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National Aeronautics and Space Administration

Lunar Sample Analysis Program

Final Report
for
Proposal No. T-75445
Apollo Missions 11-13

B.3 Mass Spectroscopy Determination for U, Th, and Pb Concentrations and Pb, U, and Th Isotope Abundances

by

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The investigations performed under Contract No. 75445 revealed:

(A) The data determined in the U-Th-Pb system on rocks collected for Apollo 11 and 12 confirm the great antiquity.

(B) U-Th-Pb data suggest that the soils were derived from source material about 4650 million years old.

(C) The large ratios of U/Pb and Th/Pb and the U-Th-Pb systematics of the lunar samples suggest that the moon was accreted from the broiled outer particles in the proto-earth-moon system about 4650 million years ago.

(D) The results of the present studies, as well as other PI's studies, also suggest that the moon was melted at the beginning stage and had a large-scale differentiation. The differentiation accumulated uranium and thorium into the surface layer.

(E) The U^{238}/U^{235} ratio in samples measured is the same as that for terrestrial uranium within experimental error (137.8 ± 0.3). The U^{234} daughter is in radioactive equilibrium with parent U^{238} in the samples; however, it could not be demonstrated that Th^{230} is in equilibrium with U^{238} in some rock samples.

The investigations performed under Contract No. T-75445 were reported in the following papers (in chronological order):

(1) Tatsumoto, M., 1969, New method for preparing ultrapure hydrofluoric acid: Anal. Chem., v. 41, p. 2088-2089.

(2) Tatsumoto, M., and Rosholt, J. N., 1970, Age of the moon: An isotopic study of uranium-thorium-lead systematics of lunar samples: Science, v. 167, p. 461-463.

(3) Tatsumoto, M., 1970, Age of the moon: An isotopic study of uranium-thorium-lead systematics of Apollo 11 lunar samples--II: Geochim. Cosmochim. Acta, Proc. Apollo 11 Lunar Sci. Conf., v. 2, p. 1595-1612.

(4) Rosholt, J. N., and Tatsumoto, M., 1970, Isotopic composition of uranium and thorium in Apollo 11 samples: Geochim. Cosmochim. Acta, Proc. Apollo 11 Lunar Sci. Conf., v. 2, p. 1499-1502.

*(5) Tatsumoto, M., 1970, U-Th-Pb age of Apollo 12 rock 12013: Earth Planet. Sci. Letters, v. 9, p. 193-200.

*(6) Tatsumoto, M., Knight, R. J., and Doe, B. R., Uranium-thorium-lead systematics of Apollo 12 lunar samples: Geochim. Cosmochim. Acta, Proc. Apollo 12 Lunar Sci. Conf. (in press).

*(7) Rosholt, J. N., and Tatsumoto, M., Isotopic composition of uranium and thorium in Apollo 12 samples: Geochim. Cosmochim. Acta, Proc. Apollo 12 Lunar Sci. Conf. (in press).

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*Reprint and preprints of papers (5), (6), and (7) listed above are attached.

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U-Th-Pb AGE OF APOLLO 12 ROCK 12013 *

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U-Th-Pb AGE OF APOLLO 12 ROCK 12013 *

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A U-Th-Pb isotopic study of three chips from lunar rock 12013 indicates that parental material of the intrusion breccia formed quite early in the moon's history, possibly 3.9 to 4.3 by ago. The U-Th-Pb characteristics of the rock are distinctly different from those of other Apollo 12 igneous rocks and suggest a different origin.

1. Introduction

Three chips from lunar rock 12013 were analyzed for lead isotopic composition and for concentrations of lead, uranium, and thorium.

Rock 12013 is a unique specimen with high uranium, thorium and potassium concentrations and extremely heterogenous texture [1]. It is an intrusion breccia in which fragments of darker colored rocks are being engulfed in lighter colored rocks. Furthermore, the rock is interesting because of its Rb-Sr total rock age of 4.6 by [2], suggested an origin for the rock very early in the moon's history.

All of the breccia is gabbroic in composition and the main cause of color differences appears to be variations in mineral percentages.

2. Experimental

2.1. *Sample description*

Three chips (about 250 mg each) from slab No. 10 of the rock 12013 were analyzed. Chips #09 and #42 were supplied in the first allocation. A second allocation, chip #45, was received one month later than the first.

Small splits (about 10 mg each) of the crushed samples #42 and #09 were subjected to a rapid petrographic examination by G.A.Izett and R.E.Wilcox, and the results are shown in the Appendix.

2.2. *Analytical procedure*

A new analytical method was developed in order to minimize lead contamination of the lead isotopic analyses. This method is briefly described here, but details will be published elsewhere. The samples from the first allocation, chips #42 and #09, were crushed in a boron-carbide mortar without any pretreatment or washing, even though a copper metallic color was observed on part of the wire-sawed surface. The sawed surfaces of #45, a light-colored chip constituting the second allocation, were abraded with a dental drill and washed quickly with double-distilled acetone using an ultrasonic vibrator. All samples were decomposed by $HClO_4$-HNO_3-HF in Teflon digestion bombs [3]. The 237-mg chip of #45 was dissolved directly, without crushing, in a digestion bomb. Samples #09A and #42 were heated at 150°C for 12 hr and #09B and #45 were heated at 200°C for 96 hr.

The content of the digestion bomb was transferred into a Teflon beaker and evaporated to dryness. The residue was again treated with HNO_3 and HF in order to facilitate isotopic equilibrium and was then taken to dryness. The residue was taken up in double-distilled concentrated nitric acid. After prolonged agitation in an ultrasonic vibrator to dissolve a white precipitate (probably metatitanic acid), the solution was divided into two parts. Although the solution appeared to be perfectly clear, it was found, following

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* Publication authorized by the Director, U.S. Geological Survey

centrifugation, that a very small amount of residue still remained. One part of the solution was used for the determination of lead isotopic composition. Following centrifugation, a $Ba(NO_3)_2$ saturated water solution was added to the supernatant [4]. The barium nitrate, which coprecipitates the lead, was centrifuged out and dissolved in water, and the lead was deposited on a platinum anode at 1.6 V in the presence of 0.5 mg of copper [5]. The deposited lead was dissolved in freshly electrolized concentrated nitric acid which contained NO_2^-. The solution was evaporated to dryness, and lead was converted to phosphate. The other part was spiked with ^{235}U, ^{232}Th, ^{207}Pb, and ^{204}Pb for isotope dilution and was dried after addition of HF. Lead was separated using the same technique employed for the composition analysis. The total lead recovery by this method was always more than 70% for the 1 μg lead range, and blanks for the lead analyses ranged from 4 to 7 ng (nanograms). Uranium and thorium were separated from the supernatant of the barium precipitation by double Dowex 1 nitrate resin treatment [4].

Lead, uranium, and thorium were analyzed in a 12-inch mass spectrometer using a Faraday cup for ion collection and a digital data read-out system. Lead was analyzed by the silica gel-phosphate method [6, 7] on a single rhenium filament, and uranium and thorium were analyzed by a triple-filament mode [8]. The standard deviations of the lead isotope ratio measurements are: $^{206}Pb/^{204}Pb$, ±1%; $^{207}Pb/^{206}Pb$ and $^{208}Pb/^{206}Pb$, ±0.1%. Mass fractionation for lead analysis was corrected by a modified version (R.J. Knight and M.Tatsumoto, unpublished data) of the double-spike method [9]. Due to a lead blank which was lower than that in the previous lunar sample analyses, the overall uncertainty for lead ratios in table 1 is less than 1% (at one standard deviation).

Sample #09B, which was a small amount of material left over from #09 powder, was spiked with ^{208}Pb, ^{235}U, and ^{230}Th before its decomposition. The lead isotopic composition was calculated, using the $^{208}Pb/^{206}Pb$ ratio of #09A. Owing to improper spiking ratio the isotopic composition has a large uncertainty, but the U, Th, and Pb concentrations are as accurate as those for samples #09A, #42 and #45. Because of such uncertainties this analysis is excluded from most of the Discussion.

Table 1

Isotopic composition of lead and concentration of lead, uranium and thorium chips of lunar rock 12013, 10 *

Fragment No.	Shade	Concentration, (ppm)			Atomic ratio							
					Raw data			Corrected for blank				
		Pb	U	Th	$\frac{^{206}Pb}{^{204}Pb}$	$\frac{^{207}Pb}{^{204}Pb}$	$\frac{^{208}Pb}{^{204}Pb}$	$\frac{^{206}Pb}{^{204}Pb}$	$\frac{^{207}Pb}{^{204}Pb}$	$\frac{^{208}Pb}{^{204}Pb}$	$\frac{^{238}U}{^{204}Pb}$	$\frac{^{232}Th}{^{238}U}$
09A	Dark	9.28_5	5.67_5	20.73	898.8	481.4	904.8	1201.7	641.6	1202.9	1612.4	3.78
09B	Dark	12.30	5.87_1	22.94				(415) **	(230) **	(423) **	(435) **	4.04
42	Light	16.31	10.80	34.29	1892.3	965.6	1631.4	2059.2	1050.2	1773.3	2792.3	3.28
45	Light	11.43	5.75_2	19.05	1202.1	643.2	1062.5	1437.6	768.1	1266.3	1510.5	3.42

* Estimated uncertainties for the concentrations are less than 1% for Pb, U and Th.

** Corrected for ^{208}Pb spike. The lead isotopic composition was calculated using $^{208}Pb/^{206}Pb$ ratio in #09A.

3. Results

The isotopic composition of lead and concentrations of lead, uranium, and thorium are given in table 1. The uranium concentrations range from 5.7 to 10.8 ppm and the thorium concentrations from 19 to 34 ppm, and indicate extreme heterogeneity of the rock. The ranges are in agreement with the values determined by Wakita and Schmitt [10]; however, their Th-U ratios of 2.3 to 3.2 are lower than the ratios (3.2–4.0) reported here. However, if LSPET's [1] ratio (3.2 ± 0.6) obtained by gamma-ray spectrometry using the total 80-g sample is valid, then Wakita and Schmitt may have analyzed samples that have a lower ratio than the ones I reported here. The highest values for uranium and thorium concentrations reported here are close to that reported by LSPET [1] for the entire rock. Since U and Th are probably concentrated in interstitial phases and in accessory minerals, such as phosphates, and the U might be enriched in zircon grains, these differences may be caused by sampling bias rather than by use of different analytical methods.

Lead concentrations which range from 9.3 to 16 ppm are 20 to 40 times higher than those in other lunar igneous rocks. The isotopic compositions are extremely radiogenic, more so than in any other lunar rocks so far examined. The analytically determined present-day ratios of $^{238}U/^{204}Pb$ are as high as 2800. This value further substantiates the conclusion derived from the Apollo 11 samples that the volatile element lead is depleted, relative to uranium and thorium, on the lunar surface. If the moon was characterized initially by a high U-Pb ratio, then at a time significantly later than its formation it becomes exceedingly difficult to apply a valid correction for the common lead generated during this interval when this common lead is incorporated in a newly formed rock.

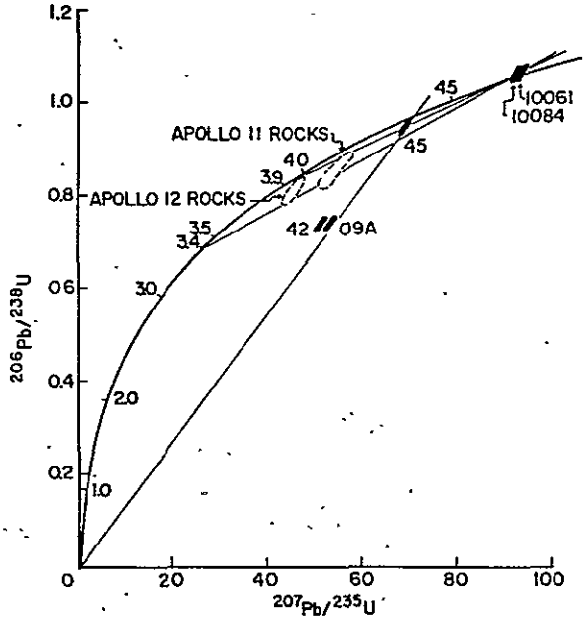

Fig. 1. U-Pb evolution diagram. Plotted points are $(^{207}Pb_{observed} - ^{207}Pb_{primordial})/^{235}U$ against $(^{206}Pb_{observed} - ^{206}Pb_{primordial})/^{238}U$. The encircled areas indicate the regions in which data of Apollo 11 [6] and Apollo 12 (unpublished data) igneous rock fall. The 10084 and 10061 are data on fines and breccia from Apollo 11. Numbers on the concordia indicate billions of years. Dimensions of the polygons indicate the analytical uncertainty.

4. Discussion

In treating U-Th-Pb data, two complicating factors must be considered before raw analytical results can be interpreted in terms of meaningful ages. (1) The amount and isotopic composition of lead incorporated into the rock at the time of crystallization (initial lead) must be known, and (2) the nature of any post-crystallization open system behavior must be determinable. No consensus of opinion in dealing with these two problems for lunar rocks is found in the recently published results of several laboratories, and difficulty in evaluating these factors still exists.

Previous studies are compatible with the assumption that the moon originally had an isotopic composition close to that of meteorite primordial lead. Therefore, in evaluating initial lead correction for lunar material, it seems to be a safe assumption that *at least* as much ^{206}Pb, ^{207}Pb, and ^{208}Pb relative to ^{204}Pb as occurs in meteorite primordial lead must be subtracted. Although a considerably larger correction may be required for lunar rocks significantly younger than the age of the moon, removal of meteorite primordial lead establishes an upper limit to the amount of the radiogenic isotopes produced by *in situ* radioactive decay in the rock (for the present, we shall ignore the additional complications arising for post-

Table 2
Apparent ages of chips of lunar rock 12013, 10 *

Fragment No.	Shade	Atomic ratios				Apparent ages (my) **			
		$\frac{^{206}Pb}{^{238}U}$	$\frac{^{207}Pb}{^{235}U}$	$\frac{^{207}Pb}{^{206}Pb}$	$\frac{^{208}Pb}{^{232}Th}$	$\frac{^{206}Pb}{^{238}U}$	$\frac{^{207}Pb}{^{235}U}$	$\frac{^{207}Pb}{^{206}Pb}$	$\frac{^{208}Pb}{^{232}Th}$
09A	Dark	0.7395	53.96	0.5296	0.1929	3602	4121	4388	3613
42	Light	0.7368	51.33	0.5073	0.1904	3582	4071	4334	3571
45	Light	0.9456	69.14	0.5306	0.2394	4330	4372	4391	4397

* Common lead used for calculation is that of troilite lead determined by Oversby [11].

** The constants used are [6]: $\lambda_{^{238}U} = 1.5369 \times 10^{-10}$ yr^{-1}; $\lambda_{^{235}U} = 9.7216 \times 10^{-10}$ yr^{-1}; $\lambda_{^{232}Th} = 4.8813 \times 10^{-11}$ yr^{-1}; $^{235}U/^{238}U = 1/137.8$.

crystallization introduction of an extraneous lead to the rock).

The data corrected for meteorite primordial lead [11, 12] are plotted on fig. 1, and the resultant apparent ages are given in table 2. The analysis for fragment #45 gives almost concordant ages, while those for fragments #42 and #09A result in significantly discordant U-Pb ages. Later, other methods of correction for initial lead will be considered; however, while this will cause some differences in age and translate the data points on the concordia diagram, the basic relationships as seen here will remain much the same. No reasonable choice of initial lead will produce a concordant age for all of the analyzed fragments, and we now turn our attention to the possible effects of postcrystallization open system conditions.

Much of our understanding of U-Th-Pb systematics derives from work done on terrestrial radioactive minerals, and it is instructive to examine the lunar mateiral in light of existing theory. It has long been known that suites of cogenetic zircon commonly show some discordancy and plot along a straight line which usually intersects concordia in two places. Several open system models have been devised to explain this relationship, among which the best known are the episodic disturbance model of Wetherill [13] and the continuous diffusion models of Tilton [14] and Wasserburg [15].

In the case of episodic disturbance, the upper (older) intersection between a linear array of sample points and concordia indicates the age of primary crystallization, and the lower (younger) intersection indicates the age of disturbance (metamorphism). Let us tentatively consider all of rock 12013 as part of one cogenetic suite of samples despite the admittedly heterogeneous character of the intrusion breccia. These data would then indicate a primary crystallization age of 4.3 by and a disturbance age of about 0.5 by for an episodic model. Such a disturbance age might record the time of brecciation, but it could likewise reflect some more subtle event in the history of the rock. By analogy with terrestrial rocks, chemical mobility within the U-Pb system may have taken place without leaving an imprint in the fabric of the rock.

If open system conditions subsequent to crystallization are responsible for the pattern of the data points in fig. 1, all of the analyzed samples apparently have preferentially lost radiogenic lead relative to uranium. With this limited number of analyses, however, it is not possible to say with certainty whether rock 12013 as a whole has been undergone a net chemical change. If the rock remained a closed system we should find somewhere fractions of it showing preferential loss of uranium relative to radiogenic lead.

As shown by Wasserburg [15], the interpretation of diffusion models becomes somewhat complex for very old systems. Nevertheless, nearly concordant samples such as #45 would still closely approximate the true crystallization age. The lower intercept of a linear array on the concordia diagram would have no physical meaning in the case of continuous diffusion.

