

# 40Ar-39Ar analysis of phlogopite in the Horoman Peridotite Complex, Hokkaido, Japan and implications for its origin

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Research Article

# $^{40}\text{Ar}$ – $^{39}\text{Ar}$ analysis of phlogopite in the Horoman Peridotite Complex, Hokkaido, Japan and implications for its origin

ICHIRO KANEOKA<sup>1\*</sup>, NATSUKO TAKAHASHI<sup>2</sup> AND SHOJI ARAI<sup>3</sup>

<sup>1</sup>*Earthquake Research Institute, University of Tokyo, Bunkyo-ku, Tokyo 113-0032, Japan*  
(email: kaneoka@eri.u-tokyo.ac.jp) <sup>2</sup>*Faculty of Science, Chiba University, Inage-ku, Chiba 263-0022, Japan,*  
<sup>3</sup>*Faculty of Science, Kanazawa University, Kakuma-machi, Kanazawa 920-1191, Japan*

**Abstract**  $^{40}\text{Ar}$ – $^{39}\text{Ar}$  analysis of phlogopite separated from a plagioclase lherzolite of the Horoman Peridotite Complex, Hokkaido, Japan, has yielded a plateau age of  $20.6 \pm 0.5$  Ma in an environment where the metamorphic fluid was characterized by an almost atmospheric Ar isotopic ratio. The age spectrum is slightly saddle-shaped, implying some incorporation of excess  $^{40}\text{Ar}$  during the formation of the phlogopite at a depth. As the phlogopite has been inferred to have formed in veins and/or interstitials during exhumation of the peridotite body, metasomatic fluids, to which ground- and sea water might have contributed, were probably involved in the formation of phlogopite in the crustal environment. A total  $^{40}\text{Ar}$ – $^{39}\text{Ar}$  age of 129 Ma of a whole rock sample of the plagioclase lherzolite, from which the phlogopite was separated and is representative of the main lithology of the Horoman Peridotite Complex, indicates the occurrence of excess  $^{40}\text{Ar}$ . Hence, the age has no geological meaning.

**Key words:** Ar–Ar age, excess Ar, Horoman Peridotite Complex, metasomatic fluids, phlogopite.

## INTRODUCTION

The Horoman Peridotite Complex, situated in the Hidaka Metamorphic Belt, Hokkaido, Japan, is regarded as an orogenic lherzolite body, which represents large upper mantle-derived fragments and has been mechanically emplaced into the continental crust (Niida 1984; Takahashi 1991a, b; Ozawa & Takahashi 1995) (Fig. 1). It is well established that the Horoman Peridotite Complex lies on granulite to amphibolite facies gneisses of the lower metamorphic sequence of the Hidaka Metamorphic Belt, that overlies metasediments of the Idon'nappu Belt along the Hidaka Main Thrust. The basal part of the peridotites is mylonitic. These structural relationships imply a tectonic emplacement into the crust. The main lithologies of the complex are that harzburgite and

lherzolite contain variable amounts of secondary phlogopite and amphibole (Niida 1975; Arai & Takahashi 1989). Phlogopites commonly occur as veinlets or interstitially in harzburgite and spinel lherzolite (Niida 1975; Arai & Takahashi 1989; Takahashi *et al.* 1989). It has been suggested that such phlogopite together with amphibole (kaersutite) might have been formed by the reaction of the 'peridotite' with fluids, that were derived from magmatic fluids which had successively released from evolving alkali basaltic magma(s) (Arai & Takahashi 1989; Takahashi *et al.* 1989). This model assumes that phlogopites formed during the latest stage in the evolution of the Horoman Peridotite Complex.

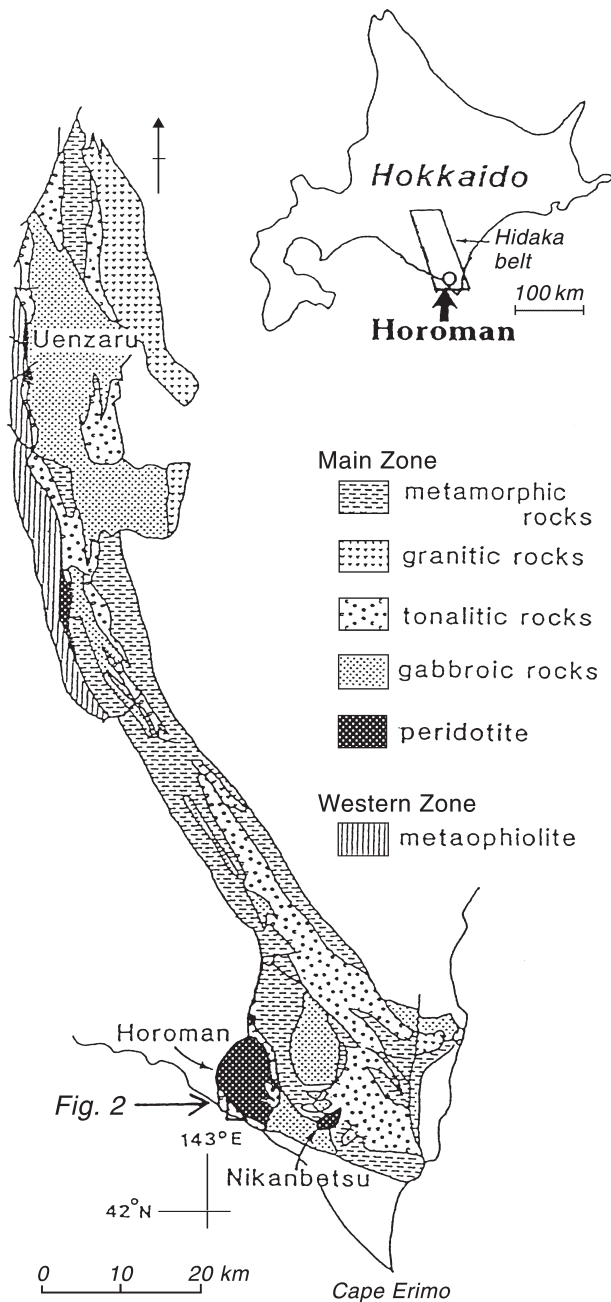
Yoshikawa *et al.* (1993) have reported a Rb–Sr age of  $23.0 \pm 1.2$  Ma for a phlogopite spinel lherzolite from the Horoman Peridotite Complex and regarded it as the age of a metasomatic event, which occurred in the wedge mantle. The initial  $^{87}\text{Sr}/^{86}\text{Sr}$  was  $0.703\,871 \pm 0.000\,007$ , which is higher than those of clinopyroxenes in the phlogopite-free peridotite (less than 0.7028) from the same area. Based on this initial value, they have inferred that

\*Correspondence.

