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^{238}U – ^{230}Th – ^{226}Ra Radioactive Disequilibria in the Lava Flows of the 1986 Eruption from Izu-Oshima Volcano, Japan

Yuichi KURIHARA, Masaomi TAKAHASHI and Jun SATO

Department of Applied Chemistry, School of Science and Technology, Meiji University
1-1-1 Higashi-mita, Tama-ku, Kawasaki-shi, Kanagawa Pref. 214-8571, Japan

Received August 18, 2008

Time scale of magmatic processes in the 1986 eruptive activity of Izu-Oshima volcano, Japan, was estimated by the ^{238}U – ^{230}Th – ^{226}Ra disequilibria observed in the 1986 lava flows. The activity ratios of $^{226}\text{Ra}/^{230}\text{Th}$ in the products were larger than unity, being enriched in ^{226}Ra relative to ^{230}Th . The $^{226}\text{Ra}/^{230}\text{Th}$ activity ratio versus $^{238}\text{U}/^{230}\text{Th}$ activity ratio and Ba/Th concentration ratio diagrams for the 1986 products showed positive correlations, suggesting that the ^{238}U – ^{230}Th – ^{226}Ra disequilibria occurred during the magma genesis of Izu-Oshima volcano by the additions of U- and Ra- (Ba-) rich fluids derived from the subducting slab by dehydration to the mantle wedge. The ^{230}Th – ^{226}Ra disequilibria in the 1986 products suggested that the time scale from the magma genesis to the eruption, including the melting of the mantle wedge, magma storage and magmatic differentiation from basalt to basaltic andesite, was less than 8 000 years.

Key Words : uranium series, radioactive disequilibrium, 1986 eruption of Izu-Oshima volcano, $^{230}\text{Th}/^{238}\text{U}$ activity ratio, $^{226}\text{Ra}/^{230}\text{Th}$ activity ratio

1. Introduction

Investigations of radioactive disequilibria among U-series nuclides in volcanic rocks have been applied to the studies of magmatic processes^{1)–3)}. During magmatic processes, from magma generation in the mantle wedge, magma ascent and storage in the crust, crystallizing differentiation in magma chamber, magma degassing, and to the eruption, the differences in chemical properties between parent and daughter nuclides will allow them to be fractionated, generating radioactive disequilibrium. As the time since fractionation can be assessed using the half-lives of the daughter nuclides (e.g., $T_{1/2} = 7.5 \times 10^4$ y for ^{230}Th , and $T_{1/2} = 1.6 \times 10^3$ y for ^{226}Ra), investigations of radioactive disequilibria among U-series nuclides in volcanic rocks can potentially provide informa-

tions on the time scale of magmatic processes. The ^{238}U – ^{230}Th disequilibria can thus be used to estimate the time scale of an event of the U–Th fractionation within the period of 3.5×10^5 years (i.e., 5 half-lives yield 97% of the equilibrium).

The majority of the arc volcanic rocks is characterized by ^{238}U -excess over ^{230}Th ^{4)–16)}. Magma genesis at the subducting zone is generally inferred to be induced by the melting of the mantle wedge by the addition of the fluid derived from the subducting slab¹⁷⁾. Because U is highly mobile element in aqueous fluids relative to Th^{18), 19)}, the excess of ^{238}U over ^{230}Th in the arc volcanic rocks has generally been ascribed to the addition of U-rich fluids derived from the subducting slab by dehydration. As both U and Th are highly incompatible elements for most minerals and have similar parti-

tion coefficients between most minerals phase and melt phase^{20)–22)}, U and Th are unlikely to be significantly fractionated from each other by the melting of the mantle wedge and the crystallizing differentiation developing during the magmatic processes. On the assumption that the inclined data point array in the ($^{230}\text{Th}/^{232}\text{Th}$) – ($^{238}\text{U}/^{232}\text{Th}$) activity ratio diagram for the most arcs represented the whole rock isochron, the ages of the magma genesis to the eruption were estimated to be in the order of 10^4 – 10^5 years^{8)–12), 15), 16)}.

The ^{230}Th – ^{226}Ra disequilibria can be used to estimate the time scale of an event of the Th–Ra fractionation within the period of 8 000 years. The investigations of the disequilibria between ^{230}Th and ^{226}Ra in fresh volcanic rocks from many arc volcanoes have shown that the majority of arc volcanic rocks are characterized by ^{226}Ra excess over ^{230}Th ^{6), 7), 13)–15), 23)–27)}. The ^{230}Th – ^{226}Ra activity ratio observed in the most arc volcanic rocks generally shows positive correlations with $^{238}\text{U}/^{230}\text{Th}$ activity ratio and Ba/Th concentration ratio (the latter is usually regarded as a good indicator of the fluid addition), which suggests that the ^{226}Ra excess over ^{230}Th may also be attributed to the addition of U- and Ra-rich fluids derived from the subducting slab^{13)–15), 23)–27)}. The arc volcanic rocks possess ^{226}Ra excess over ^{230}Th implying the time scale from the magma genesis to the eruption was less than 8 000 years.

The ^{238}U – ^{230}Th – ^{226}Ra disequilibria in the products of the 1707 eruption from Fuji volcano, Japan, have previously been observed^{16), 27)}. The 1707 eruption of Fuji volcano issued silicic magmas including andesite and dacite as well as basaltic magma^{28)–31)}. The activity ratios of $^{238}\text{U}/^{230}\text{Th}$ and $^{226}\text{Ra}/^{230}\text{Th}$ in the basaltic and andesitic products were larger than unity, being

enriched in ^{238}U and ^{226}Ra relative to ^{230}Th . The ^{230}Th – ^{226}Ra disequilibria in the 1707 eruption products suggested that the time scale from the magma genesis to the eruption, including the melting of the mantle wedge, magma storage and crystallizing differentiation from basalt to andesite, was less than 8 000 years.

