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U-Th-Pb ISOTOPIC SYSTEMATICS OF LUNAR METEORITE ASUKA-31

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Abstract: U-Th-Pb isotopic systematics indicate that Asuka-31 is of lunar origin and was formed 3940±8 Ma. The Pb isotopic composition is extremely nonradiogenic compared with those of typical Apollo mare basalts. The Pb-Pb, U-Pb, and Th-Pb ages are concordant at 3.94 Ga. The U-Pb data from maskelynitized plagioclase does not plot on the internal isochrons defined by other mineral separates, indicating that it was disturbed by a later shock event(s). The U-Th-Pb systematics of Asuka-31, combined with previous results, suggest a scenario for the early Pb isotope evolution of the Moon. According to the currently popular hypothesis for lunar origin, following a collision between the Earth and another large planetesimal, the Moon was formed mainly from the Earth's mantle and partially from the other impactor. The primary $^{238}U/^{204}Pb$ (μ) value of the Moon had been increased four to five times that of the Earth's mantle value (~8) by volatilization of Pb during the impact. Further depletion of Pb within the Moon's mantle is believed to have occurred during lunar core formation. The chalcophile behavior of Pb and large partition coefficient of Pb in silicate minerals compared to those of U and Th helped to decrease μ values of early cumulates that formed from the magma ocean and settled in the deep lunar mantle. The μ values of later cumulates gradually increased as a result of extensive fractionation. We suggest that Asuka-31 originated from partial melting of early cumulates enriched with sulfides.

1. Introduction

Eleven meteorites collected from Antarctica have so far been identified as lunar in origin (*e.g.*, YANAI and KOJIMA, 1991). Their petrology and chemistry are not totally equivalent with those of the Apollo and Luna samples, and, thus, lunar meteorites provide information on the chemistry of lunar regions unsampled during the Apollo and Luna missions. For example, the dominance of very-low-Ti (VLT) basalts in the lunar meteorites contrasts with the scarcity of these types of rocks among lunar samples (LINDSTROM *et al.*, 1991b; WARREN and KALLEMEYN, 1991).

Antarctic meteorite Asuka-31 (tentative name) was collected by the Japanese Antarctic Research Expedition party in 1988. Asuka-31 is a new type of lunar meteorite: an unbrecciated, coarse-grained gabbro that is similar to VLT basalts in bulk chemistry and mineral compositions (YANAI, 1991). Abundances of REE in Asuka-31 show a LREE-depleted pattern typically observed in VLT mare basalts, suggesting that there is no KREEP component (LINDSTROM *et al.*, 1991a).

As part of a consortium study of lunar meteorites, we have undertaken the

U-Th-Pb isotopic analysis of the Asuka-31 meteorite. We report here the U-Th-Pb systematics and age of the meteorite and discuss the genetic implications of this unique lunar gabbro.

2. Experimental Procedures

YANAI (1991) presented petrographical and mineralogical studies which showed that Asuka-31 is composed of pyroxene (59%), plagioclase (30%), ilmenite (6%), and other phases including troilite, olivine, ulvöspinel, Fe-Ni metal, apatite, and silica. He also observed shock effects in some minerals: plagioclase is completely maskelynitized, and pyroxene shows undulatory extinction.

Asuka-31,95 (1.605 g) allocated from the National Institute of Polar Research was composed of several interior fragments including one that had a sawed surface. The mineral separation procedures are schematically illustrated in Fig. 1. The sample was gently crushed with a stainless steel mortar and divided into four sized fractions (<63, 63–150, 150–300, and >300 μm) using nylon cloth sieves. Three separates (plagioclase, pyroxene, and ilmenite: IL) were handpicked from the >300 μm fraction (ilmenite and pyroxene grains were very fragile and disintegrated easily into small pieces). The plagioclase and pyroxene fractions contained poly-mineralic grains and were crushed and sieved further. Plagioclase (PL1) was handpicked from the 150–300 μm fraction. Plagioclase (PL2) and pyroxene (PX1) fractions were obtained from the first 150–300 μm fractions using heavy liquids (bromoform and methylene iodide). The PL2 fraction was a bromoform float (ρ<2.85) and PX1 was a methylene iodide sink (ρ>3.3). Three separates (plagioclase: PL3, pyroxene-rich phase: PX2, and magnetic phase: MAG) were obtained from 63–150 μm fractions using heavy liquids and a Frantz isodynamic separator equipped

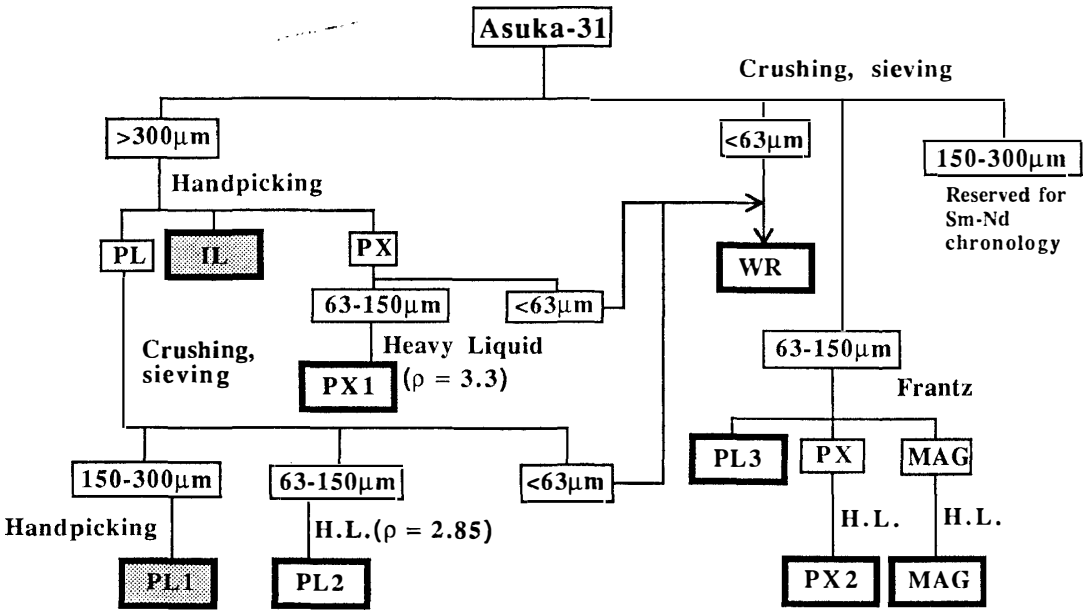

Fig. 1. Sample preparation procedure.

with an ethanol-filled chute. The MAG fraction was the most magnetic fraction from the magnetic separator and consisted of troilite and other Fe-rich components. The finest fraction ($<63\ \mu$m) was used as a whole-rock sample (WR). All samples separated using heavy liquids (PX1, PX2, PL2, and MAG) were rinsed several times with distilled acetone.

The seven separates and whole-rock sample (heavy box borders in the separation scheme in Fig. 1) were first washed twice with acetone and then five times with ethanol in an ultrasonic bath. Then, a cold 1*N* HF leaching was performed on two handpicked separates, PL1 and IL (shown by shaded boxes in Fig. 1), in order to dissolve adhering material from the grain surfaces. The other separates (PX1, PX2, PL2, PL3, MAG, and WR) were leached twice in cold 0.01*N* HBr, rinsed with distilled water, leached twice in cold 0.1*N* HBr, and finally rinsed with distilled water. The washing and leaching were done in an ultrasonic bath for 10 min each.

Washes, leaches, and residues were analyzed for U, Th, and Pb isotopes. Chemical procedures for separation of U, Th, and Pb were described in PREMO and TATSUMOTO (1991a, b). Analytical total Pb blanks were 34, 26, 95, and 43 pg for the fractions of acetone and ethanol wash, 0.01*N* HBr and 1*N* HF leaches, 0.1*N* HBr leaches, and residues, respectively. The contribution of the analytical Pb blank to total Pb in the residues was less than 1%, except for the residues of PL1 (2.9%) and PL3 (12%). However, the Pb blank contribution for some washes and leaches was more significant, especially the contribution for the 0.1*N* HBr leach for PL3 because of the low Pb abundance in the fractions. Blanks for U and Th were lower than 1.5 and 0.75 pg, respectively, for all fractions and amounted to less than 1% of the U and Th in most of the samples.

3. Results

Results of U, Th, and Pb analyses are presented in Tables 1 and 2. Pb isotopic compositions in all fractions were corrected for analytical blank ($^{206}Pb/^{204}Pb = 18.80 \pm 0.6$, $^{207}Pb/^{204}Pb = 15.65 \pm 0.2$, $^{208}Pb/^{204}Pb = 38.65 \pm 0.75$; PREMO *et al.*, 1989), mass fractionation (0.15 ± 0.02% per a.m.u.), and spike contribution (LUDWIG, 1989).

