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Notes

Activity Ratios of Uranium Isotopes in Volcanic Rocks from Izu-Mariana Island-Arc Volcanoes, Japan

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The $^{234}U/^{238}U$ activity ratios of 45 products from 6 volcanoes of Izu-Mariana island-arc ranged from 0.96 to 1.04, representing a Gaussian distribution with the average value of 0.997 and the standard deviation of 0.014. The result indicates that ^{234}U is in radioactive equilibrium with ^{238}U in the erupting magmas from island-arc volcanoes.

1. Introduction

Radioactive disequilibrium among U-series nuclides in erupting magmas has been investigated with products from volcanoes of wide localities in the world.[1, 2] As for non-isotopic members, the disequilibria of $^{230}Th < ^{238}U$ and $^{230}Th > ^{238}U$ have been observed with the volcanoes locating along the subducting zone and on the hot-spot or along the mid-oceanic ridge, respectively.[1–7]

As ^{234}U is an isotopic daughter of ^{238}U, the deviation of the $^{234}U/^{238}U$ activity ratio from unity due to chemical processes, including magmatic evolution, is far less probable than the case of such non-isotopic combinations as ^{238}U-^{230}Th.

The disequilibrium between ^{238}U and ^{234}U has been observed with such geochemical samples as underground waters, which is inferred to be raised mainly due to α-recoil phenomena within the aquifer, suggesting that the contact of liquid phase with solid phase can give rise to the disequilibrium in the liquid phase. Liquid magma interacts with surrounding solid materials for the long period from its generation to the eruption. Nishimura[1] compiled $^{234}U/^{238}U$ activity ratios in recent volcanic rocks and reported that the activity ratios ranged from 0.8 to 1.2. The $^{234}U/^{238}U$ activity ratios observed with the subduction-related volcanic products form a group within a close range around unity, ranging from 0.94 ± 0.06 to 1.06 ± 0.10 (uncertainties are expressed in 2σ),[2] indicating that they are almost in equilibrium, although some of them may be suggesting the possibility of a slight deviation from unity.

However, as the number of samples in the majority of observations with the volcanoes locating in respective region is limited, it is considered not to be sufficient enough for the discussion on the slight disequilibrium, if any, between ^{238}U and ^{234}U in erupting magmas from the particular region interested. As for Izu-Mariana island-arc, Japan, two observations on $^{234}U/^{238}U$ activity ratio were reported: 1.01 ± 0.10 for Fuji and 0.95 ± 0.10 for Izu-Oshima (uncertainties are expressed in 2σ).[2]

The present report deals with the observation of activity ratios of $^{234}U/^{238}U$ in the 45 volcanic rocks erupted recently from 6 volcanoes located along Izu-Mariana island-arc. The observed $^{234}U/^{238}U$ activity ratios were treated statistically to discuss on the radioactive equilibrium between ^{238}U and ^{234}U.

2. Experimentals

2.1. Sample Description. Observations were carried out with 45 volcanic rock samples. They are the products from Miyake-jima (1983), Nii-jima (886), Kozu-shima (838), Izu-Oshima (1986), Fuji (1707) and Asama (1108 and 1783) volcanoes which are locating along Izu-Mariana island-arc as shown in Figure 1. Figure 1 contains the depth of the subducting plate which is corresponding to the depth of the magmatic generation of respective volcano. The products from Izu-Mariana island-arc volcanes are of a wide variety of rock types ranging from basalt (for Miyake-jima and Izu-Oshima) to rhyolite (for Kozu-shima and Niijima), volcanic rocks formed in the early and late stage of the crystal differentiation of magmatic evolution, respectively. The rock type of the product from Asama is andesite which was formed in the intermediate stage of the evolution.

The products from Hawaiian island on the hot-spot and those from Iceland along the mid-oceanic ridge are also employed in the present mesurement for comparison.

The samples analyzed are listed in Table 1, and their detailed descriptions are as follows.

1) *Miyake-jima (1983)*; Five samples were taken from Ako and Awabe lava flows and one sample was collected from the scoria cone formed at Nippana.