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**Fig. 1** Geological sketch map of the Hidaka belt with the site of the Horoman Peridotite Complex.

the metasomatic fluid could have been derived from fluids caused by the dehydration of subducted oceanic sediments and altered mid-oceanic ridge basalt (MORB). In their age estimation, however, these authors assumed that isotopic equilibration among clinopyroxenes was achieved by the metasomatic fluid in the upper mantle. This was based on the similarities of the  $^{87}\text{Sr}/^{86}\text{Sr}$  among each clinopyroxene, which have higher values than those of clinopyroxenes in the phlogopite-free spinel lherzolite. The 23 Ma age is largely

dependent on the  $^{87}\text{Sr}/^{86}\text{Sr}$  value of phlogopite. If the isotopic equilibration was incomplete for clinopyroxenes with the metasomatic fluid which they used, the age might change. Hence, to substantiate the age, it is important to examine it by a different method. For this purpose, we have performed an  $^{40}\text{Ar}$ - $^{39}\text{Ar}$  analysis of phlogopite of the Horoman Peridotite Complex. As  $^{40}\text{Ar}$ - $^{39}\text{Ar}$  analyses provide an age from phlogopite alone, we can obtain its formation age directly. For comparison, we have also performed  $^{40}\text{Ar}$ - $^{39}\text{Ar}$  analysis of the host plagioclase lherzolite.

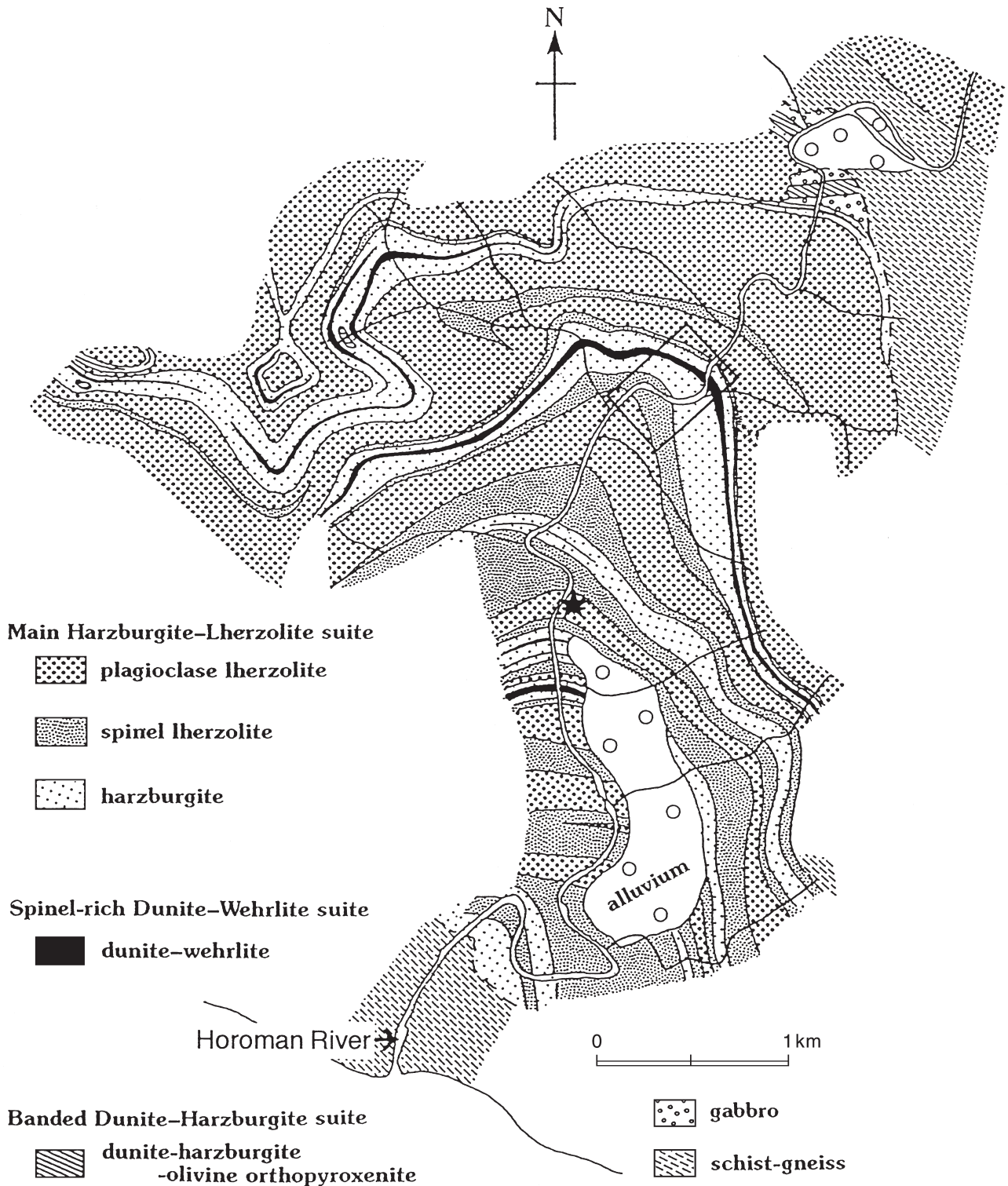
## SAMPLES

Two phlogopite samples were prepared for the present study. They were collected from the veins in the plagioclase lherzolite (Fig. 2). Such phlogopite veinlets mainly replace olivine grains and are entirely concordant with the foliation plane of peridotites (Arai & Takahashi 1989). These phlogopites are Ti rich ( $\text{TiO}_2$ ; more than 5%) and contain a small amount of Na ( $\text{Na}_2\text{O}$ ; 1.4%). Their chemical characteristics are typical of the relatively low-pressure mantle phlogopite formed in the plagioclase or spinel peridotite stability field (Arai 1984, 1986). Phlogopite grains larger than the 120 mesh were separated from host rocks, mainly by hand-picking. However, about 1–2% of kaersutite, which was probably formed at the same time as the phlogopite, could not be removed from the phlogopite separates. The kaersutite occurred near the margin or narrow parts of phlogopite-rich veinlets (Arai & Takahashi 1989). In addition, about 1–2% of chlorite and serpentine were also included in the phlogopite separates.

Although we tried to analyze two phlogopite samples, we failed to obtain reasonable data for one of them because of the leakage of sample gases through a microcrack of the reaction glass tube during analysis. Hence, we report here only the results of one sample (101212C, phlogopite) and its host rock (plagioclase lherzolite). As the host rock, the remaining part of the sample, from which phlogopites had been separated, was used. The chemical compositions of phlogopite and its host rock for the sample 101212C are given in the appendix.

## EXPERIMENTAL

Phlogopite and plagioclase lherzolite grains were wrapped in aluminium foil and vacuum-sealed in



**Fig. 2** Geological map of the southern Horoman Peridotite Complex with the sampling site for samples used in the present study (indicated with a star).

a quartz ampoule together with flux monitors (JG-1 biotite, K-Ar age,  $91.4 \pm 0.5$  Ma, Iwata 1998),  $\text{CaF}_2$  and  $\text{K}_2\text{SO}_4$ . They were irradiated with neutrons at the Japan Material Test Reactor (JMTR)

with a total fast neutron flux of about  $5 \times 10^{17}$  nvt/cm<sup>2</sup>.

