 0.03	10.05 ± 0.10	11.67 ± 0.07	8.81 ± 0.33	11.58 ± 0.22	10.69 ± 0.33	10.56 ± 0.29
FeO*	8.29 ± 0.16	8.21 ± 0.06	6.49 ± 0.11	7.60 ± 0.20	9.11 ± 0.57	9.19 ± 0.21	8.50 ± 0.17	8.36 ± 0.27	6.37 ± 0.18	10.02 ± 0.04	8.97 ± 0.14	8.20 ± 0.05	8.61 ± 0.26	7.24 ± 0.51	9.41 ± 0.45	6.27 ± 0.32
MnO	0.23 ± 0.05	0.14 ± 0.02	0.19 ± 0.03	0.19 ± 0.05	0.16 ± 0.03	0.23 ± 0.07	0.19 ± 0.03	0.19 ± 0.02	0.10 ± 0.02	0.17 ± 0.05	0.16 ± 0.03	0.13 ± 0.05	0.18 ± 0.08	0.10 ± 0.03	0.24 ± 0.07	0.24 ± 0.08
MgO	20.90 ± 0.63	16.69 ± 0.08	9.76 ± 0.06	15.02 ± 0.11	22.12 ± 0.50	15.1 ± 0.24	18.26 ± 0.11	18.79 ± 0.06	17.41 ± 0.14	15.19 ± 0.10	14.13 ± 0.12	9.49 ± 0.06	18.12 ± 0.66	8.29 ± 0.42	13.52 ± 0.91	10.38 ± 0.68
CaO	5.24 ± 0.12	6.24 ± 0.07	6.93 ± 0.03	6.21 ± 0.10	4.33 ± 0.09	6.25 ± 0.10	5.14 ± 0.03	5.04 ± 0.13	9.02 ± 0.10	6.03 ± 0.04	7.13 ± 0.07	8.34 ± 0.06	7.47 ± 0.31	10.51 ± 0.25	7.97 ± 0.23	9.99 ± 0.35
Na ₂ O	0.71 ± 0.05	1.25 ± 0.08	1.54 ± 0.00	1.32 ± 0.02	0.33 ± 0.02	1.21 ± 0.03	0.99 ± 0.03	1.27 ± 0.04	0.65 ± 0.04	1.12 ± 0.08	1.38 ± 0.06	1.65 ± 0.04	0.97 ± 0.09	1.48 ± 0.10	1.15 ± 0.09	1.27 ± 0.13
K ₂ O	0.39 ± 0.06	0.35 ± 0.03	0.26 ± 0.00	0.43 ± 0.01	0.38 ± 0.02	0.31 ± 0.03	0.24 ± 0.05	0.38 ± 0.04	0.16 ± 0.01	0.35 ± 0.04	0.41 ± 0.01	0.39 ± 0.01	0.32 ± 0.05	0.34 ± 0.01	0.32 ± 0.03	0.33 ± 0.05
Cr ₂ O ₃	0.62 ± 0.09	0.19 ± 0.04	0.28 ± 0.05	0.19 ± 0.05	0.45 ± 0.06	0.35 ± 0.06	0.77 ± 0.04	0.31 ± 0.06	1.31 ± 0.05	1.02 ± 0.10	0.65 ± 0.04	0.93 ± 0.02	0.37 ± 0.04	0.31 ± 0.10	0.54 ± 0.07	1.11 ± 0.03
H ₂ O	2.5 ± 0.5	1.9 ± 0.3	2.8 ± 0.1	3.1 ± 0.1	3.4 ± 0.6	3.2 ± 1.6	2.3 ± 0.2	2.1 ± 0.5	2.1 ± 0.1	1.8 ± 0.1	2.2 ± 0.3	1.8 ± 0.3	2.6 ± 0.7	1.2 ± 0.1	2.1 ± 0.3	1.2 ± 0.1
Total	96.81 ± 0.93	97.19 ± 0.34	94.67 ± 0.65	98.01 ± 0.22	95.55 ± 0.96	96.19 ± 0.56	97.47 ± 0.14	96.43 ± 0.21	99.41 ± 0.89	97.94 ± 0.56	99.09 ± 0.33	98.28 ± 0.25	96.37 ± 1.63	97.76 ± 0.41	97.50 ± 1.43	94.56 ± 0.08
Fe ³⁺ /ΣFe	0.09-0.20				0.09-0.20				0.11-0.24				0.10-0.22			
°C	1423-1428				1415-1419				1343-1347				1378-1384			
GPa	0.89-0.96				0.74-0.80				0.54-0.59				0.81-0.88			
Fo	95.0-95.8				94.4-95.3				95.7-96.4				94.0-95.2			
Trace element compositions [µg/g]																
Li	5.73	7.75		7.27	27.02	5.77	7.20	9.17			6.87	8.81	5.09	5.03	5.09	4.17
B	8.15	10.54		15.82	4.62	10.94	9.08	4.09	10.05		13.52	11.78	6.55	6.57	6.55	8.49
Sc	35.57	33.54		32.98	26.70	31.67	29.81	28.42	51.71		43.12	42.25	56.59	47.19	56.59	45.64