Izu-Oshima is a volcanic island located 110 km SSW of Tokyo and belonging to Izu arc. The volcanic island is composed mainly of a cone called Izu-Oshima volcano. The main cone is a stratovolcano consisting of basaltic lava flows, scoriae and ash falls^{32), 33)}. At the summit of the main cone, there is a caldera opening to the northeast formed about 1 500 – 1 300 years ago^{32), 33)}. An active central cone with a pit crater, Mt. Mihara (Lat. $34^\circ 44'$ N, Long. $139^\circ 24'$ E), is situated at the southwestern part of the summit caldera³⁴⁾. In the 20th century, several eruptions occurred at the pit crater³³⁾.

The latest eruption of Izu-Oshima volcano is the 1986 AD eruption^{35), 36)}. Similar to the 1707 AD eruption from Fuji volcano^{28)–31)}, the 1986 eruption issued the products having a variety of major chemical compositions (e.g., SiO_2 : 52 – 57%)^{37), 38)}. The variety of the chemical and mineralogical compositions suggested the existence of the separated magma chambers existed just prior to the 1986 eruption^{37), 38)}. Although the investigations of the chemical and mineralogical compositions for the 1986 products have been made, the time scale of the magmatic processes has not yet been suggested.

In succession to the previous report for ^{238}U – ^{230}Th disequilibria in the lava samples of the 1986 eruption from Izu-Oshima volcano¹⁶⁾, this report deals with the observations of the ^{230}Th – ^{226}Ra disequilibria in the 1986 lava samples, together with the major element composi-

tions and Ba concentration, in order to estimate the time scale of magmatic processes in the 1986 eruptive activities of Izu-Oshima volcano.

2. The 1986 eruption and sample description

The sequence of the 1986 eruption was described in detail by Soya et al.³⁵⁾ and Endo et al.³⁶⁾, and is briefly summarized below.

The 1986 eruption of Izu-Oshima volcano started with a fire fountain activity on November 15, 1986 at a pit crater of Mt. Mihara (A-vent). Lava filled up successively the crater and formed a lava lake, then flowed out to the caldera floor to form a lava flow (LA) on November 19. On November 21, fissure eruptions occurred in the summit caldera floor (B-vent), forming lava flows (LB I and LB III). One and a half hour later, another fissure eruptions occurred outside the northwestern caldera rim, forming a narrow lava flow (LC). The total eruptive mass was estimated to have been approximately $0.6 - 0.8 \times 10^8$ tons^{39), 40)}. The sampling localities of the 1986 lava samples are shown in Fig. 1.

The chemical and mineralogical compositions in the 1986 eruption products were well investigated by Nakano and Yamamoto³⁷⁾ and Fujii et al.³⁸⁾. These studies indicated that the magma chamber for B- and C-vent eruptions were different from the magma chamber for A-vent eruption existed just prior to the 1986 eruption.

The 1986 eruption products from A-vent have relatively uniform major element compositions, while the major elements compositions of the products from B- and C-vent are varying. The products from A-vent are plagioclase-phyric basalt; the reported SiO_2 contents are approximately 52–53%. The products from A-vent are similar to the post-caldera eruption

Fig. 1 Lava distributions of the 1986 eruption of Izu-Oshima volcano. (after Kawanabe³⁴⁾) The sampling localities are indicated by open triangles.

products from the central crater of Izu-Oshima volcano. The products from B- and C-vent are aphyric basaltic andesite and aphyric andesite; the reported SiO_2 contents range from 54 to 57%. The products from B- and C-vent have different characteristics from the post-caldera eruption products of Izu-Oshima volcano except for the 1338 AD eruption products⁴¹⁾.

Based on the major and trace element compositions along with mineralogical compositions in the 1986 eruption products, Aramaki and Fujii⁴²⁾ proposed that the magma chamber for A-vent eruption and those for B- and C-vent eruptions had existed separately just prior to the 1986 eruption. The magma of A-vent eruption was estimated to be derived directly from a deep and large magma chamber. Although the magma of B- and C-vent eruptions were de-

rived from the same source as the magma of A-vent eruption, they might undergo crystallizing differentiation in a shallow isolated magma chamber.

Nakano et al.⁴³⁾ investigated the major element, Rb, Sr, mineralogical and Sr isotopic compositions of the 1986 eruption products. They also proposed that the magma chamber for B- and C-vent eruptions had been independent of the magma chamber for A-vent eruption existed just prior to the 1986 eruption. Magmas from A-vent, and B- and C-vent were derived from the similar source with regard to the identical Sr isotopic compositions. However, based on the difference of K/Rb ratios between the products from A-vent and from B- and C-vent, the source of the magma for B- and C-vent eruptions were primarily different from the magma for A-vent eruption.

3. Experimental

3.1 Determination of major element compositions and Ba concentration in the lava samples of the 1986 eruption from Izu-Oshima volcano

Major element compositions and Ba concentration in the 1986 lava samples were determined by X-ray fluorescence spectrometry (XRF) using fused glass beads made by mixing sample with anhydrous lithium tetraborate as a flux. The dilution ratio 1 : 10 sample-to-flux was used for determination of major element compositions⁴⁴⁾. The dilution ratio 1 : 2 sample-to-flux was used for determination of Ba⁴⁵⁾. The experimental procedures have been described in detail by Nakayama and Nakamura⁴⁴⁾, and Nakayama et al.⁴⁵⁾.

