

Metasomatism of the peridotites from southern Mariana fore-arc: Trace element characteristics of clinopyroxene and amphibole

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Modal composition and mineral composition of harzburgites from the southern Mariana fore-arc show that they are highly refractory. There are a few modals of clinopyroxene (0.7 vol %) in harzburgites. Two types of amphibole are found in these harzburgites: magnesiohornblende accompanied by clinopyroxene with higher Al₂O₃ content (>7%) and lower Mg[#]; tremolite around orthopyroxene with lower Al₂O₃ content (<2%) and higher Mg[#]. Trace element of clinopyroxene and two types of amphibole are analyzed. Primitive mantle-normalised REE patterns for clinopyroxene and magnesio hornblende are very similar and both show HREE enrichment relative to LREE, while magnesiohornblende has higher content of trace element than clinopyroxene. The contents of trace element of tremolite are much lower than those of magnesiohornblende. Clinopyroxene shows enrichment of most of the trace element except HREE and Ti relative to clinopyroxene in abyssal peridotites. Petrology and trace element characteristic of clinopyroxene and two types of amphibole indicate that southern Mariana fore-arc harzburgites underwent two stages of metasomatism. The percolation of a hydrous melt led to mobility of Al, Ca, Fe, Mg, Na, and large amounts of trace element. LILE and LREE can be more active in hydrous melt than HREE and Ti, and the activities of most of the trace element except some of LILE are influenced by temperature and pressure.

trace element, clinopyroxene, amphibole, harzburgite, metasomatism, Mariana

1 Introduction

Clinopyroxene is preferred to melt over olivine and orthopyroxene during mantle melting^[1,2] and its composition can easily be influenced by percolating melts/fluids. Sometimes clinopyroxene can also be a kind of new phase formed during the interaction between wall-rock and metasomatic melt/fluids^[3]. Amphibole is commonly observed as metasomatic mineral in mantle peridotites. So, the composition of clinopyroxene and amphibole in peridotites is one of the ways for understanding the metasomatism of the mantle.

Peridotites from the Izu-Bonin-Mariana sub-arc are poorly studied^[4–8], partially because it is difficult to collect the samples and most of the samples are strongly

serpentitized. Studies of harzburgites from ODP 125 drilling of Conical Seamount in the Mariana fore-arc and Torishima Fore-arc Seamount in the Izu-Bonin fore-arc (ODP 125 harzburgites) show that these harzburgites are the residual after 15%–25% partial melting and have been metasomatised^[5–8]. Peridotites with abundant amphibole from the southern Mariana fore-arc described by Ohara and Ishii (1998) are also the residual of high partial melting. Modal compositions and whole

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rock and mineral compositions are the result of infiltration of metasomatic melt/fluid which led to enrichment in Ti, Al, Fe, K and Na^[4].

In this paper, we attempt to understand the nature of metasomatism in the southern Mariana sub-arc mantle through examining trace element of clinopyroxene and amphibole in harzburgites from southern Mariana fore-arc (SM harzburgites).

2 Geological setting

The Izu-Bonin-Mariana (IBM) arc system, where the Pacific plate and Philippine Sea plate converge, extends 2500 km south from near Tokyo, Japan, to beyond Guam, USA (Figure 1). The true subduction of the Pacific plate into the Philippine Sea plate probably began at about 43 Ma^[10]. The Izu-Bonin-Mariana arc system can be divided into two segments according to its mor-