Ti	335.77	409.02		437.78	392.14	534.31	466.18	399.03	624.10		508.74	738.16	1396.72	1214.67	1396.72	866.92
V	159.28	157.75		169.12	134.30	238.77	153.21	141.60	412.26		241.23	244.42	264.64	212.18	264.64	131.51
Co	61.65	54.87		47.33	72.58	59.56	63.84	66.84	96.43		51.21	47.45	49.71	36.89	49.71	31.70
Ni	397.66	259.95		167.02	363.20	108.96	251.07	283.56	233.43		139.76	100.60	132.83	83.60	132.83	84.28
Rb	11.67	11.60		15.21	9.68	9.39	7.01	11.38	7.34		13.35	12.60	5.21	5.05	5.21	5.71
Sr	29.54	39.46		43.52	44.50	60.53	51.22	58.17	53.36		52.24	71.98	93.20	101.86	93.20	103.34
Y	2.04	2.58		2.40	2.86	2.74	3.12	2.90	2.24		2.57	3.36	5.99	7.35	5.99	6.23
Zr	11.72	13.83		16.63	15.15	13.28	14.92	15.93	9.27		13.69	18.76	16.78	17.12	16.78	17.17
Nb	0.31	0.31		0.37	0.38	0.26	0.17	0.52	0.73		0.33	0.42	0.53	0.56	0.53	1.14
Cs	25.66	0.58		0.61	17.52	0.36	0.23	0.54			0.60	0.65	0.18	0.18	0.18	3.96
Ba	27.69	26.83		32.19	13.87	31.11	23.28	40.27	21.08		28.67	33.79	34.98	36.63	34.98	40.23
La	0.47	0.52		0.56	0.69	0.64	0.50	0.97	0.54		0.69	0.78	1.12	1.22	1.12	1.92
Ce	0.90	1.01		1.12	1.39	1.32	1.08	1.83	0.82		1.25	1.57	2.65	2.86	2.65	5.88
Pr	0.14	0.15		0.17	0.23	0.20	0.16	0.27	0.12		0.18	0.25	0.40	0.43	0.40	0.53
Nd	0.62	0.70		0.72	1.13	0.92	0.79	1.19	0.80		0.74	1.06	1.90	2.10	1.90	1.89
Sm	0.18	0.20		0.21	0.31	0.25	0.24	0.33			0.20	0.30	0.64	0.67	0.64	0.47
Eu	0.05	0.06		0.06	0.11	0.08	0.11	0.11	0.09		0.06	0.12	0.23	0.27	0.23	0.19
Gd	0.19	0.23		0.24	0.41	0.32	0.38	0.35	0.35		0.25	0.42	0.80	0.95	0.80	0.78
Tb	0.04	0.05		0.04	0.08	0.06	0.05	0.07			0.04	0.08	0.13	0.16	0.13	0.12
Dy	0.27	0.35		0.31	0.47	0.39	0.44	0.43	0.47		0.31	0.48	0.95	1.23	0.95	1.03
Ho	0.07	0.09		0.08	0.10	0.10	0.08	0.13			0.10	0.12	0.23	0.25	0.23	0.22
Er	0.28	0.33		0.28	0.37	0.34	0.33	0.32	0.36		0.32	0.42	0.73	0.83	0.73	0.70
Tm	0.04	0.05		0.05	0.05	0.05	0.07	0.05			0.06	0.06	0.12	0.13	0.12	0.10
Yb	0.35	0.43		0.36	0.44	0.40	0.40	0.41	0.41		0.37	0.44	0.74	0.83	0.74	0.70
Lu	0.07	0.08		0.07	0.08	0.07	0.07	0.07	0.07		0.07	0.12	0.13	0.13	0.13	0.13
Hf	0.31	0.37		0.45	0.52	0.38	0.40	0.49	0.44		0.47	0.61	0.46	0.50	0.46	0.59
Ta	0.03	0.02		0.02	0.03	0.02		0.03			0.02	0.02	0.03	0.03	0.03	0.07
Pb	1.66	1.39		1.20	1.70	1.50	0.99	1.64	1.02		1.13	1.59	1.07	1.05	1.07	0.87
Th	0.08	0.07		0.06	0.14	0.10	0.06	0.12			0.08	0.10	0.13	0.15	0.13	0.14
U	0.08	0.08		0.07	0.12	0.10	0.07	0.14			0.09	0.08	0.09	0.10	0.09	0.18