3.2 Determination of U and Th isotopes in the lava samples

Determination of U and Th isotopes in the 1986 lava sample was made by an isotope dilution analysis coupled with α -ray spectrometry. Uranium-232 ($T_{1/2} = 69$ y) was spiked for the U isotopes and Th reagent having different $^{230}\text{Th}/^{232}\text{Th}$ activity ratio from those in the samples was spiked for the Th isotopes. Detailed chemical separation and purification procedures of U and Th isotopes from the lava samples have previously been described in Kurihara et al.¹⁶⁾. The purified U and Th fractions were electrodeposited onto stainless steel planchets of 1 inch ϕ by Lee's method⁴⁶⁾ for α -ray counting, respectively.

3.3 Determination of ^{226}Ra in the lava samples

Determination of ^{226}Ra in the 1986 lava samples was made by non-destructive γ -ray spectrometry²⁷⁾. About 100 g of sample was packed into a canister of tin-can (76 mm ϕ , 24 mm high). The canister was sealed with epoxy resin (Araldite[®]) completely. The sample canister for the γ -ray measurement thus prepared was stored for 1 month in order that ^{214}Pb reached the equilibrium with ^{226}Ra within the canister.

3.4 Radioactivity measurement

Activity ratios of U and Th isotopes were obtained by α -ray spectrometry using a 900 mm² PIPS detector (CANBERRA, A900-25AB) coupled with an MCA system (Laboratory Equipment; ADC 2201A).

Radioactivities of U and Th isotopes were determined by the following α -rays: 4.151 MeV and 4.198 MeV for ^{238}U , 4.722 MeV and 4.775 MeV for ^{234}U , 5.263 MeV and 5.320 MeV for ^{232}U ,

Table 1 Major elements and Ba data for the lava flows of the 1986 eruption from Izu-Oshima volcano

Identification	Sample							
	OSM05061702		OSM03052801		OSM98062104		OSM98062106	
Unit	LA		LB I		LB III		LC	
<hr/>								
[%]								
SiO ₂	51.4	(0.4)	54.0	(0.7)	55.1	(0.3)	54.5	(0.2)
TiO ₂	1.18	(0.2)	1.21	(0.4)	1.18	(0.2)	1.21	(0.6)
Al ₂ O ₃	14.3	(0.5)	13.7	(0.3)	14.0	(0.3)	14.1	(0.1)
Fe ₂ O ₃ *	14.1	(0.3)	14.1	(0.2)	13.7	(0.4)	14.1	(0.1)
MnO	0.19	(0.6)	0.20	(0.3)	0.19	(0.5)	0.20	(0.3)
MgO	4.72	(0.4)	3.96	(0.4)	3.54	(0.3)	3.89	(0.3)
CaO	9.81	(0.2)	8.56	(0.2)	8.19	(0.3)	8.60	(0.2)
Na ₂ O	1.84	(0.7)	2.01	(0.7)	2.27	(0.3)	2.02	(0.8)
K ₂ O	0.40	(0.4)	0.45	(0.6)	0.54	(0.5)	0.45	(0.7)
P ₂ O ₅	0.09	(0.8)	0.10	(0.6)	0.12	(0.7)	0.10	(0.2)
Total	98.0	(0.2)	98.3	(0.4)	98.8	(0.3)	99.2	(0.1)
<hr/>								
[μg·g ⁻¹]								
Ba	239	(2.6)	277	(5.5)	326	(3.2)	261	(2.8)

* Total iron as Fe₂O₃. (): Relative standard deviation / %, n = 5

3.954 MeV and 4.013 MeV for ^{232}Th , and 4.621 MeV and 4.688 MeV for ^{230}Th , respectively.

Gamma-ray intensity was measured with an HPGe semiconductor detector (Princeton Gamma-Tec, IGC-10200 NPR) coupled with an MCA system (Aptec Engineering, 5008).

Radioactivity of ^{226}Ra was determined by the 351.9 keV γ -ray from ^{214}Pb in equilibrium with ^{226}Ra .

The determination procedures of U, Th isotopes and ^{226}Ra have previously been applied to JB-1a and JB-1b (replacement of JB-1), the geochemical reference materials of basalt, issued by Geological Survey of Japan (The National Institute of Advanced Industrial Science and Technology). The reference material is in the radioactive equilibrium among U-series nuclides, because the eruption age of JB-1 is 7.59 Ma⁴⁷⁾. The activity ratios among ^{238}U – ^{234}U – ^{230}Th – ^{226}Ra in JB-1a and JB-1b were unity²⁷⁾.

4. Results and Discussion

4.1 Major element compositions and Ba concentration in the lava flows of the 1986 eruption from Izu-Oshima volcano

Table 1 lists the major element compositions and Ba concentration in the 1986 lava samples. Figure 2 illustrates the K₂O content versus SiO₂ content diagram for the 1986 lava samples. Figure 2 contains the K₂O and SiO₂ contents for the products from the A-vent of Izu-Oshima volcano^{37), 38)}.