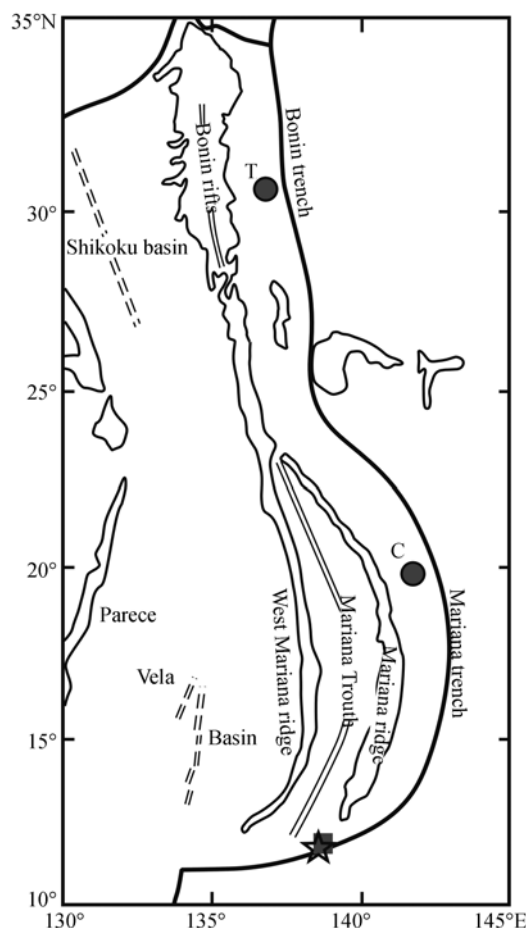


Figure 1 Major geological components of the Izu-Bonin-Mariana arc system (modified after Fryer and Pearce, 1992^[9]). Dot T is the Torishima seamount, C is the Conical seamount; the black square denotes the samples described by Ohara and Ishii (1998), while the star refers to the samples location in this paper.

phology: the Izu-Bonin segment (north of 24°N) with linear shape and the Mariana segment (south of 24°N) with arc shape. The main difference between these two segments is that the back-arc region of the Mariana segment develops an active back-arc basin known as the Mariana Trough. From north to south, the Mariana Trough changes from rifting to sea floor spreading and the period of back-arc rifting before spreading began is estimated to be less than 3 Ma^[11].

3 Samples and mineral composition

Samples come from a dredge haul (KH03-3) of the research vessel Hakuho of the University of Tokyo collected during the year 2003 in the southern part of the Mariana Trench between -4900 m and -4500 m. Mineral compositions were analyzed with EPMA-1600 electron microprobes at the Institute of Geochemistry (IG), Chinese Academy of Sciences. The operating conditions were 25 kV accelerating voltage. The analyzed results are provided in Tables 1 and 2.

Harzburgites with amphibole display porphyroclastic textures just like the Event 3 high temperature deformation described by Girardeau and Lagabriele^[7]. Primary modal compositions are 77.6% olivine, 17.3% orthopyroxene, 0.7% clinopyroxene, 3.4% amphibole and 1% spinel.

Olivines can be up to 1 cm long, while the smallest one only is 0.2 mm (enclosed by orthopyroxene). Most of them are 0.5–2 mm, and generally show wave extinction. Orthopyroxenes generally have resorption texture and show wave extinction. The size of orthopyroxenes ranges from 0.2 to 3 mm but is mainly within 0.5–2 mm. The compositions of olivine and orthopyroxene are relatively constant with the average of $Mg^{\#}$ 0.914 and 0.916, respectively, which are approximately the same as peridotites from the IBM fore-arc^[4,5]. The high $Mg^{\#}$ of olivine and orthopyroxene indicates that they are the residual of high degree melting.

Clinopyroxenes commonly range from 0.3 mm to 1.4 mm and the composition is also relatively constant with an average $Mg^{\#}$ 0.957. Compared to the primary clinopyroxene in ODP 125 harzburgites^[5], these clinopyroxenes have lower Al_2O_3 and higher $Mg^{\#}$.

Spinel commonly vary from 0.3 mm to 1 mm in size and most of them have subhedral shape. The composition of spinels exhibits a wide variation and the average $Cr^{\#}$ is 0.595. Furthermore, most of the spinels show

Table 1 Major element of mineral assemblages in harzburgites in southern Mariana fore-arc

	ol(7)		opx(11)		cpx(12)		sp2(6)		sp1	
	<i>x</i>	<i>d</i>	<i>x</i>	<i>d</i>	<i>x</i>	<i>d</i>	<i>x</i>	<i>d</i>	core	rim
SiO ₂	40.92	0.15	57.43	0.52	55.29	0.49				
Al ₂ O ₃			0.70	0.26	0.61	0.13	20.48	1.47	19.28	24.42
FeO	8.40	0.31	5.72	0.18	1.43	0.10	23.90	1.04	23.56	21.87
MnO			0.12	0.11						
MgO	49.74	0.39	34.86	0.25	17.76	0.12	10.60	0.57	10.57	11.85
CaO			0.31	0.13	24.94	0.32				
Na ₂ O					0.02	0.01				
K ₂ O										
Cr ₂ O ₃			0.15	0.11	0.30	0.08	44.78	1.96	46.21	41.41
NiO	0.31	0.11	0.03	0.03	0.01	0.01				
sum	99.37	0.35	99.32	0.42	100.36	0.42	99.75	0.52	99.62	99.55
Mg [#]	0.914	0.003	0.916	0.003	0.957	0.003				
Cr [#]							0.595	0.028	0.617	0.532

The major elements of olivine (ol), orthopyroxene (opx), clinopyroxene (cpx) and spinel 2 (sp 2) are given as average (*x*) and average absolute deviation (*d*), the analysis number is given in bracket. Spinel 1 (sp 1) is a representative major elements composition of spinel in core and rim. Mg[#]=Mg/(Mg+Fe); Cr[#]=Cr/(Cr+Al). Unit: %.