The extraction and purification procedures of Ar were almost the same as those reported in

Kaneoka and Aoki (1978) and only their outline is described here. Argon gas was extracted using an induction heater (JEOL, Tokyo, Japan) and purified following a conventional procedure at the Isotope Center of the University of Tokyo, and collected in a glass ampoule for each temperature fraction. A sample was kept for 45 min at each temperature for degassing. Argon isotopic ratios were measured on a mass spectrometer with a Faraday cup (Micromass, Cheshire, UK) at the Geological Survey of Japan, whose characteristics were described in Shibata and Takagi (1988). The amount of <sup>40</sup>Ar was calculated by assuming the sensitivity of the mass spectrometer with an uncertainty of about 10%. The measured <sup>40</sup>Ar/<sup>36</sup>Ar for the atmosphere was 297.0 ± 0.8, which was used for correcting the mass discrimination of each isotopic ratio. Apparent ages of individual step were corrected for neutron irradiation-induced interfering Ar isotopes by using the following values, which were determined from K<sub>2</sub>SO<sub>4</sub> and CaF<sub>2</sub>, irradiated at the same time with present samples:

$$\begin{aligned} (^{40}\text{Ar}/^{39}\text{Ar})_{\text{K}} &= (1.65 \pm 0.03) \times 10^{-1} \\ (^{40}\text{Ar}/^{37}\text{Ar})_{\text{Ca}} &= (1.45 \pm 0.01) \times 10^{-3} \\ (^{36}\text{Ar}/^{37}\text{Ar})_{\text{Ca}} &= (1.15 \pm 0.02) \times 10^{-4}. \end{aligned}$$

Ages were calculated by using the decay constant recommended by Steiger and Jäger (1977). Uncertainties in isotopic ratios and ages are expressed at one sigma level.

## RESULTS

<sup>40</sup>Ar–<sup>39</sup>Ar analytical results for phlogopite (101212C, phlogopite) and its host rock (101212C, plagioclase lherzolite) are given in Table 1, and their <sup>40</sup>Ar–<sup>39</sup>Ar age spectra together with the (<sup>39</sup>Ar)<sub>K</sub>/<sub>(<sup>37</sup>Ar)<sub>Ca</sub> versus <sup>39</sup>Ar release diagram and the inverse–isochron plot are shown in Figs 3 and 4.</sub>

As shown in Fig. 3, the phlogopite sample yielded a plateau age of 20.6 ± 0.5 Ma for tempera-

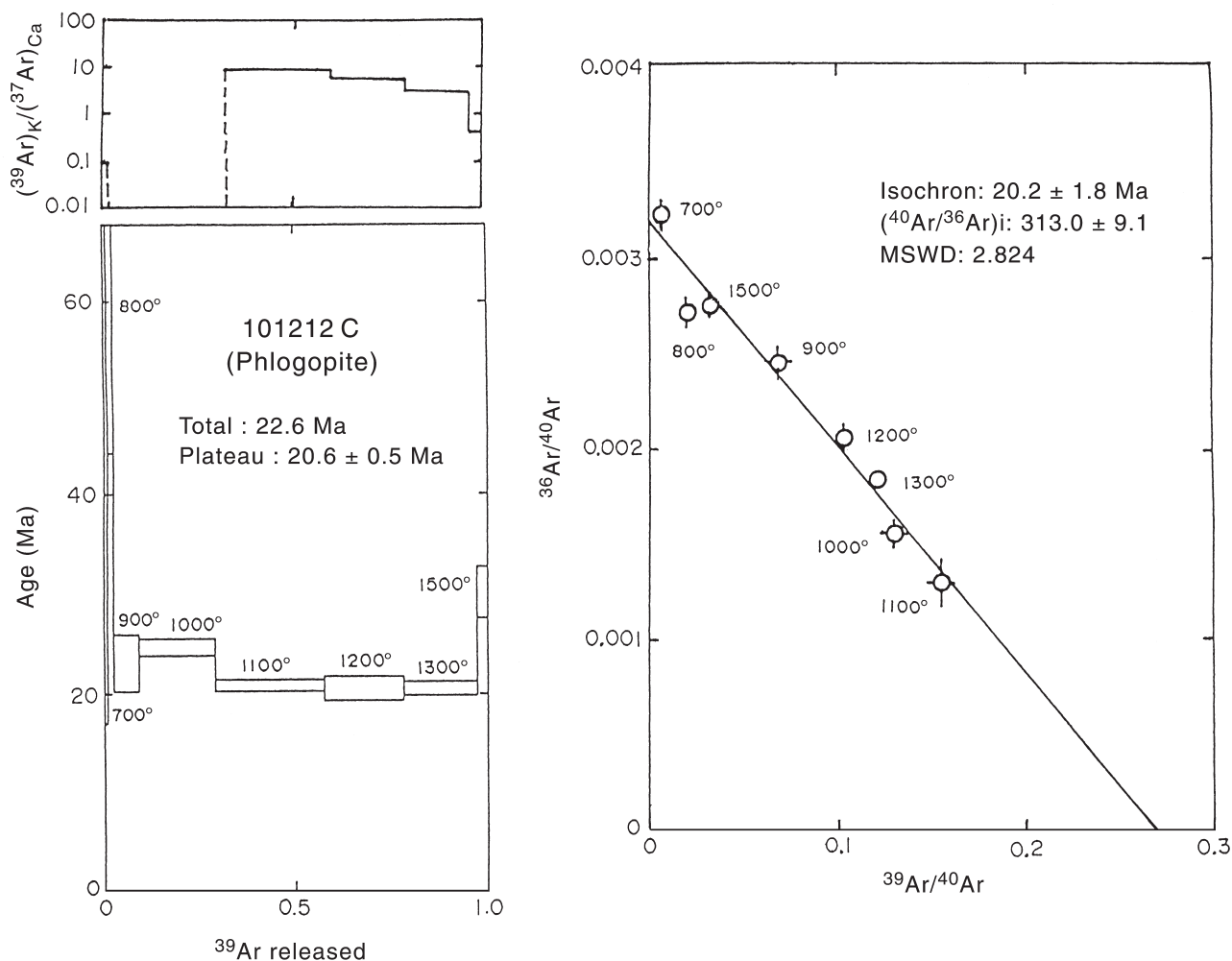
**Table 1** Ar isotopes in neutron-irradiated phlogopite and its host rock