The major element compositions of the 1986 lava samples generally agree well with the reported values of the products of the 1986 eruption from Izu-Oshima volcano. There exists a clear compositional gap between the lava sample from A-vent and the lava samples from B- and C-vent. Figure 2 shows that the K₂O content increases with increasing the SiO₂ con-

Fig. 2 K_2O content versus SiO_2 content diagram for the lava flows of the 1986 eruption from Izu-Oshima volcano. Thinly and deeply shaded zones represent data sets for the 1986 eruption products from A-vent, and from B- and C-vent, respectively. (after Nakano and Yamamoto³⁷⁾, and Fujii et al.³⁸⁾)

tents. The variations of the major element compositions are mainly attributed to the crystallizing differentiation.

The concentration of Ba in the 1986 lava samples ranges from 239 to 326 (in $\mu g \cdot g^{-1}$). These values for the lava samples are generally consistent with the published data^{38), 48)} in Table 2 for the products of the historical eruptions from Izu-Oshima volcano. Similar to the case of K_2O content, the concentration of Ba in the lava sample from the A-vent is lower than those from the B- and C-vent (Table 1).

4.2 Uranium, thorium and ^{226}Ra concentrations in the lava flows

Table 3 lists the concentrations of U and Th (in $\mu g \cdot g^{-1}$), and ^{226}Ra (in $fg \cdot g^{-1}$) and the activity concentrations of ^{238}U , ^{230}Th and ^{226}Ra (in $mBq \cdot g^{-1}$), and the concentration ratios of U/Th and Ba/Th in the 1986 lava samples. The concentrations of U, Th and ^{226}Ra range from 0.17 to 0.22¹⁶⁾ (in $\mu g \cdot g^{-1}$), from 0.19 to 0.34¹⁶⁾

Table 2 U, Th and Ba (in $\mu g \cdot g^{-1}$), and ^{226}Ra (in $fg \cdot g^{-1}$) concentrations, and $^{230}Th/^{238}U$ and $^{226}Ra/^{230}Th$ activity ratios for the products of the historical eruptions from Izu-Oshima volcano appeared in literatures

Sample	Eruption year	Concentration			Activity ratio			Reference
		U	Th	Ba	^{226}Ra	$^{230}Th/^{238}U$	$^{226}Ra/^{230}Th$	
			[$\mu g \cdot g^{-1}$]		[$fg \cdot g^{-1}$]			
Historical eruption products	10th C-1986 AD	-	0.1-0.3	150-302	-	-	-	38)
112921	1778 AD	0.21	0.36	-	-	0.88	-	7)
JB-2*	1950-1951 AD	0.18	0.35	222	-	-	-	48)
		0.16	0.22	-	-	0.64	-	50)
		0.154	0.256	-	81	0.66	2.2	51)

* JB-2 is the GSI reference rock sample of the basaltic product of the 1950-1951 eruption from Izu-Oshima volcano.

Table 3 U, Th and ^{226}Ra concentrations, and ^{238}U , ^{230}Th and ^{226}Ra activity concentrations, and U/Th and Ba/Th concentration ratios for the lava flows of the 1986 eruption from Izu-Oshima volcano

Sample identification	Unit	Concentration			Activity concentration			Concentration ratio	
		U*	Th*	^{226}Ra	^{238}U *	^{230}Th *	^{226}Ra	U/Th*	Ba/Th
		[$\mu g \cdot g^{-1}$]	[$\mu g \cdot g^{-1}$]	[$fg \cdot g^{-1}$]		[$mBq \cdot g^{-1}$]			
OSM05061702	LA	0.17 ± 0.00	0.19 ± 0.01	89 ± 3	2.12 ± 0.05	1.13 ± 0.07	3.26 ± 0.12	0.91	1258
OSM03052801	LB I	0.19 ± 0.01	0.34 ± 0.02	92 ± 3	2.41 ± 0.07	1.69 ± 0.10	3.36 ± 0.12	0.55	815
OSM98062104	LB III	0.22 ± 0.01	0.30 ± 0.02	110 ± 4	2.71 ± 0.07	1.65 ± 0.11	4.01 ± 0.13	0.72	1087
OSM98062106	LC	0.20 ± 0.01	0.29 ± 0.02	99 ± 2	2.54 ± 0.07	1.57 ± 0.09	3.61 ± 0.09	0.69	900

* Uranium and Th isotope data are taken from Table 2 of Ref. 16.

Fig. 3 Ba/Th concentration ratio versus U/Th concentration ratio diagram for the lava flows of the 1986 eruption from Izu-Oshima volcano. (U and Th data from Kurihara et al.^[6])

(in $\mu\text{g}\cdot\text{g}^{-1}$), and from 89 to 110 (in $\text{fg}\cdot\text{g}^{-1}$), respectively. Similar to the cases of K_2O content and Ba concentration, the concentrations of U, Th, ^{226}Ra in the lava sample from the A-vent are lower than those from the B- and C-vent.

Figure 3 is the Ba/Th concentration ratio versus U/Th concentration ratio diagram for the 1986 lava samples. Figure 3 shows a positive correlation and a wide variation in the Ba/Th and U/Th concentration ratios. Because Ba is also highly mobile element in aqueous fluids relative to Th^{[18], 19)}, the positive correlation is ascribed to the addition of U- and Ba-rich fluids derived from the subducting slab by dehydration. These elements are very incompatible ele-

ments for most minerals^{20)–22)} and are difficult to fractionate each other by the melting of the mantle wedge and the crystallizing differentiation developing during the magmatic processes. Therefore, as Nakano et al.^[43] noted, the magmas for B- and C-vent eruptions had primarily different compositions from the magma for A-vent eruption.

