

**Hydrogen Contents of Mantle Minerals from Peridotite
Xenoliths of Ichinomegata Volcano, NE Japan**

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Keywords : mantle xenolith, orthopyroxene, clinopyroxene, water, subduction

Abstract

Hydrogen contents of olivine, clinopyroxene, and orthopyroxene from peridotite and pyroxenite xenoliths from calc-alkaline andesite of Ichinomegata volcano have been measured by using FTIR technique. Positions of bands in the infrared spectra resemble those observed previously for the studied minerals. Olivine contains <17 ppm wt. H₂O, whereas clinopyroxene and orthopyroxene contain significant amount of H₂O (359-647 and 184-277 ppm wt., respectively). Partition coefficient of H₂O between clinopyroxene and orthopyroxene was found to vary between 1.8 and 3.2. We did not observe systematic compositional (major elements in bulk rocks and minerals) or textural dependences of hydrogen contents in minerals. The low hydrogen contents of olivine are consistent with hydrogen loss during transportation of xenoliths to the surface. The high hydrogen contents of pyroxenes are consistent with strong metasomatic enrichment of the mantle wedge beneath the Japan arc. However, hydration of nominally anhydrous pyroxenes may reflect earlier events than hydration of mantle wedge by metasomatic agents producing modal amphibole in peridotite and abundant uppermost mantle/lower crust amphibolites. Pyroxenes from Ichinomegata contain nearly maximum amount of H₂O among the others from mantle xenoliths in alkaline basalts and kimberlites.

1. Introduction

Mantle xenoliths in calc-alkaline andesite erupted at Ichinomegata volcano (Northeast Honshu arc, Japan) provide a unique opportunity to examine directly petrology of the mantle wedge. This location is very famous and studied intensively over the last 40 years (e.g. Kuno, 1967; Aoki,

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1971; Aoki and Shiba, 1973; Takahashi, 1980; 1986; Abe et al., 1998). Based on petrological characteristics of mantle and crustal xenoliths it was suggested that the lower crust of the back-arc side of NE Honshu arc is composed of hornblende gabbro and amphibolite, whereas the upper mantle is composed of amphibole-bearing plagioclase and spinel peridotite presumably equilibrated at 0.6-0.8 GPa and 600-700°C (Takahashi, 1978; 1980; 1986; Zashu et al., 1980). These data were supported by ultrasonic measurements of seismic wave velocities in the xenoliths. Laboratory studies of P -wave (V_p) velocities in xenoliths of Ichinomegata showed that low- V_p of lower crustal layer of NE Honshu (6.6-7.0 km/s, Iwasaki et al., 2001) is consistent with the thick (15 km) hydrous lower crust comprised by amphibolite and hornblende gabbro with ultrabasic composition (Nishimoto et al., 2005).

Since deep-seated xenoliths of Ichinomegata characterize hydrated island arc-related environment and contain abundant amphibole and subordinate phlogopite, it is important to study hydrogen contents of their major minerals to characterize hydrogen incorporation in nominally anhydrous minerals of the mantle wedge. In this paper we present preliminary data on water content in minerals of the mantle xenoliths from Ichinomegata volcano and compare their water contents with those from xenoliths of the other tectonic settings.

2. Analytical technique

Minerals and glasses were analyzed in polished thin sections and mounts for major elements using JEOL Superprob microanalyzer (JXA-8800) in the Institute of Mineralogy, Petrology and Economic Geology of Tohoku University in Sendai, Japan. Analyses were run with 15 keV acceleration voltages, 10 nA sample current, and 10 μm beam size. Oxides and natural and synthetic minerals were used as standards. Matrix corrections were performed by the ZAF-procedure.

Quantitative analyses of water in minerals were performed by using JEOL Diamond20 FTIR microscope. The polarized spectra were obtained on optically clear single crystals, which were double polished according to the crystallographic orientation and placed on a single crystal KBr plate. The calibration of polarizer was made by using a double-polished olivine crystal preliminarily oriented by single-crystal X-ray diffraction. Estimated error of crystal orientation by FTIR is $\sim 10^\circ$. We measured three perpendicular sections of mineral grains. We used similar approach to choose orientation of mineral grains as described by Peslier et al., (2002).