T (°C)	<sup>40</sup> Ar (× 10 <sup>-6</sup> ) cm <sup>3</sup> STP/g	<sup>39</sup> Ar* (%)	<sup>36</sup> Ar/ <sup>40</sup> Ar (× 10 <sup>-3</sup> )	<sup>37</sup> Ar/ <sup>40</sup> Ar (× 10 <sup>-3</sup> )	<sup>39</sup> Ar/ <sup>40</sup> Ar (× 10 <sup>-2</sup> )	<sup>40</sup> Ar*/ <sup>39</sup> Ar*	Age (Ma)
101 212C (phlogopite) 0.16189 g J=0.003202 ± 0.000062							
700	1.07	0.7	3.201 ± 0.102	33.25 ± 3.15	0.8091 ± 0.0201	6.649 ± 3.711	38.0 ± 21.0
800	1.01	1.6	2.731 ± 0.148	n.d.	1.920 ± 0.013	10.05 ± 2.28	57.1 ± 12.8
900	1.25	7.1	2.467 ± 0.095	n.d.	6.893 ± 0.133	3.932 ± 0.414	22.6 ± 2.4
1000	1.92	20.0	1.541 ± 0.023	n.d.	12.67 ± 0.30	4.299 ± 0.115	24.7 ± 0.8
1100	2.12	28.1	1.302 ± 0.088	16.65 ± 2.70	16.20 ± 0.20	3.636 ± 0.046	20.9 ± 0.5
1200	2.44	20.9	2.054 ± 0.049	16.40 ± 0.75	10.48 ± 0.07	3.591 ± 0.138	20.6 ± 0.9
1300	1.86	18.7	1.852 ± 0.029	25.84 ± 2.53	12.27 ± 0.14	3.530 ± 0.080	20.3 ± 0.6
1500	0.99	2.9	2.756 ± 0.049	74.38 ± 1.59	3.492 ± 0.034	5.255 ± 0.420	30.1 ± 2.5
Total	12.66	100.0	2.085	18.34	9.633	3.929	22.6
101212C (Pl. lherzolite) 0.6091 g J=0.003330 ± 0.000064							
700	1.73	1.8	3.168 ± 0.035	21.16 ± 3.06	0.3390 ± 0.0391	209.5 ± 42.8	955 ± 152
800	1.10	26.1	3.159 ± 0.070	20.49 ± 0.81	7.233 ± 0.595	9.114 ± 2.963	53.9 ± 17.3
900	0.809	6.4	3.129 ± 0.065	27.70 ± 1.37	2.438 ± 0.029	31.55 ± 7.96	180 ± 43
1000	0.575	9.5	3.226 ± 0.048	17.96 ± 2.30	5.059 ± 0.093	9.224 ± 2.815	54.6 ± 16.4
1100	0.601	11.6	3.050 ± 0.044	13.90 ± 0.72	5.873 ± 0.089	16.77 ± 2.22	98.0 ± 12.8
1200	0.498	14.8	2.997 ± 0.070	47.14 ± 3.12	9.098 ± 0.083	12.65 ± 2.29	74.4 ± 13.3
1300	0.415	5.8	3.138 ± 0.066	35.00 ± 1.34	4.270 ± 0.156	17.31 ± 4.66	101 ± 27
1500	1.21	24.0	2.684 ± 0.071	271.8 ± 4.0	6.448 ± 0.040	35.50 ± 3.48	202 ± 19
Total	6.938	100.0	3.058	67.31	4.478	22.30	129.2

n.d., not detectable. All tabulated data were corrected for the mass discrimination effect, but do not include other corrections. Uncertainties in the measured ratios represent those of the mass spectrometric analyses as 1 σ. Correction factors used to estimate <sup>40</sup>Ar\*/<sup>39</sup>Ar\* are as follows:

$$\begin{aligned} (^{40}\text{Ar}/^{39}\text{Ar})_{\text{K}} &= (1.65 \pm 0.03) \times 10^{-1}, \\ (^{39}\text{Ar}/^{37}\text{Ar})_{\text{Ca}} &= (1.45 \pm 0.01) \times 10^{-3}, \\ (^{36}\text{Ar}/^{37}\text{Ar})_{\text{Ca}} &= (1.15 \pm 0.02) \times 10^{-4}. \end{aligned}$$

<sup>40</sup>Ar\*/<sup>39</sup>Ar\* corresponds to a ratio of radiogenic <sup>40</sup>Ar(<sup>40</sup>Ar\*) to K-derived <sup>39</sup>Ar(<sup>39</sup>Ar\*) by neutron irradiation. The amount of <sup>40</sup>Ar was calculated by assuming the sensitivity of the mass spectrometer and it includes about 10% uncertainty. <sup>40</sup>Ar–<sup>39</sup>Ar age was calculated based on the standard sample JG-1 biotite (K–Ar age: 91.4 ± 0.5 Ma, Iwata 1998).

