# A LOW U/Pb SOURCE IN THE MOON: U-Th-Pb SYSTEMATICS OF LUNAR METEORITE YAMATO-793169

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Abstract: U-Th-Pb isotopic data were determined on mineral separates of the diabasic lunar meteorite, Yamato-793169, in order to define its age and characterize its magma source. Such results have been helpful toward our understanding of the magmatic/chemical evolution of the Moon. Analysis of HBr leaches revealed that the meteorite was heavily contaminated with terrestrial Pb during its residence in Antarctic ice. The three most radiogenic residues indicate a Pb-Pb age of  $3916 \pm 90$  Ma. However, this age determination is uncertain because of possible modern terrestrial Pb contamination. Residues of leached pyroxene and plagioclase separates exhibited the most radiogenic Pb compositions (206Pb/204Pb values up to 125) and the U-Pb data from these fractions plot on the lunar cataclysm array. This observation lead us to recalculate or correct all the U-Pb data for terrestrial Pb contamination in order to cause the other fractions to lie on the lunar cataclysm array as well. The contamination-corrected data and residues of pyroxene and plagioclase separates define U-Pb, Th-Pb, and Pb-Pb ages that are within error of each other. The best estimate for the Pb-Pb age of Yamato-793169 using the corrected data is  $3930 \pm 240$  Ma. The initial Pb isotopic composition obtained from U-Pb and Th-Pb isochrons is  ${}^{206}Pb/{}^{204}Pb = 11.2 \pm 0.8$ ,  ${}^{207}Pb/{}^{204}Pb = 13.1 \pm 4.3$ , and  ${}^{208}Pb/{}^{204}Pb =$  $33 \pm 11$ . Another approach to the data combines U and Th abundances removed during leaching (indigenous) with those in the residues, but disregards the Pb in the leaches (terrestrial contamination). Using this approach, the U-Pb data of the fractions plot inside the concordia curve along a discordia line, and pyroxene separates define intercept ages of 3.92 Ga with source  ${}^{238}U/{}^{204}Pb(\mu)$  of 10 between 4.56 and 3.92 Ga.

Although we cannot define the age of this diabase exactly, these calculations indicate that the age is around 3.9 Ga and not 3.3-3.6 Ga as observed for most LT and VLT mare basalts. Furthermore, the calculated initial Pb isotopic composition for this meteorite is slightly higher than the Canyon Diablo troilite Pb composition, indicating derivation from a source with a low but measurable  $\mu$  value at ~3.9 Ga. Assuming a single-stage Pb isotopic evolution model between 4.56 to 3.9 Ga, the source  $\mu$  value is estimated to be about 10. This value is much lower than those obtained for many Apollo mare basalts, suggesting that the deep lunar mantle may have had magma reservoirs with  $\mu$  values as low as that of the bulk Earth mantle.

#### 1. Introduction

Lead isotopic compositions of Apollo lunar rocks are very radiogenic compared to those of terrestrial rocks and most meteorites. This fact has led investigators to

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believe that the U/Pb ratio of the Moon is high (*e.g.*, TATSUMOTO, 1970a), commonly thought a result of depletion of volatile elements since the earliest stages of the Moon's history. Many workers have attempted to explain the loss of volatile elements, such as Pb, in various models of the Moon's origin. For example, CAMERON (1986) proposed a model of a hot lunar origin whereby the Moon formed from a cloud of vapor, dust, and particles that were blasted into Earth's orbit after the Earth collided with a roughly Mars-sized planet. The ejected silicate particles were superheated to over 5000 K during the event, resulting in volatile element depletion in the proto-Moon.

We have considered that the entire Moon had a high  ${}^{238}$ U/ ${}^{204}$ Pb ( $\mu$ ) from the onset (approximately 200) and a cumulate-remelting model (*e.g.*, PHILIPOTTS and SCHNEIZLER, 1970; NYQUIST *et al.*, 1977; FUJIMAKI and TATSUMOTO, 1984) is used to explain subsequent lunar U-Pb evolution: (1) early cumulates, including sulfides, settled from the magma ocean and are characterized by low  $\mu$  values (less than 50) due to the low partition coefficient of U and Th; (2) crust-forming anorthosite and troctolite were derived from the magma ocean with slightly elevated  $\mu$  values (300–500); and (3) residual liquids that are sources for KREEP and granite (TATSUMOTO, 1970b; COMPSTON *et al.*, 1989) have  $\mu$  values over 1000.

However, the  $\mu$  values of lunar green glass (TATSUMOTO *et al.*, 1987) and orange glass (TATSUMOTO *et al.*, 1973b; TERA and WASSERBURG, 1976) are estimated between 20 and 30. Green and orange glass are considered to have originated from the deep lunar mantle (DELANO, 1979), where metal sulfides may have accumulated. RINGWOOD (1992) argues that these glasses (lunar volcanic glasses; LVG) originated deep in the Moon's mantle and that the similarity of  $\mu$  values and abundance patterns in volatile and siderophile elements between the Moon and the Earth's mantle suggests that proto-lunar material originated from the Earth's mantle.