2) *Niijima (886)*; The 886 eruption of Niijima produced 3 eruptive units: My_1 and My_2 which consist of pumiceous volcanic ash, lapilli, and volcanic brocks (biotite rhyolitic), and My_3 which is biotite rhyolitic lava and Shiromama layer of My_1.

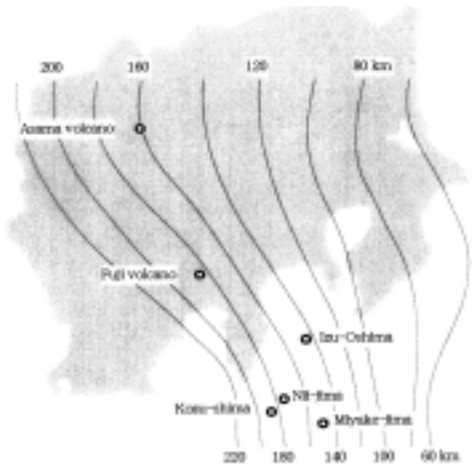

Figure 1. Localities of 6 volcanoes along Izu-Mariana island-arc. Lines on the map represent the depth of the subducting plate.

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TABLE 1: Vocanic Products from Izu-Mariana Island-Arc Volcanoes and Their $^{234}U/^{238}U$ Activity Ratios. Those from Hawaiian Islands and Iceland are also Included for Comparison. Errors on the Activity Ratios are the 1σ Uncertainties from Counting Statistics

Volcano (Eruption year)	Sample identificaftion	Rock type	Activity ratio $^{234}U/^{238}U$
Niijima volcano (886)	NJM96052602 (My_1)	rhyolite	0.99 ± 0.01
	NJM96052605 (Shiromama)	rhyolite	0.99 ± 0.01
	NJM96052607 (My_3)	rhyolite	1.00 ± 0.01
	NJM96052611 (My_2)	rhyolite	1.01 ± 0.01
(Av.)			1.00 ± 0.01
Kozu-shima volcano (838)	KZS96052702 (Tj_2)	rhyolite	0.98 ± 0.01
	KZS96052704 (Tj_2)	rhyolite	0.99 ± 0.01
	KZS96052706 (Tj_3)	rhyolite	1.00 ± 0.01
	KZS96052710 (Tj_4)	rhyolite	0.99 ± 0.01
	KZS96052711 (Tj_1)	rhyolite	1.01 ± 0.01
(Av.)			0.99 ± 0.01
Asama volcano (1108)	ASM840817-6	andesite	1.01 ± 0.01
	ASM850523-1	andesite	0.99 ± 0.01
Asama volcano (1783)	OND 17	andesite	1.03 ± 0.01
	OND 25	andesite	1.00 ± 0.01
	OND 33	andesite	0.98 ± 0.01
	OND 42	andesite	1.00 ± 0.01
	OND 45	andesite	1.00 ± 0.01
	OND 54	andesite	0.99 ± 0.01
(Av.)			1.00 ± 0.02
Miyake-jima volcano (1983)	MYK97052504 (Ako)	basalt	1.00 ± 0.01
	MYK97052505 (Awabe)	basalt	0.98 ± 0.01
	MYK97052602 (Ako)	basalt	1.00 ± 0.01
	MYK97052507 (Ako)	basalt	0.99 ± 0.01
	MYK97052509 (Awabe)	basalt	0.98 ± 0.01
	MYK97052510 (Nippana)	basalt	1.01 ± 0.01
(Av.)			0.99 ± 0.01
Izu-Oshima volcano (1986)	OSM98062101 (LBI)	basalt	0.99 ± 0.01
	OSM98062102 (LBII)	basalt	1.01 ± 0.01
	OSM98062104 (LBIII)	basalt	0.99 ± 0.01
	OSM98062106 (LC)	basalt	1.00 ± 0.01
	OSM86122007 (LA)	basalt	0.99 ± 0.01
(Av.)			0.99 ± 0.01
Fuji volcano (1707)	Hoei 1	dacite	0.99 ± 0.01
	Hoei 2	andesite	1.01 ± 0.01
	Hoei 3	andesite	0.98 ± 0.01
	Hoei 4	andesite	1.01 ± 0.01
	Hoei 5	andesite	1.01 ± 0.01
	Hoei 6	basalt	1.00 ± 0.01
	Hoei 7	basalt	0.99 ± 0.01
	Hoei 8	basalt	0.99 ± 0.01
	Hoei 9	basalt	1.01 ± 0.01
	Hoei 10	basalt	0.96 ± 0.01
	Hoei 11	basalt	0.99 ± 0.01
	Hoei 12	basalt	1.01 ± 0.01
	Hoei 13	basalt	0.99 ± 0.01
	Hoei 14	basalt	1.00 ± 0.01
	Hoei 15	basalt	1.04 ± 0.01
	Hoei 16	basalt	0.99 ± 0.01
	Hoei 17	basalt	0.99 ± 0.01
(Av.)			1.00 ± 0.02
Hawaii	TF79071901	basalt	1.01 ± 0.01
	TF79071905	trachyte	0.97 ± 0.01
(Av.)			0.99 ± 0.02
Iceland	TF80062701	basalt	0.97 ± 0.01
	TF80062002	basalt	0.99 ± 0.01
(Av.)			0.98 ± 0.01