Because our choice for the isotopic composition of initial lead used to correct the raw data was somewhat arbitrary, ages given in the above discussion are subject to some change when other initial lead is con-

Table 3
Apparent ages of chips of lunar rock 12013, 10 *

Fragment No.	Shade	Atomic ratios				Apparent ages (my)			
		$^{206}Pb/^{238}U$	$^{207}Pb/^{235}U$	$^{207}Pb/^{206}Pb$	$^{208}Pb/^{232}Th$	$^{206}Pb/^{238}U$	$^{207}Pb/^{235}U$	$^{207}Pb/^{206}Pb$	$^{208}Pb/^{232}Th$
09A	Dark	0.6421	32.92	0.3721	0.1691	3183	3567	3791	3203
42	Light	0.6760	39.04	0.4190	0.1758	3367	3771	3995	3295
45	Light	0.8298	44.68	0.3908	0.2115	3931	3931	3931	3931

* Common lead used for calculation is that obtained from #45. See text.

sidered. To examine how this factor can influence the age interpretation, let us now use an extremely radiogenic isotopic composition for this lead. For example, following the method of Tatsumoto for the Apollo 11 lunar samples [16], we may partition the radiogenic isotopes between two stages of development in such a manner as to uniquely define a concordant age for the start of the second stage. This is the so-called two-stage growth model of which the mathematical formulation was reviewed in the aforementioned paper. For this model we assume an original age of the moon of 4.6 by and an original lunar lead similar in isotopic composition to meteorite primordial lead. A concordant age is calculated for #45 by the two-stage model using the observed $^{238}U/^{204}Pb$, $^{232}Th/^{238}U$, and the analyzed lead isotopic composition. The calculated concordant age (t_1) is 3931 my. The $^{206}Pb/^{204}Pb$, $^{207}Pb/^{204}Pb$, and $^{208}Pb/^{204}Pb$ ratios at t_1 are 184.23, 278.29, and 172.95, respectively. The $^{238}U/^{204}Pb$ and $^{232}Th/^{238}U$ ratios for the first stage are 883 and 4.06, respectively. Assuming all of the material in rock 12013 to be cogenetically related, it would be reasonable to now use the isotopic composition calculated to exist at the end of the first stage for #45 to correct the other samples. The recalculated data and apparent ages obtained by using $(^{206}Pb/^{204}Pb)_{t_1}$, $(^{207}Pb/^{204}Pb)_{t_1}$, and $(^{208}Pb/^{204}Pb)_{t_1}$ from #45 are given in table 3 and are plotted in fig. 2. The $^{207}Pb/^{206}Pb$ ages calculated for this case range between 3.8 and 4.0 by. As before, samples #42 and #09A fall near a line that connects point #45 with the origin. In detail, however, points #42 and #09A are not both colinear within experimental uncertainty and now fall opposite their relative position on fig. 1. This deviation may be due either to an overcorrection for initial lead to our assumption that all three fractions are cogenetic.

We have chosen two extreme cases for making initial lead correction – meteorite primordial lead on the one hand and a lead generated by an extremely high first stage $^{238}U/^{204}Pb$ environment on the other hand. A more probable value of $^{238}U/^{204}Pb$ might be 100–300 as defined in the previous paper for Apollo 11 rocks. Correcting for such initial lead will cause the data to plot somewhere between their position on fig. 1 and fig. 2, corresponding to $^{207}Pb/^{206}Pb$ ages of 4.1 to 4.2 by. This kind of initial lead correc-

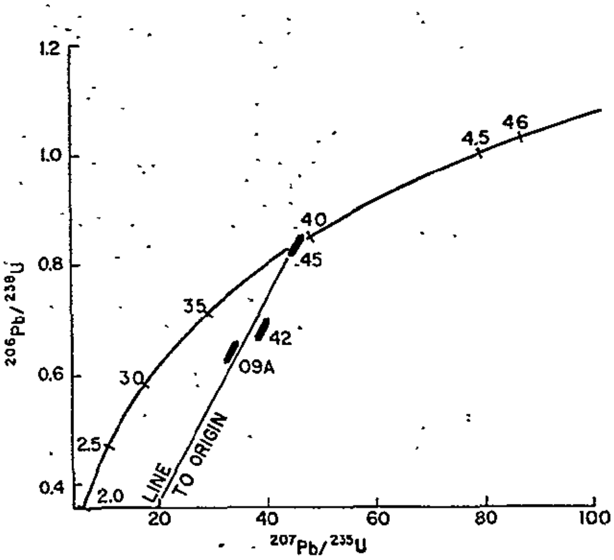

Fig. 2. U-Pb evolution diagram. Plotted points are corrected by a common lead obtained from #45. (See text.) Numbers on the concordia indicate billions of years. Dimensions of the polygons indicate the analytical uncertainty.

tion will also produce very similar $^{207}Pb/^{206}Pb$ ages for #42 and #09A, thereby eliminating the age discrepancy previously encountered for these two fractions.

Up to this point we have treated all fractions as part of one cogenetic suite; i.e., all fractions crystallized at the same time and with the same initial lead isotopic composition. If rock 12013 is truly an intrusive breccia, then it is possible that fragments of various ages and origins have been incorporated together. In such a case, it may be erroneous to assume a single age for all the fractions. Certainly, three samples are inadequate to define real petrologic groupings, but it is instructive to discuss age limitations for #42 and #09A entirely independent of #45. Without an unreasonably large and entirely contrived initial lead correction, it is difficult to attribute the extremely discordant nature of these two fragments (#09A and #42) to anything but open system conditions subsequent to some original time of crystallization. In the following discussion, we shall consider the data corrected for meteoric primordial lead as shown in fig. 1. Other reasonable methods of initial lead correction will not drastically change the conclusions to be reached.

If disturbance of the itotopic system only took place episodically, such as at the time of breccia formation, two limiting cases may be considered for the original age of the fragments and for the time of brecciation. The two limiting cases arise (1) by assuming the original age of the fragment to be as old as the moon (4.6 by) in which case the time of brecciation is approximately 2.1 by, and (2) by assuming the time of brecciation to be recent in which case the original age of the fragment is approximately 4.3 by. If mixing of radiogenic lead also occurs at the time of brecciation between samples #42 and #09A and other material, meaningful age information may be virtually masked by the unknown trajectory of their mixing line.

All of the above discussion assumes that the samples were not contaminated during sample handling and that the analytical results are accurate. If sample decompositions of #09A and #42 were incomplete, then the analytical results may be wrong. Also, if a white precipitate, which usually appears in the course of our method of the HF decomposition of lunar rock, hindered isotopic equilibrium, we could observe a discordance in the $^{206}Pb/^{238}U$ and $^{208}Pb/^{232}Th$ ages, because the white precipitate tends to include more Th than U. Such discordance was not observed on the two chips. The first two analyses, those of #42 and #09A, were performed without any special attempt to evaluate the effect of trace quantities of insoluble residue. After I was informed [2] that rock 12013 contained zircon, samples #45 and #09B were treated at a higher temperature and for a longer interval with twice the amount of HF in order to decompose the samples completely [17]. The U-Th-Pb data indicate that these samples are less discordant than the samples previously analyzed (#42, #09A on fig. 1).

Regardless of which of the several interpretations discussed above, either parental material of the intrusion breccia, if not the brecciation itself, formed quite early in the moon's history, possibly 3.9 to 4.3 by ago. Such ages are quite compatibel with those obtained by several other workers using different techniques. A ^{40}Ar-^{39}Ar age of 3.9 by has been proposed by Turner [18] and Rb-Sr internal isochron age of 4.0 by is being presented by The Lunatic Asylum [19].

If rock 12013 was transported to its present location from a highland area by impact after the mare formation, the age of the source material in the highland may be inferred as about 4 by from the age of the rock 12013. If the source of the rock were the soil overlying the mare region; the soil was felsic like rock 12013 at that time or an extensive magmatic differentiation had occurred in the mare to have produced such a felsic rock as 12013.

Even though the details are debatable, it is generally accepted that early stage differentiation of the moon resulted in a separation of an anorthositic layer at or near the surface and a mafic layer in the moon's mantle. The mafic layer has been considered to be as either gabbroic [20], clinopyroxenite or peridotite [21], or eclogite [22]. Basaltic lavas produced in the mantle have filled the maria. Because U-Th-Pb (e.g. [16]) and Rb-Sr [23] data for Apollo 11 soil indicate an age of 4.6 by, the anorthositic layer could be thought to be 4.6 by old and may represent the oldest moon surface. However, a highland area that is about 4 by old must be present, at least near the mare, if the source of the rock is in the highland.

It is interesting to note that almost all crystalline rocks from Apollo 11 and 12 fall near a chord between 4.6 by and 3.9 to 3.4 by in the U-Pb evolution diagram (fig. 1). As Papanastassiou and Wasserburg [25] have discussed, no proof has been found that large differences in age exist for different mare regions. Points for the least discordant samples from Apollo 11 and 12, including rock 12013, mainly lie on a chord between 4.6 and 3.9 by. From this observation, one could postulate that all flood-type maria were presumably filled with basaltic lava within a small span of time during the interval 3.9 to 3.4 by.

Further implications of the data on the rock 12013 will be discussed in a future paper dealing with lead isotope analyses of other Apollo 12 samples.

Acknowledgments

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Appendix

The splits of #42 and #09 were examined in random immersion mounts and selected grains were studied on the spindle stage.

Chip #42 – From the generally lighter colored parts of the rock.

Plagioclase (approximately 50–60%).

Fresh crystal fragments of plagioclase form an important part of the sample. The composition of the plagioclase as inferred from the framgnets' indices of refraction ranges widely, From An_{50} to An_{75}. Many of the plagioclase fragments show patchy areas that appear to be fractured and show some index differences from the optically uniform parts of the grains. Other fragments are clear and unfractured.

Orthopyroxene (approximately 29%)

Cleavage fragments that show positive elongation and parallel extinction and are pale light green form a significant part of the sample. No pleochroism could be seen. The optic properties are:

$$N_x = 1.712$$
$$N_y = 1.723 \pm 0.002$$
$$N_z = 1.727$$
$$N_z - N_x = 0.015$$
$$2V_x = 56 \pm 3 .$$

These properties indicate a probable content of over 50% of the Fe-end member (ferrohypersthene).

Olivine (2–10%)

Several fragments of olivine have optic properties as follows:

N_x ranges from 1.726 to1.745
N_y ranges from 1.745 to 1.780
N_z ranges from 1.765 to higher values
$N_z - N_x = 0.039.$

These properties imply compositions in the hyalosiderite and hortonolite range of the olivine series.

Sanidine (5–10%)

The optic properties of the sanidine are:

$$N_x = 1.519 \pm 0.002$$
$$N_y = 1.524$$
$$N_z = 1.526$$
$$2V_x \; 20\text{–}30.$$

The sanidine contains light green rod-shaped inclusions of pyroxene (?)

Ilmenite (approximately 0.5%)

Quartz (less than 1%)

A few grains of quartz were seen, and they show undulatory extinction and contain scattered equidimensional inclusions of an unidentified mineral.

Accessory minerals occurring in amounts of less than 0.1% were not identified in this examination. Apatite and zircon have been reported [19] in some fractions of rock 12013.

Chip #09 – From the generally darker part of the rock.

This sample contains almost the same minerals as #42, including calcic plagioclase, sanidine, olivine, and orthopyroxene, except it has less potassium feldspar and no quartz was seen. Most of the fragments are feldspathic and crystalline and are charged with many small opaque grains (ilmenite), which impart a clouded dark appearance.

References

[1] LSPET (The Lunar Sample Preliminary Examination Team), Preliminary Examination of Lunar Samples from Apollo 12, Science 167 (1970) 1325.

[2] G.J.Wasserburg, personal communication; See also paper by the Lunatic Asylum in this issue, p. 137.

[3] B.Bernas, A new method for decomposition and comprehensive analysis of silicates by atomic absorption spectrometry, Anal. Chem. 40 (1968) 1682.

[4] M.Tatsumoto, Isotopic composition of lead in volcanic rocks from Hawaii, Iwo Jima and Japan, J. Geophys, Res. 71 (1966) 1721.

[5] J.J.Lingane, Electroanalytical Chemistry (Intersci. Publ. Incl, New York, 1958) p. 375.

[6] P.A.Akishin, O.T.Nikitin and G.M.Panchenkov, A new effective ionic emitter for the isotopic analyses of lead, Geokhimiya 5 (1947) 429.

[7] A.E.Cameron, D.H.Smith and R.L.Walker, Mass spectrometry of nanogram-size samples of lead, Anal. Chem.

[8] J.N.Rosholt, B.R.Doe and M.Tatsumoto, Evolution of the isotopic composition of uranium and thorium in soil profiles, Geol. Soc. Amer. Bull. 77 (1966) 987.

[9] W.Compston and V.M.Oversby, Lead isotopic analysis using a double spike, J. Geophys. Res. 74 (1969) 4338.

[10] H.Wakita and R.A.Schmitt, personal communication and a companion paper in this publication, p. 169.

[11] C.C.Patterson, The Pb^{207}/Pb^{206} ages of some stone meteorites, Geochim. Cosmochim. Acta 7 (1955) 151.

[12] V.M.Oversby, The isotopic composition of lead in iron meteorites, Geochim. Cosmochim. Acta 34 (1970) 65.

[13] G.W.Wetherill, Discordant uranium-lead ages, 1, Trans. Amer. Geophys. Union 37 (1956) 320.

[14] G.R.Tilton, Volume diffusion as a mechanism for discordant lead ages, J. Geophys. Res. 65 (1960) 2933.

[15] G.J.Wasserburg, Diffusion processes in lead-uranium systems, J. Geophys. Res. 68 (1963) 4823.

[16] M.Tatsumoto, Age of the moon: An isotopic study of U-Th-Pb systematics of Apollo 11 lunar samples, II, Proc. Apollo 11 Lunar Science Conf., vol. 2 (1970) 1595.

[17] C.J.Allegre, personal communication on zircon decomposition by digestion bomb.

[18] G.Turner, personal communication and a companion paper in this publication, p. 177.

[19] G.J.Wasserburg, personal communication and a companion paper in this publication, p. 137.

[20] J.A.Wood, J.S.Dikey, Jr., U.B.Marvin and B.N.Powell, Lunar anorthosites and a geophysical model of the moon, Proc. Apollo 11 Lunar Science Conf., Vol. 1 (1970) 965.

[21] M.J.O'Hara, G.M.Biggar, S.W.Richardson, C.E.Ford and B.G.Jamieson, The nature of seas, mascons and the lunar interior in the light of experimental studies, Proc. Apollo 11 Lunar Science Conf., Vol. 2 (1970) 695.

[22] A.E.Ringwood and E.Essene, Petrogenesis of Apollo 11 basalts, internal constitution and origin of the moon, Proc. Apollo 11 Lunar Science Conf., Vol. 1 (1970) 769.

[23] D.A.Papanastassiou, G.J.Wasserburg and D.S.Burnett, Rb-Sr ages of lunar rocks from the Sea of Tranquillity, Earth Planet. Sci. Letters 8 (1970) 1.

[24] D.A.Papanastassiou and G.J.Wasserburg, Rb-Sr ages from the Ocean of Storms, Earth Planet. Sci. Letters 8 (1970) 269.

U-Th-Pb Systematics of Apollo 12 Lunar Samples*

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Abstract--The data determined in the U-Th-Pb system on rocks collected by the Apollo 12 mission confirm the great antiquity of and extreme depletion of common lead in the surficial lunar rocks in maria. Unlike the rocks analyzed from the Apollo 11 mission, those from Apollo 12 do not define a good Pb-Pb isochron although six of the eight rocks lie close to a 3900- to 4000-m.y. isochron. Apollo 11 rocks have a well-defined 4200-m.y. isochron, and the unusual Apollo 12 rock 12013 has a well-defined 4370-m.y. isochron. Although these Pb-Pb isochron ages for the Apollo rocks are about 10 percent older than Rb-Sr mineral isochron ages and $^{39}Ar/^{40}Ar$ whole-rock ages as determined by other workers, they are in the same relative sequence of ages. Concordia relations on whole rocks and U-Pb and Th-Pb isochrons on density fractions, even though poorly defined, are compatible with the ages determined by other methods if the values of $^{238}U/^{204}Pb$ observed for the Apollo 11 rocks and fragments from rock 12013 are proportional to the $^{238}U/^{204}Pb$ in the source materials of these rocks and if the initial lead was not homogenized in the magma-forming events. This proportionality constraint does not exist for the Apollo 12 rocks.

An attempt was made to resolve the conflict in the U-Th-Pb whole-rock data with the Rb-Sr mineral isochron and $^{39}Ar/^{40}Ar$ whole-rock age data by a concordia diagram treatment of U-Pb data on density fractions. In general, the U-Pb data as plotted by using this technique indicate an event younger

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than those events determined for the same rocks by other lead isotope or dating methods. This difference could be explained if the unknown amount of lead contamination due to the sample processing decreases with increase in the amount of sample processing, in spite of the fact that all liquids in the sample processing were distilled before use.

The agreement between apparent ages determined by U-Th-Pb methods and those ages determined by other dating techniques is much better on the soils and breccias, which are of complex origin, than it is on the rocks. In general, the U-Th-Pb data suggest that the soils were derived from source material about 4650 m.y. old that had undergone differentiation of U relative to lead no more than 4000 m.y. ago. U-Th-Pb data on Apollo 12 soils and breccia also indicate that alteration in U/Pb also has taken place at times much younger than 3000 m.y. ago by "third events" (perhaps alteration due to impact events?) which are not reflected in the Rb-Sr mineral isochron or in the $^{39}Ar/^{40}Ar$ whole-rock ages but the occurrence of which may be supported by partial alteration of whole-rock K-Ar ages. In addition, we emphasize that the occurrence of "third events" is supported by the data of all lead isotope investigators on soils and breccias, and we view this information as a most important discovery on Apollo 12 material, possibly second only in importance to the great antiquity of rock 12013.

INTRODUCTION

PRESENTED herein are the lead isotopic compositions and, as determined by isotope dilution, the uranium, thorium, and lead concentrations in some samples collected by the Apollo 12 mission (volcanic rocks 12009, 12021, 12022, 12035, 12038, 12052, 12063, 12064; igneous breccia 12013; fines 12033 and 12070; and impact breccia 12034). The new data presented here on samples collected by the Apollo 12 mission are compared primarily with data for samples returned by the Apollo 11 mission that has been previously discussed by TATSUMOTO and ROSHOLT (1970) and TATSUMOTO (1970a) to facilitate uniformity of the data used in the comparison. Certain aspects of the data on the igneous breccia 12013 are included here that were not previously included (i.e., analysis of sawdust from 12013) or that were not thoroughly discussed (i.e., data on fragment #09B) by TATSUMOTO (1970b) owing to the short time available for writing that manuscript.

DATA

Analytical procedures and accuracy

Certain facets of the U-Th-Pb dating of lunar samples conflict with the dating by other techniques. Some investigators have been tempted to attribute the conflict entirely to the U-Th-Pb system and due to analytical problems at least in part. Accordingly, a rather complete description of the aspects of the analytical procedures in question and the analytical accuracy seems warranted.