Table 2 Major and trace elements ($\times 10^{-6}$) of amphibole in harzburgites from southern Mariana fore-arc

		SiO ₂	Al ₂ O ₃	FeO	MnO	MgO	CaO	Na ₂ O	K ₂ O	Cr ₂ O ₃	NiO	sum	Mg [#]	Name
D7-64-3a1 ^{a)}	Tyep 1	46.27	9.51	2.68		19.56	11.57	2.08	0.02	1.82	0.12	93.63	0.929	Magnesio-hornblende
D7-64-5a2	Tyep 1	46.25	9.01	2.37	1.21	19.18	11.34	2.37	0.01		0.04	91.78	0.935	Magnesio-hornblende
D7-64-7a2	Tyep 1	46.32	9.31	2.58	0.03	19.47	11.42	2.36	0.05	1.85	0.04	93.43	0.931	Magnesio-hornblende
D7-64-14a1	Tyep 2	57.14	1.57	1.62		23.74	12.04	0.47			0	96.58	0.963	Tremolite
D7-64-17a1	Tyep 1	47.01	9.42	3.06	0.02	19.52	11.47	2.57	0.01	2.04	0.05	95.17	0.919	Magnesio-hornblende
	Rb	Ba	Th	U	Nb	Ta	La	Ce	Pb	Pr	Sr	Nd	Zr	Hf
D7-64-3a1	0.089	3.83	<0.005	0.006	0.113	<0.003	0.026	0.069	0.740	0.014	7.12	0.118	0.449	<0.030
D7-64-5a2	0.652	25.4	<0.007	0.009	0.266	0.010	0.029	0.095	0.770	0.019	9.04	0.091	0.424	<0.027
D7-64-7a2	0.166	4.71	<0.007	<0.007	0.097	<0.006	0.037	0.076	5.68	0.012	7.63	0.145	0.376	<0.035
D7-64-14a1	0.078	0.660	<0.007	0.142	0.011	<0.003	0.006	<0.004	1.81	0.004	10.8	<0.029	<0.041	<0.027
D7-64-17a1	0.477	17.4	<0.011	<0.011	0.223	0.010	0.046	0.112	1.72	0.014	9.69	0.135	0.463	0.037
	Sm	Eu	Ti	Gd	Tb	Dy	Y	Ho	Er	Tm	Yb	Lu	Sc	V
D7-64-3a1	0.070	0.038	258	0.211	0.044	0.455	3.58	0.152	0.492	0.100	0.790	0.124	126	410
D7-64-5a2	0.081	0.059	316	0.216	0.058	0.426	3.18	0.131	0.408	0.080	0.630	0.117	121	463
D7-64-7a2	0.066	0.038	303	0.207	0.052	0.417	3.71	0.136	0.512	0.089	0.840	0.121	129	430
D7-64-14a1	<0.023	0.009	55.5	0.026	0.003	0.044	0.244	0.006	0.039	0.009	0.042	0.010	50.1	121
D7-64-17a1	0.102	0.034	381	0.136	0.074	0.499	3.92	0.141	0.557	0.086	0.610	0.131	126	442

a) D7-64 is the sample number, 3a1 analysis number, same as in Table 3.

higher Cr[#] in the core and lower Cr[#] in the rim (Table 1). This kind of compositional zonation pattern occurs also in Type I harzburgites described by Ohara and Ishii^[4].

Amphiboles are euhedral with long columnar crystal shape. According to composition and occurrence, these amphiboles can be divided into two types (Denomination of amphibole accords the classification of Leake et al.^[12]): Type 1 is magnesiohornblende with accompanying clinopyroxene and has higher Al₂O₃ content (generally >7%) and lower Mg[#]; Type 2 is tremolite which occurs around orthopyroxene and exhibits lower Al₂O₃

content (generally <2%) and higher Mg[#] (Table 2; Figure 2).