3) *Kozu-shima (838)*; The 838 eruption of Kozu-shima produced 4 eruptive units: Tj_1 and Tj_2 which consist of pumiceous volcanic ash, lapilli, and volcanic brocks (biotite rhyolitic), and Tj_3 which is biotite rhyolitic lava and Tj_4 which is loose volcanic breccia of biotite rhyolite.
4) *Izu-Oshima (1986)*; The 1986 eruption formed 3 lava flows of LA, LB, and LC. The lava flow LB was devided into 3 branches (LB I , II, and III).
5) *Fuji (1707)*; The 1707 eruption of Fuji started with the eruption of dacitic pumice fall followed by andesitic and basaltic scoria fall, suggesting that the eruption has proceeded along the zoning layer formed in the magmatic reservoir. The air-fall tephra has formed 17 layers at Midono, Gotemba, and samples were collected from respective fall units.
6) *Asama (1108 and 1783)*; Two samples from the 1108 eruption of Asama were collected from "B" scoria fall layer, and 6 lava samples were taken from Onioshidashi lava flow of the 1783 eruption.

2.2. Chemical Separation and Radioactivity Measurement. The solvent extraction procedure using $CH_3CO_2C_2H_5$ was widely employed in isolation and purification of U from sample solution. However, as $Al(NO_3)_3 \cdot 9H_2O$, which is added in a large amount as the salting-out material in the procedure, contains ppb-levels of U impurity (0.9–12.1 ppb) in radioactive disequilibrium (the $^{234}U/^{238}U$ activity ratios were observed to range from 1.22 ± 0.07 to 4.29 ± 0.18),[8] the U impurity in the reagent can constitute a serious obstacle to the determination of U in minute concentration and to the measurement of $^{234}U/^{238}U$ activity ratios in volcanic rocks.

An application of U/TEVA·Spec resin, an extraction chromatograph material supplied by Eichrom Industries, Inc., U.S.A., to the separation procedure of U is the main improvement in this work. The extracting material is $(C_5H_{11}O)_2C_5H_{11}PO$, and it extracts hexavalent U and tetravalent actinides from $2MHNO_3$ aqueous solution and releases them into 0.02 M-HNO_3 aqueous media.

From 30 to 60 g of volcanic rocks were decomposed by the mixed acid of HF-$HClO_4$-HNO_3. A large amount of Fe was eliminated by the solvent extraction into $[(CH_3)_2CH]_2O$ from sample solution adjusted to 8 MHCl. After U fraction was separated by an anion exchange resin (Dowex 1-X8 Cl-form), U was isolated successfully by use of the U/TEVA·Spec resin. No detectable amount of U was observed in the blank test performed prior to the rock analysis.

Uranium was electrodeposited from the electrobath of a mixed solution of 0.02 $MHNO_3$ and 0.2 $M(NH_4)_2C_2O_4$ (pH = 2) onto a stainless steel planchet for α-ray counting. Activity ratios of $^{234}U/^{238}U$ were obtained by α-ray spectrometry using a 900 mm^2 Si detector (ORTEC) coupled with an MCA system (Laboratory Equipment; ADC 2201A).