4.3 ^{238}U – ^{230}Th radioactive disequilibria in the lava flows

Table 4 lists the activity ratios of $^{234}\text{U}/^{238}\text{U}$, $^{230}\text{Th}/^{238}\text{U}$, $^{238}\text{U}/^{232}\text{Th}$, $^{230}\text{Th}/^{232}\text{Th}$ and $^{226}\text{Ra}/^{230}\text{Th}$ in the 1986 lava samples. As ^{234}U in the 1986 lava samples has been reported to be in equilibrium with ^{238}U ^[49], ^{230}Th in the 1986 lava samples can be regarded as the direct decay product of ^{238}U .

Figure 4 is the $(^{230}\text{Th}/^{232}\text{Th}) - (^{238}\text{U}/^{232}\text{Th})$ activity ratio diagram for the 1986 lava samples^[16]. The $^{230}\text{Th}/^{238}\text{U}$ activity ratio is in the range of 0.53–0.70, being enriched in ^{238}U over ^{230}Th ^[16]. The previously published $^{230}\text{Th}/^{238}\text{U}$ activity ratio data^{[7), 50), 51)} in Table 2 for the products of the 1778 AD and 1950–1951 AD eruptions from Izu-Oshima volcano were also smaller than unity, being enriched in ^{238}U relative to ^{230}Th . The ^{238}U -excess observed in the 1986 lava samples was inferred to be resulted by the addition of U in the fluid derived from the subducting slab^[16].

Table 4 U–Th–Ra activity ratio data for the lava flows of the 1986 eruption from Izu-Oshima volcano

Sample identification	Unit	Activity ratio				
		$^{234}\text{U}/^{238}\text{U}^*$	$^{230}\text{Th}/^{238}\text{U}^*$	$^{238}\text{U}/^{232}\text{Th}^*$	$^{230}\text{Th}/^{232}\text{Th}^*$	$^{226}\text{Ra}/^{230}\text{Th}$
OSM05061702	LA	1.00 ± 0.02	0.53 ± 0.04	2.81 ± 0.18	1.49 ± 0.04	2.89 ± 0.22
OSM03052801	LB I	1.02 ± 0.03	0.70 ± 0.05	1.72 ± 0.10	1.21 ± 0.03	1.99 ± 0.14
OSM98062104	LB III	1.00 ± 0.02	0.61 ± 0.04	2.22 ± 0.16	1.35 ± 0.03	2.43 ± 0.19
OSM98062106	LC	1.00 ± 0.02	0.62 ± 0.04	2.12 ± 0.13	1.32 ± 0.03	2.30 ± 0.15

* Uranium and Th isotope data are taken from Table 2 of Ref. 16.

Fig. 4 $^{230}\text{Th}/^{232}\text{Th}$ activity ratio versus $^{238}\text{U}/^{232}\text{Th}$ activity ratio diagram for the lava flows of the 1986 eruption from Izu-Oshima volcano. (data from Kurihara et al.¹⁶⁾)

Fig. 5 Activity concentration of ^{226}Ra versus activity concentration of ^{230}Th for the lava flows of the 1986 eruption from Izu-Oshima volcano. (^{230}Th data from Kurihara et al.¹⁶⁾)

4·4 ^{230}Th – ^{226}Ra radioactive disequilibria in the lava flows

Activity concentration of ^{226}Ra versus activity concentration of ^{230}Th for the 1986 lava

samples is shown in Fig. 5. Figure 5 shows that the $^{226}\text{Ra}/^{230}\text{Th}$ activity ratio is in the range of 1.99–2.89, being enriched in ^{226}Ra over ^{230}Th . The $^{226}\text{Ra}/^{230}\text{Th}$ activity ratio value⁵¹⁾ for the products of the 1950–1951 AD eruption from Izu-Oshima volcano (JB-2) was also larger than unity, being enriched in ^{226}Ra over ^{230}Th .

Figure 6 is the $^{226}\text{Ra}/^{230}\text{Th}$ activity ratio versus $^{238}\text{U}/^{230}\text{Th}$ activity ratio (a) and Ba/Th concentration ratio (b) diagrams for the 1986 lava samples. Figure 6 shows $^{226}\text{Ra}/^{230}\text{Th}$ activity

ratio positively correlates with $^{238}\text{U}/^{230}\text{Th}$ activity ratio and Ba/Th concentration ratio in the lava samples. These correlations suggest that the ^{226}Ra excess over ^{230}Th in the 1986 lava samples may also be resulted by the addition of U- and Ra-rich fluids derived from the subducting slab by dehydration^{13)–15), 23)–27)}.

4·5 Time scale of magmatic processes in the 1986 eruption of Izu-Oshima volcano : Estimation from ^{238}U – ^{230}Th – ^{226}Ra radioactive disequilibria

The suite of $(^{230}\text{Th}/^{232}\text{Th})$ – $(^{238}\text{U}/^{232}\text{Th})$ data for the products from Izu-Oshima have formed a hypothetical whole rock isochron which indicated that the age of the magma was in the order of 10^4 years¹⁶⁾. However, the 1986 lava samples revealed ^{230}Th – ^{226}Ra disequilibria with ^{226}Ra excess over ^{230}Th , indicating that the ^{226}Ra -addition from the subducting slab by dehydration took place within 8 000 years. The older age of magma estimated by the

Fig. 6 $^{226}\text{Ra}/^{230}\text{Th}$ activity ratio versus $^{238}\text{U}/^{230}\text{Th}$ activity ratio (a) and Ba/Th concentration ratio (b) diagrams for the lava flows of the 1986 eruption from Izu-Oshima volcano. (Th and ^{230}Th concentrations and $^{238}\text{U}/^{230}\text{Th}$ activity ratio data from Kurihara et al.¹⁶⁾)

^{238}U – ^{230}Th disequilibria than that estimated by the ^{230}Th – ^{226}Ra disequilibria may suggest that U- and Ra-rich fluid contain some amounts of Th isotopes^{(13), (15), (25), (52)}, or that the ^{226}Ra excess reflects the time of the final stage of the possible continuous fluid addition, whereas the ^{238}U – ^{230}Th disequilibrium represents the apparent age of the whole of the magma accumulated since the beginning of the continuous fluid addition processes^{(23), (26), (53)}.