4 Trace element compositions of clinopyroxene and amphibole

Trace element compositions of clinopyroxene and amphibole were analyzed using an Agilent 7500a ICP-MS with a GeoLas 200M laser ablation system at the Key Laboratory of Continental Dynamics, Northwest University. Detection limits ranged from 1.2×10^{-9} for Ta to

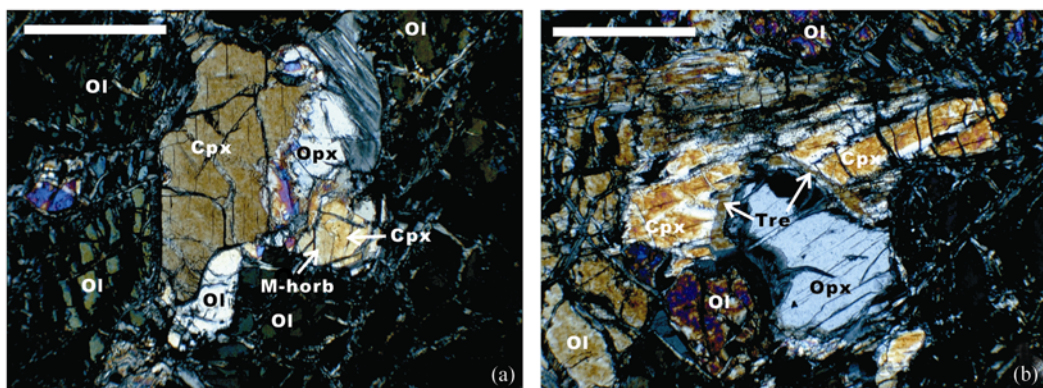


Figure 2 Microphotographies showing the correlation between pyroxene and amphibole. (a) Clinopyroxene (Cpx) accompanying magnesio-hornblende (M-horb); (b) tremolite (Tre) occurs around orthopyroxene (Opx). Scale bar is 1 mm.

Table 3 Trace element ($\times 10^{-6}$) of clinopyroxene in harzburgites from southern Mariana fore-arc

	Rb	Ba	U	Nb	La	Ce	Pb	Pr	Sr	Nd	Zr	Hf	Sm
D7-64-3a2	<0.017	0.044	0.001	<0.008	<0.008	0.012	0.247	<0.004	4.29	<0.027	0.086	<0.026	<0.041
D7-64-4a2	0.030	0.378	0.107	0.009	0.009	0.018	0.670	0.007	9.01	<0.028	0.185	<0.017	<0.017
D7-64-2a1	0.017	0.401	0.006	<0.009	0.012	0.016	0.182	<0.004	5.43	0.062	0.229	<0.018	0.038
D7-64-10a1	0.031	0.209	0.052	<0.008	0.013	0.016	0.347	0.002	4.20	0.039	0.436	<0.024	<0.018
D7-64-7a1	0.019	0.076	<0.006	0.008	0.007	0.020	0.600	<0.003	4.27	<0.026	0.098	0.010	<0.023
D7-64-6a3	0.044	0.199	0.014	<0.008	0.011	0.023	1.00	<0.004	4.86	0.030	0.124	0.025	0.030
D7-64-5a1	<0.015	0.102	<0.003	0.006	0.007	0.015	1.23	<0.004	4.07	0.029	0.064	<0.027	0.027
D7-64-17a2	0.035	0.293	<0.008	<0.012	<0.007	0.017	1.91	<0.007	4.54	<0.039	0.122	<0.034	0.043
	Eu	Ti	Gd	Tb	Dy	Y	Ho	Er	Tm	Yb	Lu	Sc	V
D7-64-3a2	<0.009	60.6	0.038	0.012	0.082	0.459	0.019	0.061	0.017	0.095	0.019	60.7	123
D7-64-4a2	0.008	83.2	0.029	0.010	0.071	0.486	0.020	0.052	0.011	0.135	0.021	67.2	142
D7-64-2a1	0.017	77.4	0.031	0.013	0.167	0.900	0.038	0.102	0.024	0.158	0.033	66.5	178
D7-64-10a1	<0.008	52.6	0.055	0.011	0.093	0.611	0.021	0.061	0.017	0.125	0.024	55.9	106
D7-64-7a1	<0.007	67.8	<0.024	0.011	0.054	0.510	0.023	0.065	0.012	0.096	0.029	59.5	141
D7-64-6a3	0.013	86.4	0.077	0.017	0.138	0.867	0.029	0.138	0.024	0.170	0.026	64.8	164
D7-64-5a1	0.007	62.1	0.034	0.005	0.061	0.419	0.017	0.062	0.010	0.104	0.017	54.8	102
D7-64-17a2	<0.011	82.4	<0.032	0.008	0.050	0.544	0.014	0.074	0.015	0.119	0.022	62.2	119

0.5×10^{-9} for Ti. The NIST 610 glass standard was used for calibration of relative element sensitivities and each analysis was normalized to CaO values determined by an electron microprobe. The analyzed results are provided in Tables 2 and 3. In the following sections, all the trace element ratios have been calculated using data normalized to the primitive mantle values^{13]}.