Sample preparation. All samples except the igneous breccia 12013 and the soils were first briefly washed with acetone by use of an ultrasonic vibrator. Fragments #42 and #09 from rock 12013 and the soil samples were analyzed without any precleaning. Sawed surfaces on fragment #45 of rock 12013 were abraded with a dental drill and washed briefly with double-distilled acetone using an ultrasonic vibrator. For most samples, a small chunk was crushed in a boron carbide mortar and one portion of the powder was used for determination of the lead isotopic composition and the other for determination of the uranium, thorium, and lead concentrations. Fragment #45 of rock 12013 was treated directly without crushing. Rock 12064 was processed by use of a tungsten carbide mortar rather than a boron carbide mortar. The whole-rock analysis of this sample was made on about 4.4 g of the sample, and the density separations were made on the remaining 13 g. The portion used for density separations was analyzed by the same procedures given previously by TATSUMOTO (1970a, p. 1596) except that the isodynamic separator used on sample 12064 was that in the clean mineral separation room of L. T. SILVER at California Institute of Technology. The sample weights of the density fractions are:

10084, fraction 0 (acetone suspension, Apollo 11 samples only)--0.2196 g, hand magnet fraction weight on this sample was not record; sample 12064, hand magnet fraction--0.008g, fraction 1 (acetone suspension)--0.1216 g, fraction 2 (bromoform floats)--2.4261 g, fraction 3 (methylene iodide floats--1.9635 g, fraction 4 (methylene iodide sinks, nonmagnetic)--5.8065, fraction 5 (methylene iodide sinks, magnetic)--1.6460 g; sample 12033, hand magnet fraction--a few grains, fraction 1--0.1226 g, fraction 2--0.1029, fraction 3--1.0351 g, fraction 4 (total methylene iodide sinks)--0.2510; sample 12070, hand magnet fraction -- a few grains, fraction 1--0.1816; fraction 2--0.2154; fraction 3--0.9758 g; fraction 4 (methylene iodide sinks)--0.2510.

Density fractions of samples of rocks 10017 and 12064 and soils 10084, 12033, and 12070 were made by the same techniques as those previously described, which utilized immediately vacuum-predistilled heavy-density reagents and doubly-distilled acetone (TATSUMOTO, 1970a). There are still some reasons, nevertheless, to suspect lead contamination from the heavy liquids. For Apollo 12 materials, fraction 1 refers to material that remained in suspension in acetone after 2 minutes of ultrasonic agitation. On Apollo 11 (TATSUMOTO, 1970a, p. 1600), fraction 1 refers to bromoform floats, which is equivalent to fraction 2 on Apollo 12 materials; however, the acetone suspension was made on Apollo 11 samples. Analysis of this fraction on soil 10084 will be reported here.

Analysis. The lead contents of the lunar rocks are so little that we have had to develop new analytical techniques that have lowered the level of lead contamination to 4 to 7 nanograms (10^{-9} g) per analysis (TATSUMOTO, 1970b). Even so, this level of contamination is great enough that it still limits the size of the samples which can be analyzed. Dissolution of samples was made by a $HClO_4$-HNO_3-HF mixture in a Teflon digestion bomb followed by treatment of the residue with HNO_3 and HF in a Teflon beaker in a Teflon hood under nitrogen atmosphere. In most concentration determinations by isotope dilution, the ^{204}Pb, ^{207}Pb (and, for density fraction 1 of 12064 and 12033 and fragment #09B of rock 12013, the ^{208}Pb), and the ^{235}U, and ^{230}Th "spikes" were added to the bomb along with the powdered samples. Fragments #09A, #42, and #45 of rock 12013 were taken into solution first, then aliquoted, and spiked. Tests have been made on spiking before and after dissolution on oceanic tholeiites (which have lead contents comparable to those of lunar samples) and the reproducibility of the U/Pb is about 0.3 percent of the ratio. Therefore, there should be no problem of equilibration of the sample with the "spike," but there always is a small amount of white precipitate left in the lunar sample dissolution (metatitanic acid?) so that some problems in determinations of concentrations cannot be completely ruled out, nor can it be predicted at this time whether the effect would result in values of U/Pb or Th/Pb that are too great or too small. The bombs were also tested for memory by making sequential runs of ^{204}Pb and ^{207}Pb spikes usually used for the blank determination followed by ^{208}Pb spike, but no memory was observed.

The lead, which is coprecipitated from concentrated HNO_3 by $Ba(NO_3)_2$ and dissolved in water, is electrodeposited onto a platinum anode, converted to the phosphate, and is then analyzed by the silica gel-phosphate method (TATSUMOTO, 1970b). Early in the use of this technique the electrodeposition was sometimes not too efficient and the lead blank from the silica gel-phosphate method (0.03 ng) was more significant than it was later (where this blank was significant will be noted in the text where appropriate). The standard deviations of the mass spectrometric measurements of the lead isotope ratios are less than 1 percent for ^{206}Pb/^{204}Pb and less than 0.1 percent for ^{207}Pb/^{206}Pb and ^{208}Pb/^{206}Pb. The overall lead blank is about 5 ng; this amount leads to an uncertainty in the lead isotope ratios for most samples of less than 3 percent.

Trace elements

The lead, uranium, and thorium concentrations in the Apollo 12 samples are shown in Table 1. The samples are basaltic and doleritic rocks whose concentrations range from 0.270 to 0.653 ppm Pb, 0.157 to 0.404 ppm U, and 0.615 to 1.41 ppm Th. In addition, we analyzed two fines and two breccia samples, of which rock 12013 is called an igneous breccia and 12034 is an impact breccia (WARNER, 1970). The lead, uranium and thorium concentrations in the impact breccia and the fines of Apollo 12 are about 10 or more times greater than those of the igneous rocks.

The concentrations of lead, uranium and thorium in the igneous breccia 12013 are exceedingly great--20 to 40 times greater than in the lunar igneous rocks--and, in addition, they reflect the extremely heterogeneous nature of the rock as previously stated (TATSUMOTO, 1970b). The values of Th/U reported by WAKITA and SCHMITT (1970) are low as compared to our values, their highest ratio of 3.0 is 10 percent less than our lowest ratio of 3.3. The difference in Th/U seems to be due to their systematically low thorium contents which appear to be low on the average by 10 to 20 percent as compared to our values; however, the rock does seem to be heterogeneous in Th/U. The LSPET (1970) value on the whole rock is 3.2, and the possibility remains that we analyzed fragments greater in Th/U than the average and that WAKITA and SCHMITT analyzed fragments less than average in Th/U. Use of the low Th/U values with our lead values will, however, always give ages by Th-Pb methods lower than U-Pb ages. The newly analyzed sawdust must be contaminated with lead from some source apparently different from the copper wire used in the wire saw inasmuch as the sawdust contains 2.65 percent copper and the saw wire only 0.6 ppm lead, values which would result in a negligible lead addition. More likely the contamination comes from the vacuum cleaner bags involved in vacuuming up the sawdust. If all the ^{204}Pb in the sawdust comes from contamination, the contaminant in the sample is 14.8 ppm (versus about 0.2 ppm common-type lead in the three largest fragments analyzed from rock 12013) of the 25.3 ppm lead in the total sawdust sample. The isotopic composition of the contaminant lead in this calculation was assumed to be similar to that found in dust gathered from the filters of our clean laboratory ($^{206}Pb/^{204}Pb$: 18.48; $^{207}Pb/^{204}Pb$: 15.73; $^{208}Pb/^{204}Pb$: 38.38; T. J. CHOW, written commun., 1970). On this basis, the corrected lead content is found to be comparable to that in the fragments. The sawdust was thought to be a representative, homogenized sample of this igneous breccia; however, the uranium and thorium concentrations in the sawdust (two elements that are probably not contaminated) are 5 and 17 ppm respectively, contents that are about half the reported values obtained by gamma spectrometry as reported by LSPET (1970) on the whole rock and about 10 to 20 percent lower than the concentrations reported in other chips by TATSUMOTO (1970b). Rather than attributing the low results to dilution from extreme contamination (to avoid this, considerable time was spent in handpicking fibers from the sawdust which were probably introduced by the vacuum cleaner bags), we feel that uranium- and thorium-rich small particles (such as phosphates) were probably not recovered in the sawdust. At any rate, as analyzed, the sawdust is not a representative sample of the igneous breccia; and data on it must be interpreted with care.

A comparison of the lead, uranium, and thorium concentrations between samples returned by the Apollo 11 and Apollo 12 missions is given in Table 2. The contents of the three elements and the value of Th/U for the Apollo 12 mission rocks are similar to those of the Group II rocks from the Apollo 11 mission (potassium poor) but distinctly lower than those of the Group I rocks returned by the Apollo 11 mission (which are highly potassic). The Group I and Group II classification used here is adopted from COMPSTON _et al_. (1970). The lead, uranium and thorium contents of impact breccia and fines from Apollo 12 are two to five times greater than in the equivalent rock types from Apollo 11, but are between those contents of the igneous rocks and the igneous breccia 12013 suggesting that the fines and impact breccia are a mixture of rocks and the igneous breccia. SCHNETZLER et al. (1970) and SCHNETZLER and PHILPOTTS (1970) have pointed out that the dark phase of rock 12013 is a good candidate for the non-meteoritic "KREEP-like component" in the lunar soils. WOOD et al. (1971) has also identified crystallized material of similar composition in the coarse fines of the Apollo 12 mission and calls them norite, but some of this norite has as much as 2 percent K_2O. Potassium-rich, rare earth element-rich, and phosphorous-rich glass and rock fragments (KREEP material) which were also reported at the Conference (particularly by MEYER et al., 1971; HUBBARD et al., 1971), are rather similar in composition to the dark phase of rock 12013. Most investigators have compared KREEP-like material with common terrestrial basalts; however, the major element compositions of these materials are rather equivalent to those of more unusual terrestrial basalts called shoshonites (basalts with coexisting plagioclase and sanidine in the groundmass). Analyses of two samples of Cenozoic shoshonites for the U-Th-Pb system have been given by PETERMAN et al. (1970) [2.56 and 2.63 ppm uranium, 9.54 and 11.01 ppm thorium, and 22.6 and 20.0 ppm lead]. Comparison of the shoshonite data with those on the dark phase of rock 12013 indicates that uranium and thorium in rock 12013 is enriched relative to the shoshonite data available by a factor of about 2, whilst the common lead content still follows the lunar depletion pattern found for the other igneous rock types and is depleted in rock 12013 by about a factor of 100 relative to the amount in the available shoshonite data (the lead in lunar samples is mainly radiogenic and the common lead component in fragment 12013 is only about 0.2 ppm). This lead depletion factor is actually larger than that found for other lunar rock types as compared to their terrestrial equivalents which is about a factor of 10 to 20.

The lead content of lunar samples is sufficiently small that some workers have attributed all the common lead found to contamination at some stage of sample handling on the moon, in the Lunar Receiving Laboratory, or in the lead isotope analyses procedures. Thallium, for example, which like lead is a "volatile element" that is less abundant on the moon than on the earth, is found on the moon (0.3 to 1.1 ppb; GANAPATHY et al., 1970) at about 1/10 to 1/100 that in terrestrial basalts (about 20 ppb; GANAPATHY et al., 1970). Indeed, our computed common lead contents of lunar rocks are just about the values expected from the thallium depletion factor. We have reason to believe, therefore, that a common or initial lead correction of some sort is required for the lunar samples.

Isotope ratios

The isotopic compositions of Apollo 12 samples (Tables 3 and 4) are extremely radiogenic and the observed $^{238}U/^{204}Pb$ values--so-called μ values--range from 470 to 2700. These great μ values indicate that the volatile element lead was greatly depleted relative to uranium and thorium on the lunar surface as compared to that on the surface of the earth, as has been shown in the Apollo 11 papers (TATSUMOTO, 1970a; SILVER, 1970; GOPALAN et al., 1970). The isotopic compositions of the rocks fall into two groups at least: one group (12009, 12022, 12035, 12038, and 12064) that has a value of about 300 to 450 for $^{206}Pb/^{204}Pb$ and $^{208}Pb/^{204}Pb$ and another group (12021, 12052, and 12063) that has values of about 700 for the $^{206}Pb/^{204}Pb$ ratio and 650 to 800 for $^{208}Pb/^{204}Pb$. Sample 12070 ("contingency fines") has a value of

$^{206}Pb/^{204}Pb$ intermediate between the values for the two groupings of igneous rocks, but fines sample 12033, the impact breccia 12034, and the igneous breccia 12013 have exceedingly great values for the ratio.

A comparison of the isotopic compositions of the Apollo 12 samples with those of Apollo 11 is given in Table 5. The two groupings of Apollo 12 rocks by $^{206}Pb/^{204}Pb$ and $^{208}Pb/^{204}Pb$ are rather similar to those in rocks from the Apollo 11 mission where the Group I (high-potassium) rocks have values of $^{206}Pb/^{204}Pb$ and $^{208}Pb/^{204}Pb$ that are greater than 400 and the Group II (potassium-poor) rocks have values for $^{206}Pb/^{204}Pb$ of about 300 to 400 and for $^{208}Pb/^{204}Pb$ of 300 to 450. No parallelism is found, however, in the lead, uranium and thorium contents of the Apollo 12 rocks. WARNER and ANDERSON (1971) have further subdivided the Group II rocks into porphyritic basalts (also olivine-rich) [of which 12009, 12021, 12022, and 12052 are included in this study] and granular or ophitic basalts (also olivine-poor) [of which 12035, 12038, 12063, and 12064 are included in this study]; however, no correlation of $^{206}Pb/^{204}Pb$ with this subgrouping is found, either. Two of four rocks from the Apollo 12 mission classified as granular and ophitic (Group II for Apollo 11 material) are in the great $^{206}Pb/^{204}Pb$ category, whereas one of four porphyritic basalts is in the low $^{206}Pb/^{204}Pb$ category. Apparently the subdivision of Group II is not of significance in the U-Th-Pb system.

DISCUSSION

Isochron age methods

There are four isochron techniques of dating in the U-Th-Pb system, three of which are independent--two of three for the U-Pb system (^{207}Pb/^{204}Pb--^{206}Pb/^{204}Pb, ^{207}Pb/^{204}Pb--^{235}U/^{204}Pb, ^{206}Pb/^{204}Pb--^{238}U/^{204}Pb) and one for the Th-Pb system (^{208}Pb/^{204}Pb--^{232}Th/^{204}Pb). The Pb-Pb isochron, which was the first dating technique to give precise ages of meteorites, will be discussed first because of the simplicity of the method, its proven reliability, and the insensitivity of the method to a variety of analytical uncertainties. This discussion is followed by discussions of the ^{206}Pb/^{204}Pb-^{238}U/^{204}Pb, ^{207}Pb/^{204}Pb-^{235}U/^{204}Pb, and ^{208}Pb/^{204}Pb-^{232}Th/^{204}Pb isochrons which are analogous to the ^{87}Sr/^{86}Sr-^{87}Rb/^{86}Sr or BPI (Bernard Price Institute) isochron dating techniques.

Pb-Pb isochron

Owing to the fact that ^{238}U/^{235}U in lunar and terrestrial bodies is, for all practical purposes, a physical constant of 137.8 (see ROSHOLT and TATSUMOTO, 1970, for measurements on lunar samples), the Pb-Pb isochron has several advantages over other isochron methods. For example, this dating technique is not sensitive to analytical uncertainties in the determination of U/Pb and Th/Pb, and, in fact, the uranium, thorium, and lead contents need not be determined at all! As in all isochron treatments, no initial lead composition need be determined or estimated if several coeval samples with the same lead isotopic compositions are available that had a spread in their ratios of radioactive parent to the stable isotope of the daughter element, ^{204}Pb, for long periods of time. Even better, isochron lines that pass near the origin as do those for many lunar samples are insensitive to blank correction because the lunar samples are very old and highly radiogenic, whereas the isotopic composition of the blank lead is always near the origin (however, our blanks on whole-rock and soil analyses are less than one percent the lead in the sample analyzed).

The only effect of the blank is to determine the position of the sample on the isochron, but it does not affect the slope (i.e., age) of the isochron. Lastly, the lunar samples are so old that errors in ^{204}Pb measurement (the least abundant lead isotope) also have little effect.

Naturally, the technique has some constraints. These are that all samples must be of the same age, have started with the same initial lead isotopic composition and have a spread in the lead isotope ratios. The linear relationship should not be the result of mixing.

If there is no spread in the isotope ratios--for example, if you have only one sample--an initial lead must be assumed (this statement was oversimplified in TATSUMOTO, 1970a, p. 1598). The samples also should not have evolved in greater than two-stage systems if the third and higher order stages occurred at times significantly greater than t = 0. If these constraints are not met, irregularities in the isochron development will probably occur. Other aspects of the Pb-Pb isochron will be discussed where appropriate.

In comparison of rock-lead dating with Rb-Sr dating on terrestrial materials, good agreement has generally been obtained between the two methods. ROSHOLT _et al_. (1970) get 1820 ± 110 m.y. for the Pb-Pb isochron and 1810 ± 50 m.y. for the Rb-Sr isochron on a sample of granite from Saskatchewan, Canada. The U-Pb and Th-Pb isochrons are also in good agreement with the ages determined by the other techniques. FARQUHARSON and RICHARDS (1970) report ages of 1553 ± 29 m.y. for the U-Th-Pb isochrons and 1565 ± 52 m.y. for the Rb-Sr isochrons for a microgranite and pegmatite from Mount Isa, Australia. PETERMAN _et al_. (1971) give ages of 2950 ± 110 m.y. for the Pb-Pb isochron and 2925 ± 80 m.y. for the Rb-Sr isochron on a paragneiss from the Granite Mountains, Wyoming, and their age of 2610 ± 70 m.y. by the Rb-Sr isochron is within analytical uncertainties of the 2820-m.y. age obtained by the Pb-Pb and Th-Pb isochrons (ROSHOLT and BARTEL, 1969) on a recently altered granite that has also suffered metamorphism and intrusion at about 1600 m.y. (ROSHOLT *et al.*, 1969; PETERMAN *et al.*, 1971).