3.1. Leaching procedures

In order to remove any terrestrial contamination and to strip the grain surface of any adsorbed Pb component, we carried out acid leach experiments on all separates. As previously mentioned, attaining primary crystalline ages of lunar samples has been plagued by a variety of factors, including brecciation, metamorphism, secondary Pb adsorption, meteoritic contamination, and terrestrial contamination (TATSUMOTO *et al.*, 1987; PREMO and TATSUMOTO, 1991a, b). More importantly, a major problem with U-Th-Pb chronology of lunar samples is the failure to measure precise initial isotopic compositions.

Contamination of lunar samples can occur by addition of lunar labile Pb and/or meteoritic Pb by meteorite impact on the lunar surface and by terrestrial Pb con-

Table 1. U-Th-Pb analytical data for the Asuka-31 meteorite.

Sample	Weight* (mg)	^{238}U (pmol/g)	^{232}Th (pmol/g)	^{204}Pb (pmol/g)
Acetone & Ethanol washes				
PL1	112.90	0.7604	2.776	0.7406
PX1	93.54	2.629	12.80	0.9426
WR	98.52	4.533	21.48	0.4767
IL	27.11	3.213	7.673	0.0502
PL2	121.14	1.478	4.272	13.07
PX2	123.42	3.180	14.28	1.336
PL3	12.68	0.5611	1.949	1.050
MAG	29.68	11.03	26.77	6.225
0.01*N* HBr (1*N* HF) leaches#				
PL1	112.90	1.134	1.170	0.0859
PX1	93.54	35.84	62.42	6.301
WR	98.52	36.77	71.77	1.203
IL	27.11	116.5	29.22	0.2374
PL2	121.14	12.94	40.35	11.29
PX2	123.42	104.8	172.2	21.47
PL3	12.68	31.81	113.0	1.325
MAG	29.68	207.7	539.5	34.86
0.1*N* HBr leaches				
PX1	112.90	13.29	85.77	1.510
WR	98.52	76.53	215.6	0.8251
PL2	121.14	12.83	62.15	2.710
PX2	123.42	16.82	88.78	1.797
PL3	12.68	2.814	24.83	0.1343
MAG	29.68	221.9	1011	13.23
Residues				
PL1	110.99	12.41	62.83	0.4524
PX1	89.97	6.348	325.2	1.469
WR	95.09	262.5	1244	2.746
IL	24.14	573.6	2540	1.129
PL2	116.57	57.71	303.8	2.864
PX2	121.82	166.5	983.9	8.909
PL3	11.31	18.87	108.2	1.270
MAG	28.38	331.2	1544	20.92

* Th, and Pb concentration of washes and leaches are shown against the starting weight.

\# PL1 and IL were leached in 1*N* HF.

tamination. For Antarctic meteorites, the terrestrial contamination can be incurred both from the Antarctic ice and laboratory handling, including the analytical blank. In order to eliminate the contamination, we employed stepwise washing and leaching procedures. In several previous meteorite and lunar sample studies, it has been shown that stepwise leaching can eliminate contamination (*e.g.*, BROUXEL and TATSUMOTO, 1990; PREMO and TATSUMOTO, 1991a, b). However, a shortcoming of this procedure is that leaching also quite possibly extracts indigenous elements along with contamination. Thus, preferential elemental extraction of indigenous portions may occur during leaching, and chronologic interpretations using parent-

Table 2. Isotope data for the Asuka-31 meteorite.

Sample	Weight (mg)	$^{206}Pb/^{204}Pb$‡	$^{207}Pb/^{204}Pb$‡	$^{208}Pb/^{204}Pb$‡	$^{238}U/^{204}Pb$‡	$^{232}Th/^{204}Pb$‡	$^{206}Pb/^{238}U$‡¶	$^{207}Pb/^{235}U$‡¶	$^{208}Pb/^{232}Th$‡¶
Acetone & Ethanol washes									
PL1	112.90	18.637	15.954	38.414	1.027	3.749	9.087	760.0	2.385
		(0.0860) §	(0.0815)	(0.113)	(2.57)	(2.38)	(2.56)	(2.52)	(2.45)
PX1	93.54	20.845	16.529	41.507	2.789	13.58	4.137	308.3	0.8862
		(0.252)	(0.234)	(0.266)	(2.26)	(2.27)	(2.06)	(2.15)	(2.15)
WR	98.52	26.035	18.881	47.009	9.510	45.07	1.759	124.5	0.3890
		(0.863)	(0.554)	(0.589)	(3.61)	(3.62)	(2.67)	(2.81)	(2.68)
IL	27.11	58.7	31.4	67.0	64	150	0.772	45.6	0.246
		(76)	(56)	(47)	(130)	(130)	(45)	(50.9)	(50.2)
PL2	121.14	18.879	15.706	38.874	0.1131	0.3270	84.63	6598	28.74
		(0.552)	(0.0715)	(0.0892)	(0.669)	(1.15)	(0.672)	(0.687)	(1.19)
PX2	123.42	20.508	16.423	40.689	2.380	10.69	4.707	355.2	1.049
		(0.0994)	(0.0837)	(0.131)	(1.20)	(1.22)	(1.11)	(1.14)	(1.19)
PL3	12.68	19.187	15.971	39.003	0.534	1.86	18.5	1460	5.13
		(0.564)	(0.331)	(0.469)	(17.7)	(15.4)	(17.3)	(17.3)	(15.2)
MAG	29.68	18.998	15.930	38.908	1.772	4.300	5.470	438.6	2.194
		(0.752)	(0.0858)	(0.111)	(1.78)	(3.59)	(1.77)	(1.77)	(3.61)
0.01*N* HBr (1*N* HF) leaches#									
PL1	112.90	24.15	18.83	45.06	13.2	13.62	1.12	89.1	1.14
		(2.50)	(1.83)	(1.59)	(14)	(14)	(11)	(11)	(11)
PX1	93.54	20.946	16.595	41.260	5.688	9.907	2.046	152.7	1.189
		(0.0598)	(0.0727)	(0.112)	(0.380)	(0.557)	(0.367)	(0.391)	(0.642)
WR	98.52	32.057	21.272	53.639	30.56	59.64	0.7445	49.53	0.4052
		(0.373)	(0.316)	(0.271)	(1.06)	(1.13)	(0.695)	(0.832)	(0.817)
IL	27.11	140	65	140	491	123	0.261	15.3	0.890
		(14)	(12)	(12)	(16)	(16)	(2.50)	(2.73)	(2.64)
PL2	121.14	19.585	15.920	39.679	1.145	3.573	8.973	685.9	2.856
		(0.258)	(0.263)	(0.269)	(0.509)	(0.629)	(0.499)	(0.654)	(0.979)
PX2	123.42	21.657	16.798	42.082	4.881	8.022	2.530	183.7	1.571
		(0.207)	(0.214)	(0.219)	(0.726)	(0.827)	(0.714)	(0.779)	(0.956)
PL3	12.68	37.93	23.42	57.05	24.003	85.270	1.192	75.37	0.3234
		(3.13)	(2.04)	(1.99)	(0.698)	(0.697)	(3.73)	(4.05)	(3.76)
MAG	29.68	22.221	17.144	42.357	5.9569	15.470	2.168	158.6	0.8325
		(0.146)	(0.166)	(0.166)	(0.442)	(0.540)	(0.428)	(0.506)	(0.657)
0.1*N* HBr leaches									
PX1	112.90	19.169	49.328	49.328	8.799	56.79	2.203	139.1	0.3496
		(0.835)	(0.492)	(0.588)	(3.25)	(3.27)	(2.32)	(2.46)	(2.27)
WR	98.52	85.7	42.7	111	92.75	261.3	0.8239	48.13	0.3116
		(3.58)	(2.89)	(2.98)	(4.71)	(4.72)	(1.22)	(1.33)	(1.23)
PL2	121.14	22.831	17.285	43.962	4.733	22.93	2.857	203.7	0.6318
		(0.236)	(0.159)	(0.187)	(1.50)	(1.55)	(1.25)	(1.32)	(1.31)
PX2	123.42	27.214	19.108	48.405	9.360	49.40	1.913	129.8	0.3832
		(0.539)	(0.318)	(0.358)	(2.15)	(2.10)	(1.59)	(1.68)	(1.51)
PL3	12.68	40	27	67	21	180	1.5	110	0.20
		(230)	(180)	(180)	(520)	(520)	(230)	(230)	(200)
MAG	29.68	31.660	21.111	52.872	16.78	76.46	1.333	88.91	0.3060
		(0.415)	(0.289)	(0.303)	(1.15)	(1.22)	(0.774)	(0.804)	(0.871)
Residues									
PL1	110.99	42.621	32.875	66.731	27.44	138.9	1.214	113.5	0.2683
		(0.188)	(0.175)	(0.141)	(3.68)	(3.72)	(1.78)	(1.70)	(1.80)
PX1	89.97	61.133	34.014	86.113	43.23	221.4	1.1990	75.663	0.25579
		(0.865)	(0.674)	(0.697)	(1.39)	(1.45)	(0.660)	(0.684)	(0.793)
WR	95.09	105.24	53.019	130.59	95.568	452.90	1.0038	61.640	0.22326
		(0.526)	(0.455)	(0.458)	(0.735)	(0.819)	(0.373)	(0.378)	(0.523)
IL	24.14	507.4	216.0	506.9	508.1	2250	0.98033	55.824	0.21218
		(5.36)	(5.16)	(5.14)	(5.57)	(5.59)	(0.339)	(0.355)	(0.557)
PL2	116.57	36.708	25.156	60.942	20.150	106.10	1.3598	101.69	0.29664
		(0.308)	(0.272)	(0.272)	(0.666)	(0.771)	(0.445)	(0.470)	(0.616)
PX2	121.82	37.206	23.920	59.692	18.693	110.40	1.4925	100.50	0.27361
		(0.0902)	(0.0896)	(0.121)	(0.328)	(0.545)	(0.291)	(0.304)	(0.550)
PL3	11.31	32.70	25.90	56.86	14.9	85.2	1.574	144.8	0.3215
		(4.60)	(4.21)	(3.43)	(13)	(13)	(7.71)	(7.37)	(7.32)
MAG	28.38	33.080	22.207	53.568	15.829	73.790	1.5019	103.77	0.32651
		(0.135)	(0.118)	(0.124)	(0.427)	(0.612)	(0.340)	(0.360)	(0.574)