Furthermore, our recent U-Th-Pb studies of the lunar meteorite, Asuka(A)-881757 (MISAWA *et al.*, 1992: Asuka(A)-31 by tentative number), showed a somewhat different story, indicating that the meteorite was derived from a source with a considerably lower  $\mu$  (approximately 10) than we previously estimated. A-881757 is an unbrecciated gabbro and its bulk chemistry is similar to low titanium (LT) or very low-Ti (VLT) basalt, also thought to have originated from deep-seated olivine-orthopyroxene cumulates.

In this paper, we report the U-Th-Pb analyses of lunar meteorite, Yamato(Y)-793169, as a part of the consortium study. Y-793169 is an unbrecciated diabase and, similar to A-881757, resembles LT or VLT basalt (YANAI and KOJIMA, 1991). The major purpose of this study is to determine the age and history of the meteorite as well as characterize its source. The TiO<sub>2</sub> and REE abundances in LT and VLT basalts are lower than high-Ti (HT) basalts, indicating that the source magma was chemically less fractionated from primitive mantle than those of HT mare basalts. Therefore, U-Th-Pb analyses of this meteorite may give us more information about the primordial lunar mantle.

### 2. Experimental Procedure

Y-793169,43, allocated from the National Institute of Polar Research, was a single interior fragment, weighing 0.104 g. This meteorite consists mainly of pyroxene and

plagioclase with a small amount of ilmenite, spinel and glass (YANAI and KOJIMA, 1991). Since the amount of sample available for analysis was small, the sample was separated into only four fractions. First, the sample was gently crushed in a stainless steel mortar and divided into two size fractions,  $< 63 \,\mu\text{m}$  (Fines) and  $63-150 \,\mu\text{m}$ , using nylon cloth sieves. The  $63-150 \,\mu\text{m}$  fraction was separated into three using heavy liquids. A heavy fraction ( $\rho > 3.3$ ) was first separated from the  $63-150 \,\mu\text{m}$  fraction using methylene iodide. This fraction consists mainly of pyroxene (PX1), but also included minor ilmenite. The methylene iodide float fraction was then treated with bromoform; the fraction that sank in bromoform ( $2.85 < \rho < 3.3$ ), consisted mainly of pyroxene and other minerals (PX2). The floating fraction ( $\rho < 2.85$ ) consisted of plagioclase (PL).

Lunar meteorites recovered from Antarctic ice contain some Pb other than indigenous Pb, including adsorbed Pb from residence at the lunar surface, meteoritic Pb contributed from impactors, modern terrestrial Pb introduced during the meteorite's residence in the ice, and laboratory contamination (blank Pb) introduced during sample preparation. In order to remove these secondary Pb components, the fractions were washed with acetone and alcohol and leached with 0.01 N HBr and 0.1 N HBr. However, these procedures may have also removed some degree of indigenous U, Th, and Pb as well as secondary Pb contamination. The procedure has no guarantee for the perfect removal of secondary Pb.

The four separates (PX1, PX2, PL, and Fines) were first washed twice with acetone, and then ten times with ethanol, and then rinsed with distilled water (the acetone, ethanol, and water were combined and are hereafter referred to as acetone and ethanol washes). In order to remove any terrestrial Pb contamination or adsorbed surficial Pb components, the separates were leached twice with cold 0.01 N HBr, and rinsed twice with water (here after referred to as 0.01 N HBr leaches), and then finally leached twice with cold 0.1 N HBr and rinsed twice with distilled water (here after referred to as 0.1 N HBr leaches). The washing and leaching were carried out in an ultrasonic bath for 15 minutes at each step. The residues were then dissolved with concentrated HF and HNO<sub>3</sub>.

The U, Th, and Pb concentrations and Pb isotopic compositions were determined on all washes, leaches, and residues. Chemical separation procedures of U, Th, and Pb are described in PREMO and TATSUMOTO (1991, 1992a). Blank Pb levels were 42, 15, 13, and 32 pg total Pb for the washes, 0.01 N HBr leaches, 0.1 N HBr leaches, and residues, respectively. U and Th blanks were 3 pg or less for all the washes, leaches, and residues. All the Pb isotopic data were corrected for an analytical blank Pb with compositions of  ${}^{206}\text{Pb}/{}^{204}\text{Pb} = 19.76 \pm 0.20$ ,  ${}^{207}\text{Pb}/{}^{204}\text{Pb} = 15.79 \pm 0.15$ ,  ${}^{208}\text{Pb}/{}^{204}\text{Pb} = 38.79 \pm 0.35$ (average of four measurements), mass fractionation (0.15 ± 0.02% per a.m.u.), and spike contribution (LUDWIG, 1989).