3. Result and Discussion

Table 1 contains the $^{234}U/^{238}U$ activity ratios for the volcanic rock samples analyzed. Measurements were carried out so that all of the statistical errors (1σ) accompanying respective measurement were arranged to be ± 0.01, because it is convinient to compare the distribution of the set of individual data with the result of the statistical treatment of the whole data.

The $^{234}U/^{238}U$ activity ratios of rhyolitic magmas from Niijima and Kozu-shima varies from 0.98 to 1.01, those of andesitic magma from Asama from 0.98 to 1.03, and those of basaltic magma from Miyake-jima and Izu-Oshima from 0.98 to 1.01. The activity ratio along the fall units of sedimentation layer formed by the 1707 eruption of Fuji ranges from 0.98 to 1.04. The observed activity ratio showed no connection either with the rock type or with the order of eruptive sequence.

While the $^{234}U/^{238}U$ activity ratio of the 45 samples ranges from 0.96 to 1.04, the average value of the $^{234}U/^{238}U$ activity ratios of volcanic products from each volcano ranges from 0.99 to 1.00. The histogram for the distribution of whole observed values are shown in Figure 2.

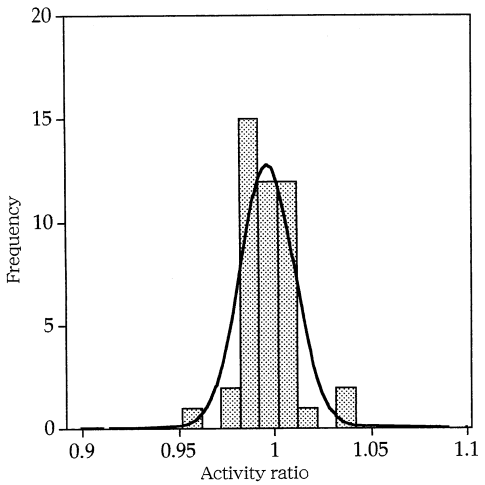

Figure 2. Histogram of $^{234}U/^{238}U$ activity ratios of the present 45 volcanic rock samples from Izu-Mariana island-arc volcanoes, Japan. The whole data represent a Gaussian distribution with the average value of 0.997 and the standard deviation of 0.014.

The histogram represents a Gaussian distribution having the average value of 0.997 and the standard deviation of 0.014 which is corresponding to the statistical errors of the individual measurements, indicating that the ^{234}U is in radioactive equilibrium with ^{238}U: the degree of the possible disequilibrium in the magma due to the crystal differentiation and/or the interaction with surrounding solid phase materials is estimated to be smaller than ± 0.01 which is corresponding to the detection limit of the present measurement.

Table 1 also contains the $^{234}U/^{238}U$ activity ratios for volcanic rock samples from Hawaiian islands and Iceland. The values of magmas from the hot-spot and mid-oceanic ridge ranged from 0.97 to 1.01 which is in a good agreement with the reported value[6] and is in the range of the Izu-Mariana island-arc volcanes. Uranium-234 in those magmas are also regarded to be in radioactive equilibrium with ^{238}U, though the number of samples are limited.

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References

(1) S. Nishimura, Earth Planet. Sci. Lett. **8**, 293 (1970).
(2) J. B. Gill and R. W. Williams, Geochim. Cosmochim. Acta **54**, 1427 (1990).
(3) S. Newman, J. D. Macdougall, and R. C. Finkel, Nature **308**, 268 (1984).
(4) M. Condomines, Ch. Hemond, and C. J. Allègre, Earth Planet. Sci. Lett. **90**, 243 (1988).
(5) M. Condomines, M. Barnat, and C. J. Allègre, Earth Planet. Sci. Lett. **33**, 122 (1976).
(6) B. L. K. Somayajulu, M. Tatsumoto, J. N. Rosholt, and R. J. Knight, Earth Planet. Sci. Lett. **1**, 387 (1966).
(7) M. Condomines, P. Morand, C. J. Allègre, and G. Sigvaldason, Earth Planet. Sci. Lett. **55**, 393 (1981).
(8) Y. Kanai, Bunseki Kagaku (Analytical Chemistry) **41**, T83 (1992).