‡ Data corrected for mass fractionation, spike contribution, and laboratory blank (LUDWIG, 1989).
¶ Data corrected for the primordial Pb ($^{206}Pb/^{204}Pb$=9.307, $^{207}Pb/^{204}Pb$=10.294, $^{208}Pb/^{204}Pb$=29.476: TATSUMOTO *et al.*, 1973).
§ Error at 95% confidence limits (in percent).
# PL1 and IL were leached in 1*N* HF.

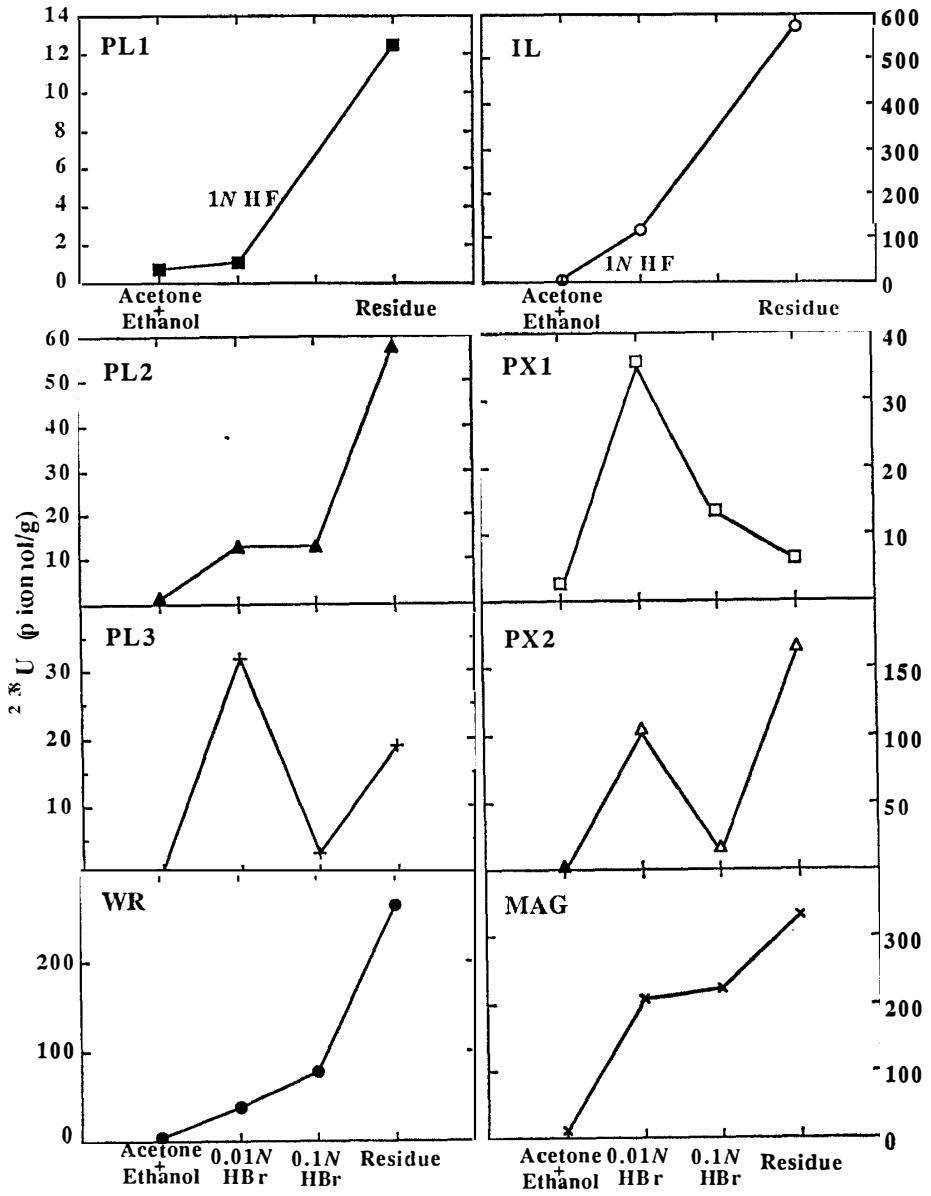

Fig. 2a. Abundances of ^{238}U in the eight fractions from Asuka-31 at different washing and leaching steps.

daughter isotope pairs must be made with great care.

U, Th, and Pb abundances for eight separates at each step of the washing and leaching procedures are shown in Figs. 2a, b, and c. In Fig. 3, $^{206}Pb/^{204}Pb$ ratios of washes, leaches, and residues for eight separates are shown as an index of terrestrial Pb removal. Significant amounts of Pb (20–40% of total Pb) were removed from plagioclase separates (PL1, PL2, and PL3) during the acetone and ethanol washing steps. Their $^{206}Pb/^{204}Pb$ values in the organic reagent washings are essentially terrestrial. Ethanol washes for the three PL fractions were slightly turbid, and their $^{206}Pb/^{204}Pb$ ratios were somewhat higher than the blank value. In the $^{207}Pb/^{206}Pb$ *versus* $^{204}Pb/^{206}Pb$ diagram (Fig. 4), the data points for leaches and