Assuming that ^{226}Ra excess over ^{230}Th was generated at the magma genesis, the time scale from the magma genesis to the eruption, including the melting of the mantle wedge, magma storage and crystallizing differentiation from basalt to basaltic andesite, is less than 8 000 years.

Acknowledgments

The authors gratefully acknowledge Prof. T. Nakamura, Meiji University, for his continuous encouragement, and Dr. K. Nakayama, Meiji University, for his cooperation in the X-ray

fluorescence analysis, and Dr. T. Saito, Osaka University, for his discussion and Dr. T. Nakano-Ohta, Kyoto University, and Mr. H. Takahashi, The University of Tokyo, for the cooperation in sampling the 1986 lava samples from Izu-Oshima volcano.

References

- 1) Ivanovich, M. and Harmon, R. S., Uranium-series Disequilibrium: Applications to Earth, Marine, and Environmental Sciences, pp.1-910, Oxford University Press, New York (1992)
- 2) Bourdon, B., Henderson, G. M., Lundstrom, C. C. and Turner, S. P., Uranium-Series Geochemistry, pp.1-656, Geochemical Society, Mineralogical Society of America, Washington, D. C. (2003)
- 3) Peate, D. W. and Hawkesworth, C. J., U series disequilibrium: Insights into mantle melting and the timescales of magma differentiation, *Rev. Geophys.*, **43**, 1-43 (2005)
- 4) Somayajulu, B. L. K., Tatsumoto, M., Rosholt, J. N. and Knight, R. J., Disequilibrium of the ^{238}U series in basalt, *Earth Planet. Sci. Lett.*, **1**, 387-391 (1966)
- 5) Nishimura, S., Disequilibrium of the ^{238}U series in

- recent volcanic rocks, *Earth Planet. Sci. Lett.*, **8**, 293-300 (1970)
- 6) Condomines, M., Hemond, Ch. and Allègre, C. J., U-Th-Ra radioactive disequilibria and magmatic processes, *Earth Planet. Sci. Lett.*, **90**, 243-262 (1988)
 - 7) Gill, J. B. and Williams, R. W., Th isotope and U-series studies of subduction-related volcanic rocks, *Geochim. Cosmochim. Acta*, **54**, 1427-1442 (1990)
 - 8) Turner, S., Hawkesworth, C., van Calsteren, P., Heath, E., Macdonald, R. and Black, S., U-series isotopes and destructive plate margin magma genesis in the Lesser Antilles, *Earth Planet. Sci. Lett.*, **142**, 191-207 (1996)
 - 9) Turner, S., Hawkesworth, C., Rogers, N., Bartlett, J., Worthington, T., Hergt, J., Pearce, J. and Smith, I., ^{238}U - ^{230}Th disequilibria, magma petrogenesis, and flux rates beneath the depleted Tonga-Kermadec island arc, *Geochim. Cosmochim. Acta*, **61**, 4855-4884 (1997)
 - 10) Elliott, T., Plank, T., Zindler, A., White, W. and Bourdon, B., Element transport from slab to volcanic front at the Mariana arc, *J. Geophys. Res.*, **102**, 14991-15019 (1997)
 - 11) Turner, S., McDermott, F., Hawkesworth, C. and Kepezhinskas, P., A U-series study of lavas from Kamchatka and Aleutians: constraints on source composition and melting processes, *Contrib. Mineral. Petrol.*, **133**, 217-234 (1998)
 - 12) Turner, S. P., Peate, D. W., Hawkesworth, C. J., Eggins, S. M. and Crawford, A. J., Two mantle domains and the time scales of fluid transfer beneath the Vanuatu arc, *Geology*, **27**, 963-966 (1999)
 - 13) Chabaux, F., Hémond, C. and Allègre, C. J., ^{238}U - ^{230}Th - ^{226}Ra disequilibria in the Lesser Antilles arc: implications for mantle metasomatism, *Chem. Geol.*, **153**, 171-185 (1999)
 - 14) Turner, S. and Foden, J., U, Th and Ra disequilibria, Sr, Nd and Pb isotope and trace element variations in Sunda arc lavas: predominance of a subducted sediment component, *Contrib. Mineral. Petrol.*, **142**, 43-57 (2001)
 - 15) Yokoyama, T., Kobayashi, K., Kuritani, T. and Nakamura, E., Mantle metasomatism and rapid ascent of slab components beneath island arcs: Evidence from ^{238}U - ^{230}Th - ^{226}Ra disequilibria of Miyakejima volcano, Izu arc, Japan, *J. Geophys. Res.*, **108**, doi: 10.1029/2002JB 002103 (2003)
 - 16) Kurihara, Y., Takahashi, M. and Sato, J., ^{238}U - ^{230}Th radioactive disequilibria in the volcanic products from Izu arc volcanoes, Japan, *RADIO-ISOTOPES*, **56**, 795-809 (2007)
 - 17) Tatsumi, Y., Migration of fluid phases and genesis of basalt magmas in subduction zones, *J. Geophys. Res.*, **94**, 4697-4707 (1989)
 - 18) Brenan, J. M., Shaw, H. F., Ryerson, F. J. and Phinney, D. L., Mineral-aqueous fluid partitioning of trace elements at 900 °C and 2.0 GPa: Constraints on the trace element chemistry of mantle and deep crustal fluids, *Geochim. Cosmochim. Acta*, **59**, 3331-3350 (1995)
 - 19) Keppler, H., Constraints from partitioning experiments on the composition of subduction-zone fluids, *Nature*, **380**, 237-240 (1996)
 - 20) Beattie, P., The generation of uranium series disequilibria by partial melting of spinel peridotite: constraints from partitioning studies, *Earth Planet. Sci. Lett.*, **117**, 379-391 (1993)
 - 21) Green, T. H., Experimental studies of trace-element partitioning applicable to igneous petrogenesis—Sedona 16 years later, *Chem. Geol.*, **117**, 1-36 (1994)
 - 22) Blundy, J. and Wood, B., Mineral-melt partitioning of uranium, thorium and their daughters, *Rev. Mineral. Geochem.*, **52**, 59-123 (2003)
 - 23) Turner, S., Bourdon, B., Hawkesworth, C. and Evans, P., ^{226}Ra - ^{230}Th evidence for multiple dehydration events, rapid melt ascent and the time scale of differentiation beneath the Tonga-Kermadec island arc, *Earth Planet. Sci. Lett.*, **179**, 581-593 (2000)
 - 24) Turner, S., Evans, P. and Hawkesworth, C., Ultrafast source-to-surface movement of melt at island arcs from ^{226}Ra - ^{230}Th systematics, *Science*, **292**, 1363-1366 (2001)
 - 25) Sigmarsson, O., Chmieleff, J., Morris, J. and Lopez-Escobar, L., Origin of ^{226}Ra - ^{230}Th disequilibria in arc lavas from southern Chile and implications for magma transfer time, *Earth Planet. Sci. Lett.*