Rocks. All the samples analyzed from the Apollo 12 mission are plotted in Fig. 1, and an enlarged diagram of part of Fig. 1 is given on Fig. 2. The data on Fig. 1 are dominated by those for the igneous breccia 12013 and discussion of this sample first will help to illustrate the controversy existing between the different kinds of dating techniques. All samples of 12013, including the sawdust, lie very close to an isochron of 4370 m.y. that passes near the common lead field *no matter whether the analyses are or are not corrected for blank lead*, as if the lead initially present in the rock developed in a source with a small value of ^{238}U/^{204}Pb. Even though fragment #42 departs from the isochron beyond analytical uncertainties, the departure is not great and could be due to a minor process ~~transient effect~~. This age of 4370 m.y. is in conflict with the two ages given by the Rb-Sr dating system (4500 m.y. for a whole-rock isochron composed of dark fragments and 4000 m.y. for a mineral isochron; LUNATIC ASYLUM, 1970) and the age given by the ^{39}Ar/^{40}Ar method (3900 m.y.; TURNER, 1970b). In any controversy about the dating, however, we should not forget that all data indicate the sample is old, probably not younger than 4000 m.y. nor much older than 4600 m.y. Although the 4370-m.y. age by the Pb-Pb isochron is a valid age estimate, the intermediate age shown by the method relative in Rb-Sr dating could be explained if the $^{238}U/^{204}Pb$ that developed in each fragment at 4000 m.y. and existing to the present (second stage) is exactly proportional (enriched in ^{238}U over ^{204}Pb by a factor of about 2) to the $^{238}U/^{204}Pb$ existing in all fragments except #42 between 4600 m.y. and 4000 m.y. (first stage). For fragment #42, the $^{238}U/^{204}Pb$ in the second stage is enriched in ^{238}U over ^{204}Pb by a factor of about 2.4 and therefore departs only a little from that proportionality constant that governs the other samples. In this approach to resolving the age discrepancy, the values of $^{206}Pb/^{204}Pb$ and $^{207}Pb/^{204}Pb$ in the source of the magmas also cannot have been homogenized 4000 m.y. ago. If such systematics are the answer to the age discrepancies, then we feel that the most logical interpretation of the history of rock 12013, of the two interpretations given from Rb-Sr dating (LUNATIC ASYLUM, 1970), is that the entire rock was formed at one time (perhaps 4500 m.y.) and internally reconstituted by some thermal event (impact?) at a later time (probably 4000 m.y.) rather than that an old gabbro was intruded by a younger granite. This kind of interpretation is also supported by the petrographic study of JAMES (1971). If such regularities as described above are considered in the U-Pb system, they should also be considered in the Rb-Sr system. For example, if some lead was volatilized from 12013 in a proportional manner relative to uranium by a heating event, volatilization of some rubidium relative to strontium in a proportional manner is also reasonable (CLIFF *et al.*, 1971, report volatilization of both lead and rubidium). Such a process would result in an age of rock formation that is too young on the Pb-Pb isochron and too old on a Rb-Sr isochron (see discussion in SILVER, 1970, p. 1571). If the age of rock 12013 is older than that given by the Pb-Pb isochron (4370 m.y.), it is also probably younger than that given by the Rb-Sr "whole-rock" isochron (4500 m.y.). The best estimate of the age of formation of rock 12013 seems therefore to be in the range 4370 to 4500 m.y.

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In the rocks collected by the Apollo 11 mission, the lead isotope data plot into two groups. Those with the greater value of $^{206}Pb/^{204}Pb$ are the Group I, high-potassium basalts and those with the lesser values of $^{206}Pb/^{204}Pb$ are the Group II, low-potassium basalts. As was found for rock 12013, an isochron line connecting the two groups of samples also passes through the common lead field with an indicated age of close to 4200 m.y. (Fig. 2), again as if the initial lead developed in a source with a small value of $^{238}U/^{204}Pb$. (The data on Fig. 2 of this paper has not been corrected for initial lead. TATSUMOTO (1970a) had corrected for a radiogenic initial lead on his Fig. 3 where he obtained a 4000-m.y. age.) The 4000-m.y. age given by TATSUMOTO (1970a) is a lower limit on the calculated age. Note that if one of the two groups of basalt is on the 3980-m.y. isochron (by using its observed $^{206}Pb/^{204}Pb$ but changing the $^{207}Pb/^{204}Pb$ until the point falls on that isochron) and the other remains on the 4200-m.y. isochron, a line connecting the two groups would not pass close to the common lead field. We, therefore, hardly appear to be mixing rocks of significantly different ages. Just as in rock 12013, because of the arrangement of the data, we would quite confidently interpret the rocks as being 4200 m.y. old in the absence of other information, and that they started with a nonradiogenic initial lead isotopic composition similar to terrestrial common lead. Again, however, the lead isotope data are in conflict with the age of the rocks determined by other means, strongly so for Group I basalts because the value of Rb/Sr does not appear to be close to equal to that in its source (the whole-rock Rb-Sr data for the Group I basalts do not lie near the 4500-m.y. whole-rock isochron). As for rock 12013, the Pb-Pb isochron age is older than the Rb-Sr mineral isochron age (PAPANASTASSIOU *et al.*, 1970) and the $^{39}Ar/^{40}Ar$ whole-rock age (TURNER, 1970a). The lead isotope data could be also interpreted to agree with the ages determined by other methods through assuming that the values of $^{238}U/^{204}Pb$

in the rocks are proportional to those values in the source material for the rocks, as for rock 12013. In this interpretation, the enrichment factor for ^{238}U/^{204}Pb of Apollo 11 rocks would be about 2.7 for all rocks (using an age of formation of 3800 m.y.). This is the interpretation preferred by COMPSTON et al. (1970) and considered by SILVER (1970) and by TATSUMOTO (1970a, p. 1606) in his concordia discussion for these rocks. The disturbing thing about this interpretation is the precise enrichment factor required. We have to choose, then, between two rather unsatisfying hypotheses: one, that the rocks are about 4200 m.y. old and that they had a nonradiogenic initial lead; and the other, that the rocks are of some age (perhaps between 3600 and 3800 m.y. old), and that they had a very precise enrichment factor for ^{238}U/^{204}Pb. Of the two hypotheses, we still tend to favor the first (4200-m.y. age); because of the high quality of the isochron and because ~~because~~ several lines of information suggest that the initial lead in the rocks may not have been very radiogenic. We realize that this interpretation leaves a conflict with those arrived at by other dating techniques.

Consideration of one greater aspect in complexity of the Apollo 11 rocks is instructive. Suppose that the Group I rocks are about 200 m.y. younger than the Group II rocks (for our calculations, 3600 m.y. rather than 3800 m.y.). A Pb-Pb isochron for all rocks that goes through the common lead field could still be obtained by judicious regulation of the values of ^{238}U/^{204}Pb between the time of rock formation and the present. For this case, the enrichment factor in ^{238}U/^{204}Pb for the Group II rocks would still appear to be about 2.7 as before, but for the Group I rocks it would be only about 2.4. Amazingly, the enrichment factors would compensate for the age difference just the right amount to allow the isochron to still pass through the common lead field rather than above it or below it. This aspect is better illustrated in Apollo 12 rocks.

The lead isotopic data for rocks returned from the Apollo 12 mission (other than rock 12013) contrast markedly to those for rock 12013 and to those for rocks collected by the Apollo 11 mission because of the irregularity of the data on a Pb-Pb isochron plot. Data on six rocks lie near an isochron of about 3900 to 4000 m.y. that passes through the common lead field, but data on rock 12064 lie well above that isochron and those on rock 12035 lie well below it. These last two rocks clearly could not have had the same initial lead as the others, and they could be of a different age. If the samples are of about the same age (as suggested by the Rb-Sr mineral isochron from the LUNATIC ASYLUM (1971) and $^{39}Ar/^{40}Ar$ whole-rock data from TURNER (1971), the value of $^{238}U/^{204}Pb$ initially in the rock was also not proportional, at least for 12064 and 12035, to that in its source material. Note that a line passed either through 12064 or 12035 and through any other sample does not pass through the common lead field. Thus, in the analyzed rock assemblage from Apollo 12, there is clear failure in the assumptions necessary for using the Pb-Pb isochron. The data do establish, however, that proportionality of $^{238}U/^{204}Pb$ in a magma relative to the source of the magma is not necessarily the rule. It may be worth noting that the isochron that passes through the bulk of the data and the common lead field indicates a younger age than that found for the rocks collected by the Apollo 11 mission which in turn are younger than rock 12013. The order is the same as that established by either of the other dating methods and there is some similarity to the intervals between apparent ages. We know of no obvious criteria to cause us to discard either rock 12064 and 12035 from the Pb-Pb isochron treatment, and conclusions regarding Apollo 12 rocks achieved solely by this technique are suspect. Sufficient sample was obtained for rock 12064 to permit some investigation of it for a possible internal isochron.

Density fractions on rocks 10017 and 12064

The isotopic compositions of the density fractions separated from rocks 10017 (TATSUMOTO, 1970a) and 12064 (Table 4) are plotted on Fig. 3. Both sets of density-fraction data have a considerable spread in their isotopic ratios that permit construction of Pb-Pb isochrons. In both rocks the slope of the isochron is markedly less than the slopes for the whole-rock specimens, and the resultant apparent ages are 3950 m.y. for 10017 (versus 4200 m.y. for the whole-rock isochron of Apollo 11) and 3650 m.y. for 12064 (versus 3900 to 4000 m.y. for an isochron through primordial lead and the whole rock). A similar line drawn through the data on a concentrated HNO_3 leach of 10017 and its residue indicates an apparent age of about 3950 m.y., similar to the age on the density fractions on the same sample obtained by TATSUMOTO (1970a). While these ages are closer to the Rb-Sr mineral isochron ages (3400 m.y. for Apollo 12 rocks by PAPANASTASSIOU and WASSERBURG, 1970) and $^{39}Ar/^{40}Ar$ whole-rock ages (TURNER, 1971), they are still high by 5 to 10 percent. A disturbing feature of both Pb-Pb isochrons arising from density fractions is that neither isochron includes its respective whole-rock analysis. Such a relationship could occur in nature if the whole rock contained a relict phase rich in lead that either was lost ~~as~~ in the unanalyzed hand-magnet fraction, lost as very fine particles that remained in suspension (a candidate might be phosphate minerals), or ~~was~~ dissolved and lost in the heavy liquids (the sum of the leads in the density fractions does not equal the lead content of the whole rock). Unfortunately, all heavy liquids were combined after use, so this explanation cannot be tested. Even though the liquids were distilled before use, the heavy liquids more likely have contributed a greater lead blank to the density fractions than we have estimated.

Fraction 2 of rock 10017, for example, lies above the density-fraction isochron that is described by the rest of the fractions, and it lies along a line that connects the whole rock with the common lead field. This relationship suggests difficulties in blank correction. The fact that all density fractions are less radiogenic in $^{206}Pb/^{204}Pb$ than their whole rocks is also compatible with a contamination problem. The greater age for the Pb-Pb isochron is suggestive that the amount of lead contamination is inversely proportional to the observed $^{206}Pb/^{204}Pb$ values in the fractions.

The density-fraction data would have agreed with the Rb-Sr data had they fallen on a line of 3600-m.y. slope for 10017 that passes through the whole-rock value, and on a line of 3400-m.y. slope for rock 12064 that passes through the whole-rock value. Considering the uncertain state of the blank, we cannot now say that such is not true. The approximate values of $^{206}Pb/^{204}Pb$ for which lines through the common lead field and the observed density-fraction data intersect the isochron of approximate age that includes the whole-rock value are: for 10017, fraction 1: 930; 2: 400 (same as the whole rock), 3: 540; and 4: 670; and for 12064, fraction 1: 300; 2: 600; 3: 900; 4: 1100; and 5: 1400. The data do support the concept that the mineral-fraction data lie on an isochron of less slope, and therefore of younger age, than the isochron for the whole rocks. Otherwise, all the density-fraction data would lie along lines connecting the whole rock to the common lead field.

The contamination explanation does seem to require two baffling coincidences, however, that continuously seem to be needed to obtain ages compatible with the Rb-Sr mineral isochron and $^{39}Ar/^{40}Ar$ whole-rock ages. First, the contamination during our mineral separation must have occurred in a rather regular manner for the density-fraction data to retain linearity. Both random scatter of data and even negative slopes of isochrons are possible. Second, the data of SILVER (1970) on the whole-rock 10017 lie near our density-fraction isochron for 10017 and his data on whole-rock 12064 (SILVER, 1971) plot very near our mineral isochron for rock 12064. Both of Silver's whole rocks do lie near lines connecting our respective analyses and the common lead field so that coincidental amounts of contamination could account for the differences--but this could hardly have been predicted. Therefore, the presence of a randomly distributed relict component cannot yet be completely abandoned. Also, we are not yet prepared to say whether the lead-isotope data on density fractions either agree or disagree with the ages determined by other methods.

Soils

The lead-isotope data on soils 12033 and 12070 and breccia 12034 returned by the Apollo 12 mission are shown on Figs. 1 and 2. All these soil and breccia data clearly lie below the isochron drawn between the Apollo 11 soils and breccia and the common lead field. There is perhaps some irony in the fact that the model whole-soil age reported by PAPANASTASSIOU et al. (1970) of 4670 m.y. is in excellent agreement with the Pb-Pb isochron age (4660 m.y.) on the Apollo 11 soils by TATSUMOTO (1970a) inasmuch as the soil and breccia are clearly complex material that contain components from local rocks, rock detritus thrown in by impacts at some distance (including

(21a follows)

a "KREEP-like component"), and meteorites. If such agreement is surprising for these complex materials, the irony is only compounded by the excellent agreement between the model "whole-soil" Rb-Sr age on soil sample 12070 of 4440 m.y. (PAPANASTASSIOU and WASSERBURG, 1970) and the Pb-Pb isochron model age determined by us of 4470 m.y. Other soil (12033) and breccia (12034) samples from the Apollo 12 mission are younger in their model Pb-Pb isochron ages, down to 4200 m.y. The close agreement is probably more representative of excellent mixing of components in the soil rather than of the age of soil formation. (There is some conflict in the model Rb-Sr ages reported on these samples in the unpublished proceedings of the Apollo 12 Lunar Science Conference. Both MURTHY *et al.* (1971) and CLIFF *et al.* (1971) state that 12070 has a model age of 4600 m.y. within their analytical uncertainties.) For 12033, MURTHY *et al.* give a model age of 4200 m.y., which is satisfyingly close to the Pb-Pb model age of 4320 m.y.(6)

Density fractions on 10084, 12070, and 12033

Density separations were carried out on three soil samples--10084 from the Apollo 11 mission (TATSUMOTO, 1970a) and 12070 and 12033 from the Apollo 12 mission. Soil data are plotted for 10084 and 12070 on Fig. 3 and for 12033 on Fig. 1. The data on the Apollo 12 soils are included in Table 3 and those for 10084 may be obtained from the original report (TATSUMOTO, 1970a). The data on 10084 lie reasonably close to the 4650-m.y.-old isochron drawn through primordial lead and the whole-soil. The new analysis of the acetone suspension fraction of 10084 (Table 4) is less radiogenic than the whole soil and is in good agreement with the most easily leachable lead reported by SILVER (1970). Soil 12033 gives a Pb-Pb isochron in agreement with the primordial lead-whole-soil Pb-Pb isochron. Sample 12070 contains two fractions that express the heterogeneity of this soil sample. Fraction one (acetone suspension) could represent a mobilized lead component such as that postulated by SILVER (1970) and TATSUMOTO (1970a, p. 1606) and fraction 3 could represent some of the KREEP-like component. These components apparently balance each other in the whole soil.

Parent-Daughter Isochron Relationships

Unlike the Pb-Pb isochron, whole-rock lead isotope data on U-Pb and Th-Pb diagrams (Fig. 4) do not define any definite isochrons, although the ages involved are clearly very old. The reasons for the scatter are not entirely understood. We feel at this time that the scatter is more likely to be due to volatilization of some lead from the rocks caused by the relatively recent impact event responsible for the rock now being on the surface rather than to exceedingly great analytical uncertainties. However, the impact may cause redistribution of the lead within the rock rather than volatilization of lead from the rock. If this surmise is correct, some parts of the rock may have become enriched in lead, and others may have been depleted of lead.

Conceivably, we might learn something by passing "best-fit" lines through the scatter of the data. For rock 12013, the ages are then indicated to be about 3900 m.y. for the ^{238}U-^{206}Pb isochron, 4200 m.y. for the ^{235}U-^{207}Pb isochron, and 4000 m.y. for the ^{208}Pb-^{232}Th isochron, the limiting uncertainties are drawn on Fig. 4. The average of these crude ages (4030 m.y.) is surprisingly close to the ages determined by the Rb-Sr mineral isochron (LUNATIC ASYLUM, 1970) and ^{39}Ar/^{40}Ar methods (TURNER, 1970b). When the same best-fit lines are drawn through the scatter of the data on the density fractions for rock 10017 (TATSUMOTO, 1970a), the ages are about 3660 m.y., 3950 m.y., and roughly about 2390 m.y. (little variation in the ^{208}Pb/^{204}Pb), respectively, in the same order as given for 12013. The average of the U-Pb isochrons (3800 m.y.) is in general agreement with the Rb-Sr mineral isochron ages (PAPANASTASSIOU <u>et al</u>., 1970) and the ^{39}Ar/^{40}Ar whole-rock ages (TURNER, 1970a). For rock 12064 density fractions, these ages are 3320 m.y., 3550 m.y., and 3570 m.y., respectively. Again, the average isochron age (3480 m.y.) is in remarkable agreement, considering the scatter, with the Rb-Sr mineral isochron generally observed in Apollo 12 rocks (COMPSTON <u>et al</u>., 1971; MURTHY <u>et al</u>., 1971; CLIFF <u>et al</u>., 1971; LUNATIC ASYLUM, 1971) and ^{39}Ar/^{40}Ar whole-rock ages (TURNER, 1971).