4.1 Clinopyroxene

Primitive mantle-normalised REE patterns for clinopyroxenes in SM harzburgites show a smooth downward slope from HREE to MREE with a concave up-trend to La (Figure 3(b)). The values of La/Yb and La/Ce are 0.04–0.07 and 0.90–2.15, respectively. Primitive mantle-normalised trace element patterns display strong positive U (U/Nb: 368.97), Pb (Pb/Pr: 162.08–267.08) and Sr (Sr/Nd: 5.50–10.18) anomalies (Figure 3(a)). Rb

and Ba are enriched compare to Ce in clinopyroxenes with 2.67–5.62 for Rb/Ce and 0.96–6.24 for Ba/Ce, although they display no evident positive anomalies. Most of pyroxenes display strong negative Zr (Zr/Nd: 0.26–0.49) and weak negative Ti (Ti/Gd: 0.43–1.30) anomalies.

The contents of trace element in clinopyroxenes in SM harzburgites are far lower than the ones in abyssal peridotites from the Indian Ocean ridge^[14], except Sr (Figure 4(a)). But these two display strong negative Zr and weak negative Ti anomalies.

The trace element patterns of clinopyroxenes in SM harzburgites have a closed shape similar to clinopyroxenes in ODP 125 harzburgites, while the former have relatively high Sr content, much lower Ce content and a little lower other trace element contents (such as Zr, Ti, Dy and Yb) (Figure 4(a)).

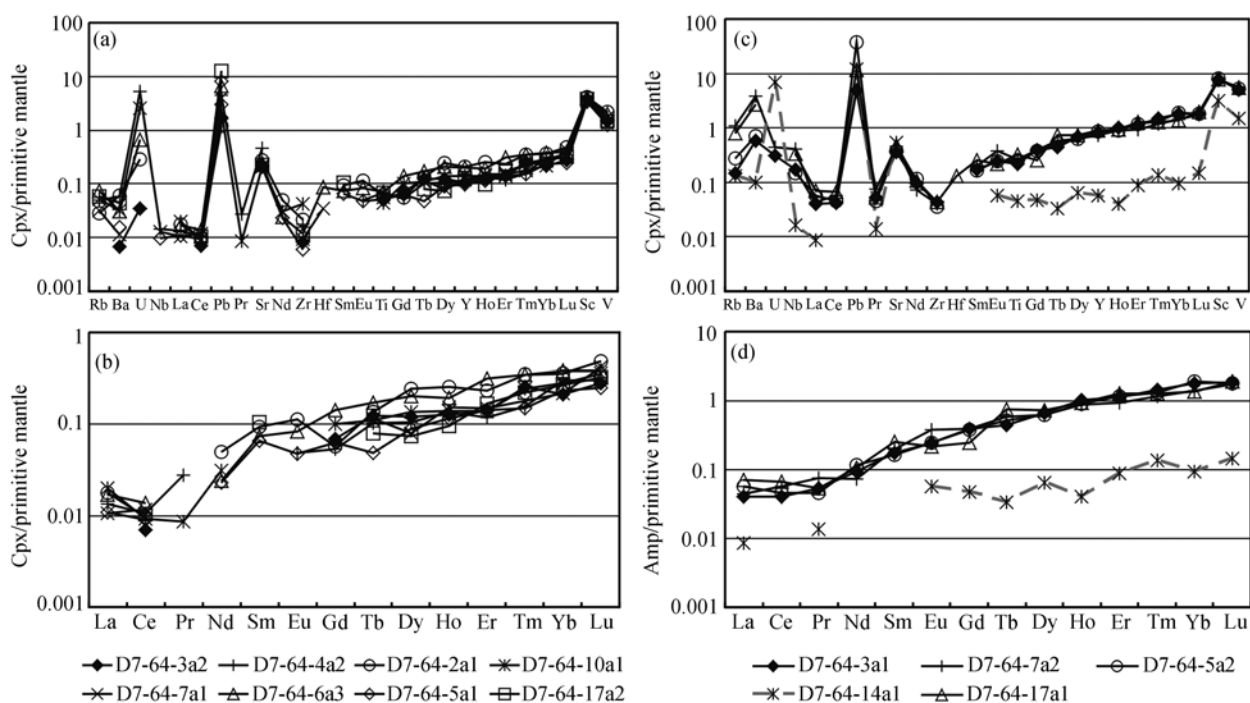


Figure 3 Trace element and REE patterns for clinopyroxene (Cpx) and amphibole (Amp).