- 196, 189-196 (2002)
- 26) Dosseto, A., Bourdon, B., Jordon, J. -L. and Dupré, B., U-Th-Pa-Ra study of the Kamchatka arc : New constraints on the genesis of arc lavas, *Geochim. Cosmochim. Acta*, **67**, 2857-2877 (2003)
 - 27) Kurihara, Y., Takahashi, M. and Sato, J., ^{238}U – ^{230}Th – ^{226}Ra radioactive disequilibria in the products from 1707 eruption of Fuji volcano, Japan, *RADIOISOTOPES*, **57**, 471-483 (2008)
 - 28) Tsuya, H., Geological and petrological studies of Volcano Fuji, V : On the 1707 eruption of Volcano Fuji, *Bull. Earthq. Res. Inst. Univ. Tokyo*, **33**, 341-383 (1955)
 - 29) Nakamura, T., Banju, M., Sato, J. and Takahashi, H., Compositional variation within the 1707 fall-out tephra from Fuji volcano, *Bull. Volcanol. Soc. Japan*, **31**, 253-264 (1986) (in Japanese with English abstract)
 - 30) Yoshimoto, M., Fujii, T., Kaneko, T., Yasuda, A. and Nakada, S., Multiple magma reservoirs for the 1707 eruption of Fuji volcano, Japan, *Proc. Japan Acad. Ser. B*, **80**, 103-106 (2004)
 - 31) Watanabe, S., Widom, E., Ui, T., Miyaji, N. and Roberts, A. M., The evolution of a chemically zoned magma chamber : The 1707 eruption of Fuji volcano, Japan, *J. Volcanol. Geotherm. Res.*, **152**, 1-19 (2006)
 - 32) Nakamura, K., Volcano-stratigraphic study of Ōshima volcano, Izu, *Bull. Earthq. Res. Inst. Univ. Tokyo*, **42**, 649-728 (1964)
 - 33) Isshiki, N., Geology of the Ōshima district. Quadrangle series, scale 1 : 50000, pp. 1-133, Geol. Surv. Japan (1984) (in Japanese with English abstract)
 - 34) Kawanabe, Y., Geological map of Izu-Ōshima volcano 1 : 25000 scale, Geol. Surv. Japan (1998) (in Japanese with English abstract)
 - 35) Soya, T., Sakaguchi, K., Uto, K., Nakano, S., Hoshizumi, H., Kamata, H., Sumii, T., Kaneko, N., Yamamoto, T., Tsuchiya, N., Suto, S., Yamazaki, H., Yamaguchi, Y., Okumura, Y. and Togashi, S., The 1986 eruption and products of Izu-Ōshima volcano, *Bull. Geol. Surv. Japan*, **38**, 609-630 (1987) (in Japanese with English abstract)
 - 36) Endo, K., Chiba, T., Taniguchi, H., Sumita, M., Tachikawa, S., Miyahara, T., Uno, R. and Miyaji, N., Tephrochronological study on the 1986-1987 eruptions of Izu-Oshima volcano, Japan, *Bull. Volcanol. Soc. Japan*, **33**, S32-S51 (1988) (in Japanese with English abstract)
 - 37) Nakano, S. and Yamamoto, T., Major element chemistry of products of the 1986 eruption of Izu-Ōshima volcano, *Bull. Geol. Surv. Japan*, **38**, 631-647 (1987) (in Japanese with English abstract)
 - 38) Fujii, T., Aramaki, S., Kaneko, T., Ozawa, K., Kawanabe, Y. and Fukuoka, T., Petrology of the lavas and ejecta of the November, 1986 eruption of Izu-Oshima volcano, *Bull. Volcanol. Soc. Japan*, **33**, S234-S254 (1988) (in Japanese with English abstract)
 - 39) Hayakawa, Y., Eruption volume and eruptive rate of the 1986 eruption of Izu-Oshima volcano, *Monthly Earth*, **9**, 366-371 (1987) (in Japanese)
 - 40) Nagaoka, M., Geomorphological characteristics of volcanic products of the 1986 eruption of Izu-Ōshima volcano and its volume measured by photogrammetry, *Bull. Volcanol. Soc. Japan*, **33**, S7-S15 (1988) (in Japanese with English abstract)
 - 41) Nakano, S., Yamamoto, T., Takada, A. and Soya, T., Chemical variation of fissure eruption products at post-caldera Y₅ stage of Izu-Oshima volcano, *Bull. Volcanol. Soc. Japan*, **33**, 31-35 (1988) (in Japanese)
 - 42) Aramaki, S. and Fujii, T., Petrological and geological model of the 1986-1987 eruption of Izu-Oshima volcano, *Bull. Volcanol. Soc. Japan*, **33**, S297-S306 (1988) (in Japanese with English abstract)
 - 43) Nakano, S., Togashi, S. and Yamamoto, T., Bulk and mineral chemistry of products of the 1986 eruption of Izu-Oshima volcano, *Bull. Volcanol. Soc. Japan*, **33**, S255-S264 (1988) (in Japanese with English abstract)
 - 44) Nakayama, K. and Nakamura, T., X-ray fluorescence analysis of rare earth elements in rocks using low dilution glass beads, *Anal. Sci.*, **21**, 815-822 (2005)
 - 45) Nakayama, K., Shibata, Y. and Nakamura, T., Glass beads/x-ray fluorescence analyses of 42 components in felsic rocks, *X-ray Spectrom.*, **36**,