We used following equation of the Beer-Lambert law to calculate H_2O contents of minerals: $C_{\text{H}_2\text{O}} = A_i/\mu_i$, where $C_{\text{H}_2\text{O}}$ is the water concentration in ppm wt, $A_i = A_a + A_b + A_c$, the total integrated absorbance of OH stretching vibration in the three perpendicular directions of the optical

indicatrixes normalized to 1 cm thickness taken between 3750 and 3000 cm^{-1} for clinopyroxene, between 3750 and 2800 cm^{-1} for orthopyroxene, and between 3650 and 3200 cm^{-1} for olivine, and μ_i is the integrated molar absorption coefficient (7.09 $\text{ppm}^{-1}\text{cm}^{-2}$ for clinopyroxene, 14.84 $\text{ppm}^{-1}\text{cm}^{-2}$ for orthopyroxene; Bell et al., 1995; and 5.32 $\text{ppm}^{-1}\text{cm}^{-2}$ for olivine, Bell et al., 2003). Uncertainties of individual FTIR measurements were estimated to be better than 10%.

3. Host volcanics and variety of mantle xenoliths

Megata volcanoes are located at the Oga Peninsula (Akita, Japan). There are three craters, Ichinomegata, Ninomegata, and Sannomegata, all of which provide mafic to ultramafic xenoliths. It is related to the NE Honshu island arc with back-arc marginal sea basin on the convergent plate boundary between the Pacific and North American plates. Volcanism of the NE Honshu arc is related to the westward subduction of the Pacific plate with an approximate dip angle of 30° (e.g. Nishimoto et al., 2005). Thickness of extensional Cenozoic sediments on the back arc side of NE Honshu is estimated to be more than 5 km (Sato, 1994). The host magma of Ichinomegata xenoliths is calc-alkaline andesite to dacite dated at 80,000-60,000 yr (Kitamura, 1990) or 10,000 yr according to earlier estimations (Horie, 1964; Aoki, 1971).

Megata volcanic group is well known due to abundant mantle and crustal xenoliths. Major and trace elements and isotopic studies of peridotite and gabbro xenoliths have been published in many papers (e.g. Kuno, 1967; Aoki, 1971; Takahashi, 1980; 1986; Abe, 1998). Major mafic lithologies include hornblende gabbro, gabbro, pyroxenite, and amphibolite. Peridotite varies from harzburgite to lherzolite. Unlikely to other arc-related localities dunite has been rarely found (e.g. Arai and Takahashi, 1987). Peridotites form complex veined xenoliths with feldspatic (gabbro) and pyroxenitic rocks. They often contain amphibole (pargasite) and rarely phlogopite (e.g. Arai, 1986).

4. Major element chemistry and thermobarometry

Representative compositions of minerals from Ichinomegata xenoliths studied for H_2O contents are shown in Table 1. Fig.1 shows $\text{Cr}\#$ ($=100\text{Cr}/(\text{Cr}+\text{Al})$) of spinel versus Fo content of olivine for Ichinomegata xenoliths from our collection. $\text{Cr}\#$ of spinel varies between 7 and 57 and is in general agreement with previous works (e.g. Abe et al., 1998). Olivine in majority of spinel- and amphibole-bearing xenoliths has Fo_{88-91} composition. Some Fe-bearing peridotites, which may be strongly hydrated according to Abe et al. (1998), contain olivine with Fo_{84-87} . These peridotites deviate from major sequence of olivine-spinel mantle array proposed by Arai (1987, 1991) (Fig.1). Similarly, pyroxenes of Fe-bearing lherzolites have low $\text{Mg}\#=85-90$ relative to other lherzolites

Table 1. Major element compositions (wt.%) of minerals from Ichinomegata xenoliths.