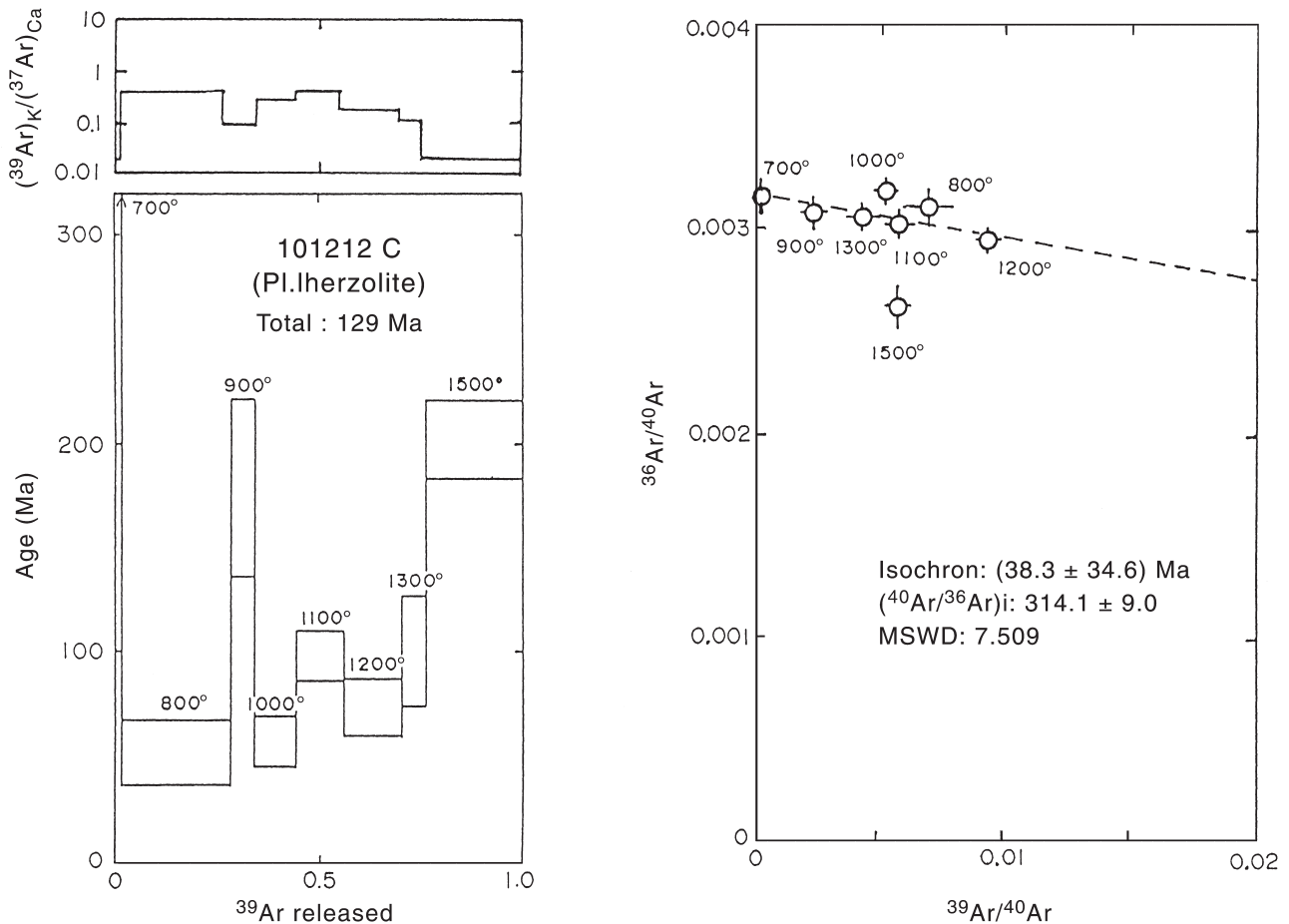


**Fig. 3**  $^{40}\text{Ar}$ - $^{39}\text{Ar}$  age spectrum (left) and inverse-isochron plot (right) for the phlogopite sample. Degassing temperatures are indicated in degrees Celsius. The heights of bars indicate the degree of uncertainties at one sigma level in the age spectrum. In the inverse-isochron plot, the isochron indicates an isochron age based on the data for all temperature fractions.  $(^{40}\text{Ar}/^{36}\text{Ar})_i$  indicates an initial  $^{40}\text{Ar}/^{36}\text{Ar}$ . Error bars for each plot in the isochron plot correspond to the one sigma uncertainty. In the above left, the  $(^{39}\text{Ar})_{\text{K}}/(^{37}\text{Ar})_{\text{Ca}}$  is plotted against the  $^{39}\text{Ar}$  release, where  $(^{39}\text{Ar})_{\text{K}}$  designates K-derived  $^{39}\text{Ar}$  and  $(^{37}\text{Ar})_{\text{Ca}}$  Ca-derived  $^{37}\text{Ar}$  induced by neutron irradiation.

ture fractions 1100–1300°C, which cover about 68% of the  $^{39}\text{Ar}$  release. However, the age spectrum has higher  $^{40}\text{Ar}$ - $^{39}\text{Ar}$  ages in the lower and higher temperature fractions, a signature of excess  $^{40}\text{Ar}$  as a saddle-shaped pattern (Kaneoka 1974; Lanphere & Dalrymple 1976). In contrast, on an inverse-isochron plot in Fig. 3, each temperature fraction is not so much deviated from an isochron of  $20.2 \pm 1.8$  Ma, which goes through an initial  $^{40}\text{Ar}/^{36}\text{Ar}$  of  $313.0 \pm 9.1$ . The initial  $^{40}\text{Ar}/^{36}\text{Ar}$  is slightly higher than the present atmospheric  $^{40}\text{Ar}/^{36}\text{Ar}$  of 295.5, but may not be so different as when assigned uncertainty has to be taken into account. Hence, the main trapped Ar component can be regarded to be of almost atmospheric origin. This point will be discussed in more detail later.

In contrast, the host rock shows a strongly disturbed age spectrum with a total  $^{40}\text{Ar}$ - $^{39}\text{Ar}$  age of 129 Ma, which is much higher than that of phlogopite (22.6 Ma). This clearly indicates that the host rock reflects mostly the trapped component of Ar with abundant excess  $^{40}\text{Ar}$ .

Furthermore, although microprobe analyses of phlogopites apparently show no presence of Ca, the neutron-irradiated phlogopite indicates the existence of about 1% Ca. This might have been caused by incorporation of impurities such as kaersutite in the phlogopite separate. However, these impurities do not affect the obtained  $^{40}\text{Ar}$ - $^{39}\text{Ar}$  plateau age defined by the fractions of 1100–1300°C. Based on the  $(^{39}\text{Ar})_{\text{K}}/(^{37}\text{Ar})_{\text{Ca}}$  versus  $^{39}\text{Ar}$  release diagram in Fig. 3, kaersutite is inferred to have degassed mostly in the 1500°C fraction.



**Fig. 4**  $^{40}\text{Ar}$ - $^{39}\text{Ar}$  age spectrum (left) and the inverse-isochron plot (right) for the host rock (plagioclase Iherzolite, 101212C). Degassing temperatures are indicated in degrees Celsius. The heights of bars indicate the degree of uncertainties at one sigma level in the age spectrum. In the inverse-isochron plot, the isochron indicates an isochron age based on the data for all temperature fractions.  $(^{40}\text{Ar}/^{36}\text{Ar})_i$  indicates an initial  $^{40}\text{Ar}/^{36}\text{Ar}$ . Error bars for each plot in the isochron plot correspond to the 1 sigma uncertainty. In the above left, the  $(^{39}\text{Ar})_K / (^{37}\text{Ar})_{Ca}$  is plotted against the  $^{39}\text{Ar}$  release, where  $(^{39}\text{Ar})_K$  designates K-derived  $^{39}\text{Ar}$  and  $(^{37}\text{Ar})_{Ca}$  Ca-derived  $^{37}\text{Ar}$ . The isochron age has no geological meaning as shown by a large uncertainty in the age with the large value of mean square of weighted deviations.