- 130-140 (2007)
- 46) Lee, M. H. and Lee, C. W., Preparation of alpha-emitting nuclides by electrodeposition, *Nucl. Instrum. Methods, A*, **447**, 593-600 (2000)
- 47) Uchiumi, S., Uto, K. and Shibata, K., K-Ar ages of rock reference materials, *Mass Spectroscopy*, **37**, 375-381 (1989) (in Japanese with English abstract)
- 48) Imai, N., Terashima, S., Itoh, S. and Ando, A., 1994 compilation values for GSJ reference samples, "Igneous rock series", *Geochem. J.*, **29**, 91-95 (1995)
- 49) Sato, J. and Endo, M., Activity ratios of uranium isotopes in volcanic rocks from Izu-Mariana Island-Arc volcanoes, Japan, *J. Nucl. Radiochem. Sci.*, **2**, N1-N3 (2001)
- 50) Fukuda, S. and Nakai, S., $^{238}\text{U}/^{230}\text{Th}$ disequilibrium measurement for volcanic standard rock samples using a multiple-collector ICPMS, *Geochem. J.*, **36**, 465-473 (2002)
- 51) Yokoyama, T., Kuritani, T., Kobayashi, K. and Nakamura, E., Geochemical evolution of a shallow magma plumbing system during the last 500 years, Miyakejima volcano, Japan: Constraints from ^{238}U - ^{230}Th - ^{226}Ra systematics, *Geochim. Cosmochim. Acta*, **70**, 2885-2901 (2006)
- 52) Tepley III, F. J., Lundstorm, C. C., Gill, J. B. and Williams, R. W., U-Th-Ra disequilibria and the time scale of fluid transfer and andesite differentiation at Arenal volcano, Costa Rica (1968-2003), *J. Volcanol. Geotherm. Res.*, **157**, 147-165 (2006)
- 53) Turner, S., Bourdon, B. and Gill, J., Insights into magma genesis at convergent margins from U-series isotopes, *Rev. Mineral. Geochem.*, **52**, 255-315 (2003)

要 旨

伊豆大島火山 1986 年溶岩流の ^{238}U - ^{230}Th - ^{226}Ra 放射非平衡

栗原雄一, 高橋賢臣, 佐藤 純

明治大学理工学部応用化学科

214-8571 神奈川県川崎市多摩区東三田 1-1-1

伊豆大島火山 1986 年溶岩流の ^{238}U - ^{230}Th - ^{226}Ra 放射非平衡から, 1986 年の噴火活動におけるマグマ発生から噴出に至る時間を推定した。伊豆大島火山 1986 年溶岩流では, ^{230}Th - ^{226}Ra 間は, ^{230}Th に比べて ^{226}Ra が多い $^{226}\text{Ra}/^{230}\text{Th} > 1$ の放射非平衡であった。 $^{226}\text{Ra}/^{230}\text{Th}$ 放射能比と $^{238}\text{U}/^{230}\text{Th}$ 放射能比及び Ba/Th 濃度比との間で, 伊豆大島火山 1986 年溶岩流のプロットが正の相関を示したことにより, 1986 年溶岩流の ^{238}U - ^{230}Th - ^{226}Ra 放射非平衡は, マグマ発生時に起こったものであると考えられる。伊豆大島火山 1986 年溶岩流が ^{230}Th - ^{226}Ra 間の放射非平衡を保持していることより, 1986 年に噴出したマグマの発生から噴出までの時間は, 玄武岩から玄武岩質安山岩までの結晶分化の時間も含めて, 8 000 年以内であると推定された。