Sample	Ig-16				Ig-36				Ig-31			
	Sp lherzolite		Cpx		Sp lherzolite		CPx		Amp lherzolite		Cpx	
Min	Ol	OPx	Cpx	Sp	Ol	OPx	CPx	Sp	Ol	OPx	Cpx	Amp
SiO ₂	40.60	55.29	52.54	0.02	41.14	56.37	52.41		40.50	55.47	51.62	43.99
TiO ₂		0.09	0.21	0.13		0.05	0.45	0.05		0.04	0.30	0.72
Al ₂ O ₃		3.40	3.25	48.90	0.02	3.21	4.32	53.53		2.56	3.81	14.21
Cr ₂ O ₃	0.02	0.32	0.64	16.41	0.00	0.33	0.98	13.42	0.01	0.48	0.90	0.08
FeO	10.05	6.80	2.44	16.29	9.76	6.45	2.52	11.70	12.04	9.76	2.76	6.83
MnO	0.17	0.15	0.09	0.16	0.12	0.14	0.10	0.08	0.23	0.23	0.09	0.09
MgO	48.38	33.01	17.07	17.52	49.40	33.73	15.91	20.21	47.35	31.47	16.57	16.72
CaO	0.02	0.54	23.45		0.06	0.46	22.70		0.02	0.52	22.80	11.67
Na ₂ O	0.02	0.01	0.32	0.02	0.00	0.03	0.68		0.01		0.32	2.30
K ₂ O												0.73
NiO	0.40	0.08	0.04	0.33	0.39	0.10	0.03	0.34	0.40	0.07	0.05	0.11
Total	99.65	99.69	100.1	99.78	100.9	100.8	100.1	99.33	100.6	100.6	99.22	97.44
Mg#	89.6	89.6	92.6	65.7	90.0	90.3	91.8	75.5	87.5	85.2	91.5	81.4

Sample	Ig-21				Ig-24				Ig-4			
	Amp lherzolite		Cpx		Sp harzburgite		CPx		Sp harzburgite		Cpx	
Min	Ol	OPx	Cpx	Amp	Ol	OPx	CPx	Sp	Ol	OPx	CPx	Sp
SiO ₂	41.03	56.18	52.23	41.91	41.49	56.98	53.41		40.77	56.57	52.91	0.02
TiO ₂		0.08	0.28	1.61		0.02	0.06	0.16		0.02	0.05	0.07
Al ₂ O ₃		3.16	4.32	14.52		2.25	2.48	36.47	0.01	2.67	2.97	45.44
Cr ₂ O ₃	0.02	0.37	0.65	0.02		0.39	0.86	29.63	0.02	0.39	0.78	20.78
FeO	10.69	6.74	3.51	13.17	8.33	5.16	2.28	14.51	9.18	6.26	2.74	14.85
MnO	0.16	0.18	0.09	0.23	0.10	0.13	0.07	0.11	0.16	0.14	0.07	0.13
MgO	48.64	33.68	16.47	13.04	50.68	34.39	17.12	18.21	49.46	34.11	17.12	18.52
CaO	0.08	0.54	21.97	10.27	0.07	0.47	23.00		0.06	0.57	22.32	
Na ₂ O	0.02	0.02	0.36	2.51	0.00	0.03	0.29	0.01		0.03	0.44	0.02
K ₂ O				0.30								
NiO	0.37	0.10	0.05		0.41	0.09	0.02	0.26	0.40	0.11	0.02	0.33
Total	101.0	101.0	99.93	97.58	101.0	99.90	99.58	99.36	100.0	100.9	99.42	100.2
Mg#	89.0	89.9	89.3	63.8	91.6	92.2	93.1	69.1	90.6	90.7	91.8	69.0

Sample	Ig-42			Ig-45		Ig-47			
	Gabbro		OPx	Plag	Amp	Amphibolite		Amp*	Phl*
Min	CPx	OPx	Plag	Amp	OPx	Amp*	Phl*	Mt	
SiO ₂	52.34	56.01	46.27	42.01	55.11	39.20	36.97	0.07	
TiO ₂	0.25	0.05	0.01	1.79	0.08	4.98	7.23	6.01	
Al ₂ O ₃	2.71	3.40	34.38	14.67	4.14	13.41	14.76	7.01	
Cr ₂ O ₃		0.35			0.25	0.26	0.19		
FeO	8.12	6.67	0.24	13.61	7.36	10.50	12.55	79.85	
MnO	0.41	0.18		0.18	0.16	0.07	0.01	0.28	
MgO	13.58	33.32	0.01	12.98	32.12	14.27	14.90	2.31	
CaO	22.61	0.50	17.90	10.39	0.89	10.86	0.05		
Na ₂ O	0.39	0.02	1.27	2.55	0.01	2.68	0.26		
K ₂ O			0.03	0.35		1.18	9.23		
NiO	0.01	0.08	0.01		0.08	0.00	0.00		
Total	100.4	100.6	100.1	98.52	100.2	98.03	97.21	95.52	
Mg#	74.9	89.9		63.0	88.6	70.8	67.9		