### 3. Results and Discussion

The results of the analyses are given in Table 1. Laboratory Pb contamination in each fraction is generally small, less than 1%, except for the PL-residue, PL 0.1 N-HBr leach, PX1-wash, and Fines-wash, which contain less than 1 ng Pb. U, Th, and Pb abundances of all the separates at each step of the washing and leaching procedures are shown in Figs. 1a, b, and c. In Fig. 2, <sup>206</sup>Pb/<sup>204</sup>Pb values in relevant fractions are

Sample	Weight (mg)	Blank Pb (%)	Pb (ppb)	U (ppb)	Th (ppb)	<sup>206</sup> Pb/ <sup>204</sup> Pb	<sup>207</sup> Pb/ <sup>204</sup> Pb	<sup>208</sup> Pb/ <sup>204</sup> Pb	<sup>207</sup> Pb/ <sup>206</sup> Pb	<sup>208</sup> Pb/ <sup>206</sup> Pb	$^{238}U/^{204}Pb$	<sup>232</sup> Th/ <sup>204</sup> Pb
Residue	S											
PX1	36.96	0.23	376	175	770	124.6 (7)	58.89 (30)	153.2 (8)	0.47250 (34)	1.2289 (9)	135.9 (10)	617 (5)
PX2	16.47	0.37	530	170	728	56.70 (24)	31.60 (12)	79.99 (31)	0.55621 (46)	1.4107 (13)	46.88 (31)	207.7 (16)
PL	3.91	4.9	167	69.9	274	83 (6)	43.4 (25)	107 (6)	0.522 (6)	1.289 (14)	85 (8)	342 (32)
Fines	17.53	0.14	1274	159	646	25.212 (16)	18.386 (16)	45.29 (4)	0.72925 (34)	1.7963 (8)	9.673 (27)	40.70 (21)
0.1 N H	Br leach											
PX1	36.96	0.60	58.9	6.57	30.8	21.80 (7)	17.02 (5)	41.86 (13)	0.78076 (20)	1.9202 (8)	7.88 (7)	38.09 (30)
PX2	16.47	0.74	106	12.2	56.4	21.805 (38)	17.094 (31)	41.78 (9)	0.78392 (33)	1.9160 (26)	8.11 (7)	38.80 (35)
PL	3.91	6.6	50.4	5.35	39.4	22.04 (11)	17.16 (8)	42.24 (19)	0.7788 (10)	1.9165 (33)	7.6 (5)	57.5 (35)
Fines	17.53	0.4	184	6.83	54.6	19.22 (6)	16.02 (5)	39.02 (13)	0.8338 (4)	2.0304 (21)	2.421 (23)	18.87 (13)
0.01 N H	<b>HBr</b> leach											
<b>PX1</b>	36.96	0.18	230	17.2	40.2	19.521 (23)	16.077 (21)	39.51 (6)	0.82359 (39)	2.0241 (15)	4.924 (17)	11.88 (6)
PX2	16.47	0.42	216	20.8	46.9	20.051 (10)	16.269 (19)	40.013 (35)	0.8114 (8)	1.9955 (9)	6.410 (35)	14.95 (10)
PL	3.91	0.84	459	27.6	62.2	18.559 (31)	15.810 (37)	38.40 (8)	0.8519 (14)	2.0691 (21)	3.835 (32)	8.94 (8)
Fines	17.53	0.13	661	47.8	100.5	19.60 (5)	16.14 (4)	39.32 (11)	0.82334 (26)	2.0062 (11)	4.752 (21)	10.32 (6)
Acetone	& ethano	l wash										
PX1	36.96	3.8	29.8	0.54	1.72	19.293 (27)	15.984 (23)	39.36 (6)	0.8285 (8)	2.0403 (21)	1.18 (6)	1.72 (6)
PX2	16.47	0.41	620	11.4	25.0	19.49 (8)	16.13 (7)	39.87 (17)	0.82752 (31)	2.0459 (22)	1.212 (14)	2.754 (25)
PL	3.91	0.30	3580	25.3	49.5	18.229 (22)	15.684 (21)	38.20 (5)	0.8604 (4)	2.0957 (9)	0.447 (4)	0.902 (6)
Fines	17.53	5.9	40.3	0.90	2.36	19.27 (9)	16.04 (8)	39.25 (19)	0.8320 (13)	2.0365 (31)	1.47 (8)	3.96 (21)

Table 1. U-Th-Pb analytical data for Yamato 793169\*.

\* Concentrations and isotopic ratios are corrected for blank Pb using the method of LUDWIG (1989, 1991). The concentrations for washes and leaches are calculated using the original weight of the sample fractions. Numbers in the parentheses are 2σ errors (95% confidence level) for the last degits.



Fig. 1. U, Th, and Pb abundances in fractions from Y-793169 at different leaching steps. (a) U, (b) Th, and (c) Pb. Concentrations of each element are shown in units of 10<sup>-12</sup> mole/g for the untreated (original) sample weight.



Fig. 2. Variations of <sup>206</sup> Pb/<sup>204</sup> Pb values in fractions from Y-793169 at different leaching steps.

also used as an index for terrestrial Pb contamination.

### 3.1. Leaching procedures

Generally, the amounts of U and Th removed by washing and leaching of Apollo samples are much less than the abundances of these elements in the residues. In contrast, for Y-793169, a significant amount of Pb was removed by the washing and leaching procedures: especially for PL and PX2, where the amount of Pb removed during acetone and ethanol washing was much higher than the amounts of Pb in the residues (Fig. 1c) The Pb isotopic compositions of all the washes and leaches are close to modern terrestrial Pb compositions, indicating that the washing and leaching procedures effectively removed terrestrial Pb contamination. Even for the 0.1N HBr leaches, the Pb isotopic compositions are very close to those of blank Pb. Therefore, these fractions seem to be heavily contaminated with terrestrial Pb.