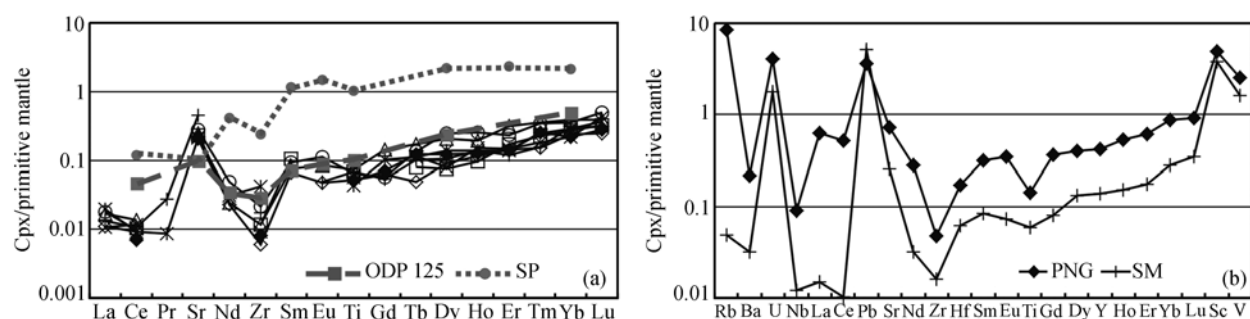


Figure 4 (a) Trace element patterns of clinopyroxene in harzburgites from the southern Mariana fore-arc (black lines), compared with clinopyroxene in abyssal peridotite (SP) and ODP 125 harzburgites. (b) Diagram illustrating the close similarities of primitive mantle normalized trace element patterns for clinopyroxene between the southern Mariana fore-arc harzburgite (SM) and the harzburgite from the Lihir fore-arc, Papua New Guinea (PNG). The legend of SM in (a) refers to Figure 3(a). ODP 125, SP in (a) and PNG, SM in (b) are given in average.

4.2 Amphibole

Magnesianhornblends have much higher contents of trace element than clinopyroxenes in SM harzburgites. Primitive mantle-normalised REE patterns for magnesianhornblends show a smooth downward slope from HREE to LREE (La/Yb: 0.02–0.05) (Figure 3(d)) and essentially parallel the patterns for clinopyroxene, but without La to Ce enrichment (La/Ce: 0.78–1.26). Strong positive Ba (Ba/U: 1.90–8.69), Pb (Pb/Pr: 67.21–829.15) and Sr (Sr/Nd: 3.31–6.24) anomalies are displayed in primitive mantle-normalised trace element patterns for magnesianhornblends (Figure 3(c)). High incompatible elements, such as Rb, U and Nb, are

enriched compared to Ce with 3.60–19.16 for Rb/Ce, 7.41–7.82 for U/Ce and 3.25–7.13 for Nb/Ce. Positive Zr anomaly is evident while Ti shows no anomaly.

REE contents, especially MREE and HREE, of tremolites are much lower than magnesianhornblends in SM harzburgites. Primitive mantlenormalised REE patterns for tremolites show a much shallower downward slope from HREE to LREE (La/Yb: 0.09) than magnesianhornblends. Primitive mantle-normalised trace element patterns for tremolites display strong positive U (U/Nb: 422.27), Pb (Pb/Pr: 875.70) and Sr (Sr/Pr: 39.42) anomalies and Rb (Rb/La: 15.04), Ba (Ba/La: 11.57) and Nb (Nb/La: 809.43) to La enrichments.

5 Discussion

5.1 Mantle metasomatism

The modal composition and the high $Mg^\#$ of olivine and orthopyroxene indicate these SM harzburgites are solid residues by high degrees of partial melting. The characteristics of clinopyroxenes show that they are more depleted than the ODP 125 harzburgites (such as much lower REE content).