*Amphibole and phlogopite contain also BaO = 0.04 and 0.13 wt.%, F = 0.59 and 0.93 wt.% respectively. Ol, olivine; OPx, orthopyroxene; CPx, clinopyroxene; Sp, spinel; Amp, amphibole; Plag, plagioclase; Phl, phlogopite; Mt, Ti-magnetite.

where pyroxenes have Mg#=90-93 (Fig.2). However, by other components pyroxenes of the Fe-bearing lherzolites resemble the composition of normal ones. Clinopyroxene contains less Na₂O (0.2-0.8 wt.%) relative to majority of clinopyroxenes in xenoliths from intraplate alkaline basalts worldwide.

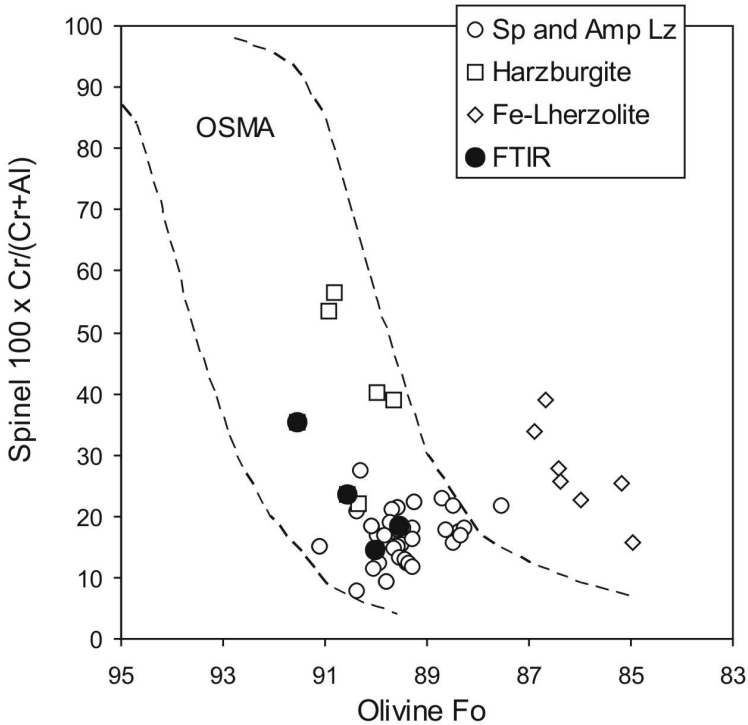


Figure 1. Relationships between the Fo content of olivine and Cr/(Cr+Al) atomic ratio of Cr-spinel in Ichinomegata xenoliths. OSMA, olivine-spinel mantle array (e.g. Arai, 1987; 1991).

Amphibole from lherzolite has typically higher Mg# (64-82) and contain 0.7-2.0 wt.% TiO₂ relative to those from amphibolites and Fe-pyroxenites and gabbros (Mg#=54-72) and 1.5-5.9 wt.% TiO₂. However, their compositions are significantly intersected indicating cognate origin of amphibole.

Fig.3 shows correlation between temperature and the oxidation state of Ichinomegata peridotites. Most of them correspond to relatively oxidized conditions of $\Delta\log fO_2(QFM)$ from -0.5 to +2.0 (comparing to majority of xenoliths from intraplate alkaline basalts ($\Delta\log fO_2(QFM)$) = -3 to 0, e.g. Litasov and Taniguchi, 2002). Two-pyroxene temperatures vary from 770 to 1050°C, with majority of estimations at 850-950°C, which are significantly higher than earlier values by Takahashi (1986).

In Figs. 1-2 we marked compositions of minerals from xenoliths, which were selected for FTIR study. They are related to harzburgite, spinel- and amphibole-bearing lherzolites, and gabbroic rocks (pyroxenite and amphibolite) (Table 1).