## DISCUSSION

### AGE OF THE PHLOGOPITE FORMATION

As shown in the previous section, stepheating of phlogopite yields an  $^{40}\text{Ar}$ - $^{39}\text{Ar}$  plateau age of  $20.6 \pm 0.5$  Ma with about 68% degassing of the integrated  $^{39}\text{Ar}$  for the three temperature fractions (1100–1300°C). As three temperature fractions are concordant at one sigma level, we can interpret the plateau age as approximating the formation age of the phlogopite. In this case, we propose that the span between the time of crystallization of phlogopite and the time when it reached the closure temperature for the K-Ar system was relatively short compared to the succeeding geological time to the event. If such a situation did not hold, we would not have a plateau pattern in the age spectra

as a result of successive Ar loss. To calculate the age, we have assumed the atmospheric  $^{40}\text{Ar}/^{36}\text{Ar}$  as the initial ratio. As the phlogopite shows the occurrence of some amount of excess  $^{40}\text{Ar}$  in the lower and higher temperature fractions, the age spectrum seems to show a slight saddle-shaped pattern. In such a case, the intermediate age might represent the maximum age of the formation, because we cannot exclude the possibility of occurrence of excess  $^{40}\text{Ar}$  in the intermediate temperature fractions (e.g. Kaneoka 1974). However, the initial  $^{40}\text{Ar}/^{36}\text{Ar}$  of the phlogopite is regarded to be close to the atmospheric value as revealed in the inverse-isochron plot (Fig. 3). Furthermore, the isochron age is almost the same as the plateau age within their analytical uncertainties. Hence, even if the excess  $^{40}\text{Ar}$  affected the plateau age, the effect would be small with no significant change in

the obtained plateau value. As the phlogopite of the Horoman Peridotite Complex has been regarded to have been formed by metasomatism (Arai & Takahashi 1989; Takahashi *et al.* 1989), the age would correspond to the time of metasomatism in this area.

Yoshikawa *et al.* (1993) interpreted a Rb–Sr age of  $23.0 \pm 1.2$  Ma ( $2\sigma$ ), based on an isochron plot composed of a phlogopite, four clinopyroxenes and a whole rock as the formation age of the phlogopite. Although both  $^{40}\text{Ar}$ – $^{39}\text{Ar}$  and Rb–Sr ages show similar values, the former indicates a slightly younger value than the latter, even if we take their analytical uncertainties into account. One of the reasons may be because of the difference in the closure temperatures of phlogopite for K–Ar and Rb–Sr systems. Although the closure temperature for phlogopite for the K–Ar system is reported to be in the order of 400–470°C (Geyh & Schleicher 1990), no data have been reported for that of the Rb–Sr system. However, for muscovite, the closure temperatures are reported to be  $350 \pm 50^\circ\text{C}$  and  $500 \pm 50^\circ\text{C}$  for the K–Ar and Rb–Sr systems, respectively (Geyh & Schleicher 1990). If the same trend holds for phlogopite, an  $^{40}\text{Ar}$ – $^{39}\text{Ar}$  age for phlogopite would correspond to a later cooling stage than a Rb–Sr age. In this case, it took about 1 million years to have cooled down from the closure temperature of phlogopite for the Rb–Sr system to about 400–470°C during the metasomatism which formed the phlogopite.

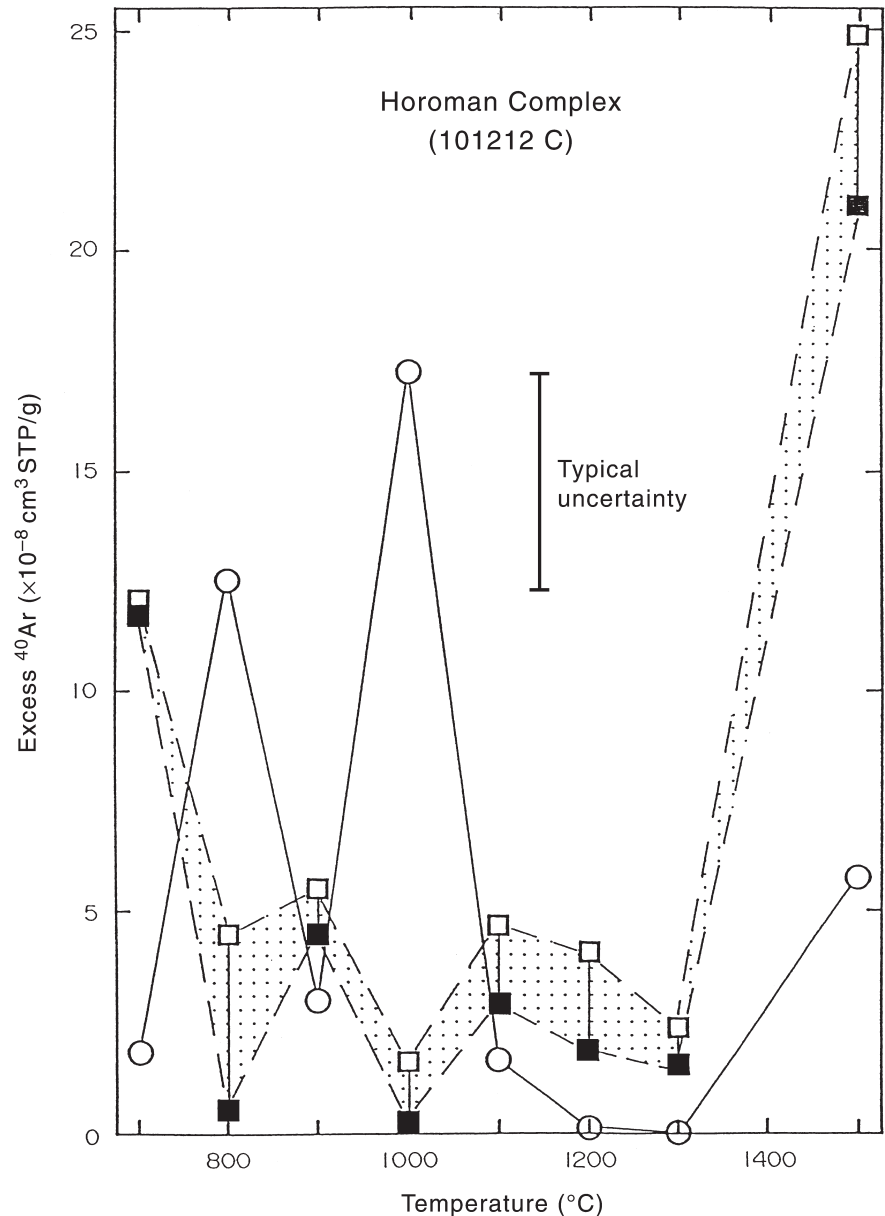
Yoshikawa *et al.* (1993) assumed that clinopyroxenes had achieved isotopic equilibrium among them during the metasomatic event. The assumption was made because the clinopyroxenes were taken within 60 mm from the phlogopite vein and they showed similar but definitely higher  $^{87}\text{Sr}/^{86}\text{Sr}$  than clinopyroxenes in the phlogopite-free spinel lherzolite. Even if the clinopyroxenes had been affected by the metasomatism, but were not completely homogenized isotopically, the calculated Rb–Sr age would have been slightly older than the age of the metasomatic event. Furthermore, the closure temperature of clinopyroxene might be different from that of phlogopite in the Rb–Sr system. If the cooling process during the metasomatism occurred rather slowly, it would also have caused some ambiguities in the Rb–Sr systematics. At present, we cannot give the details of the metasomatic process. Hence, the age of the metasomatic event might be better approximated by the  $^{40}\text{Ar}$ – $^{39}\text{Ar}$  phlogopite plateau age than by the Rb–Sr isochron age, although apparent difference is small.