The density-fraction isochron data on the Apollo 11 rocks, the rock 12064 and igneous breccia 12013, although not of as good quality as the whole-rock data, do appear to support the contention that we may be dealing with an acute initial lead problem with some coincidental proportionalities of the ^{238}U/^{204}Pb values in the magma and its source rocks, but we feel that the data are not of sufficient quality to compel this conjecture.

CONCORDIA AGE RELATIONSHIPS

Rocks

The U-Pb system is plotted on a U-Pb concordia diagram (Fig. 5). Using this diagram requires knowledge of the uranium and lead contents as well as of the lead isotopic composition. An initial lead must also be determined or assumed. This last requirement is not as difficult to meet for lunar samples as might be imagined, inasmuch as the initial lead in the magmas is primordial lead plus some radiogenic addition. The systematics of the U-Pb system are such that correction of the observed ratios for primordial lead will give a data point that still lies on the straight line drawn between the age of the source material and the time of formation of the magma but to the right of the position where it would lie if corrected for the true initial lead. This line, called a discordia line, is drawn for times representing the two limiting conditions of the data, i.e.-between 4650 and 3400-3900 m.y. The Apollo 11 rocks lie in one patch and the Apollo 12 rocks (except for 12035 and 12064) in another patch. Though there is not a straightforward relationship between the ^{206}Pb-^{207}Pb isochron and the concordia diagram, the two patches reflect the younger age shown by the ^{206}Pb-^{207}Pb isochron for the Apollo 12 rocks relative to that for the Apollo 11 rocks. For the concordia ages to agree with the ^{40}Ar/^{39}Ar and internal Rb/Sr isochron ages, data for rock 12013 should lie on a line between about 4650 m.y. and 4,000 m.y., rocks of Apollo 11 near a line between 4650 m.y. and about 3700 m.y., and rocks of Apollo 12 near a line from 4650 m.y. and about 3400 m.y. assuming that the age of all the source rocks for these materials is a primordial source and that only two stages have been involved. The data do not clearly reflect what was expected because rock 12013 data are on a distinctly different discordia line and the data for Apollo 11 rocks lie on or below a 3900- to 4650-m.y. discordia line. Note that the sawdust from 12013 lies very near the concordia curve when corrected for modern lead ($^{206}Pb/^{204}Pb$: 18.8; $^{207}Pb/^{204}Pb$: 15.8; $^{208}Pb/^{204}Pb$: 38.8) as the initial lead in spite of the abundant lead contamination. There seem to be two groups in the Apollo 12 igneous rocks (except 12035 and 12064) as shown by the dashed lines in Fig. 5. The groupings also shown in the parent-daughter diagram (U-Pb, Th-Pb), may indicate that at least two parent magmas were involved. The data for Apollo 12 rocks scatter considerably about the 4650- to 3400-m.y. discordia line but they may lie within all the uncertainties as estimated from the multiple analyses of 12063 and 12052 (sample inhomogeneity plus laboratory treatment) that affect the data of the 4650- to 3400-m.y. discordia line. Rock 12035 lies drastically below such a discordia line, but we are somewhat concerned about this sample. It is very friable and perhaps some key material in the U-Th-Pb system was lost by the third events (impact?) or in the collection and processing of the material. Such loss would be an episodic event at time zero and would move the data point towards zero. The loss cannot account for the lateral offset of 12035 from the grouping of six analyses.

If it is assumed that all rocks are concordant and that the problem is one of not knowing what the initial lead really was in the rock, the initial lead and $^{238}U/^{204}Pb$ in the source rocks may be calculated (Table 6). If the uranium content in each of the magmas and source rocks is assumed to be constant, then the Apollo 12 rocks have lost about five-sixths of their lead relative to that in the source rocks. For comparison, Apollo 11 rocks have lost about two-thirds of their lead relative to that in source rocks.

As pointed out previously (TATSUMOTO, 1970b), the data on rock 12013 clearly do not fit the 4650- to 3400- or 3900-m.y. discordia lines but seem to reflect some event much younger than 3000 m.y. and are evidence of third event systems on the moon (it should be kept in mind that chiplet #09B was a separate piece from chip #09A, both of which came from fragment #09).

Density Fractions on Rock 12064

The data on rock 12064 are given in Fig. 6. The density fraction data for this rock appear to lie reasonably well along a 4400- to 2500-m.y. trend that would also include rock 12035 reasonably well. The 4400-m.y. intersection is also common to that for the igneous breccia 12013 and the impact breccia 12034 and the soil 12033. Though we expected that the data would lie on a 4650- to 3400-m.y. discordia line, the data cannot easily be made to fit such a line. Fraction 1 (acetone suspension) lies well above it and fractions 3, 4, and 5 lie below it. Even though the observed arrangement of the data could be due to the possible contamination, previously discussed, if the degree of contamination is the reciprocal of the amount of sample handling, again we are surprised at the regularity of the data if contamination is the cause. The 2500-m.y. intercept, however, does not seem to be reflected in the results determined by any of the other treatments of the U-Th-Pb system or in those determined by the Rb-Sr mineral isochron or $^{39}Ar/^{40}Ar$ methods. The only other information suggesting an approximate age of 2500 m.y. is the K-Ar ages in the Apollo 12 preliminary report (LSPET, 1970), we, however, tend to interpret those apparent ages as intermediate ages between the time of volcanism and the younger age of impact throwout.

Soils

In contrast to the rocks of Apollo 11 and 12 and soils and breccias of Apollo 11 which seem to approximate single- (no alteration of $^{238}U/^{204}Pb$ since the moon formed) or two-stage development (line t_1-t_o in Fig. 7), the soils and breccias of Apollo 12 show distinct development of a third much younger stage (Fig. 5) and are more accurately represented by such lines as t_2-Q_1 in Fig. 7. All other lead isotope data reported in the unpublished proceedings of the Apollo 12 Lunar Science Conference are in agreement on this point (CLIFF _et al._, 1971; SILVER, 1971; and HUEY _et al._, 1971). The only alternative explanation is that some new phase, not dissolved or leached of its uranium and lead, is present in Apollo 12 soil and breccias but is not present in any other rock of either Apollo 11 or Apollo 12 or in Apollo 11 soil and breccia. No such phase was reported at the Second Lunar Sample Conference, and in fact, one possibly troublesome phase, ilmenite, is less in the Apollo 12 samples. Some unusual occurrences did happen in rock 12013 that are suggestive of a third event, as already discussed. In these events, the KREEP-like component either may be young, much less than 3000 m.y., or may have been altered at a young age. Third events are the most likely explanation for the configuration of the 12013 data and almost certainly is the explanation for the soil and impact breccia data from the Apollo 12 mission. We do not know the precise age or ages of the young, third-stage event or events;

conceivably, they might be related to formation of impact glasses and alteration of the U/Pb ratio in some rocks, such as 12013, by some relatively young impact such as Copernicus, a ray from which the Apollo 12 samples may in fact have been collected.

Density Fractions of 12033 and 12070

Some insight into the complexity of Apollo 12 soils and breccias may be given by the density-fraction data for soils 12070 and 12033 (Fig. 8). The data point of the extremely fine fraction of 12070 (fraction 1) falls nicely on a t_1-t_0 line. The data represented by this line could be for soil that might have been derived from 4550-m.y.-old source material at about 3200 m.y. ago at which time fraction 1 was enriched somewhat in lead over uranium. Fractions 3 (MI float) and 4 (MI sink) also fall on this line. Fraction 2 (BF float) falls on a line drawn through it and one other bit of fraction data that is much younger. The oldest intersection with concordia for this younger event would be a line connecting fractions 1 and 2 which would intersect concordia at about 1.5 b.y. Fraction 2 primarily contains feldspar and scoriaceous glass which may be a younger (or relatively recently altered) KREEP-like material (note the great lead, uranium and thorium concentrations for fraction 2 in Table 4). The behavior of the fractions from 12033 differs markedly from the behavior of the density-fractions of 12070. It is almost as if 12033 was largely reconstituted in terms of the U-Pb system at some fairly recent time rather than just containing some added definable component. Even though the soil and breccia data of Apollo 12 are interpreted only with great difficulty to any degree of precision, evidence clearly indicates a component greater than 4 b.y. in age and another component much younger than 3 b.y. in age. Neither the $^{39}Ar/^{40}Ar$ method nor the internal Rb/Sr isochron methods is reflecting this younger third event; however, these methods are applied to the soil only with great difficulty.

CONCLUSIONS

In lunar samples, differences in the ages obtained by the Rb-Sr and $^{39}Ar/^{40}Ar$ methods and those obtained by U-Th-Pb dating seem to be attributed, by some investigators, mainly to analytical problems in the U-Th-Pb system and complexities in the interpretation of that system. The Pb-Pb isochron is, however, the dating method least sensitive to analytical uncertainties. In addition, if the U/Pb and Th/Pb values in the volcanic rocks have been enriched over those in the isotopically inhomogeneous source rocks by volatilization of lead from the magma, depletion of the magma in rubidium relative to strontium is also likely. The general effects of such volatilization would be Pb-Pb whole-rock isochron ages that are younger than the age of the source rocks for the magma and Rb-Sr whole-rock isochron ages that are older than the age of the source rocks. If the high quality Pb-Pb isochron for the Apollo 11 rocks and rock 12013 is due to a coincidental constant enrichment factor for U/Pb in the volcanic rocks relative to the source rocks, the approximate agreement of the low-potassium whole-rock Rb-Sr isochron ages (which are interpreted as ages of the magma source rocks) with the "age of the moon" also may be a coincidence. We feel that the Pb-Pb isochrons on the Apollo 11 rocks and rock 12013 are of sufficiently high quality that abandonment of a 4200-m.y.-old age for Apollo 11 rocks and 4370-m.y.-old age for rock 12013 is premature. The lead isotope data on Apollo 12 rocks, other than 12013, does not form a simple Pb-Pb isochron so that ages must be derived from concordia diagrams. In the absence of other kinds of data, we would interpret the concordia relations as suggesting that the Apollo 12 rocks were derived about 3000 m.y. ago from a 4650-m.y.-old source. There is sufficient scatter in these data, however, to permit a derivation of the rocks at 3400 m.y. from a 4650-m.y.-old source, a derivation age that is in essential agreement with the Rb-Sr mineral isochron and ^{39}Ar/^{40}Ar whole-rock methods. Although the lead isotope data on the lunar soils are subject to alternative interpretations, the 4650 ± 50 m.y. age determined by U-Th-Pb methods for the Apollo 11 soils is probably a consequence of the Apollo 11 soils being a well-mixed composite of lead enriched and depleted materials, and the age still furnishes the best working hypothesis for the age of the moon. The most significant feature of the lead isotope data on Apollo 12 soils is evidence of "third events" in these soils that are much less than 3000 m.y. in age. These "third events" are probably attributable to formation of or volatilization of lead from components in the soil due to young impacts. These "third events" are not clearly shown in the ^{39}Ar/^{40}Ar or Rb-Sr dating methods.

A speculation on the origin of the moon is not discussed in this paper because of page limitations and because of the rapid advance of physical astronomy; however, our concept of the lunar origin is not much different from those of other investigators (for example, GANAPATHY <u>et al.</u>, 1970; ANDERS, 1970-1971) and the symposium on the evolution of the solar system (SHIMAZU, 1967). This concept is that the moon originated from the beginning as a satellite of the earth in the process of solar evolution.

The radiation effect of the proto-sun at the Hayashi phase (high luminosity stage; HAYASHI <u>et al.</u>, 1962) could account for the depletion of lead and other volatile elements when the particle-size in the proto-earth-moon system reached millimeter to meter size (ONO and FUJIMOTO, 1967). The moon, which was formed from the outer particles, became more depleted in volatile elements than the earth, which was accreted from interior particles of the proto-earth-moon system.

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REFERENCES

ANDERS E. (1970) Water on the moon? Science 169, 1309-1310.

ANDERS E. (1971) Meteorites and the early solar system (preprint). Ann. Rev. Astron. Astrophys. 9 (in press).

CLIFF R. A., LEE-HU C. and WETHERILL G. W. (1971) Rb-Sr and U, Th-Pb measurements on Apollo 12 material. Apollo 12 Lunar Science Conference (unpublished proceedings).

COMPSTON W., CHAPPELL B. W., ARRIENS P. A. and VERNON M. J. (1970) The chemistry and age of Apollo 11 lunar material. In Proc. Apollo 11 Lunar Sci. Conf., Geochim. Cosmochim. Acta Suppl. 1, Vol. 2, 1007-1027. Pergamon.

COMPSTON W., BERRY H., VERNON M. J., CHAPPELL B. W. and KAYE M. J. (1971) Rubidium-strontium chronology and chemistry of lunar material from the Ocean of Storms. Apollo 12 Lunar Science Conference (unpublished proceedings).

FARQUHARSON R. B. and RICHARDS J. R. (1970) Whole-rock U-Th-Pb and Rb-Sr ages of the Sybella microgranite and pegmatite, Mount Isa, Queensland. J. Geol. Soc. Aust. 17, 53-58.

GANAPATHY R., KEAYS R. R., LAUL J. C. and ANDERS E. (1970) Trace elements in Apollo 11 lunar rocks: Implications for meteorite influx and origin of moon. In Proc. Apollo 11 Lunar Sci. Conf., Geochim. Cosmochim. Acta Suppl. 1, Vol. 2, 1117-1142. Pergamon.

GOPALAN K., KAUSHAL S., LEE-HU C. and WETHERILL G. W. (1970) Rb-Sr and U, Th-Pb ages of lunar materials. In Proc. Apollo 11 Lunar Sci. Conf., Geochim. Cosmochim. Acta Suppl. 1, Vol. 2, 1195-1205. Pergamon.

HAYASHI C., HOSHI R. and SUGIMOTO D. (1962) Evolution of the stars. Suppl. Prog. Ther. Phys. 22, 183 p.

HUBBARD N. J., GAST P. W. and MEYER C. (1971) The origin of the lunar soil based on REE, K, Rb, Ba, Sr, P, and Sr^{87}/Sr^{86} data. Apollo 12 Lunar Science Conference (unpublished proceedings).

HUEY J. M., IHOCHI H., BLACK L. P., OSTIC R. G. and KOHMAN T. P. (1971) Lead isotopes and volatile transfer in the lunar soil. Apollo 12 Lunar Science Conference (unpublished proceedings).

JAMES O. B. (1971) Petrology of lunar microbreccia 12013,6. U.S. Geol. Survey-NASA Interagency Rpt., Astrogeol. 23, 39 p.

LSPET (LUNAR SAMPLE PRELIMINARY EXAMINATION TEAM) (1970) Preliminary examination of the lunar samples from Apollo 12. Science 167, 1325-1339.

LUNATIC ASYLUM (1970) Mineralogic and isotopic investigations on lunar rock 12013. Earth Planet. Sci. Lett. 9, 137-163.

LUNATIC ASYLUM (1971) Rb-Sr ages, chemical abundance patterns and history of lunar rocks. Apollo 12 Lunar Science Conference (unpublished proceedings).

MEYER C., Jr., AITKEN F. K., BRETT R., McKAY D. and MORRISON D. (1971) Rock fragments and glasses rich in K, REE, P in Apollo 12 soils: Their mineralogy and origin. Apollo 12 Lunar Science Conference (unpublished proceedings).

MURTHY V. R., EVENSON N. M., JAHN B.-M. and COSCIO M. R., Jr. (1971) Rb-Sr isotopic relations and elemental abundances of K, Rb, Sr, and Ba in Apollo 11 and 12 samples. Apollo 12 Lunar Science Conference (unpublished proceedings).

ONO S. and FUJIMOTO Y. (1967) Origin of the solar system (in Japanese). KAGAKU 39, 547-551.

PAPANASTASSIOU D. A., WASSERBURG G. J. and BURNETT D. S. (1970) Rb-Sr ages of lunar rocks from the Sea of Tranquillity. Earth Planet. Sci. Lett. 8, 1-19.

PAPANASTASSIOU D. A. and WASSERBURG G. J. (1970) Rb-Sr ages from the Ocean of Storms. Earth Planet. Sci. Lett. 8, 269-278.

PETERMAN Z. E., DOE B. R. and PROSTKA H. J. (1970) Lead and strontium isotopes in rocks of the Absaroka volcanic field, Wyoming. Contrib. Mineral. Petrol. 27, 121-130.

PETERMAN Z. E., HILDRETH R. A. and NKOMO I. (1971) Precambrian geology and geochronology of the Granite Mountains, central Wyoming. Geol. Soc. Amer. Abstracts with Programs 3, no. 5,

ROSHOLT J. N. and PETERMAN Z. E. (1969) Uranium, thorium, lead systematics in the Granite Mountains, Wyoming. Geol. Soc. Amer. Abstracts with Programs, Part 5, 70.

ROSHOLT J. N., PETERMAN Z. E. and BARTEL A. J. (1970) U-Th-Pb and Rb-Sr ages in granite reference sample from southwestern Saskatchewan. Can. J. Earth Sci. 7, 184-187.

ROSHOLT J. N. and TATSUMOTO M. (1970) Isotopic composition of uranium and thorium in Apollo 11 samples. In Proc. Apollo 11 Lunar Sci. Conf., Geochim. Cosmochim. Acta Suppl. 1, Vol. 2, 1499-1502. Pergamon.

SCHNETZLER C. C. and PHILPOTTS J. A. (1970) Trace element abundances in Apollo 12 samples. Geol. Soc. Amer. Abstracts with Programs 2, no. 7, 676.

SCHNETZLER C. C., PHILPOTTS J. A. and BOTTINO M. L. (1970) Li, K, Rb, Sr, Ba, and rare-earth concentrations, and Rb-Sr age of lunar rock 12013. Earth Planet. Sci. Lett. 9, 185-192.

SHIMAZU Y. [editor] (1967) Symposium of solar system (in Japanese): Kagaku 37, 514-571.

SILVER L. T. (1970) Uranium-thorium-lead isotopes in some Tranquillity Base samples and their implications for lunar history. In Proc. Apollo 11 Lunar Sci. Conf., Geochim. Cosmochim. Acta Suppl. 1, Vol. 2, 1533-1574. Pergamon.

SILVER L. T. (1971) U-Th-Pb relations in Apollo 11 and 12 lunar samples. Apollo 12 Lunar Science Conference (unpublished proceedings).