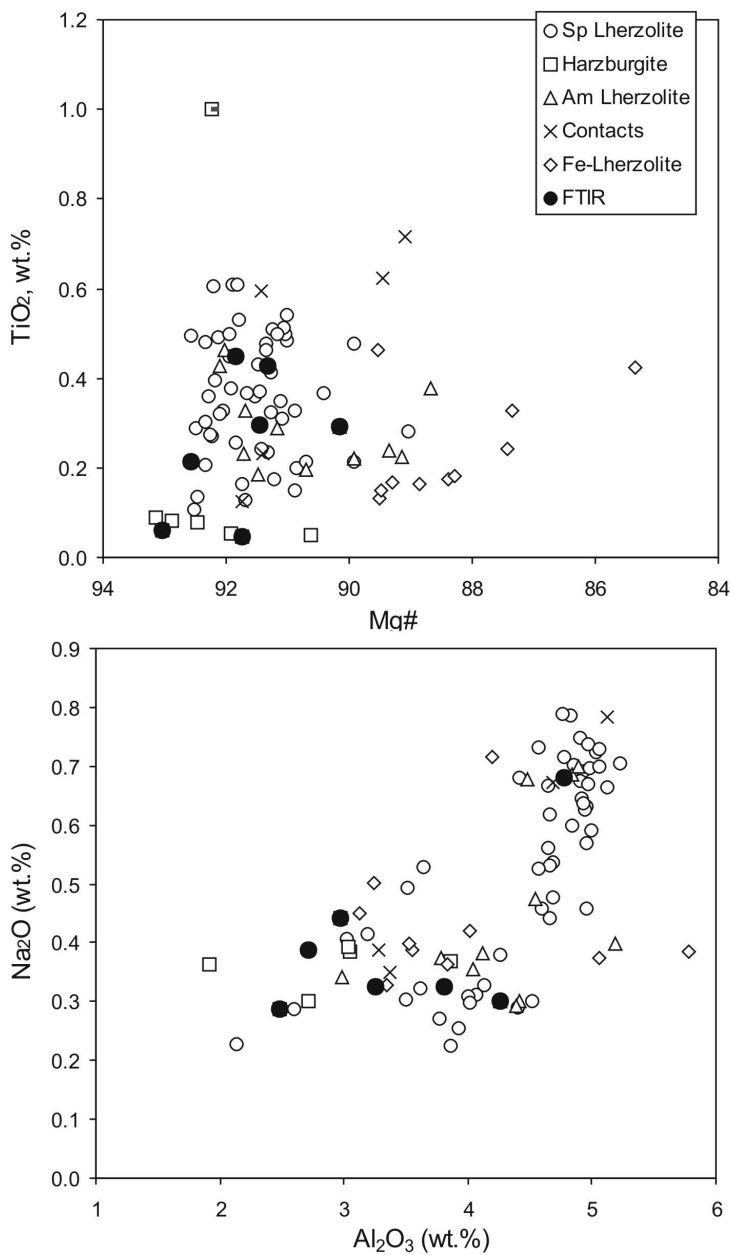


Figure 2. Composition of clinopyroxenes from Ichinomegata peridotite xenoliths.

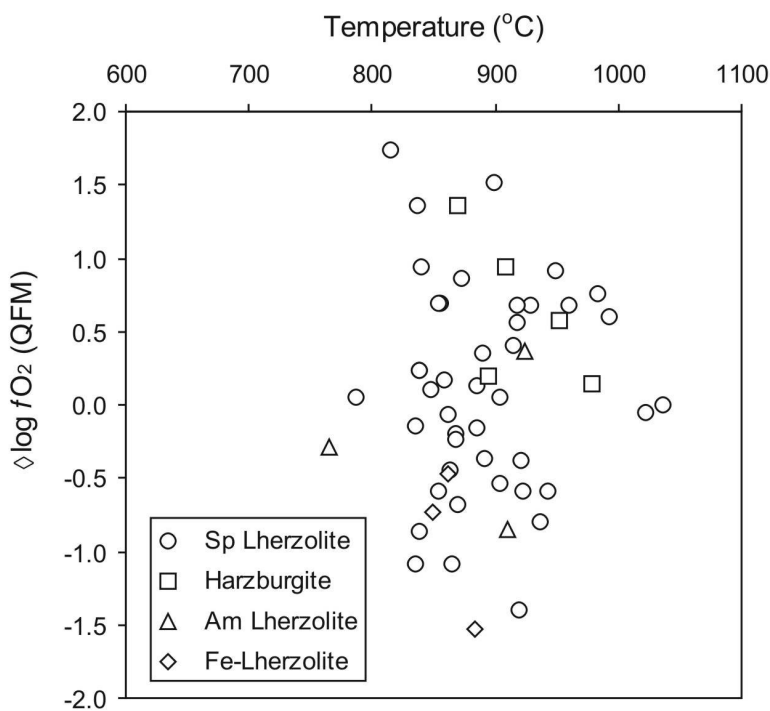


Figure 3. Temperature versus oxidation state for Ichinomegata peridotite xenoliths. The combination of two-pyroxene thermometer of Brey and Kohler (1990) and oxybarometer of Wood et al. (1990) was used.