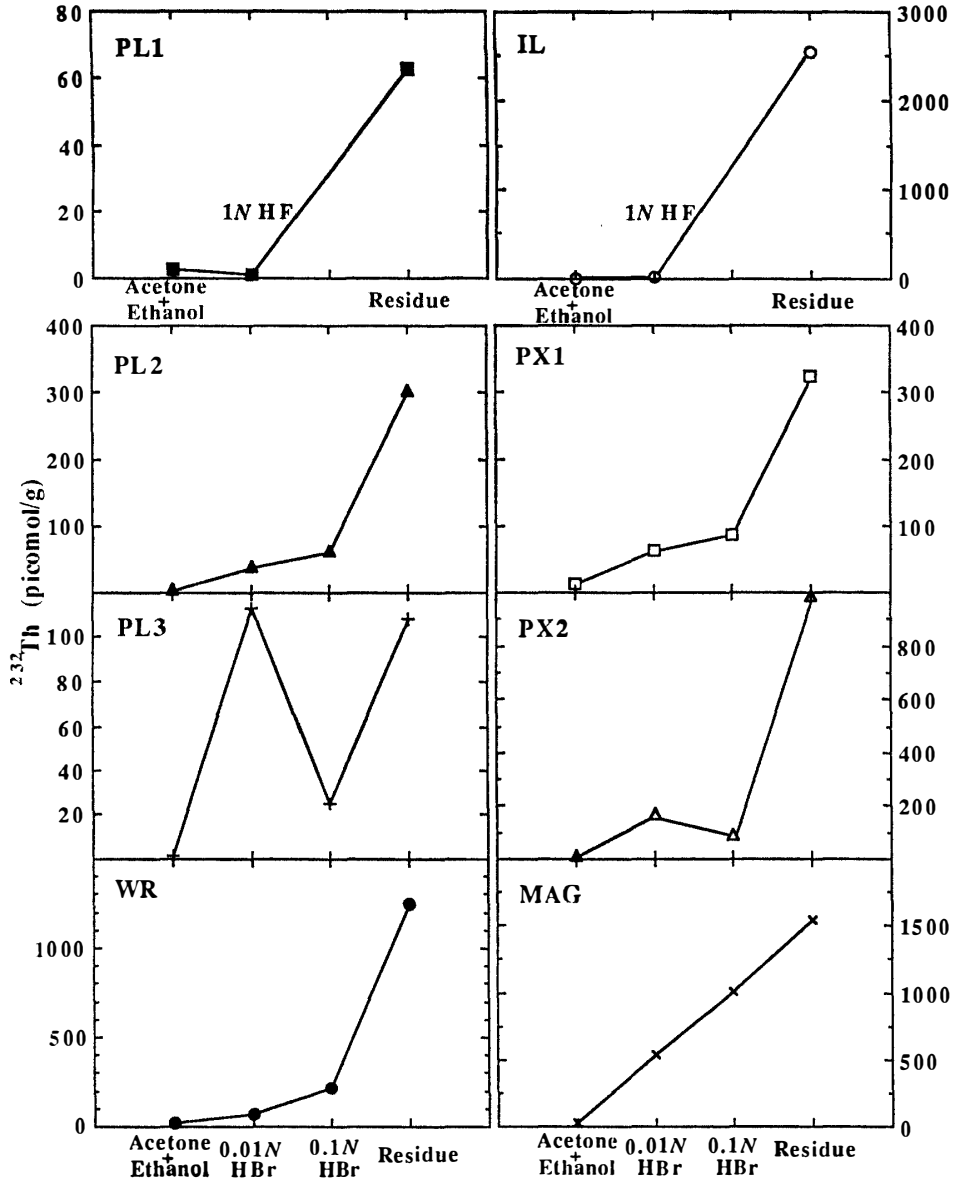

Fig. 2b. Abundances of ^{232}Th in the eight fractions from Asuka-31 at different washing and leaching steps.

residues of the three plagioclase fractions are not colinear between the blank and the residues, suggesting that Pb isotopic compositions of plagioclase separates are not two-component mixtures of the residues and blank Pb.

Except for PL3, the Pb in residues is more radiogenic than that in the washes and leaches. Approximately 35% of the Pb was leached during the 0.01*N* HBr treatment of PL2, which is less radiogenic than Pb in the 0.1*N* HBr leach and the residue. Pb in the 0.01*N* HBr leach of PL3, which is about 40% of the total Pb, is more radiogenic than Pb in the residue, indicating that this separate contains another U-rich component (phosphate or mesostasis phase ?) that easily dissolved in dilute HBr, or that secondary radiogenic Pb was adsorbed onto plagioclase grains

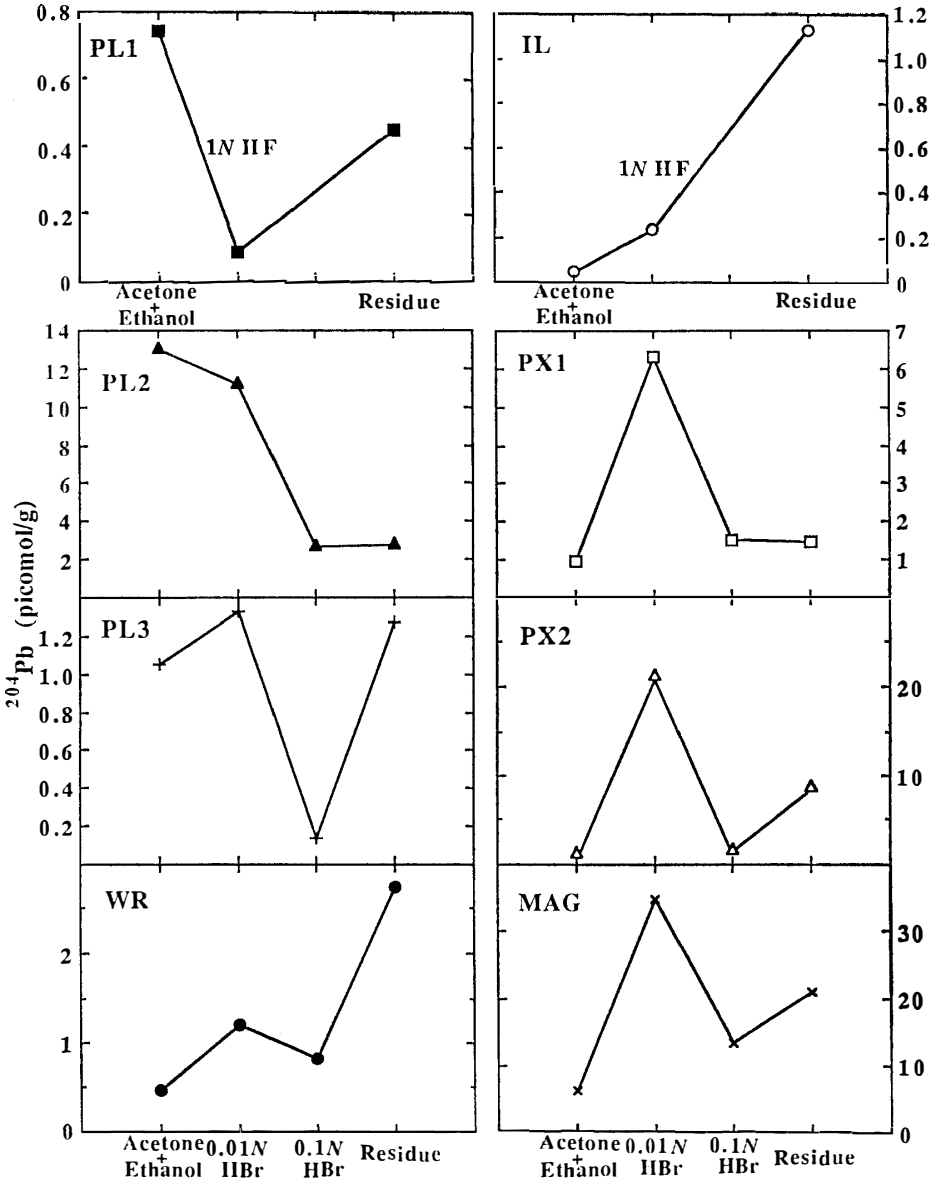

Fig. 2c. Abundances of ^{204}Pb in the eight fractions from Asuka-31 at different washing and leaching steps.

during impact events on the lunar surface. About 50% of U and Th of PL3 was also removed in 0.01*N* HBr.

3.2. Pb isotopes

All residues exhibit more radiogenic Pb-isotopic compositions than blank Pb; especially WR and IL, which were extremely radiogenic, with ^{206}Pb/^{204}Pb ratios of 105 and 507, respectively.

The Pb isotopic data from washes, leaches, and residues along with our laboratory blank Pb composition and primordial Pb composition (Cañon Diablo troilite: CDT; TATSUMOTO *et al.*, 1973) are shown in ^{207}Pb/^{206}Pb *versus* ^{204}Pb/^{206}Pb diagrams