Total concentrations of U and Th in each separate are ~0.2 ppm U and 0.8 ppm Th for PX1, PX2 and Fines, and ~0.1 ppm U and 0.38 ppm Th for PL. These concentrations are about 3 to 10 times higher than those in A-881757 (MISAWA *et al.*, 1992) and Luna 24 VLT mare basalt (UNRUH and TATSUMOTO, 1978), but similar to three Apollo 12 LT mare basalts (12009, 12022, and 12064; TATSUMOTO *et al.*, 1971). However, observed  $\mu$  and  ${}^{206}$ Pb/ ${}^{204}$ Pb values of Y-793169 separates are much lower than those of Apollo 12 LT mare basalts, but closer to those of A-881757 and Luna 24 VLT basalt.  ${}^{206}$ Pb/ ${}^{204}$ Pb values are ~400 for Apollo 12 LT mare basalts compared to 20–150 for Y-793169 residues, between 22 and 50 for Luna 24170, and between 30 and 100 for A-881757 (except for a value of 507 from an ilmenite separate). These  $\mu$  values indicate that the relative abundance of  ${}^{204}$ Pb in the lunar meteorites is higher than that in Apollo 12 LT mare basalts.

#### 3.2. Pb isotopic systematics

The Pb isotopic data for all the fractions are shown in Fig. 3. The residues exhibit



Fig. 3.  ${}^{204}Pb/{}^{206}Pb$  vs.  ${}^{207}Pb/{}^{206}Pb$  diagram for Y-793169 data. Squares indicate PX1, diamonds = PX2, circles = PL, and triangles = Fines. Filled symbols are data from residues and open symbols are washes and leaches. The solid line was calculated from the three residues, PX1, PX2, and PL. The age calculated from the Y-intercept (0.402±0.024), the radiogenic  ${}^{207}Pb/{}^{206}Pb$  ratio of the sample, is  $3916\pm90$  Ma using the regression method of LUDWIG (1991).

Pb isotopic compositions that are much more radiogenic than the washes and leaches (except for the Fines residue). As shown in Table 1, the Pb isotopic composition of Fine-residue is closer to a terrestrial Pb composition than the other residue compositions. Its Pb concentration is the highest among the residues, indicating that the Fine fraction probably still contained some adsorbed terrestrial Pb even after leaching. For this reason, the Pb-Pb age of  $3916 \pm 90$  Ma (Fig. 3) is calculated using three residues and excluding the Fines. Although it is believed that terrestrial Pb contamination was taken off by leaching, there is no way of knowing how much terrestrial Pb might have been remained in these fractions and this Pb -Pb age is still uncertain.

## 3.3. U-Th-Pb systematics

The U-Pb, Th-Pb isochron plots for the residues and leaches are shown in Figs. 4a, b, and c. The data from washes, leaches, and the Fines residue are close to terrestrial Pb compositions, as their  ${}^{238}\text{U}/{}^{204}\text{Pb}$  or  ${}^{232}\text{Th}/{}^{204}\text{Pb}$  values approach zero, indicating the removal of terrestrial Pb contamination. The residues show linear trends, yielding ages calculated from the three most radiogenic fractions;  $3655 \pm 44 \text{ Ma}$  ( ${}^{238}\text{U}-{}^{206}\text{Pb}$  age),  $3826 \pm 16 \text{ Ma}$  ( ${}^{235}\text{U}-{}^{207}\text{Pb}$  age), and  $3324 \pm 53 \text{ Ma}$  ( ${}^{232}\text{Th}-{}^{208}\text{Pb}$  age). These ages do not agree with each other, and the initial ratios calculated from these isochrons,  ${}^{206}\text{Pb}/{}^{204}\text{Pb} = 20.9 \pm 0.7$ ,  ${}^{207}\text{Pb}/{}^{204}\text{Pb} = 17.2 \pm 0.3$ ,  ${}^{208}\text{Pb}/{}^{204}\text{Pb} = 43.0 \pm 0.9$ , are close to modern terrestrial Pb isotopic compositions. In addition to post-crystallization Pb loss or gain relative to U and Th, perhaps during the impact events that caused maskelynitization and produced the meteorite, there are important effects. Two probable effects disturbing these isochrons are (1) terrestrial Pb contamination remaining in the residues, and (2) elemental fractionation due to preferential leaching of U, Th, or Pb during HBr treatments (PREMO and TATSUMOTO, 1992b). Because a significant amount



Fig. 4. The U-Pb and Th-Pb isochron diagrams for fractions from Y-793169. (a)  ${}^{238}U/{}^{204}Pb vs. {}^{206}Pb/{}^{204}Pb$ , (b)  ${}^{235}U/{}^{204}Pb vs. {}^{207}Pb/{}^{204}Pb$ , (c)  ${}^{232}Th/{}^{204}Pb vs. {}^{208}Pb/{}^{204}Pb$ . The symbols are the same as those in Fig. 3.

of both U and Th was leached out during HBr treatment, it is possible that the leaching procedure caused artificial effects on the U-Pb and Th-Pb systematics of the residues. To correct for leaching effects on the data, the leach data can be added back to the residue data. However, because of the large amount of terrestrial Pb in most of the leaches, this approach may also cause the erroneous results for these fractions.