Table 2. H₂O contents of minerals and PT-*f*O₂ parameters for the xenoliths.

	Ol, H ₂ O*	Opx, H ₂ O*	Cpx, H ₂ O*	T, Ca-OPx (°C)	T, 2-Px (°C)	Δlog <i>f</i> O ₂ (QFM)
Ig-16	< 5	277	603	889	838	+0.23
Ig-36	< 10	241	449	857	858	+0.16
Ig-31	< 17	256	633	882	867	
Ig-21	< 11	202	647	885	1001?	
Ig-24	< 5	184	537	860	907	+0.94
Ig-4	< 7	241	581	898	952	+0.58
Ig-42			359	871	887	

T, Ca-OPx - Ca-in-orthopyroxene and T, 2-Px - two pyroxene thermometers of Brey and Kohler (1990).

Δlog *f*O₂ (QFM) – olivine-spinel oxybarometer of Wood et al. (1990) combined with T, 2-Px.

* The H₂O contents are in ppm wt.

5. FTIR spectra and hydrogen contents

The H₂O contents were measured in minerals of 10 xenoliths (Table 2). The OH infrared bands occur between 3750 and 2800 cm⁻¹ in Ichinomegata pyroxenes, and positions of peaks and their

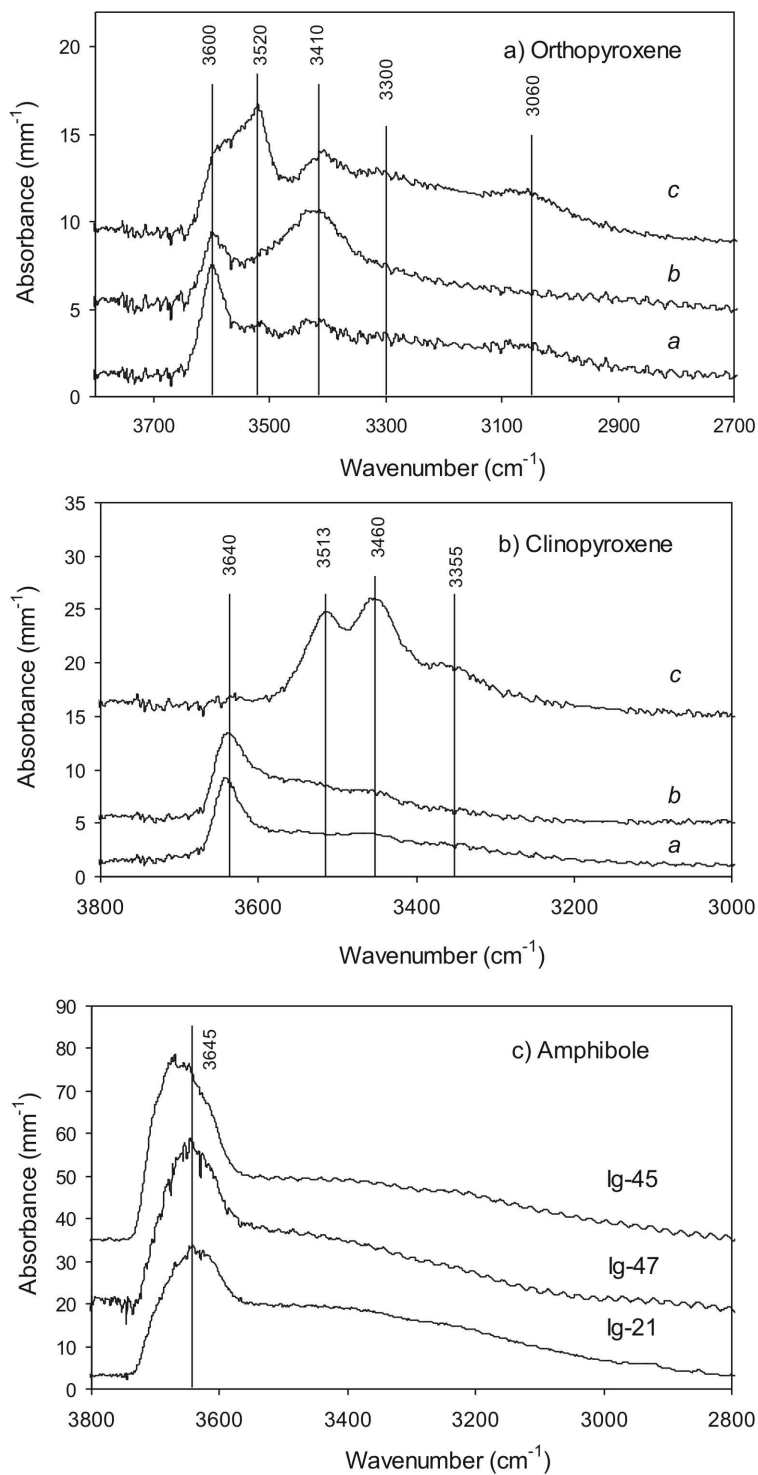


Figure 4. Representative FTIR spectra of minerals from Ichinomegata xenoliths.

intensities are similar with those reported for pyroxenes from Mexican and Simcoe (WA, USA) peridotite xenoliths described by Peslier et al. (2002) and those from pyroxenites reported by Bell et al. (1995). Orthopyroxene has several absorption bands at 3600 and 3410 cm^{-1} in the spectra with the electric field vector (E) parallel to the a and b axes and at 3520, 3410, 3300, and 3060 cm^{-1} in the spectra with $E // c$. Clinopyroxene has sharp bands at 3640 cm^{-1} in the spectra with the $E // a$ and b (which are nearly equal) and at 3513 and 3460 cm^{-1} in the spectra with $E // c$ (with smaller feature at 3355 cm^{-1}) (Fig.4). There are no significant differences in intensity of the individual bands of pyroxene spectra from lherzolite and pyroxenite. Olivine reveals only two minor bands at 3343 and 3570 cm^{-1} , which are just slightly above the baseline of unpolarized spectra. Amphibole reveal broad band at 3645 cm^{-1} , which may be shifted to 3660 cm^{-1} in amphibolites (Fig.4).