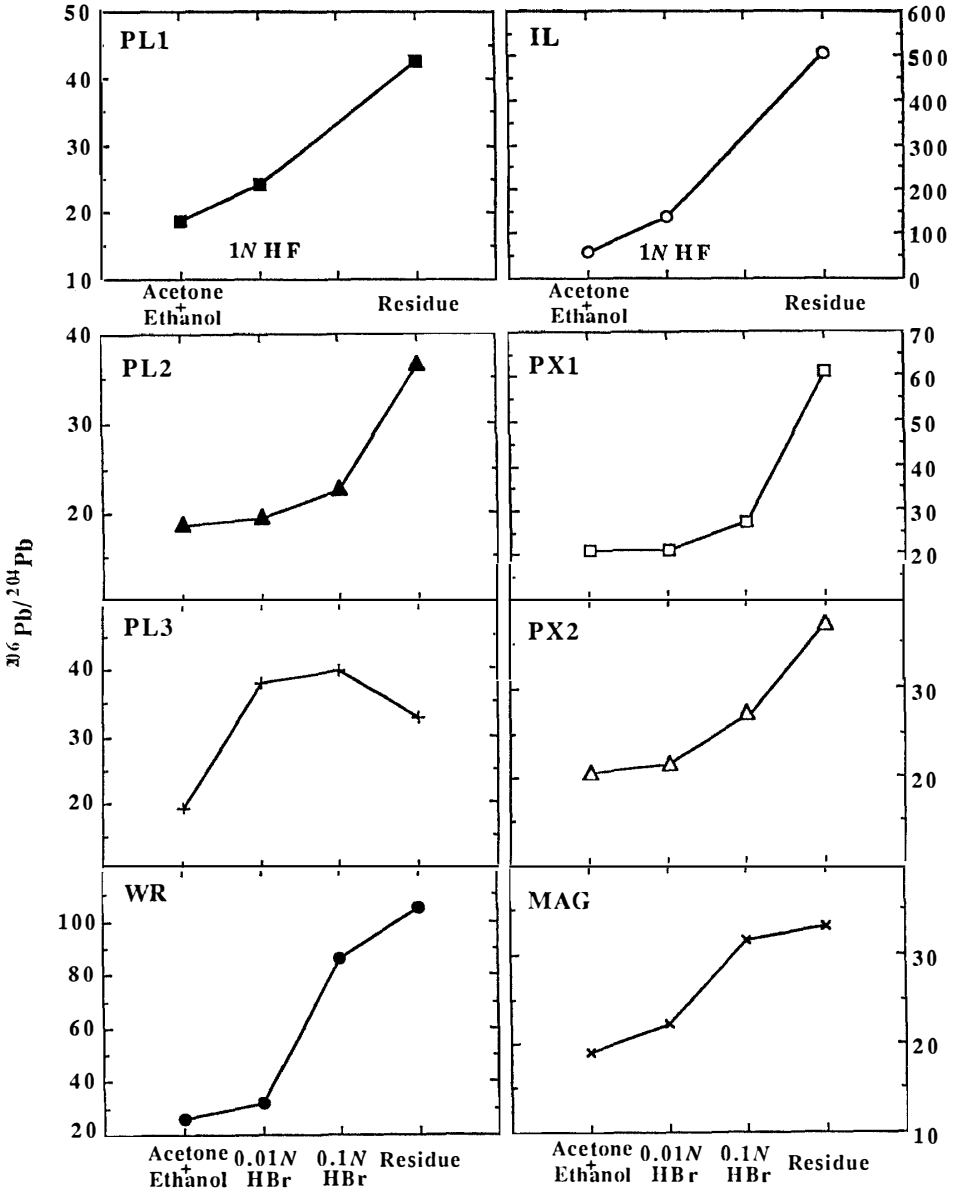

Fig. 3. Variations of $^{206}Pb/^{204}Pb$ values in the eight fractions of Asuka-31 at different washing and leaching steps. Pb isotopic compositions in acid residues are more radiogenic than those in washes and leaches except for PL3. The value of the 0.1N HBr leach for PL3 contains large experimental errors.

(Figs. 5a and b). The Pb data from all residues (eight fractions) define a Pb-Pb isochron age of 3930±52 Ma. Plagioclase and whole-rock fractions (PL1, PL2, PL3, and WR) do not plot on the line; this suggests open system behavior from a disturbance in the U-Pb system. Excluding these plagioclase and whole-rock fractions, the remaining four residues (PX1, PX2, IL, and MAG) yield a Pb-Pb isochron age of 3940±8 Ma. This isochron does not pass through the value of our laboratory Pb blank or CDT.

Whole-rock residue, however, does not plot in the data field of Apollo mare basalts. The Pb isotopic compositions of Asuka-31 are less radiogenic than those

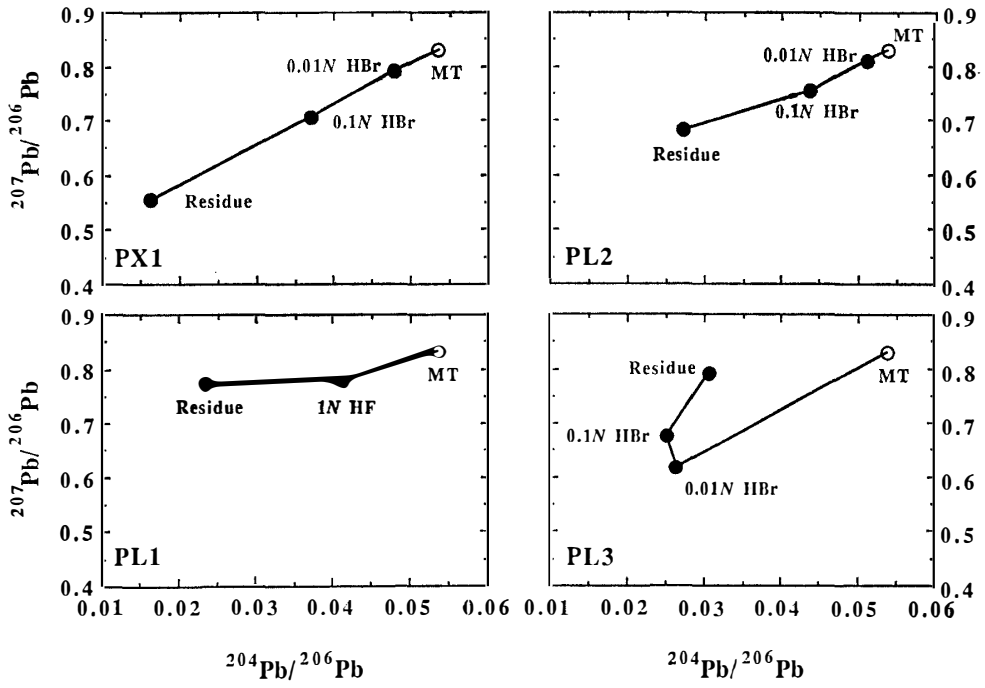

Fig. 4. Pb isotope correlation diagrams for plagioclase separates (PL1, PL2, and PL3) and one pyroxene separate (PX1). The data points for leaches and residues of the plagioclase fractions are not colinear between the blank and the residues. The Pb isotopic compositions for leaches of PX1 are two-component mixtures of blank Pb and radiogenic Pb of the residue. "MT": blank Pb.

of typical Apollo mare basalts, indicating that this lunar meteorite is a unique specimen (see the low μ value for Asuka-31 in Fig. 5b). The ^{206}Pb and ^{207}Pb are daughters of U (238 and 235, respectively), therefore, preferential elemental extraction during the leaching procedure does not affect the Pb-Pb isochron age. Furthermore, Pb's of PL and MAG residues are extremely non-radiogenic, and the isochron does not extend to blank (modern terrestrial) Pb. Thus, it appears that the isochron is not affected by secondary Pb or terrestrial Pb contamination and represents the true age.

On the other hand, if we combine the Pb removed during leaching with that in the residues, the four points yield a Pb-Pb isochron age of 3950±3 Ma. This age is similar to that for four residue fractions. The similarity indicates that acetone and ethanol washing were effective for removing the secondary Pb and that the contribution of terrestrial contamination in the leaches is small (refer to Fig. 2c), although Pb from the PL fractions is somewhat large. The age obtained here is similar to those of some lunar mare basalts and lunar basin-forming events at 3.8–4.0 Ga (Imbrium 3.87–3.90 Ga; Crisium 3.90 Ga; Humorum 3.92–3.95 Ga; Nectaris 3.98 Ga; and Serenitatis 3.97 Ga; JESSBERGER *et al.*, 1974) and indicates that the gabbro was formed from a magma related to a basin excavation event(s), as were lunar basalts. However, it must be emphasized that the gabbro is less radiogenic than Apollo mare basalts.