In Fig. 5a, a modified concordia diagram shows the U-Pb behavior of leaches and residues corrected for initial Pb composition using the composition of Canyon Diablo troilite (CDT; TATSUMOTO *et al.*, 1973a). Except for the PX1-residue and PL-residue, all of the data plot outside of the concordia curve, indicating that there is an excess of radiogenic Pb unsupported by U and Th radioactive decay, possibly as a result of terrestrial Pb contamination. The most radiogenic samples, PX1- and PL-residues plot close to the cataclysm array for lunar rocks (TERA and WASSERBURG, 1974). This feature indicates the probability that Pb contamination in these fractions was removed during leaching, suggesting that the other U-Pb data from Y-793169 separates would lie on the cataclysm array if this meteorite had not been contaminated while residing in Antarctic ice. Note that PX-2 is above concordia and has much low <sup>206</sup>Pb/<sup>204</sup>Pb and much higher <sup>204</sup>Pb than PX-1.

#### 3.4. Correction for terrestrial Pb contamination

In order to discuss the U-Th-Pb systematics of this meteorite, we must make some assumptions with regard to the terrestrial Pb contamination. In the following, we consider two approaches to the U-Pb data from Y-793169. The first approach assumes that any deviation of the total U-Pb data (combined leach and residue for each fraction) from the lunar cataclysm array on a concordia diagram is due entirely to terrestrial Pb contamination. This assumption is reasonable if we consider that the two most radiogenic residues, PX1 and PL, already lie on the array (Fig. 5a) and the fact that the Pb-Pb data for residues give a 3.9 Ga-age. The lunar cataclysm array is defined by the U-Pb data from many Apollo terra rocks, including some mare basalts (TERA and WASSERBURG, 1974). As is commonly observed, plagioclase in many of these samples plots outside of the concordia curve, indicating that this mineral contains some "old" (apparently  $\sim$  4.42 Ga) Pb component. In the case of this  $\sim$  3.9 Ga-old meteorite, the "old" Pb may have been introduced into the plagioclase during an impact event that partially transformed it to maskelynite. The Pb composition of the maskelynite subsequently remains unchanged because of the very low concentrations of U and Th. Similar results were observed from the U-Pb data of maskelynite from leached residues of lunar meteorite, A-881757 (MISAWA et al., 1992) and norite 78235 (PREMO and TATSUMOTO, 1991).

When we plot the total U-Pb data, all the fractions plot outside the concordia curve (Fig. 5b, filled symbols). Assuming the only cause of the deviation is from terrestrial Pb contamination, we note that large amounts of modern terrestrial Pb must be removed (*e.g.* more than 70% of the <sup>204</sup>Pb in the Fines fraction) in order to correct the data to the lunar cataclysm array (Fig. 5b, open symbols). The present average crustal Pb composition ( $^{206}$ Pb/ $^{204}$ Pb = 18.70,  $^{207}$ Pb/ $^{204}$ Pb = 15.63, and  $^{208}$ Pb/ $^{204}$ Pb = 38.63; STACEY and KRAMERS, 1973) was used as the modern terrestrial Pb composition. The results are given in Table 2.



Fig. 5a. Modified concordia diagram showing the U-Pb isotopic behavior of leaches and residues of Y-793169.

Filled symbols indicate residues and open symbols indicate leaches. All the data are corrected using the initial Pb isotopic composition of Canyon Diablo troilite. Lunar cataclysm array (TERA and WASSERBURG, 1974) is also shown (solid line), which intersects the concordia curve at 4.42 and 3.9 Ga.



Fig. 5b. Modified concordia diagram showing the contamination-corrected total U-Pb data. Filled symbols indicate uncorrected total U-Pb data and open symbols indicate contamination corrected data. Squares indicate PX1, diamonds = PX2, circles = PL, and triangles = Fines. Percentages equated to each corrected fraction indicate the amounts of <sup>204</sup> Pb contamination removed from the uncorrected data. The calculated  $\mu$ -value for each fraction is also shown.

Although a large amount of Pb is subtracted from the total data as contamination, the  $^{238}U/^{204}Pb$  ratios of the corrected total data of PX1 and PL are lower than those using only residue data. This observation indicates that either some low- $\mu$  materials, such as troilite, may have been dissolved during leaching with dilute HBr or the residue