The calculated H_2O contents of minerals from Ichinomegata xenoliths are listed in Table 2. Olivine contains less than 20 ppm wt. H_2O , which is close to the detection limit of FTIR microscope used (ca. 10 ppm wt. H_2O). Accordingly, we could not be able to calculate possible diffusion profiles in olivines, which were observed in the range of 0 to 6 ppm wt H_2O by Peslier et al. (2006). Clinopyroxene contains 359-647 ppm wt. H_2O , whereas orthopyroxene contains 184-277 ppm wt. H_2O . We did not estimate H_2O contents of amphibole, since there is no high-quality calibration reported for mantle amphibole. We did not observed significant dependence of H_2O contents of minerals from composition, texture and type of xenoliths. However, it should be noted that we measured H_2O contents only in limited amount of samples. Partition coefficient of H_2O between clinopyroxene and orthopyroxene $\text{DC}_{\text{px}/\text{Opx}} = 1.8\text{-}3.2$. It is almost in the same range with that reported by Peslier et al. (2002), $\text{DC}_{\text{px}/\text{Opx}} = 2.0\text{-}2.9$.

6. Discussion and conclusions

Olivine from Ichinomegata xenoliths contains surprisingly less amount of water than it could be expected according to significant hydration of the mantle wedge and data for solubility of H_2O in olivine at PT-conditions (e.g. Lemaire et al., 2004; Zhao et al., 2004) approximately corresponding to those for the mantle wedge beneath Ichinomegata (1 GPa and 900°C). However, pyroxenes contain significant amount of H_2O (Fig.5). Low H_2O contents of olivine may be explained by possible loss of hydrogen during transportation of xenoliths to the surface (e.g. Peslier et al., 2002; Peslier and Luhr, 2006; Demouchy et al., 2006). Dehydration experiments have shown that hydrogen diffusion in olivine and clinopyroxene is very fast (e.g. Ingrin et al., 1995; Hercule and Ingrin, 1999; Mackwell and Kohlstedt, 1990; Demouchy and Mackwell, 2003). Thus, H_2O contents measured in