TATSUMOTO M. (1970a) Age of the moon: An isotopic study of U-Th-Pb systematics of Apollo 11 lunar samples--II. In Proc. Apollo 11 Lunar Sci. Conf., Geochim. Cosmochim. Acta Suppl. 1, Vol. 2, 1595-1612. Pergamon.

TATSUMOTO M. (1970b) U-Th-Pb age of Apollo 12 rock 12013. Earth Planet. Sci. Lett. 9, 193-200.

TURNER G. (1970a) Argon-40/argon-39 dating of lunar rock samples. In Proc. Apollo 11 Lunar Sci. Conf., Geochim. Cosmochim. Acta Suppl. Vol. 2, 1665-1684. Pergamon.

TURNER G. (1970b) ^{40}Ar-^{39}Ar age determination of lunar rock 12013. Earth Planet. Sci. Lett. 9, 177-180.

TURNER G. (1971) ^{40}Ar-^{39}Ar ages from the lunar maria. Apollo 12 Lunar Science Conference (unpublished proceedings).

WAKITA H. and SCHMITT R. A. (1970) Elemental abundances in seven fragments from lunar rock 12013. Earth Planet. Sci. Lett. 9, 169-176.

WARNER J. [compilor] (1970) Apollo 12 lunar-sample information. NASA TR R-353.

WARNER J. L. and ANDERSON D. H. (1971) Lunar crystalline rocks--Petrography, geology, and origin. Apollo 12 Lunar Science Conference (unpublished proceedings).

WOOD J. A., DICKEY J. S., Jr., MARVIN U. B. and POWELL B. N. (1970) Lunar anorthosites and a geophysical model of the moon. In Proc. Apollo 11 Lunar Sci. Conf., Geochim. Cosmochim. Acta Suppl. 1, Vol. 1, 965-988.

WOOD J. A., MARVIN U., REID J. B., TAYLOR G. J., BOWER J. F., POWELL B. N. and DICKEY J. S., Jr. (1971) Relative proportions of rock types, and nature of the light-colored lithic fragments in Apollo 12 soil samples. Apollo 12 Lunar Science Conference (unpublished proceedings).

.. Lead, uranium and thorium concentrati

Sample No.	Type*	Description	Pb (p
12009,22	A	Porphyritic olivine basalt	0.4
12021,122	B	Pigeonite dolerite, pegmatite	0.4
12022,37	B	Olivine dolerite	0.3
12035,10,a	B	Troctolite	0.3
12035,10,b	B	do.	0.2
12038,42	A	Basalt	0.2
12052,66,a	A	Olivine basalt	0.6
12052,66,b	A	do.	0.5
12052,66,c	A	do.	0.6
12063,49,a	A	do.	0.3
12063,49,b	A	do.	0.3
12064,21	B	Dolerite with crystobalite	0.5
12034,16,a	C	Crystal impact breccia with glass	4.1
12034,16,b	C	do.	3.82
12033,53,a	D	Fines	4.0
12033,53,b	D	do.	4.43
12070,56	D	Fines (contingency)	3.16
12070,56	D	do.	3.86
12013,10	A	Igneous breccia	
#09A			9.28
#09B			12.30
#42			16.31
#45			11.43
Sawdust			25.34
			(10.7

FOLDOUT FRAME

*Type: A, fine-grained igneous rock; B, medium-grained i

**Corrected for lead contamination in order to get a reaso n in Apollo 12 samples

m)	U (ppm)	Th (ppm)	Th^{232}/U^{238}	Approx. initial Pb (ppb) (after blank)
04	0.243	0.881	3.74	27
19	0.261	0.932	3.69	11
09	0.198	0.710	3.70	23
15	0.240	0.801	3.45	26
70	0.199	0.682	3.54	--
83	0.157	0.615	4.05	19
53	0.365	1.282	3.63	20
85	0.347	1.231	3.67	--
35	0.404	1.411	3.61	--
32	0.191	0.679	3.67	13
02	0.191	0.637	3.45	--
32	0.278	0.977	3.64	30
5	3.576	13.00	3.76	82
2	3.497	13.29	3.93	--
	2.670	9.700	3.75	96
	3.269	12.14	3.84	--
	1.641	6.020	3.79	190
	2.103	7.700	3.78	--
	5.675	20.73	3.78	200
	5.871	22.94	4.04	760
	10.80	34.29	3.28	200
	5.752	19.05	3.42	220
	5.158	16.88	3.38	--
)**				

FOLDOUT FRAM

2

eous rock; C, breccia; D, soil.

ble U-Th-Pb system as compared to that for other chips.

Table 2. Lead, uranium and thorium concentrations in Apollo 11 and 12 samples

Samples	Pb (ppm)	U (ppm)	Th (ppm)	Th^{232}/U^{238}
		Apollo 11		
Rocks				
Group II (3)*	0.29 ~ 0.51	0.16 ~ 0.27	0.53 ~ 1.02	3.53 ~ 3.99
Group I (3)*	1.56 ~ 1.74	0.85 ~ 0.87	3.30 ~ 3.43	4.03 ~ 4.08
Breccia (1)*	1.7	0.67	2.6	3.94
Fines (1)*	1.4	0.54	2.1	3.97
		Apollo 12		
Rocks (8)*	0.28 ~ 0.64	0.16 ~ 0.40	0.61 ~ 1.41	3.61 ~ 3.92
Breccia (1)*	4.2	3.6	13.0	3.76
Fines (2)*	3.2 ~ 4.00	1.6 ~ 2.7	6.0 ~ 9.7	3.75 ~ 3.79
Igneous breccia [12013]** (4)***	9.2 ~ 16.3	5.7 ~ 10.8	19.1 ~ 34.3	3.28 ~ 4.04

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*Number is number of samples analyzed.
**Number is sample number.
***Number is number of chips analyzed.

Table 4. Isotopic compositio
and thorium in mine

Sample No.	Fraction	Pb (ppm)	U (ppm)	Th (ppm)
12033	Whole rock	4.00	2.67	9.70
	1. Very fine (float in acetone)	4.65	2.95	11.69
	2. $\rho < 2.9$ plag.(~98%)	4.24	2.60	8.91
	3. $2.9 < \rho < 3.3$ mixture (glass ~70%;compound grain 30%)	3.70	3.13	11.06
	4. $3.3 < \rho$ ilm. (Py ~85%;01 10% others)	1.05	0.700	1.38
	Σ Sum of fractions	3.09	2.68	9.37
12070	Whole rock	3.16	1.64	6.02
	1. Very fine (float in acetone)	6.39	2.44	9.35
	2. $\rho < 2.9$ plag. (>95%)	12.62	8.58	31.42
	3. $2.9 < \rho < 3.3$ mixture (glass ~60%; compound grains ~40%)	1.26	0.588	2.16
	4. $3.3 < \rho$ ilm. (Py 40%; 01 ~40%)	0.606	0.343	1.10
	Σ Sum of fractions	3.17	1.79	6.59
12064	Whole rock	0.532	0.278	0.977
	1. Very fine (float in acetone)	1.68	0.451	1.58
	2. $\rho < 2.9$ plag.(plag. >95%)	0.307	0.144	0.462
	3. $2.9 < \rho < 33$ mixture (compound grains)	0.579	0.353	1.14
	4. $3.3 < \rho$ ilm. non-magnetic (Py ~60%, ilm. ~40%)	0.216	0.147	0.520
	5. $3.3 < \rho$ ilm. magnetic (ilm. ~80%)	0.759	0.497	1.77
	Σ Sum of fractions	0.384	0.231	0.793
10084	0. acetone suspension	3.68	0.850	3.26

*Corrected for Pb^{208} spike ($^{206}Pb/^{208}Pb$ is assumed to be the same as for th

n of lead and concentration of lead, uranium,
ral concentrates

Th^{232}/U^{238}	Corrected for blank Pb^{206}/Pb^{204}	Pb^{207}/Pb^{204}	Pb^{208}/Pb^{204}	U^{238}/Pb^{204}
3.75	1134	577.6	1028	1581
4.10	237.5	124.3	231.8	317.1
3.54	224.4	128.0	218.8	293.4
3.66	1391	711.6	1271	2453
2.04	79.44	42.77	75.52	108.9
3.42				
3.79	433.4	251.7	415.6	495.5
3.97	449.2	285.4	437.8	394.2
3.79	158.3	93.97	165.2	247.3
3.79	484.6	268.5	469.0	496.1
3.32	249.9	126.3	231.4	286.4
3.73				
3.64	449.6	234.9	444.6	510.6
3.61	(42.79)*	(30.76)*	(41.76)*	(28.56)*
3.31	124.7	64.47	142.8	136.6
3.35	152.7	68.78	163.2	204.7
3.65	187.4	79.39	203.6	279.8
3.69	354.3	135.8	364.3	487.7
3.54				
3.96	83.1	66.5	98.8	50.0

e whole rock).

Table 5. Lead isotopic composition of Apollo 11 and 12 samples

Samples	Pb^{206}/Pb^{204}	Pb^{207}/Pb^{204}	Pb^{208}/Pb^{204}
Apollo 11			
Rocks			
Group II (3)*	280 ~ 420	140 ~ 200	290 ~ 450
Group I (3)*	410 ~ 1240	190 ~ 590	440 ~ 1280
Breccia (1)*	260	170	270
Fines (1)*	260	170	270
Apollo 12			
Rocks (5)*	390 ~ 410	160 ~ 170	400 ~ 430
(3)*	670 ~ 720	260 ~ 300	640 ~ 710
Breccia (1)*	1390	660	1220
Fines [12070]** (1)*	430	252	416
[12033] (1)*	1130	580	1030
Igneous breccia [12013] (3)***	1200 ~ 2060	640 ~ 1050	1200 ~ 1770

*Number is number of samples analyzed.
**Number is sample number.
***Number is number of chips analyzed.

Table 6. Calculated first stage parameters for concordant age (t_1 in million years) by two-stage model*

Sample	Concordant t_1	$(U^{238}/Pb^{204})_{t_o \sim t_1}$	$(Pb^{206}/Pb^{204})_{t_1}$	$(Pb^{207}/Pb^{204})_{t_1}$
12009	3214	161.3	73.63	89.10
12021	3401	183.2	73.57	93.79
12022	3770	74.05	28.02	37.66
12035	2908	67.89	41.51	46.29
12038	3725	61.90	25.72	33.90
12052	3511	256.0	91.67	121.3
12063,a	3742	153.0	49.14	68.04
12064	3087	302.9	139.6	164.1

?

*Assuming $t_o = 4.63$. For $t_o = 4.60$ and $t_o = 4.65$, the calculated ages change less than 30 m.y.

Captions to Illustrations

Fig. 1. The $^{206}Pb/^{204}Pb$-$^{207}Pb/^{204}Pb$ evolution for Apollo 12 whole rock and soil samples, density fractions (x) of soil 12033, and a meteorite (Nuevo Laredo). The heavy isochron line is drawn through the data on igneous breccia 12013. Data for rock 12013 would lie on the 4650-4000 m.y. isochron if it evolved in a U-Pb system between 4650 and 4000 m.y. with a $^{238}U/^{204}Pb$ of zero since 4000 m.y. ago. Isochron labeled 4650-3400 m.y. is included for comparison. Isochrons labeled 4000-0 m.y. define the locus of points of the $^{238}U/^{204}Pb > 0$ since 4000 m.y. ago. The intersection of the 4000-0 m.y. isochron with the 4650-4000 m.y. isochron would represent the lead isotopic composition initially in the rock at 4000 m.y. ago. See text for discussion. Numbers on data points are final digits of sample number and a's and b's indicate duplicate determinations. S.D. is sawdust. Size of boxes indicates the experimental error.

Fig. 2. Enlarged $^{206}Pb/^{204}Pb$-$^{207}Pb/^{204}Pb$ evolution diagram showing data on Apollo 12 rocks in greater detail. The heavy isochron lines are drawn from primordial lead for several different ages. Data for the rocks would lie on the 4650-3400 m.y. isochron if they evolved in a U-Pb system during that time period and subsequently had a value of $^{238}U/^{204}Pb$ of zero. Isochron labeled 4650-4000 m.y. is included for comparison. Isochrons labeled 3400-0 m.y. define the locus of points if the $^{238}U/^{204}Pb > 0$ since 3400 m.y. ago. The intersection of

the 3400-0 m.y. isochron with the 4650-3400 m.y. isochron would represent the isotopic composition of the lead initially in the rock. See Fig. 1 caption for additional explanation of symbols.

Fig. 3. The $^{206}Pb/^{204}Pb-^{207}Pb-^{204}Pb$ evolution for density fractions from selected Apollo 12 samples. For each density fraction on the rocks, 10017 and 12064, the left symbol represents the raw data and the symbol more to the right is the data corrected for chemistry blank. The data represented by + are for whole-rock 10017, Silver (1970) and for whole-rock 12064, Silver (unpublished proceedings of the Apollo 12 Lunar Science Conference).

Fig. 4. The $^{206}Pb-^{238}U$, $^{207}Pb-^{235}U$, and $^{208}Pb-^{232}Th$ isochrons for whole rocks of Apollo 12 samples. Numbers on data points are final digits of sample numbers, and appended a's and b's indicate duplicate determinations. The size of the symbols approximate the analytical error (1 σ).

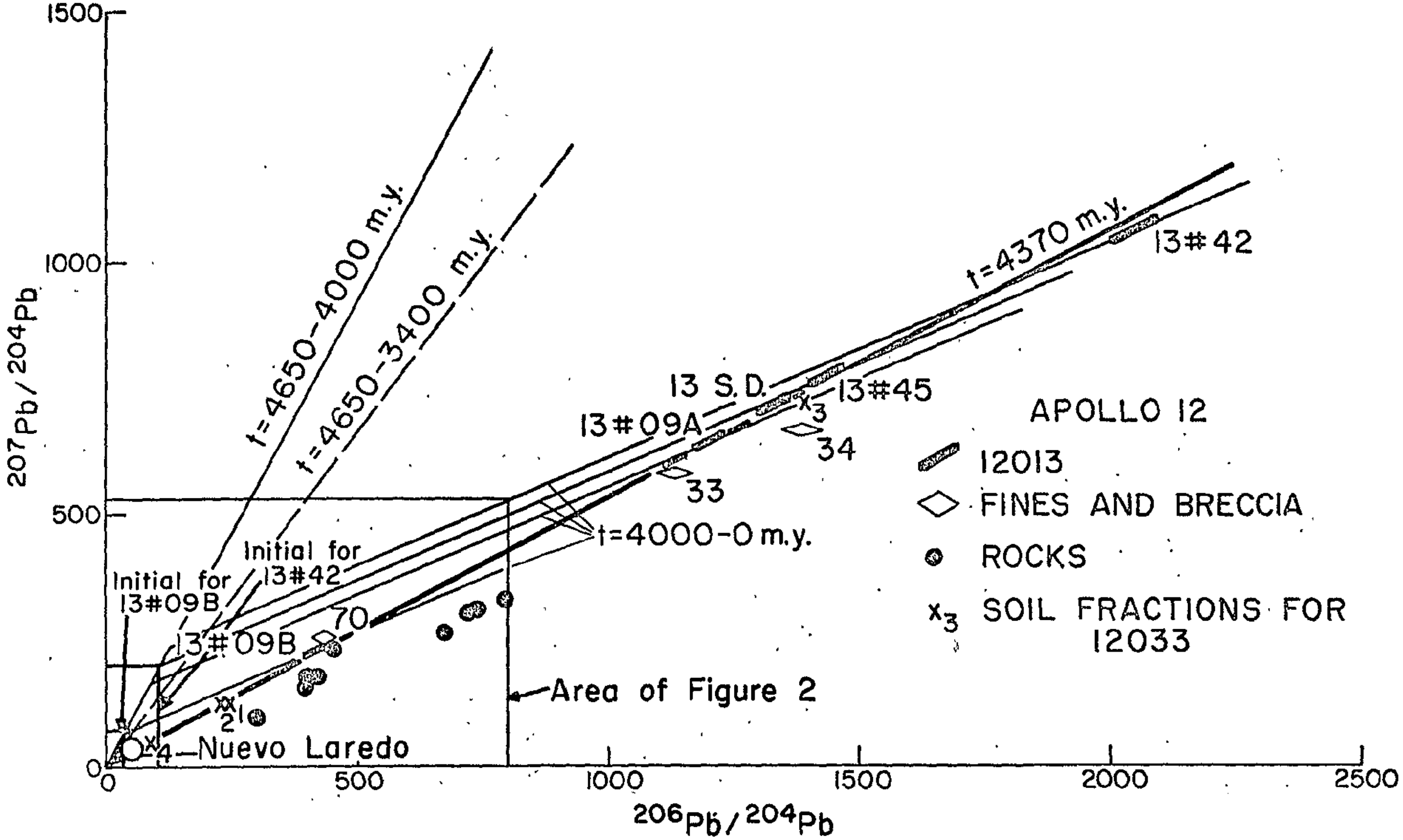
Fig. 5. The U-Pb evolution diagram. Plotted points are $(^{207}Pb_{observed} - {}^{207}Pb_{primordial})/^{235}U$ against $(^{206}Pb_{observed} - {}^{206}Pb_{primordial})/^{238}U$. Numbers on data points are final digits of sample numbers, and numbers on concordia indicate 10^9 years. Discordia lines connect limiting values for Apollo 11 (4660-3900 m.y.) and Apollo 12 (4660-3400 m.y.) rocks according to Rb-Sr mineral isochrons (Papanastassiou et al., 1970); all soil samples (10061, 10084, 12033, and 12064); and fragments

of igneous breccia 12013 that also includes impact breccia 12034. Dashed discordia lines give the limiting $^{207}Pb/^{206}Pb$ slopes of all Apollo 12 rocks except 12035 and 12064.

Fig. 6. The U-Pb evolution diagram for rock 12064 and its separated density fractions. Dashed lines are drawn to fit the data. For comparison, rock 12035, the field encompassing most Apollo 12 rocks, and the trend of the fragments of igneous breccia 12013 are given. The definitions of the density fractions 1-5 are given in Table 4. The bars express the estimated analytical uncertainties.

Fig. 7. U-Pb evolution diagram illustrating processes involved in second and third stage events (for detailed explanation see Tatsumoto, 1970a).