Sample	<sup>204</sup> Pb 10 <sup>-12</sup> mole/g	Terrestrial <sup>204</sup> Pb (%)	<sup>238</sup> U/ <sup>204</sup> Pb	<sup>232</sup> Th/ <sup>204</sup> Pb	<sup>206</sup> Pb/ <sup>204</sup> Pb	<sup>207</sup> Pb/ <sup>204</sup> Pb	<sup>208</sup> Pb/ <sup>204</sup> Pb	<sup>207</sup> Pb/ <sup>206</sup> Pb	$^{238}U/^{206}Pb^{*}$	<sup>207</sup> Pb*/ <sup>206</sup> Pb*
Total data <sup>††</sup>										
PX1	23.4		35.43 (25)	154.6 (13)	43.99 (20)	26.043 (79)	65.95 (20)			
PX2	34.9		24.25 (15)	102.75 (75)	36.25 (13)	23.058 (53)	57.65 (13)			
PL	36.4		11.79 (77)	44.5 (31)	24.95 (49)	18.53 (12)	45.21 (28)			
Fines	122		7.284 (39)	28.16 (19)	22.71 (13)	17.39 (10)	42.63 (25)			
Contamination corrected										
<b>PX</b> 1	13.3	43.0	62.15 (44)	271.1 (22)	63.06 (35)	33.90 (14)	86.56 (35)	0.5376 (37)	1.156 (11)	0.4392 (38)
PX2	13.4	61.5	62.99 (38)	266.9 (19)	64.27 (33)	34.93 (14)	88.03 (35)	0.5434 (35)	1.1460 (97)	0.4481 (37)
PL	18.2	50.0	23.6 (15)	89.0 (62)	31.19 (98)	21.43 (23)	51.79 (56)	0.687 (23)	1.077 (77)	0.509 (27)
Fines	34.2	72.0	26.01 (14)	100.57 (68)	33.00 (47)	21.90 (37)	52.91 (89)	0.664 (15)	1.098 (56)	0.490 (24)

Table 2. Contamination correction for the total U-Th-Pb data<sup>†</sup>.

<sup>†</sup> The numbers in the parentheses are  $2\sigma$  errors for the last degits, which include the errors corresponding to calculation of total data and contamination correction.

<sup>††</sup> Sum of residue, 0.1 N HBr leach, and 0.01 N HBr leach.

\* Radiogenic Pb, which were corrected for initial Pb composition using CDT Pb composition.

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may have been enriched with a high- $\mu$  mineral such as ilmenite which may be resistant to leaching. We believe the latter is more probable, because this meteorite actually contains ilmenite (YANAI and KOJIMA, 1991). The  $\mu$  values obtained from this contamination-correction method seem to be reasonable. For the corrected total data, the  $\mu$  values of PX1 and PX2 are similar and higher than PL (usual for pyroxene and plagioclase in the same rock), whereas the  $\mu$  value for the PL-residue is higher than that for the PX2-residue. The whole rock  $\mu$  value may be somewhere around 40, which is similar to that for A-881757 (MISAWA *et al.*, 1992) and Luna 24170 (UNRUH and TATSUMOTO, 1978), but far lower than those previously estimated for mare basalts



Fig. 6a.  ${}^{238}U/{}^{204}Pb$  vs.  ${}^{206}Pb/{}^{204}Pb$  diagram using contamination-corrected data. Open symbols are contamination-corrected total U-Pb data and filled symbols are uncorrected data. The age obtained using contamination-corrected total data is  $3928 \pm 54$  Ma with an initial  ${}^{206}Pb/{}^{204}Pb = 11.2 \pm 0.8$ .



Fig. 6b.  ${}^{235}U/{}^{204}Pb vs. {}^{207}Pb/{}^{204}Pb$  diagram using contamination-corrected data. Symbols are the same as those appeared in Fig. 6a. The age obtained using the corrected total data is  $3930 \pm 220 \text{ Ma}$  with initial  ${}^{207}Pb/{}^{204}Pb = 13.1 \pm 4.3$ .



Fig. 6c.  ${}^{232}Th/{}^{204}Pb$  vs.  ${}^{208}Pb/{}^{204}Pb$  diagram using contamination-corrected data. Symbols are the same as those appeared in Fig. 6a. The age obtained using the corrected total data is  $3730 \pm 780$  Ma with an initial  ${}^{208}Pb/{}^{204}Pb = 33 \pm 11$ .



Fig. 7. <sup>204</sup>Pb/<sup>206</sup>Pb vs. <sup>207</sup>Pb/<sup>206</sup>Pb diagram using contamination-corrected data. In this figure, all the residues (large filled symbols) and the most contaminated fraction, PL-wash (small filled circle), are plotted for comparison. The best fit line for the contamination-corrected data, corresponds to a Pb-Pb age of 3930±240 Ma. Note that contaminated fractions, PL-wash and Fines-residue (filled triangle), are off the line, but the deviation of these data is small compared to the uncertainty in the estimate of the best fit line. Also, another contaminated fraction, PX2-residue, lies on the line. Pb contamination in these samples does not cause a large deviation from the best fit line, because the modern terrestrial Pb isotopic composition (cross marked, MT) is also very close to the line.

(between 300 and 1000; TATSUMOTO *et al.*, 1977), indicating the source of Y-793169 was less enriched than those of most mare basalts.