Major element oxides are listed in wt%. 1σ of major element oxides are standard deviations of multiple analyses, except the primary Low-Si-D melt inclusion (*) for which 1σ is given as statistic errors of X-ray count.

Primary boninite compositions of the four geochemical types are denoted by asterisks (*) and are listed with the estimated T-P conditions and Fo contents at olivine and orthopyroxene multiple saturation.

Considering the uncertainties of H₂O contents and f_{O2}, T-P estimates are shown as ranges including 1σ of H₂O and f_{O2} of ΔFMQ 1±1 in log unit.

Table DR-2. Repeated analyses of standard glass NIST610, NIST612, and BIR. Reference values are from Jochum and Nohl (2008).

NIST610	Li	B	Sc	Ti	V	Co	Ni	Rb	Sr	Y	Zr	Nb	Cs	Ba	La	Ce	Pr	Nd	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu	Hf	Ta	Pb	Th	U	
Reference value [$\mu\text{g/g}$]	485	356	441	434	442	405	444	431	497	450	440	419	361	424	457	448	430	431	451	461	420	443	427	449	426	420	462	435	418	377	413	451	457	
Average of 24 anal. [$\mu\text{g/g}$]	491	250	511	502	488	445	405	454	579	584	537	506	405	489	500	519	517	500	525	508	533	549	533	558	558	528	546	551	521	532	463	528	508	
Standard deviation [$\mu\text{g/g}$]	17	61	36	18	12	11	24	9	27	55	42	19.2	10.2	15.0	27.3	16.3	23.3	27.7	31.2	29.9	40.5	50.0	48.3	53.9	51.8	59.0	48.5	62.0	54.9	35.9	12.9	44.2	13.8	
RSD (%)	3.4	24.2	7.0	3.6	2.5	2.5	6.0	2.0	4.7	9.4	7.9	3.8	2.5	3.1	5.5	3.1	4.5	5.5	5.9	5.9	7.6	9.1	9.1	9.7	9.3	11.2	8.9	11.3	10.5	6.7	2.8	8.4	2.7	
Accuracy (%)	1.4	-42.5	13.7	13.6	9.4	9.0	-9.7	5.1	14.1	23.0	18.0	17.1	10.8	13.3	8.6	13.8	16.8	13.8	14.1	9.2	21.3	19.3	19.9	19.4	23.7	20.4	15.4	21.1	19.9	29.2	10.8	14.7	10.0	
NIST612	Li	B	Sc	Ti	V	Co	Ni	Rb	Sr	Y	Zr	Nb	Cs	Ba	La	Ce	Pr	Nd	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu	Hf	Ta	Pb	Th	U	
Reference value [$\mu\text{g/g}$]	42	43	44	45	46	47	48	49	50	51	52	53	54	55	56	57	58	59	60	61	62	63	64	65	66	67	68	69	70	71	72	73	74	
Average of 9 anal. [$\mu\text{g/g}$]	42	26	41	43	41	40	36	33	84	44	42	40	46	41	38	43	41	39	41	38	41	43	41	42	44	42	43	42	41	42	43	41	41	
Standard deviation [$\mu\text{g/g}$]	1	2	0	1	0	0	1	0	1	1	1	0.6	0.7	0.3	0.6	0.3	0.4	0.3	0.3	0.4	0.7	0.5	0.7	1.1	1.2	0.6	0.6	0.7	0.1	0.0	0.4	0.3	0.4	
RSD (%)	2.5	6.2	1.1	2.2	0.9	0.8	3.5	1.5	0.9	2.0	2.3	1.5	1.6	0.6	1.6	0.8	1.0	0.9	0.7	1.1	1.7	1.2	1.8	2.6	2.7	1.5	1.4	1.6	0.2	0.0	0.9	0.7	1.1	
Accuracy (%)	1.0	-63.2	-5.8	-3.5	-10.2	-16.5	-31.9	-46.0	41.0	-14.3	-21.8	-31.9	-16.8	-32.2	-45.3	-32.4	-39.3	-50.2	-46.4	-60.6	-51.4	-45.6	-55.6	-51.9	-50.5	-58.8	-57.7	-62.2	-70.3	-66.7	-66.4	-77.6	-79.4	
BIR-1G	Li	B	Sc	Ti	V	Co	Ni	Rb	Sr	Y	Zr	Nb	Cs	Ba	La	Ce	Pr	Nd	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu	Hf	Ta	Pb	Th	U	
Reference value [$\mu\text{g/g}$]	3		43	5400	326	52	178	0.2	109	14	14	0.52	0.01	6.50	0.61	1.89	0.37	2.37	1.09	0.52	1.85	0.35	2.55	0.56	1.70	0.24	1.64	0.25	0.57	0.04	3.70	0.03	0.02	
Average of 13 anal. [$\mu\text{g/g}$]	3	2	41	5740	327	54	143	0	110	14	14	1	0	7	1	2	0	110	2	1	1	2	0	2	1	2	0	2	0	1	0	4	0	0
Standard deviation [$\mu\text{g/g}$]	0	1	1	114	8	1	12	0	3	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
RSD (%)	3.1	53.0	2.3	2.0	2.3	2.0	8.5	5.5	2.4	3.0	2.5	6.2	71.4	3.7	4.5	3.1	4.2	4.4	3.4	3.8	4.2	6.8	5.0	6.2	3.5	5.6	5.0	9.0	8.4	27.9	3.7	16.9	30.8	
Accuracy (%)	-0.7		-4.4	5.9	0.4	4.5	-24.3	-0.3	0.8	0.2	-2.9	-1.9	-36.8	1.4	-3.3	3.8	0.3	-1.0	-0.2	0.1	-9.0	-2.9	-2.9	-5.7	-4.8	-3.2	-2.3	-7.3	-3.8	3.2	-3.4	-6.2	-37.2	