The deviation of the analyses of plagioclase residues from the isochron is

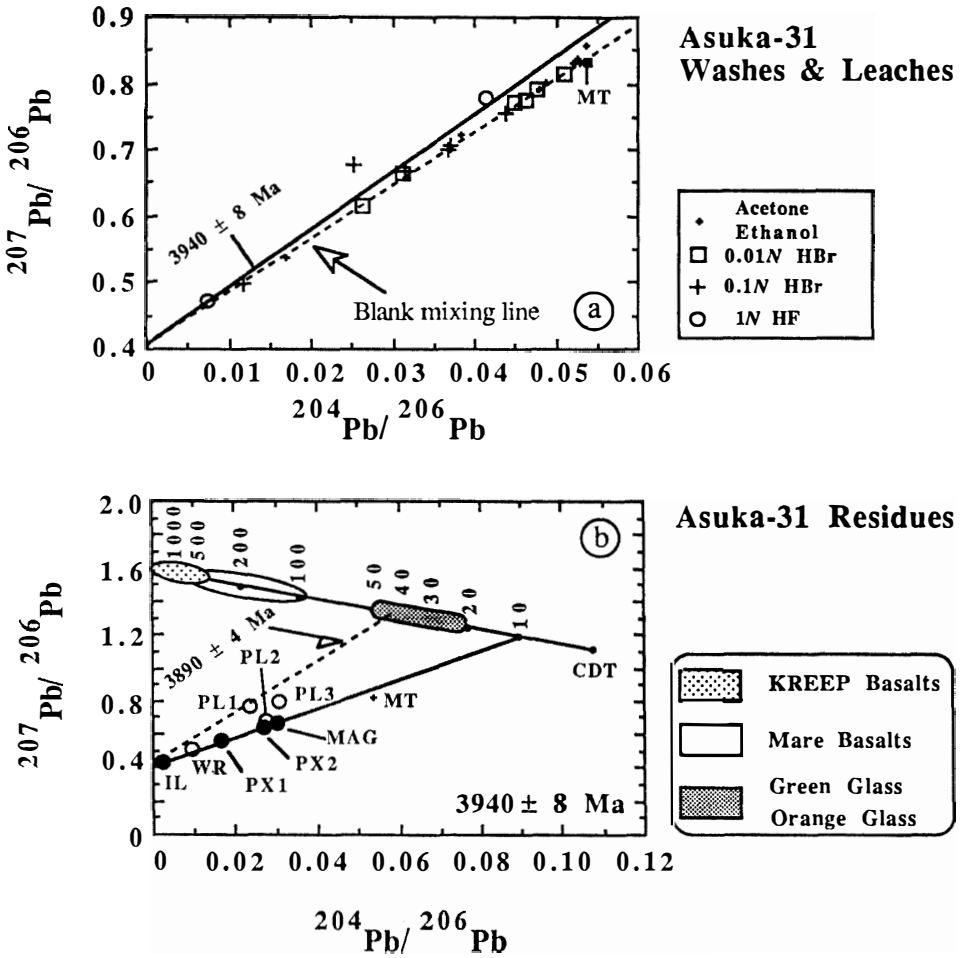

Fig. 5. Pb isotope correlation diagrams for samples from Asuka-31. (a) Acetone and ethanol washes, 0.01N HBr leaches, 1N HF leaches, and 0.1N HBr leaches. (b) Residues; "MT": laboratory blank; "CDT": Cañon Diablo troilite Pb (TATSUMOTO et al., 1973). All eight fractions yield an age 3930±52 Ma using regression treatments of LUDWIG (1991). Without three plagioclase fractions (PL1, PL2, and PL3) and a whole-rock (WR), four fractions (PX1, PX2, IL, and MAG) yield an age of 3940±8 Ma, which indicates a reference isochron. The tie line between PL1 (the most pure separate of plagioclase) and IL yields an age of 3890±4 Ma. The straight line across the top of the diagram represents all possible closed system Pb compositions evolving in a single stage between 4.56 and 3.94 Ga for various values of μ ($^{238}U/^{204}Pb$) assuming initial CDT Pb.

difficult to explain, but it is not caused by terrestrial Pb contamination because they all plot on the left side of the 3.94 Ga isochron and not on the right side, toward the blank Pb composition. Due to a significant amount of plagioclase, the data point for the residue of WR also deviates from the isochron. Among lunar plutonic rocks so far analyzed (norite 78235 and troctolite 76535), some maskelynite fractions lie on isochrons, and the Pb from them is interpreted as the least affected during shock events (PREMO and TATSUMOTO, 1991a, b). However, because only the maskelynite fractions deviate from our isochron, we favor a disturbance of the

U-Pb system in maskelynitized plagioclase during a shock event(s) or some later events. If PL fractions were affected by these events more than other fractions, then their $^{207}Pb/^{206}Pb$ ratios should be lower than other fractions since "old" Pb with a high $^{207}Pb/^{206}Pb$ value would have been lost during shock events. Because the PL fractions plot above the isochron, the only *ad hoc* reason conceivable is that an "old" Pb from older lunar highland rocks or soils was incorporated into plagioclase during maskelynitization. It is not conceivable that ^{206}Pb was preferentially leached out by weathering in Antarctica or laboratory procedures. The tie line between PL1 (the most pure separate of plagioclase) and IL yields a younger age of 3890±4 Ma. This age corresponds to the maximum age for a shock event, if we assume Pb loss. All but three data points of washes and leaches plot below the 3.94 Ga isochron, indicating that they are mixtures of the blank Pb and radiogenic Pb components (Fig. 5a). However, the values of the three points that are off the trend contain large measurement errors.

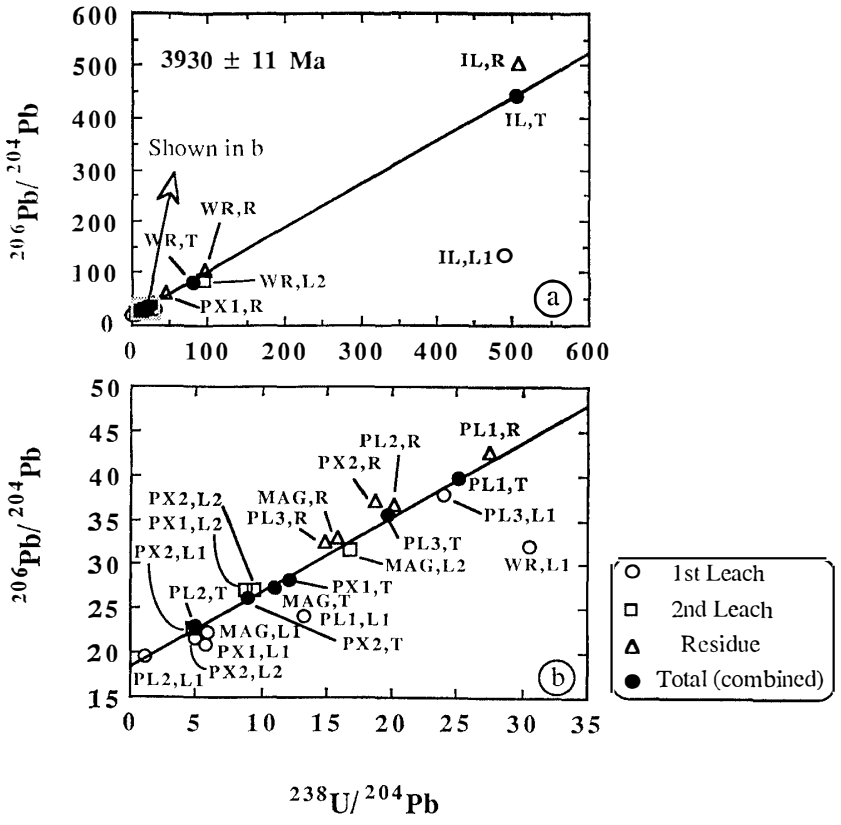

Fig. 6. *$^{206}Pb/^{204}Pb$ versus $^{238}U/^{204}Pb$ correlation diagram. The line through eight separates (calculated values from leaches plus residue) has a slope corresponding to a U-Pb age of 3930±11 Ma. First leaches from PL1, PX1, PX2, WR, IL, and MAG and second leach from WR do not plot on this line. Residues from PX1, PX2, WR, IL, and MAG also do not plot on the line. The line for the four residues (PX1, R; PX2, R; IL, R; and MAG, R) yields an age of 4340±38 Ma. "L1": first leach (0.01N HBr or 1N HF), "L2": second leach (0.1N HBr), "R": residue, "T"; calculated values from leaches plus residue.*

3.3. *U-Th-Pb systematics*

On a $^{206}Pb/^{204}Pb$ *versus* $^{238}U/^{204}Pb$ diagram (Fig. 6), the data points of the first (0.01*N* HBr or 1*N* HF) leaches plot on the U-rich side, suggesting that U was preferentially leached over Pb during this procedure. In contrast, the residue fractions plot on the U-deficient side. When the U and Pb in leaches (but not washes) are combined with those in the residues, data points of the calculated values define an isochron age of 3930±11 Ma and a y-intercept of 18.30±0.59. On a $^{208}Pb/^{204}Pb$ *versus* $^{232}Th/^{204}Pb$ diagram (Fig. 7), the data points of 0.01*N* HBr leach of IL and 0.1*N* HBr leach of WR plot on the Th-poor side, indicating preferential leaching of Pb over Th during this procedure. Similar to the U-Pb result, we obtain an isochron age of 3940±9 Ma and a y-intercept of 38.62±0.39 for the combined values of the leaches plus the residues. Within the error range, the U-Pb, Pb-Pb, and Th-Pb ages are concordant. As is the case with the Pb-Pb age, these U-Pb and Th-Pb ages are strongly governed by the most radiogenic separate, ilmenite (IL). On the other hand, the y-intercept values of U-Pb and Th-Pb plots ($^{206}Pb/^{204}Pb = 18.30\pm0.59$, $^{207}Pb/^{204}Pb = 17.04\pm2.28$, $^{208}Pb/^{204}Pb =$

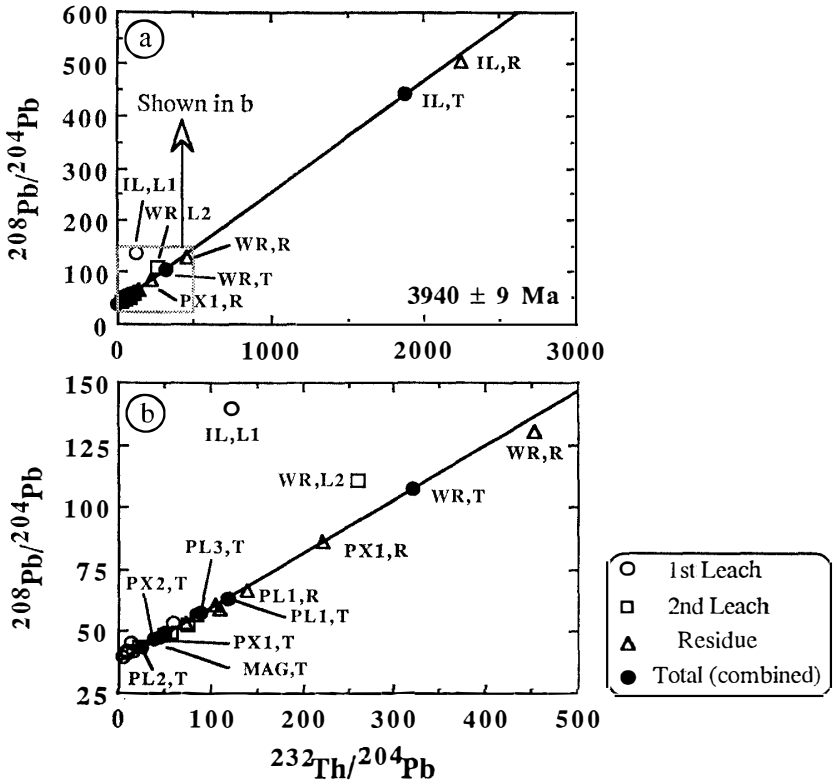

Fig. 7. *$^{208}Pb/^{204}Pb$ versus $^{232}Th/^{204}Pb$ correlation diagram. The line through eight separates (calculated values from leaches plus residue) has a slope corresponding to a Th-Pb age of 3940±9 Ma. First leach from IL is displaced far from the correlation line. The line for the four residues (PX1, R; PX2, R; IL, R, and MAG, R) yields an age of 3830±88 Ma. "L1": first leach (0.01N HBr or 1N HF), "L2": second leach (0.1N HBr), "R": residue, "T": calculated values from leaches plus residue.*