Using these contamination-corrected data, the U-Pb, Th-Pb isochron ages were recalculated, and the results are shown in Figs. 6a, b, and c. These ages are all close to 3.9 Ga, similar to the lower-intercept age of the lunar cataclysm array. The Th-Pb

data are more scattered than the U-Pb data and the calculated Th-Pb age is consequently somewhat younger but within error of the U-Pb isochron ages, perhaps supporting the validity of our contamination-correction procedure. The initial Pb isotopic composition obtained from these isochrons is  ${}^{206}$ Pb/ ${}^{204}$ Pb = 11.2 ± 0.8,  ${}^{207}$ Pb/ ${}^{204}$ Pb = 13.1 ± 4.3, and  ${}^{208}$ Pb/ ${}^{204}$ Pb = 33 ± 11. These values are much lower than those obtained from a number of lunar rocks, and closer to the CDT Pb composition, which defines the lowest values observed in the solar system. They indicate that this meteorite was formed from a source magma with a  $\mu$  value significantly lower than to most Apollo mare basalts. A low  $\mu$ source is also indicated by the contamination-corrected  ${}^{204}$ Pb/ ${}^{206}$ Pb- ${}^{207}$ Pb/ ${}^{206}$ Pb data (Fig. 7). The Pb-Pb age calculated from these data is  $3930 \pm 240$  Ma, consistent with the U-Th-Pb isochron ages using the same data set. The least contaminated residues, PX1 and PL, that are coincident with the lunar cataclysm array, also lie on this isochron.

Another approach to correcting the data would be to assume that the HBr leaching removed some indigenous U and Th while leaching out Pb contamination. When we combine the leached U with that in the residues and calculate using only the Pb in residues (TATSUMOTO and PREMO, 1991), then the U-Pb data of the separates plot inside the concordia curve (except Fines). The CDT-corrected data do not produce a reasonable concordia intercept age but they plot near 4.1 Ga. However, if we use initial Pb values calculated using  $\mu = 10$  at 3.92 Ga and assuming 4.56 Ga of the age of the Moon, the PX1 and PX2 data point plot around the chord connecting 3.92 with lower intercept close to 0 Ma (Fig. 8). This exercise suggests that Y-793169 formed around 3.9 Ga,



Fig. 8.  $^{238}U/^{206}Pb^*$  vs.  $^{207}Pb^*/^{206}Pb^*$  diagram showing the behavior of U (leach plus residue) vs. residue Pb data. Filled symbols are CDT initial Pb corrected data and open symbols are data corrected using evolved initial Pb values from sources with  $\mu = 10$  and  $\mu = 13$ , applying single-stage Pb evolution between 4.56 and 3.9 Ga. Except for the Fines fraction, all the data plot inside the concordia curve. Excess radiogenic Pb in the Fines fraction is the result of terrestrial Pb contamination. When using the initial Pb correction from a source with  $\mu = 13$ , the slope of the tie line between PX2, PX1, and PL is negative, indicating that this initial Pb correction is not reasonable. A more realistic source  $\mu$  value might be as low as 10. Data corrected using initial Pb values calculated from a source with  $\mu = 10$  correspond to an age of 3.9 Ga, although a more precise age could not be obtained.



Fig. 9. Pb-Pb data from separates of Y-793169 and their best fit line indicate a low model source  $\mu$ -value of ~ 10 (Pb-Pb data are the same as Fig. 7). Pb-Pb data from A-881757 (pyroxene, ilmenite and whole rock; MISAWA et al., 1992) also lie on the same line, indicating that these two lunar meteorites were formed at approximately the same time from source magmas that have similar  $\mu$ -values. The solid line indicates Pb isotopic compositions at 3.9 Ga for various  $\mu$ -values assuming a single-stage Pb evolution from a CDT Pb composition at 4.56 Ga. These data indicate that these lunar rocks were derived from an environment where the average  $\mu$ -value was quite low (~10) even over the period between 4.56 and 3.9 Ga. Two-stage Pb evolution models (e.g.,  $\mu$ =8 between 4.56 to 4.4 Ga, lunar primary differentiation age) yield source  $\mu$  values similar to single-stage calculations.

although the exact formation age is not uniquely defined by the U-Th-Pb isotopic systematics.

Using a Pb-Pb diagram, the source  $\mu$  value for this lunar meteorite is estimated by a single-stage Pb evolution model. In Fig. 9, the Pb isotopic composition of the source at 3.9 Ga is shown for various  $\mu$  values over the time period between 4.56 and 3.9 Ga. The Pb-Pb results indicate a single-stage  $\mu$ value of 10 (between 4 and 15) for the Y-793169 source. This value is much lower than the source  $\mu$ -value for most mare basalts (between 100 and 300; TATSUMOTO *et al.*, 1977). It is also lower than some of the lowest estimates of source  $\mu$  values for LVG, between 20 and 30 (TATSUMOTO *et al.*, 1973b; 1987; Tera and WASSERBURG, 1976), and anorthosite 60025, about 35 (HANAN and TILTON, 1987).

It is interesting that the U-Pb data of another lunar meteorite, A-881757 (MISAWA et al., 1992), shows a similar age and  $\mu$  value (Fig. 9), although U and Th abundances in A-881757 are lower than those in Y-793169. This observation suggests to us that these two lunar meteorites were derived from sources that had similar  $\mu$  values at nearly the same time. The age of 3.9 Ga obtained from these lunar meteorites is older than those of Apollo LT or VLT mare basalts (3.3–3.6 Ga, e.g., NYQUIST and SHIH, 1992). Even though the major- and trace-element compositions and mineralogy of these lunar meteorites are similar to LT or VLT mare basalts, their petrogenesis and site of

derivation on the Moon may be quite different. The older ages as well as low source  $\mu$ -values indicate that these two lunar meteorites represent more primitive lunar mantle-derived rocks.