As for the formation age of the Complex itself,  $^{40}\text{Ar}$ – $^{39}\text{Ar}$  analysis did not result in plateau age as a result of the occurrence of excess  $^{40}\text{Ar}$  in the host rock. For a sample with excess  $^{40}\text{Ar}$ , the youngest age in the intermediate temperature fractions might be least affected by the incorporation of excess  $^{40}\text{Ar}$  and represent the upper limit for the formation age of the sample (Kaneoka 1974). If this is the case for the present sample, the lowest apparent age of  $54 \pm 17$  Ma for the plagioclase lherzolite may give a measure of the upper limit for the formation age of the Complex.

#### OCCURRENCE OF EXCESS $^{40}\text{Ar}$ IN THE PHLOGOPITE AND IMPLICATION FOR ITS FORMATION AT A DEPTH

In Fig. 5, the degassing temperature for the phlogopite is plotted against the amount of excess  $^{40}\text{Ar}$  for each step, which was calculated based on the inferred age of the metasomatism, following the manner by Kaneoka (1974). In the case of the host rock plagioclase lherzolite, the formation age of 50 Ma was assumed tentatively to show the release pattern of excess  $^{40}\text{Ar}$  and to compare it with that of phlogopite. The case for a formation age of 20.6 Ma was also calculated for comparison, but no large difference is observed, although slightly higher values of excess  $^{40}\text{Ar}$  were obtained in the latter case.

As shown in Fig. 5, the phlogopite phase definitely contains excess  $^{40}\text{Ar}$  in the lowest and in the highest temperature fraction. This indicates that the phlogopite was formed at a depth, where the metasomatic fluid still contained some amounts of excess  $^{40}\text{Ar}$ . Apparently high concentrations of excess  $^{40}\text{Ar}$  in the 800°C and 1000°C fractions derived from the phlogopite might have been caused because of the enrichment of excess  $^{40}\text{Ar}$  together with the other incompatible elements such as K through metasomatic fluids. Such excess  $^{40}\text{Ar}$  has also been observed in phlogopites from kimberlite inclusions (Kaneoka & Aoki 1978; Allsopp & Roddick 1985; Phillips & Onstott 1988; Phillips 1991). As excess  $^{40}\text{Ar}$  is also observed in different temperature fractions of the phlogopite from the intermediate temperature fractions, the trapping sites of such  $^{40}\text{Ar}$  might be some inclusions in the phlogopite for the components observed in the lower temperature fractions, and probably in the lattice itself for the component observed in the highest temperature fraction, but not at the K-site. Excess  $^{40}\text{Ar}$  may be heterogeneously distributed in grains with core-rim gradients as revealed by laser probe single grain



**Fig. 5** Release patterns of excess  $^{40}\text{Ar}$  for phlogopite and its host, plagioclase Iherzolite. Excess  $^{40}\text{Ar}$  has been calculated, assuming formation ages of 20.6 Ma and 50 Ma for the host rock and 20.6 Ma for phlogopite. The bar of typical uncertainty is shown as a measure for the uncertainty of concentration in each temperature fraction, but analytical uncertainties in relative concentrations among different temperature fractions are less compared to it. (○), phlogopite ( $t=20.6$  Ma); (□), Pl. Iherzolite ( $t=20.6$  Ma); (■), Pl. Iherzolite ( $t=50$  Ma). Dots indicate the possible range for the estimated amount of excess  $^{40}\text{Ar}$  as a result of the difference in the assumed formation of each phase.

studies of mantle phlogopite (Phillips & Onstot 1988). In contrast, the host rock of the phlogopite contains excess  $^{40}\text{Ar}$  in all temperature fractions, especially at the highest temperature (1500°C). Such characteristics in the release pattern of excess  $^{40}\text{Ar}$  agree with a deep origin of the plagioclase Iherzolite.

The phlogopite is proposed to have been formed at a shallower depth compared with the host rock. This is because it contains much less excess  $^{40}\text{Ar}$  than the host rock in the highest temperature fraction than what is expected from the amount of excess  $^{40}\text{Ar}$  observed in the lower temperature fractions of the phlogopite when it was formed at

a similar depth with the host rock. Thus, the occurrence and the amount of excess  $^{40}\text{Ar}$  in the phlogopite suggests its origin at a depth, where the metasomatic fluids contain excess  $^{40}\text{Ar}$ . However, the formation depth would be shallower than the host rock, though it is difficult to identify the definite depth. Saheki *et al.* (1995) obtained K-Ar biotite ages of 17.2–18.5 Ma and 26.9 Ma for hornblende from metamorphic and igneous rocks in the southern part of the Hidaka Belt. Although K-Ar ages do not permit detailed interpretations as  $^{40}\text{Ar}$ - $^{39}\text{Ar}$  ages due to loss of radiogenic  $^{40}\text{Ar}$  and/or occurrence of excess  $^{40}\text{Ar}$ , the ages are comparable with the age obtained on phlogopite in the present

study. This may be used as an additional indication that the metasomatism occurred in the crustal environment. Arita *et al.* (1993) also provided K–Ar data that might be indicative of the exhumation of the Hidaka Oeridotite Complex.

#### THE TRAPPED $^{40}\text{Ar}$ IN THE PHLOGOPITE AND ITS CONSTRAINT ON THE ENVIRONMENTS OF THE METASOMATISM

Although the occurrence of excess  $^{40}\text{Ar}$  in the phlogopite suggests that it was formed at a depth, it is inferred to have been formed at a shallower depth than that of its host rock. As shown in the inverse–isochron plot for the phlogopite (Fig. 3), most plotted data for each temperature fraction lie almost on an isochron of  $20.2 \pm 1.8$  Ma, which goes through the  $^{40}\text{Ar}/^{36}\text{Ar}$  with the value of  $313.0 \pm 9.1$  at the ordinate. This implies that the metasomatic fluid might have contained trapped Ar with a  $^{40}\text{Ar}/^{36}\text{Ar}$  value close to the atmospheric value. Although no blank correction was applied for each temperature fraction, the blank level of about  $1 \times 10^{-8}$  cm<sup>3</sup> standard pressure and temperature (STP)  $^{40}\text{Ar}$  with the atmospheric  $^{40}\text{Ar}/^{36}\text{Ar}$  for each temperature fraction have precluded a possibility of any significant effect of the blank to the observed values.

If the trapped Ar in the phlogopite is almost atmospheric, it gives a constraint on the origin of the metasomatic fluid. If the metasomatic fluid was derived from the mantle, its  $^{40}\text{Ar}/^{36}\text{Ar}$  is expected to show a definitely higher  $^{40}\text{Ar}/^{36}\text{Ar}$  than the atmospheric value of 296. The  $^{40}\text{Ar}/^{36}\text{Ar}$  value of trapped Ar is quite variable from place to place, ranging from around 350–30 000, depending on their environmental conditions (e.g. Ozima & Podosek 1983).