38.62±0.39) are probably influenced by the less radiogenic Pb of PX2 and terrestrial contamination because 0.01*N* HBr leach fractions presumably still contain some terrestrial Pb. Therefore, these y-intercept values may not be the proper initial Pb isotopic composition of this rock. However, the amount of contaminated Pb may be small compared with the indigenous Pb because most data points plot on the left side of the concordia in the U-Pb modified concordia diagram (see Fig. 8). If all U and Th in leaches and olny Pb in 0.1*N* HBr leaches (1*N* HF leaches for PL1 and IL) are combined with those in the residues (assuming that most Pb in the 0.01*N* HBr leach is contamination), the y-intercept values of U-Pb and Th-Pb plots become smaller ($^{206}Pb/^{204}Pb = 12.40 \pm 5.66$, $^{207}Pb/^{204}Pb = 15.35 \pm 3.19$, $^{208}Pb/^{204}Pb = 32.75 \pm 5.53$).

In Fig. 8, the relation of $^{238}U/^{206}Pb$ and radiogenic $^{207}Pb/^{206}Pb$ for the first (L1) and second (L2) leaches, residues (R), and the combined values (T) for leaches and residue of each fraction is shown on a modified concordia diagram (TERA and WASSERBURG, 1972). The data are corrected for initial Pb using CDT Pb composition. All but three (first and second leaches of WR and first leach of

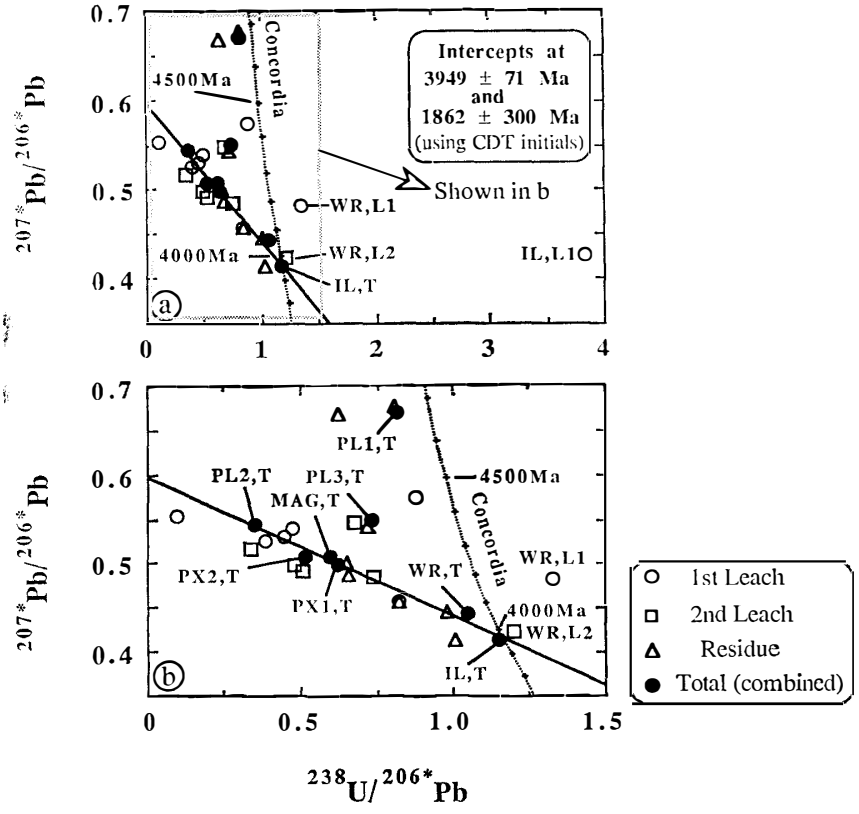

Fig. 8. *$^{207*}Pb/^{206*}Pb$ versus $^{238}U/^{206*}Pb$ modified concordia diagram after TERA and WASSERBURG (1972). Radiogenic Pb is plotted after correction for analytical blank and CDT Pb (TATSUMOTO et al., 1973) as primordial Pb isotopic compositions. A chord is drawn through the calculated values (leaches plus residue) of PX1, PX2, IL, and MAG. "L1": first leach (0.01N HBr or 1N HF), "L2": second leach (0.1N HBr), "R": residue, "T": calculated values from leaches plus residue.*

IL) plot on the left of the concordia curve, indicating an excess of radiogenic Pb. A chord through U-Pb data points for four residues (MAG, PX1, PX2, and IL) intercepts concordia at 2713 and 2545 Ma with large errors. The intercept values do not match with the Pb-Pb age, indicating that preferential elemental extraction disturbed the U-Pb system. A chord through four data points (PX1, T; PX2, T; IL, T; and MAG, T) intercepts concordia at 3949±71 and 1862±300 Ma. The chord is unlike those for typical mare basalts (the cataclysm array of TERA *et al.*, 1974), with an upper intercept on the concordia curve of ~4.4 Ga and a lower intercept of ~3.9 Ga. A shallow slope for the Asuka-31 chord is rather similar to that for the ferroan anorthosite 60025 (HANAN and TILTON, 1987).

The upper intercept age appears to be identical to the Pb-Pb age of 3940± 8 Ma. However, the CDT Pb values are not valid for the initial Pb correction of this sample, and the true initial Pb values must be more evolved. If we assume the age of the rock is 3940±8 Ma and a single stage Pb evolution from CDT Pb at 4.56 Ga as the age of the Moon, the most evolved initial Pb values are $^{206}Pb/^{204}Pb = 10.609$, $^{207}Pb/^{204}Pb = 12.364$, and $^{208}Pb/^{204}Pb = 30.493$ at 3.94 Ga. These values correspond to a source μ value of 7. *This estimated μ value is extremely low compared with those for lunar mare basalts.* Moreover, these initial Pb values are within the error range of those obtained from the y-intercepts of U-Pb and Th-Pb plots when all U and Th in leaches and Pb in 0.1*N* HBr (or 1*N* HF) leaches are combined with those in residues.

4. Discussion

4.1. The age of the Asuka-31 gabbro

As mentioned above, differential leaching of U, Th, and Pb in our procedures probably has altered the U/Pb and Th/Pb ratios in the residues. Thus, in the following discussion, we will first use the combined data of acid leaches plus residues of each fraction for calculation of U-Pb and Th-Pb isochron ages. Because a slight amount of terrestrial Pb may be included in the acid leaches, a second calculation is made by combining the U and Th in the leaches with those in the residues, and using only the residue Pb (assuming Pb in leaches is mostly contamination). The true solution must be between the two treatments. In either case, we will show that the combined U-Pb data show similar ages with the Pb-Pb residue age of 3940±8 Ma, which we believe is the age of Asuka-31. The concordance at 3.94 Ga among the different systems (U-Pb age of 3930±11 Ma and Th-Pb age of 3940± 9 Ma) with the Pb-Pb age strongly suggests that this is the formation age of the Asuka-31 gabbro.

Plagioclase separates, however, show an apparent disturbance in the U-Pb system. YANAI (1991) reported that plagioclase is completely maskelynitized and that pyroxene grains show cracks and exhibit undulatory extinction. The shock event(s) recorded in the meteorite may be responsible for the disturbance of the U-Th-Pb systems. U-Th-Pb systematics of lunar maskelynite did not appear to be disturbed in norite 78235 (PREMO and TATSUMOTO, 1991a). Based on U-Th-Pb systematics, CHEN and WASSERBURG (1986) also suggested that the shock process

that produced maskelynite in shergottites was not sufficient to erase primary igneous characteristics. The behavioral difference in maskelynite between previous and present studies is difficult to explain. However, we guess that, because the plagioclase data of the Asuka-31 plot in the older, 207-rich side of the isochron, an old Pb component has been incorporated into plagioclase during maskelynitization. This "old" Pb component could have originated from the impactor or from lunar-surface Pb.