During early differentiation of the lunar magma ocean, Pb was probably lost by volatilization from the magma ocean, scavenged by sulfides that sank to the lunar core, and further removed to the lunar crust by anorthosite flotation (*e.g.*, TATSUMOTO *et al.*, 1987), resulting in post-magma ocean, high- $\mu$ , mantle magma reservoirs. These scenarios are possible due to the fact that partitioning of U and Th into common mantle minerals is about five to ten times lower than Pb, and Pb partitioning into feldspar is large. Thus, we suggest that non-radiogenic Pb was preferentially transported into the deep, lunar interior (NUNES *et al.*, 1974; TERA and WASSERBURG, 1974, TATSUMOTO *et al.*, 1987, Ringwood, 1992). If Fe-Ni sulfides sank from a partially molten magma ocean, then  $\mu$  values for the deep lunar mantle from which VLT basalts, the A-881757 gabbro, and Y-793169 diabase may be generated, could have substantially lower  $\mu$  values than other higher level lunar plutonic rocks.

The low  $\mu$  value estimated in this study implies that the source(s) of two lunar basalts (A- 881757 and Y-793169) are either enriched in Pb or depleted in U relative to sources for Apollo mare basalts. By analogy with VLT basalts, we suggest that the source materials for the two meteorites are early-stage, mafic cumulates precipitated from a lunar magma ocean along with Fe-Ni sulfides. WARREN and KALLEMEYN (1991) and others argue from a geochemical standpoint that lunar meteorites most likely were derived from five or more different lunar craters and that VLT basalts should be more abundant in other areas than the Apollo and Luna landing sites. Their results indicate that the Moon may not have had a vast chemically homogeneous magma ocean, resulting in regional variations in  $\mu$  values. Alternatively, the initial  $\mu$  value of the bulk Moon may be lower than we have previously estimated, perhaps as low as that of the terrestrial mantle ( $\mu = 8$ , e.g., TATSUMOTO, 1978). Repeated differentiation caused by successive planetesimal bombardment may have increased  $\mu$  values around the Oceanus Procellarum—Fra Mauro region by progressive depletion of volatile elements.

### 4. Conclusions

The Pb isotopic composition of leached residues of mineral separates from Y-793169 are radiogenic compared to the near-terrestrial compositions of the leaches, indicating heavy terrestrial Pb contamination of this meteorite probably during its residence in the Antarctic ice. The Pb-Pb age obtained from three radiogenic residues is  $3916 \pm 90$  Ma. However, because of the severe terrestrial Pb contamination, it is difficult to discuss the U-Th-Pb systematics of this meteorite without using some assumptions.

Two approaches to the data are applied to estimate the contamination-free U-Pb components. Assuming that the contamination-free (original sample) U-Pb data must lie on the lunar cataclysm array on a concordia diagram, the total data were corrected for terrestrial Pb contamination. The amounts of <sup>204</sup>Pb assigned to the separates were large (between 40 and 70%). Using the contamination-corrected data and the least contaminated residues, PX1 and PL, the Pb-Pb age and U-Th-Pb ages were calculated. The results are as follows;  $3930\pm240$  Ma for <sup>207</sup>Pb-<sup>206</sup>Pb age,  $3928\pm54$  Ma for

<sup>238</sup>U-<sup>206</sup>Pb age,  $3930 \pm 220$  Ma for <sup>235</sup>U-<sup>207</sup>Pb age, and  $3730 \pm 780$  Ma for <sup>232</sup>Th-<sup>208</sup>Pb age. These age results are consistent with each other and the same as the lower-intercept age of the lunar cataclysm array, supporting the validity of our assumption regarding the contamination of this meteorite. Since the <sup>207</sup>Pb-<sup>206</sup>Pb age did not change significantly by the contamination correction, we think this age,  $3930 \pm 240$  Ma, represents the formation age of this meteorite. The initial Pb isotopic compositions of Y-793169 are <sup>206</sup>Pb/<sup>204</sup>Pb=11.2±0.8, <sup>207</sup>Pb/<sup>204</sup>Pb=13.1±4.3, and <sup>208</sup>Pb/<sup>204</sup>Pb=33±11. These values correspond to source  $\mu$ -value at 3.9 Ga of 10, by applying a single-stage Pb evolution model from CDT initial Pb compositions at 4.56 Ga.

As applied to other lunar U-Pb data (TATSUMOTO and PREMO, 1991), the other approach, which assumes HBr leaching removes indigenous U and Th as well as Pb contamination, yields U-Pb data indicating a formation age of ~3.9 Ga from a source with  $\mu = 10$ . Although a more precise age could not be defined by the U-Th-Pb method, these results indicate that the deep lunar mantle may have  $\mu$ -values as low as that of Earth's mantle. If LT and VLT basalts are the dominant type of lunar basalt in lunar areas other than the Apollo sites (WARREN and KALLEMEYN, 1991), then  $\mu$  of the bulk Moon may be considerably lower than previously estimated.

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