Fig. 8. U-Pb evolution diagram for soils 12033 and 12070 and their density fractions. The definitions of 1-4 density fractions are given in Table 4. The bars express the estimated analytical uncertainties.


1500
1000
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$^{207}Pb/^{204}Pb$
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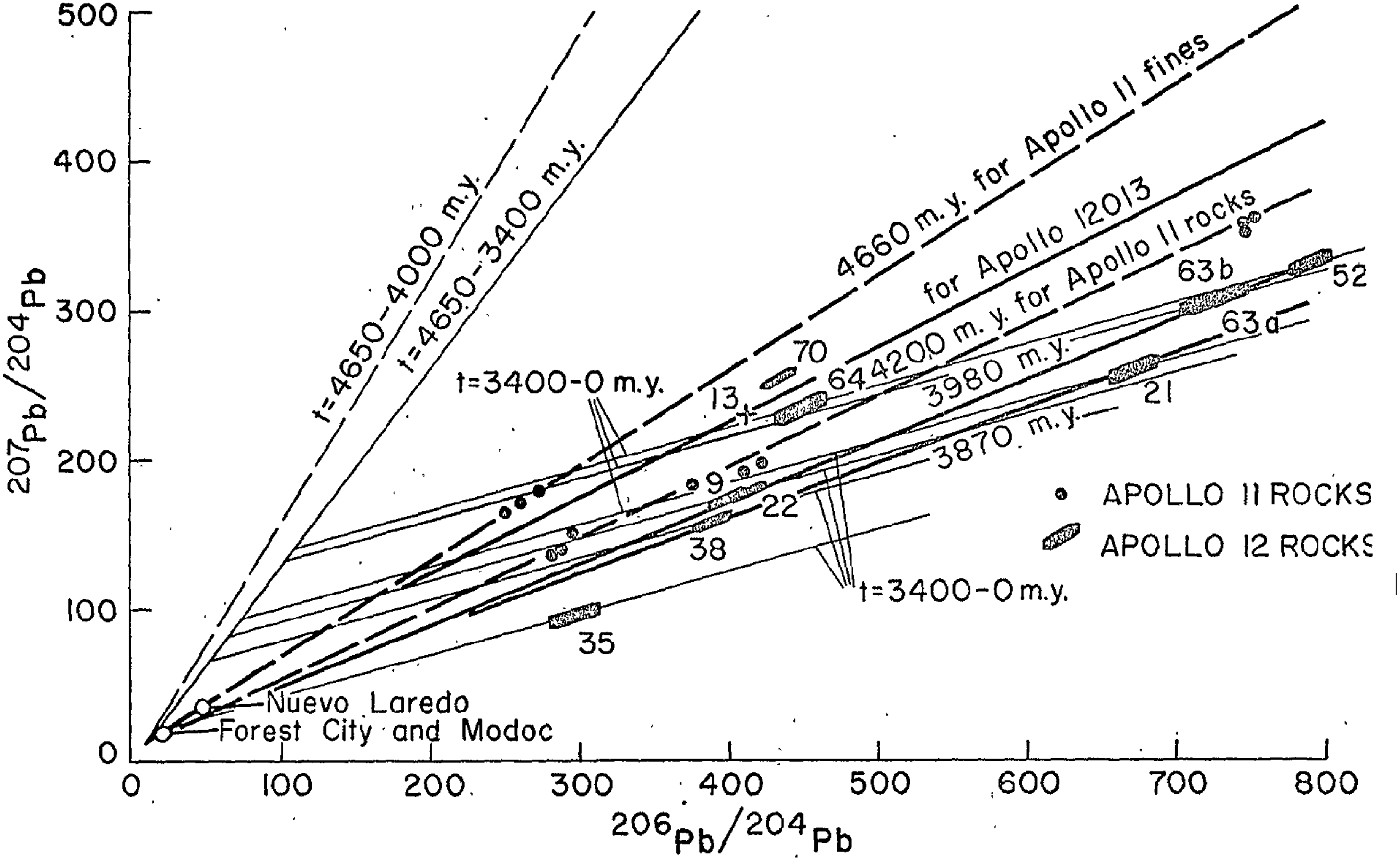
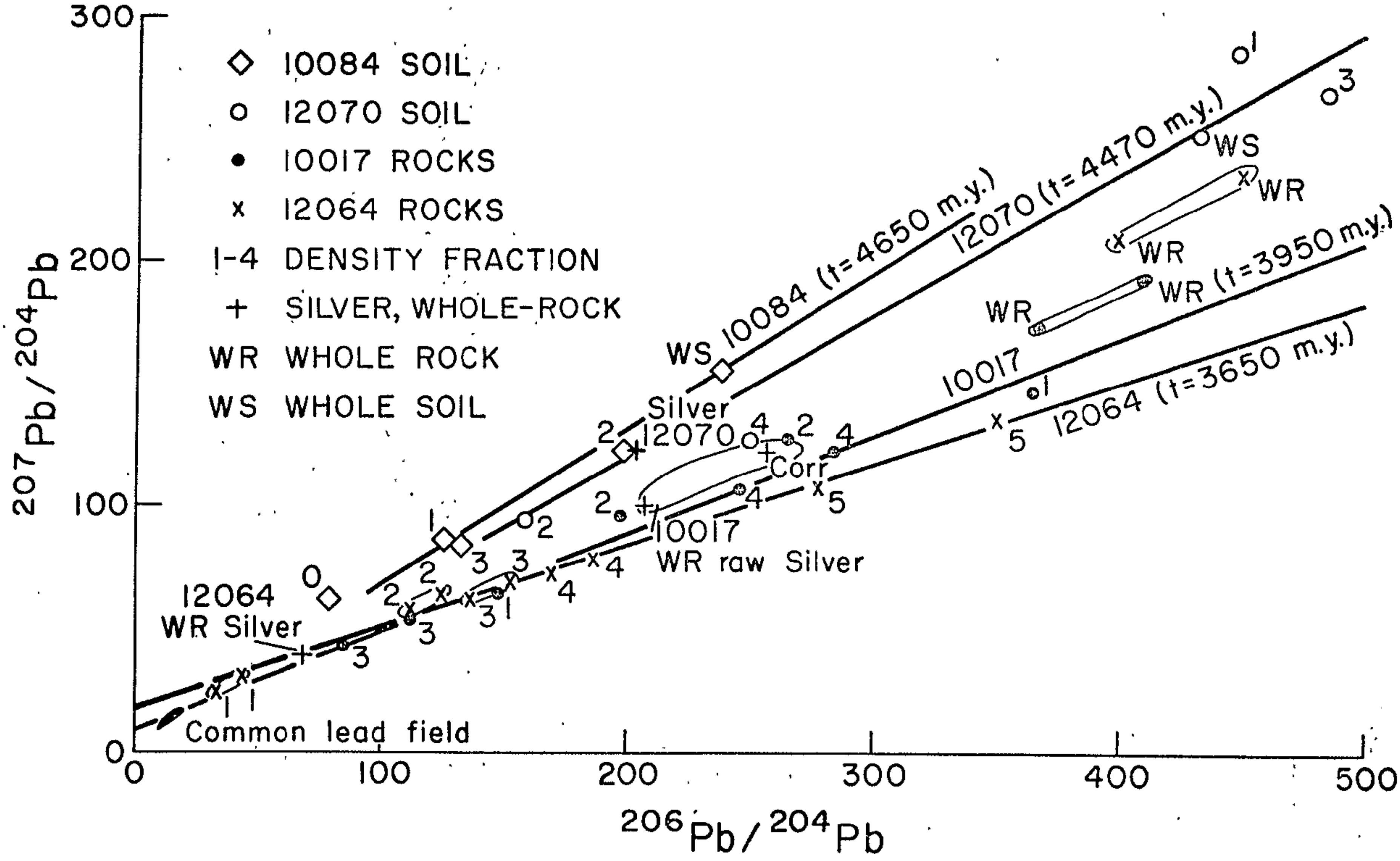
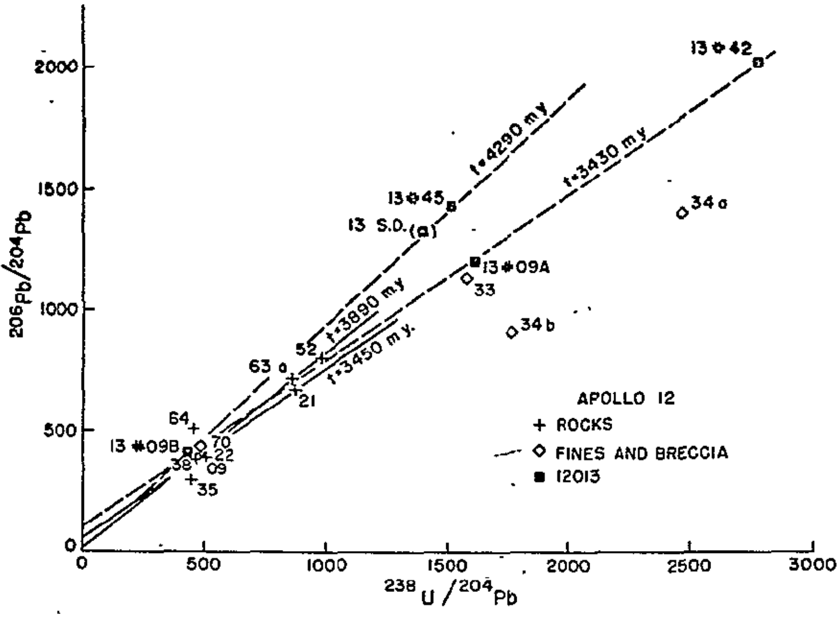
$^{206}Pb/^{204}Pb$
t=4650-4000 m.y.
t=4650-3400 m.y.
t=4370 m.y.
t=4000-0 m.y.
13#42
13 S.D.
13#45
13#09A
x_3
34
33
Initial for 13#42
Initial for 13#09B
13#09B
70
x_2
x_1
x_4
Nuevo Laredo
Area of Figure 2
APOLLO 12
12013
FINES AND BRECCIA
ROCKS
x_3 SOIL FRACTIONS FOR 12033



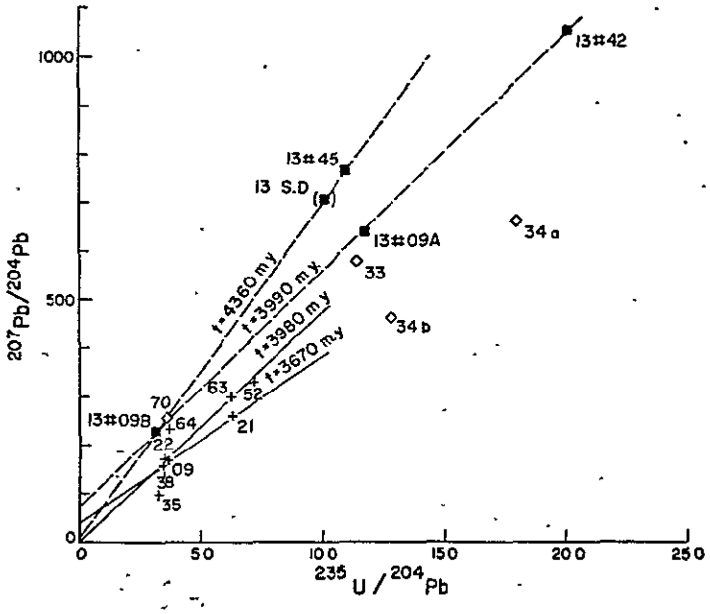
500
400
300
200
100
0
0
100
200
300
400
500
600
700
800
$^{207}Pb/^{204}Pb$
$^{206}Pb/^{204}Pb$
t=4650-4000 m.y.
t=4650-3400 m.y.
4660 m.y. for Apollo 11 fines
for Apollo 12013
4200 m.y. for Apollo 11 rocks
t=3400-0 m.y.
70
13
64
3980 m.y.
3870 m.y.
63b
52
63a
21
9
22
38
35
t=3400-0 m.y.
APOLLO 11 ROCKS
APOLLO 12 ROCKS
Nuevo Laredo
Forest City and Modoc



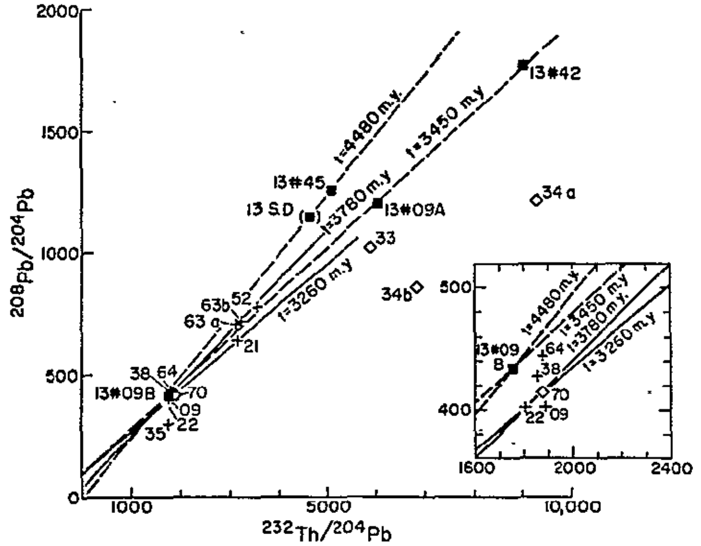
◇ 10084 SOIL
○ 12070 SOIL
● 10017 ROCKS
x 12064 ROCKS
1-4 DENSITY FRACTION
+ SILVER, WHOLE-ROCK
WR WHOLE ROCK
WS WHOLE SOIL
WS 10084 (t=4650 m.y.)
12070 (t=4470 m.y.)
WR (t=3950 m.y.)
10017
12064 (t=3650 m.y.)
Silver
12070
Corr
10017
WR raw Silver
12064
WR Silver
Common lead field
WS
WR
WR
WR
WS
0
1
2
3
4
5
^{207}Pb/^{204}Pb
^{206}Pb/^{204}Pb
0
100
200
300
400
500



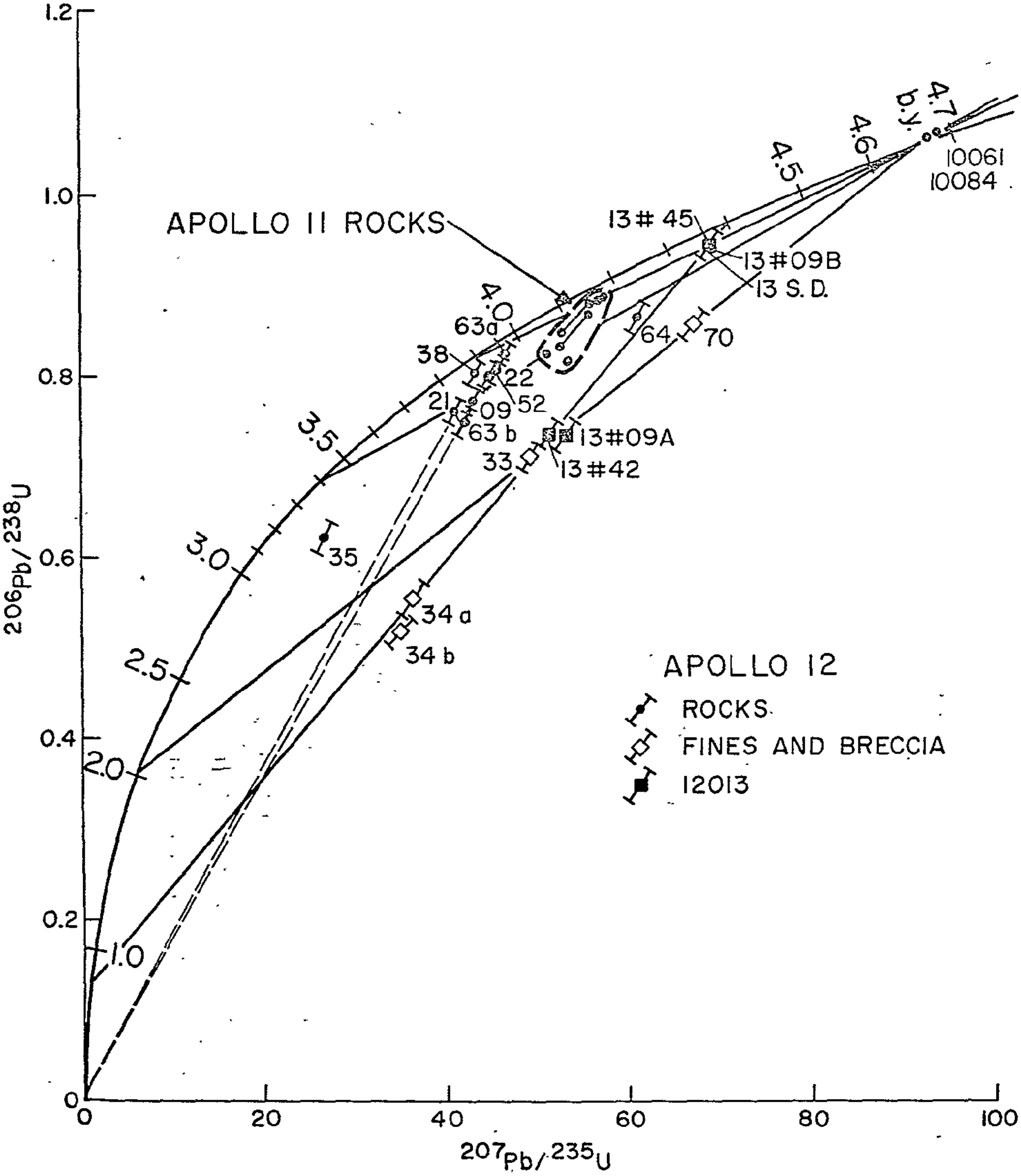
2000
1500
1000
500
0
^{206}Pb/^{204}Pb
13#42
t=4290 m.y.
t=3430 m.y.
13#45
13 S.D.
34a
13#09A
33
34b
52
t=3890 m.y
t=3450 m.y.
63a
21
APOLLO 12
+ ROCKS
◇ FINES AND BRECCIA
■ 12013
64
13#09B
70
22
38
09
35
0
500
1000
1500
2000
2500
3000
^{238}U/^{204}Pb