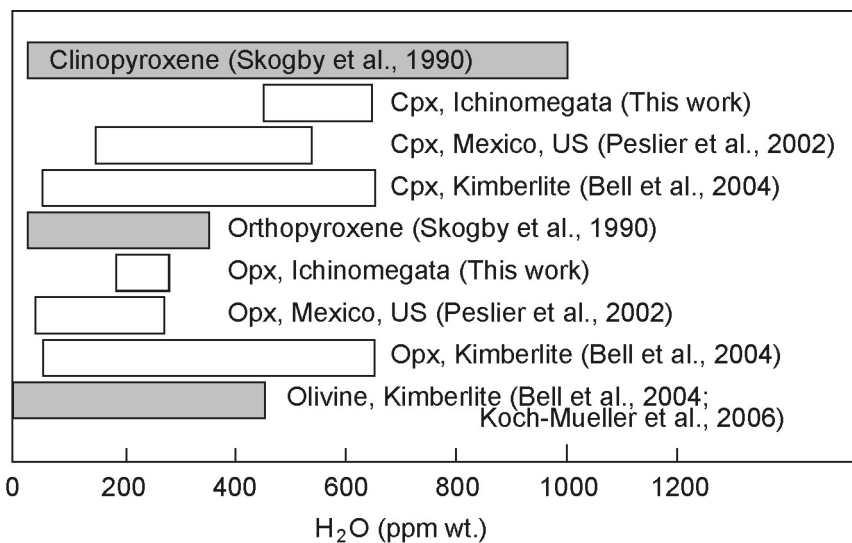


Figure 5. Comparison of hydrogen contents in minerals from Ichinomegata xenoliths with those from other locations worldwide.

xenolith minerals may underestimate the original mantle abundances.

Peslier et al. (2002) showed the correlation between H₂O contents of pyroxenes and xenolith compositions (such as bulk Al₂O₃, Na₂O in clinopyroxene, Fe³⁺ in spinel), and argued that hydrogen loss during xenolith entrainment to the surface is not important for pyroxene. Instead, H₂O content of pyroxenes from xenoliths may, in particular, reflects original water content of the rock regulated by metasomatic or partial melting processes in the mantle. At the same time, significant negative correlation between calculated Fe³⁺/ΣFe ratios in spinel and H₂O contents of pyroxenes suggest that the oxidation state of mantle can also control the amount of water present in pyroxenes. This is consistent with the prevailing mechanism of H incorporation in pyroxenes by reduction of iron by the reaction (Skogby and Rossman, 1989):



The H₂O contents of pyroxenes from Ichinomegata, which are more oxidized than subarc xenolith from Mexico and Simcoe (Peslier et al., 2002), is high relative to those for mentioned locations. This may indicate that it is either independent of the oxidation state of the uppermost mantle or simply that mantle wedge beneath Ichinomegata has higher degree of hydration.

We did not observe significant dependence of H₂O contents of minerals from the composition and texture of xenoliths. Pyroxenes from amphibole-bearing peridotites do not reveal higher water contents relative to anhydrous peridotites (note that major element composition of pyroxenes also

do not deviate from normal in amphibole-bearing peridotites, Fig.2). As well, pyroxenes from amphibole-bearing pyroxenites contain even less amounts of H₂O relative to peridotitic ones (Table 2). This facts are most likely suggest that H₂O in pyroxenes is not related to late metasomatic events formed amphibole in peridotite and abundant amphibolites, but corresponds to earlier metasomatic events in geological history of the mantle wedge beneath Ichinomegata. Besides, we can argue that peridotite of the mantle wedge acts more as an easy medium for water to be transported to the surface by arc magmas, rather than being a major contributor in the subduction zone water budget.

Here we reported first data for H₂O contents in minerals of mantle-wedge related peridotites of Ichinomegata volcano, however additional systematic study might be necessary to clarify various dependences of these H₂O contents from major and trace element composition and P-T-fO₂ parameters of the xenoliths.

Acknowledgements

KL thanks to Y. Ito for help in EPMA and acknowledges a Center for Northeast Asian Studies (CNEAS), Tohoku University, Japanese Society for Promotion of Sciences and 21th Century COE program at Tohoku University for Research Fellowships during 1999-2007. The constructive comments by anonymous referees were very helpful to improve the quality of the manuscript.

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