Figure 5 shows the correlation between the $Mg^\#$ and abundance of various trace elements for clinopyroxenes. Almost all the trace element show a good negative correlation with $Mg^\#$ for clinopyroxenes in abyssal peridotites which are commonly considered solid residues from dry melting. In clinopyroxenes in SM harzburgites, those less incompatible elements, such as Ce, Sr, Nd, Zr,

Sm and Eu, are relatively enriched than in abyssal peridotites while the more incompatible elements, such as Ti and HREE show no relative enrichments. The same characteristics of trace element in clinopyroxenes of ODP 125 harzburgites had been described by Parkinson et al.^[6] (Figure 5). Bizimis et al.^[15] suggest that because these harzburgites underwent hydrous melting, that is, an hydrous fluid originating from the subducted slab flowed promoting the melting of peridotites in the sub-arc mantle wedge and enriched them with less incompatible elements, such as LREE, MREE, Zr, Sr, and so on.

The occurrence of amphibole in SM harzburgites also indicates that these harzburgites have undergone hydrous metasomatism. Previous studies show that the content of Al in amphibole increases with the rise of

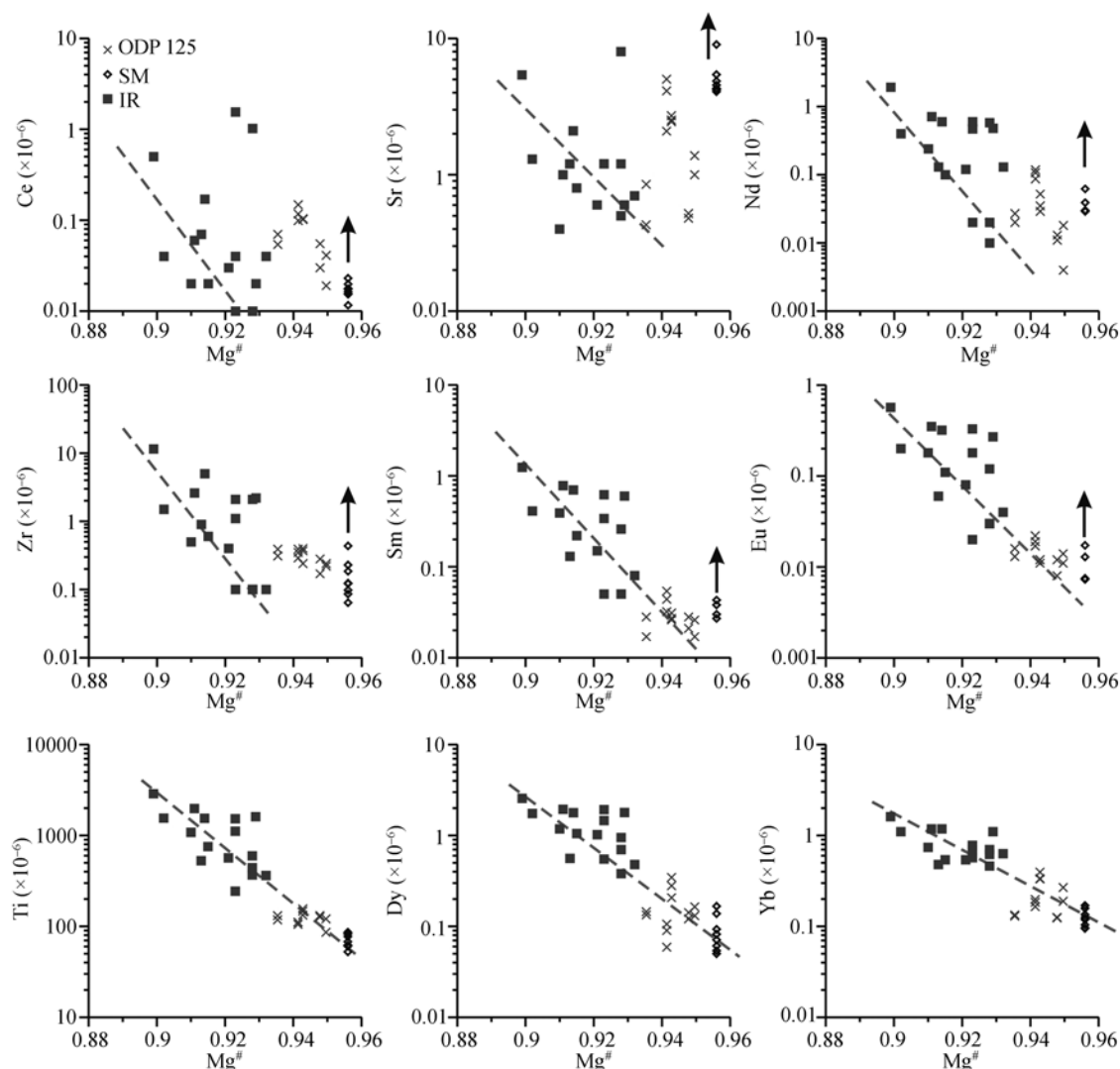


Figure 5 Relation between the average $Mg^\#$ and trace element of clinopyroxene. Clinopyroxene in abyssal peridotites (IR)^[14], ODP 125 harzburgites^[6] are shown for comparison. Dot line shows the depleted direction during dry melting; arrow line indicates enrichment by metasomatism.

temperature and pressure^[16], so the two types of amphibole represent two stages of hydrous metasomatism:

(1) In higher temperature and pressure, hydrous metasomatic agent infiltrated southern Mariana sub-arc mantle wedge and formed magnesiohornblendes when reacting with clinopyroxenes. The similarity of trace element and REE patterns between clinopyroxenes and magnesiohornblendes indicates that they belong to the same stage of hydrous reaction with mantle wedge peridotites.

(2) In relatively low temperature and pressure, hydrous agent replaced orthopyroxenes in SM harzburgites and formed tremolites.

5.2 Characteristics of metasomatic agent

Figure 4(b) illustrates the close similarities of trace element patterns between clinopyroxenes in SM harzburgites and harzburgites from the Lihir fore-arc, Papua New Guinea (PNG harzburgites)^[17]. Their trace element patterns display similar shapes and the same positive Rb, U, Pb and negative Zr, Ti anomalies. McInnes et al.^[18] proposed that PNG harzburgites have been metasomatised by a high-density hydrous melt which induced the mobility of Al, Si, Na, K and S. Grégoire et al.