4.2. *The source of the Asuka-31 gabbro*

As previously discussed (*e.g.*, NUNES *et al.*, 1975), the Pb isotopic composition at the time of primary lunar differentiation may have been more radiogenic than CDT Pb values. Thus, calculated U-Pb ages using the CDT Pb as the initial Pb are maxima.

Single stage evolution for the initial Pb — Using the Pb-Pb age of 3940±8 Ma obtained for four residues and assuming 4.56 Ga for the age of the Moon, we can estimate a μ value ($^{238}U/^{204}Pb$) for the source material of the Asuka-31 gabbro during the interval from 4.56 to 3.94 Ga ago. Using initial Pb values corresponding to $\mu=7$ yields a chord defining intersect ages of 3945±4 and 365±248 Ma (Fig. 9). Initial Pb values, calculated when μ is greater than 8, produce a negative lower intercept. As μ approaches a value of 17, the chord becomes flat. The upper intercept age is not sensitive to the choice of μ values because ilmenite plots close to concordia and its Pb is sufficiently radiogenic so that assuming different initial Pb values has little effect on its plot. The chord connecting the four data points (PX1, T; PX2, T; IL, T; and MAG, T) is, therefore, strongly controlled by the ilmenite data point. Assuming the initial Pb values of $\mu=7$ and using the combined value of residue plus leach U and residue Pb values, the four data points (PX1; PX2; IL; and MAG) plot on the right side (*i.e.*, inside) of the concordia curve, and a chord through these points intercepts concordia at 3938±673 Ma. Similarly, if we combine all U in leaches and residue, and Pb in 0.1*N* HBr (or 1*N* HF) leaches with residues, intersect ages are 3943 and 1744 Ma with large errors. In either case, it appears that the gabbro was derived from a source with a considerably low μ value.

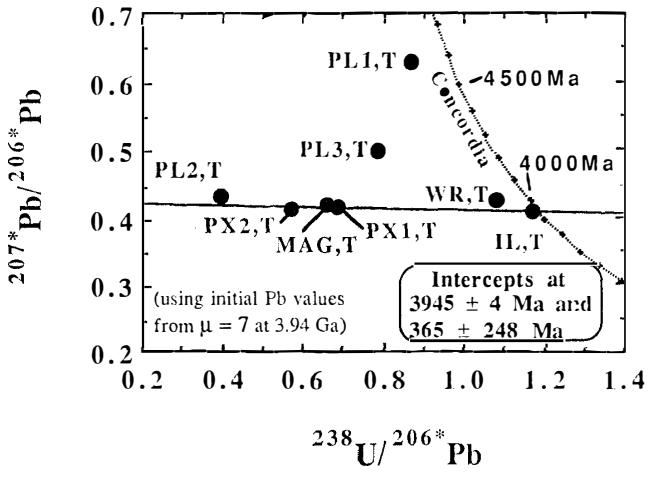

Fig. 9. $^{207*}Pb/^{206*}Pb$ vs. $^{238}U/^{206*}Pb$ modified concordia diagram from regression of the calculated values (leaches plus residue) for PX1, PX2, IL, and MAG using calculated initial values, corresponding to $\mu=7$, assuming a single stage Pb evolution from CDT Pb compositions at 4.56 Ga as the age of the Moon.

Two-stage evolution — It is generally accepted that the Moon underwent a primary differentiation that formed a lunar core, mantle, and crust from an initial magma ocean stage. The age and duration of the primary lunar differentiation is in dispute. We assume a two-stage Pb evolution starting with CDT Pb values at 4.56 Ga and a secondary stage between 4.35 Ga (primary lunar differentiation from the lunar magma ocean) to 3.94 Ga with μ_2. Assuming a low $\mu_2=1$ for the gabbro source, the highest μ_1 value for the first stage is about 40. The μ_1 value becomes 98 if μ_2 is 0.1. If this gabbro is the plutonic equivalent of lunar mare basalts, this low μ value for the source of Asuka-31 is surprising compared with the high μ values (100–300) for the sources of typical Apollo mare basalts (*e.g.*, TATSUMOTO *et al.*, 1977). This low μ value is smaller than the estimated μ for the source of anorthosite Yamato-86032 ($\mu=50$–60; TATSUMOTO and PREMO, 1991) but similar to those estimated for the source of 60025 anorthosite ($\mu=16$–55; HANAN and TILTON, 1987), for the source of green glass 15426 ($\mu=19$–55; TATSUMOTO *et al.*, 1987), or for Apollo 17 orange glass 74220 ($\mu=35$; NUNES *et al.*, 1974, $\mu<29$; TERA and WASSERBURG, 1976; Fig. 5b).

PREMO and TATSUMOTO (1991b) argued that Hanan and Tilton's U-Pb data for ferroan anorthosite 60025 can be interpreted such that the anorthosite is an early-formed float from the lunar primary magma ocean of $\mu\approx35$ at ~4.44 Ga (the Sm-Nd age; CARLSON and LUGMAIR, 1988). Thus, the low μ of ~40 for the source of Asuka-31 may be possible. However, it must be noted that this low μ value for Asuka-31 can be obtained only by assuming a very low μ_2 for the second stage. One may speculate that the source region may have contained a large amount of sulfides enriched in chalcophile Pb.

Three-stage evolution — The giant impact hypothesis (CAMERON, 1986) for the lunar origin has attained considerable popularity. Although many workers are in disagreement about the origin of the material that formed the Moon (the Earth's mantle or the mantle of a giant impacting planetesimal), we can assume a μ_1 of 8 (value for the Earth's mantle) starting from 4.56 Ga until the Moon's accretion at ~4.44 Ga (the formation age of 60025: the oldest known lunar rock). We assume further a second stage between 4.44 to 4.35 Ga (the source cumulate age from the primary magma ocean) with μ_2, and the third stage between 4.35 to 3.94 Ga with $\mu_3=1$. In this three-stage model, we get an upper intercept age of 3942 Ga with $\mu_2=90$. The radiogenic portion of the PX2, T fraction disappears when $\mu_2\approx180$, and with $\mu_3=0.1$, it disappears when $\mu_2\approx230$. One may argue that the cumulation age of 4.35 Ga would be too young. Nonetheless, these calculations indicate that the primary μ for the Asuka-31 source could have been increased as large as those for lunar mare basalts (100–300) but requires adjusting the ages for the Moon's formation and differentiation.

Major element chemistry and rare earth element abundances suggest that Asuka-31 is related to VLT basalts (YANAI, 1991; LINDSTROM *et al.*, 1991a). On the basis of REE and Sc abundances and modal melting calculations, WENTWORTH *et al.* (1979) conclude that the source rocks for the Apollo 17 VLT basalts were deep-seated olivine-orthopyroxene cumulates. In spite of the similarity in major and trace element signatures to VLT mare basalts and the similarity to typical mare

basalts in its solidification age, the μ value for the source of Asuka-31 is very low ($\mu<17$) compared with typical mare basalts.

As discussed previously (TATSUMOTO *et al.*, 1987), during early lunar differentiation, Pb was lost by volatilization and was scavenged by sulfides that sunk to the lunar core. Pb was further removed into the lunar crust by anorthosite flotation. Thus, μ values drastically increased during the early stages of lunar differentiation. Based on ion probe analytical data for two granite clasts, COMPSTON and WILLIAMS (1989) also suggest that there was very early Pb loss from large volumes of the Moon. On the other hand, the refractory lithophile, incompatible elements, U and Th, were concentrated in the residual liquid (source of KREEP basalts) because the partition of U and Th is about ten times lower than Pb in common mafic minerals. It is suggested that non-radiogenic Pb was transported in the deep interior of the Moon more than U and Th (NUNES *et al.*, 1974; TERA and WASSERBURG, 1974; TATSUMOTO *et al.*, 1987). If Fe-Ni sulfides sank from the partially molten magma ocean, then μ of the deep lunar mantle from which VLT basalts and the Asuka-31 gabbro were generated should be substantially lower than μ values of subsequent lunar mantle-derived rocks.

The low μ value estimated in this study implies that the source of Asuka-31 is either enriched in Pb or depleted in U relative to the source for mare basalts. By analogy with the formation process of VLT basalts, we suggest that the source materials of Asuka-31 are early-stage mafic cumulates precipitated from a lunar magma ocean. The source of Asuka-31 may be of deep origin and may be enriched in non-radiogenic Pb, as is the case for green glass and orange glass. If the source was not saturated with sulfur (*i.e.*, a reservoir of sulfides was not so large and/or a degree of partial melting is relatively large), the sulfide melt could not be segregated from the silicate melt during partial melting and could not remove chalcophile elements effectively during gabbro formation. If this is the case, it is also suggested that some amount of Pb was sealed in the deep mantle of the Moon, and we should not only relax the lower limits on the bulk-Moon contents of refractory lithophiles (WARREN and KALLEMEYN, 1991) but also reexamine the upper limits of volatile chalcophiles.

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