In the case of the Japanese Islands, the magmatic gas is expected to contain trapped Ar with an  $^{40}\text{Ar}/^{36}\text{Ar}$  of more than 350, based on the analytical results of some plagioclase phenocrysts (e.g. Takaoka 1989; Hanyu & Kaneoka 1997). Hence, if the metasomatic fluid is derived from such a mantle, it would also indicate a value of at least 350. As the atmosphere and/or the water that equilibrated with the atmosphere contain relatively large amounts of Ar (about 0.93% of Ar in the atmosphere and  $(3\text{--}5) \times 10^{-4}$  cm<sup>3</sup> STP/g of Ar in ground- and seawater) (Ozima & Podosek 1983), it easily affects the original trapped Ar. In the relatively deep interior of the Earth, direct incorporation of the atmosphere is unlikely. Hence, incorporation of the atmospheric Ar through water

is the most likely way to change the original mantle signature of Ar to an almost atmospheric value of the metasomatic fluid. The present results require at least such components in the metasomatic fluid from which the phlogopite was formed.

Then, what kinds of environments are proposed from the present result? Arai and Takahashi (1989) have envisaged that phlogopite and amphibolite were probably formed from magmatic volatiles, which were successively released from evolving alkali basaltic magmas. In such a case, the magma should have already been contaminated by some materials which contain relatively large amounts of water and at least original magmatic signatures of the mantle origin might have been almost erased. In the case of Yoshikawa *et al.* (1993), based on a probably increased  $^{87}\text{Sr}/^{86}\text{Sr}$  for the metasomatic fluid, they assumed that the fluids were derived from the dehydration of subducted oceanic sediments and altered MORB. At present, we have no definite idea about the  $^{40}\text{Ar}/^{36}\text{Ar}$  in the fluids dehydrated from oceanic sediments and altered MORB. As described in the increased  $^{87}\text{Sr}/^{86}\text{Sr}$  for the fluids, it is almost certain that they contain radiogenic  $^{40}\text{Ar}$ , which might increase the  $^{40}\text{Ar}/^{36}\text{Ar}$  to some extent. However, the value is completely dependent on the rate between the incorporated atmospheric Ar and the production of radiogenic  $^{40}\text{Ar}$ . For example, if the sediment retains 10% of the seawater, it contains about  $(3\text{--}5) \times 10^5$  cm<sup>3</sup> STP  $^{40}\text{Ar}$ /g. However, a sample with a K–content of 1% produces about  $5 \times 10^{-6}$  cm<sup>3</sup> STP of radiogenic  $^{40}\text{Ar}$  per 100 Ma. If such a sediment is decomposed to form a part of metasomatic fluids, the fluids would have an  $^{40}\text{Ar}/^{36}\text{Ar}$  of around 330, a definitely higher value than the atmospheric one. Hence, even if oceanic sediment and/or altered MORB were related to the formation of the metasomatic magma, it would be insufficient to explain our findings of trapped component with an  $^{40}\text{Ar}/^{36}\text{Ar}$  close to the atmospheric value. Furthermore, if such metasomatic fluids are homogenized with the surrounding mantle material, its  $^{40}\text{Ar}/^{36}\text{Ar}$  would become higher than 350, as mentioned before.

An alternative explanation is that the phlogopite was formed at a relatively shallow depth where the ground- or seawater might have affected the metasomatic fluids, but not so shallow that the inherited Ar, which was derived from the source region of the metasomatic fluids, could partially degas. In this case, we cannot disregard the possibility that the phlogopite might have been formed

in the crustal circumstances, because it is difficult to imagine that the groundwater might have penetrated into the mantle directly. Enrichment of Sr isotopic ratio as observed by Yoshikawa *et al.* (1993) might have occurred due to the incorporation of components either derived from the continental crust or from subducted oceanic sediments and/or altered MORB. Thus, the metasomatic fluids, which formed the phlogopite, might have included multiple components, reflecting a rather complicated formation process. The present result has indicated the possibility that the metasomatic fluids might have been affected to some extent by the ground- or seawater in the crustal environments. It has been reported that the pressure-temperature (P-T) evolution based on mineral equilibria of parts of the peridotite massif points to two stages (one at 900–950°C, 2 GPa and a second of 950–750°C at 0.5 GPa (Takazawa *et al.* 1996). The P-T evolution points to decompression and exhumation of the peridotites and P-conditions of the second stage (0.5 GPa, i.e. 15–17 km) are clearly crustal. Such data are also compatible with the present result on the formation of phlogopite in the crustal environments.

## SUMMARY

- (1) The phlogopite separated from a phlogopite lherzolite from the Horoman Peridotite Complex shows an  $^{40}\text{Ar}$ - $^{39}\text{Ar}$  age of  $20.6 \pm 0.5$  Ma (1 sigma), which might approximate the time of metasomatic event which formed the phlogopite.
- (2) The phlogopite includes some amount of excess  $^{40}\text{Ar}$ , suggesting its origin at a depth, where excess  $^{40}\text{Ar}$  has been kept in the metasomatic fluid.
- (3) The inverse-isochron plot indicates that the trapped Ar in the phlogopite has an almost atmospheric value, though the amount of excess  $^{40}\text{Ar}$  might have been increased due to the enrichment of Ar during the phlogopite formation together with some incompatible elements.
- (4) The phlogopite was formed at a depth, where excess  $^{40}\text{Ar}$  was kept to some extent, but not so deep, as the ground- or seawater had not affected the metasomatic fluids. Hence, the metasomatic fluids which formed the phlogopite might have reflected the crustal environments to some extent.
- (5) The host rock of the phlogopite, plagioclase lherzolite, might surely reflect the environments of deep origin, indicating the occurrence of excess  $^{40}\text{Ar}$  in all releasing temperature fractions.

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**Appendix** Chemical compositions of samples used for  $^{40}\text{Ar}$ – $^{39}\text{Ar}$  analyses

Sample No.	101212C (Pl lherzolite)	101212C (Phlogopite)
SiO <sub>2</sub> (wt%)	44.94	39.19
TiO <sub>2</sub>	0.15	5.80
Al <sub>2</sub> O <sub>3</sub>	3.05	16.12
Cr <sub>2</sub> O <sub>3</sub>	0.35	1.85
Fe <sub>2</sub> O <sub>3</sub>	8.59	–
FeO	–	2.98
MnO	0.15	0.07
MgO	38.81	21.43
NiO	0.31	0.21
CaO	2.97	0.00
Na <sub>2</sub> O	0.25	1.40
K <sub>2</sub> O	0.04	7.67
Total	99.61	96.73

Whole rock was analyzed by the X-ray fluorescence method and phlogopite was analyzed with an electron microprobe analyzer at the Kanazawa University. Fe was calculated as total iron.