^[17] argued that this kind of hydrous melt is enriched in LILE and LREE and relatively depleted in Zr, Ti and HREE. The similar trace element characteristics of clinopyroxenes observed between the SM harzburgites and the PNG harzburgites suggest that the upper mantle beneath southern Mariana sub-arc and Lihir sub-arc has undergone a metasomatism related to percolation of a probably similar hydrous melts. In fact, during the first stage of metasomatism in SM harzburgites, the occurrence of magnesiohornblendes indicates that the hydrous agent is also a kind of highly dense melt and induced the mobility of Al, Si, Ca, Fe, Mg, Cr and Na., the enrichments of LILE (such as Rb, Ba, U, Pb and Sr) and LREE in clinopyroxenes suggest that the hydrous melt was enriched in LILE and LREE relative to HREE and Ti. This phenomenon is consistent with the results of recent high temperature and pressure experiments which suggest that the activity of Ti and HREE in subducting fluid/melt is lower than LILE and LREE^[19]. Furthermore, the formation of magnesiohornblendes suggests that this hy-

drous melt also carries a little amount of HREE and Ti. The main difference between SM harzburgites and PNG harzburgites corresponds to the lower trace element content of the clinopyroxenes in SM harzburgites which may be related to a more refractory character of SM harzburgites prior to the metasomatism.

During the second stage metasomatism, the formation of tremolites replacing orthopyroxenes indicates that the hydrous metasomatic agent induced the mobility of Al, Ca, Fe, Mg and Na which means that the hydrous agent may also be a highly dense melt. The difference of trace element between tremolite and magnesiohornblende indicates that the temperature and pressure may influence the activity of trace element in hydrous melt. The rise of temperature and pressure increases REE (especially MREE and HREE) and Ti activity in hydrous melt. But there is no clear influence on some of LILE (such as U, Pb, Sr).

6 Conclusion

Southern Mariana fore-arc harzburgites are highly refractory in composition and modified by subduction-related mantle metasomatism processes. The metasomatism can be divided into two stages: (1) In higher temperature and pressure, hydrous melt reacted with clinopyroxenes in harzburgites which enriched clinopyroxenes in most of the trace elements except Ti and HREE and formed magnesio-hornblendes. (2) In relatively low temperature and pressure, hydrous melt reacted with orthopyroxenes in harzburgites leading to the formation of tremolites.

The percolation of hydrous melt leads to mobility of Al, Ca, Fe, Mg, Na, and so on. The trace element signatures of clinopyroxene and amphibole indicate that LILE and LREE can be more active in hydrous melt than HREE and Ti, and the activities of most of the trace elements except some of LILE are increased by the rise of temperature and pressure.

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