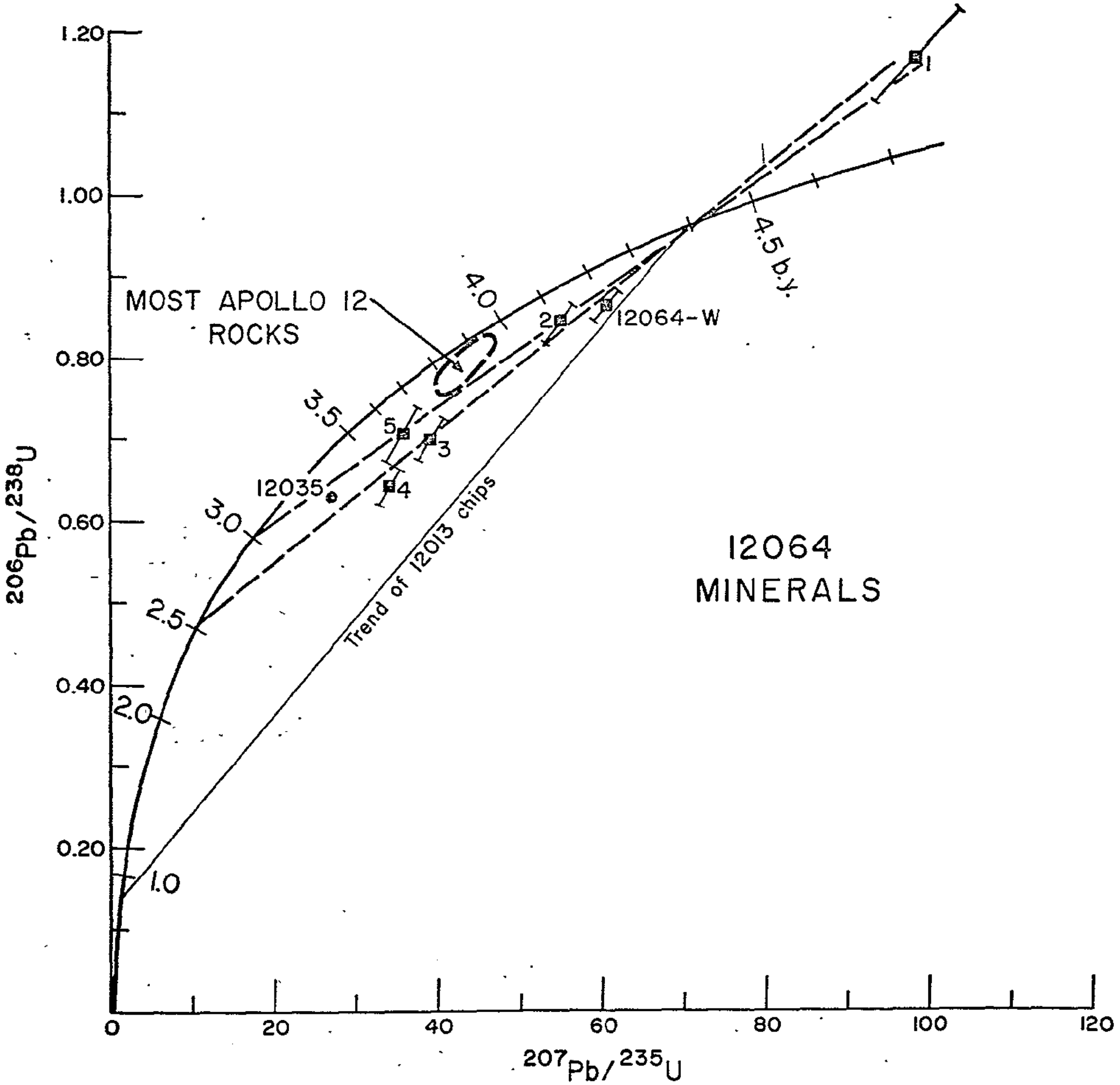
1000
500
0
^{207}Pb/^{204}Pb
13#42
13#45
13 S.D
13#09A
33
34a
34b
t=4360 m.y
t=3990 m.y
t=3980 m.y
t=3670 m.y
63
52
21
70
13#09B
64
22
09
38
35
0
50
100
150
200
250
^{235}U/^{204}Pb



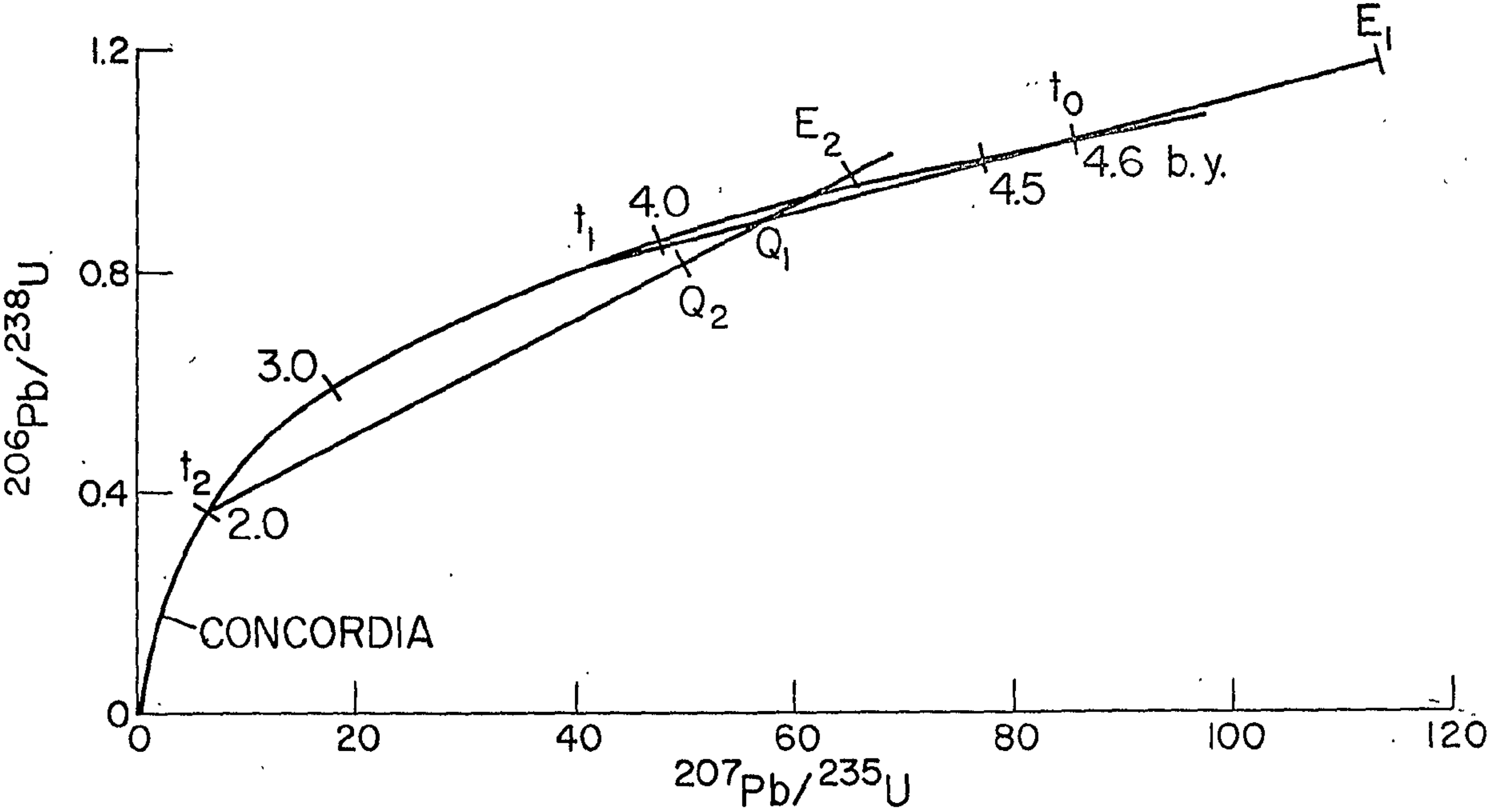
2000
1500
1000
500
0
^{208}Pb/^{204}Pb
t=4480 m.y.
t=3450 m.y
13#42
13#45
13 S.D
t=3780 m.y.
13#09A
33
34a
34b
t=3260 m.y
63b
52
63a
21
38
64
13#09B
70
09
35
22
500
400
13#09 B
t=4480 m.y
t=3450 m.y
t=3780 m.y
t=3260 m.y
64
38
70
22
09
1600
2000
2400
1000
5000
10,000
^{232}Th/^{204}Pb



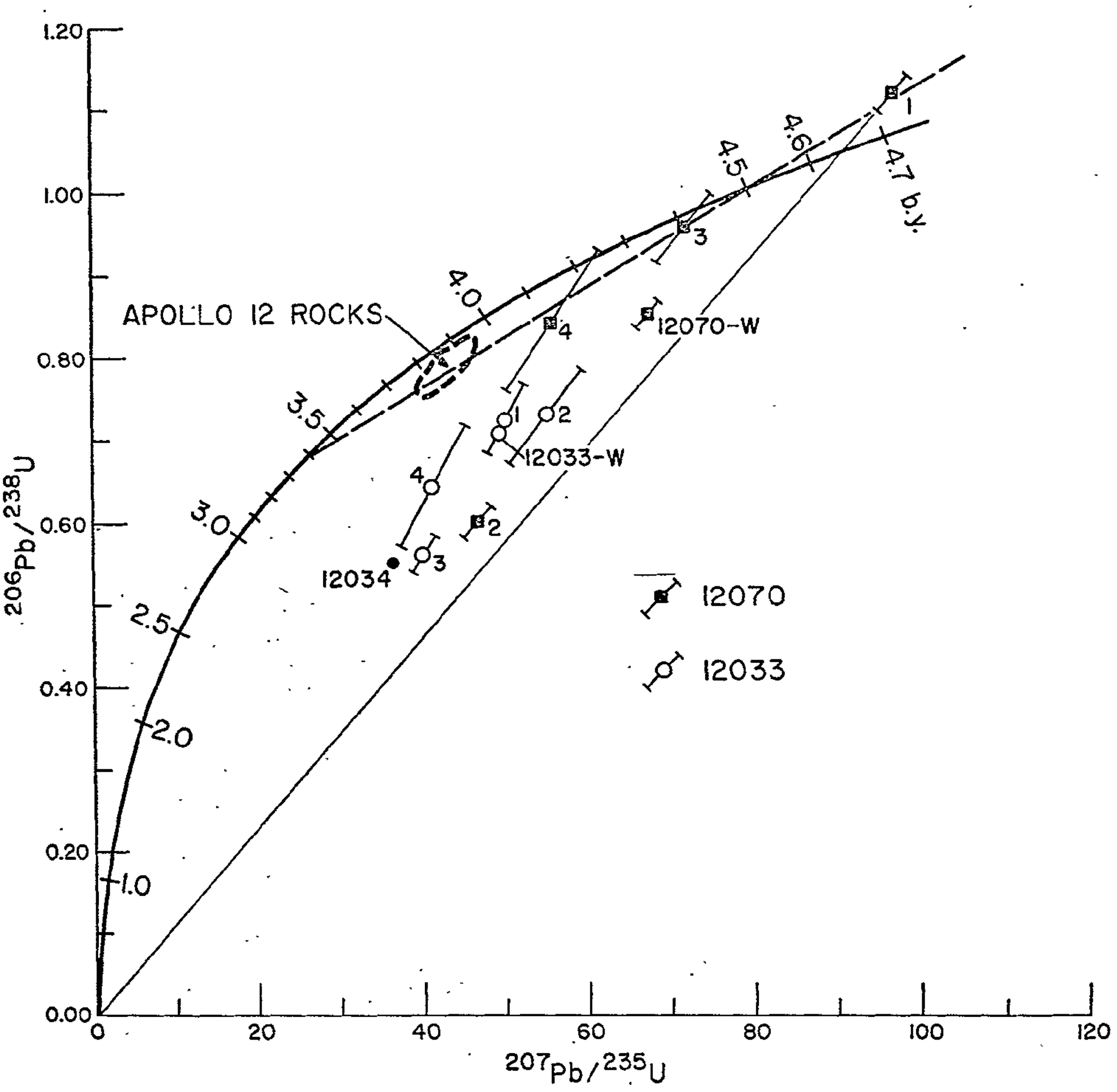
1.2
1.0
0.8
0.6
0.4
0.2
0
0
20
40
60
80
100
$^{206}Pb/^{238}U$
$^{207}Pb/^{235}U$
APOLLO 11 ROCKS
1.0
2.0
2.5
3.0
3.5
4.0
4.5
4.6
4.7
b.y.
10061
10084
13 # 45
13 # 09B
13 S.D.
64
70
63a
38
22
52
21
09
63 b
13 # 09A
33
13 # 42
35
34 a
34 b
APOLLO 12
ROCKS
FINES AND BRECCIA
12013



12064
MINERALS
MOST APOLLO 12
ROCKS
12035
12064-W
Trend of 12013 chips
4.5 b.y.
4.0
3.5
3.0
2.5
2.0
1.0
1
2
3
4
5
$^{206}Pb/^{238}U$
$^{207}Pb/^{235}U$
1.20
1.00
0.80
0.60
0.40
0.20
0
20
40
60
80
100
120



1.2
0.8
0.4
0
$^{206}Pb/^{238}U$
0
20
40
60
80
100
120
$^{207}Pb/^{235}U$
CONCORDIA
t_2
2.0
3.0
t_1
4.0
Q_2
Q_1
E_2
4.5
t_0
4.6 b.y.
E_1



$^{206}Pb/^{238}U$
$^{207}Pb/^{235}U$
1.20
1.00
0.80
0.60
0.40
0.20
0.00
0
20
40
60
80
100
120
1.0
2.0
2.5
3.0
3.5
4.0
4.5
4.6
4.7 b.y.
APOLLO 12 ROCKS
12070-W
12033-W
12034
1
2
3
4
12070
12033

Submitted to Geochim. et Cosmochim. Acta, Proc. Apollo 12 Lunar Sci. Conf.

Isotopic composition of thorium and uranium in Apollo 12 samples*

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Abstract--The isotopic composition of uranium and thorium was determined by mass spectrometry and alpha spectrometry in 14 lunar samples from Apollo 12. The U^{238}/U^{235} ratio in samples measured is the same as that for terrestrial uranium within experimental error (137.8 ± 0.3). The U^{234} daughter is in radioactive equilibrium with parent U^{238} in the samples; however, it could not be demonstrated that Th^{230} is in equilibrium with U^{238} in some rock samples as measured by alpha spectrometry.

One interpretation of the α-particle spectral data of thorium would suggest the existence of a significant amount of a shape isomer of Th^{232} (Th^{232m}) accompanied by a decay series collateral to the regular decay series for Th^{232}. Different modes are postulated for the production of Th^{232m} as a fraction of primordial thorium and for the production of radiogenic Th^{232} by decay of Pu^{244} progenitor.

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*Publication authorized by the Director, U.S. Geological Survey.

The isotopic composition of uranium and thorium was determined in 14 samples from Apollo 12. The isotopic ratios of uranium (U^{238}, U^{235}, U^{234}) were identical, within experimental error, with those of terrestrial reference samples in radioactive equilibrium. However, unexpected variations in the isotopic ratios of thorium (Th^{232}, Th^{230}, Th^{228}) may provide clues to the variable cosmogenic history of thorium.

The description of chemical separations and the measurements for uranium and thorium were given in ROSHOLT and TATSUMOTO (1970) and TATSUMOTO (1970a). After the separation of lead for the determination of the isotopic abundances of lead isotopes (TATSUMOTO, 1970b), uranium and thorium were separated from the lunar samples.

Both mass spectrometry and alpha spectrometry were used in determination of the isotopic composition of uranium. Radioactivity ratios of U^{234} to U^{238} were determined first; then uranium was removed from the platinum counting disk and U^{238}/U^{235} ratios were determined by mass spectrometry on 0.4-microgram quantities of uranium. The results (Table 1) indicate that U^{234} is in radioactive equilibrium with U^{238}, and the U^{238}/U^{235} ratio in samples measured is the same as terrestrial uranium (137.8 ± 0.3). Uncertainties in radioactivity ratios are standard deviations based on counting statistics.

The radioactivity ratios of Th^{232}/Th^{230} in lunar samples (Table 2) were determined by α-particle spectrometry on aliquots of the same solution of the rock sample used in determining the concentrations of uranium and thorium (TATSUMOTO, 1970b); however, separate splits of the fines were used for determination of the concentrations of uranium and thorium.

Th^{228} data is not included in this paper because, to evaluate properly its relation to Th^{232}/Th^{230} variations, more time is required to measure Th^{228} decay and Ra^{228} growth from Th^{232}. Thorium-232 has been used as a natural tracer for the determination of the state of radioactive equilibrium between Th^{230} and parent U^{238} in terrestrial rocks (ROSHOLT _et al._, 1967). The 4.0 Mev α-particle emitted from Th^{232} was measured to determine its radioactivity. A typical α-particle spectra of lunar crystalline rock is shown in Fig. 1. If Th^{230} is in radioactive equilibrium with U^{238}, the activity ratio expected for thorium isotopes can be calculated, using the atomic ratio of Th^{232}/U^{238} as determined by isotope dilution (TATSUMOTO, 1970b) and using the decay constants for Th^{232} and U^{238}. The expected activity ratio is

$$(Th^{232}/Th^{230})_{expected} = (Th^{232}/U^{238})_{atom} \times (\lambda_{232}/\lambda_{238}),$$

where λ_{232} and λ_{238} are the decay constants for Th^{232} and U^{238}. Values for the measured activity ratios of thorium isotopes and the expected activity ratios of Th^{232}/Th^{230} are shown in Table 2.

Comparison of the expected ratio/measured ratio (Table 2) indicates that fragments (12013,10-42 and 12013,10-45), breccia (12034), and fines (12033) are similar in thorium isotopic composition to terrestrial reference samples; however, the sample of the dark portion of rock 12013 (12013,10-09) has a low value for this comparison. A split of contingency fines (12070) has an excess of radioactivity over that expected at the α-particle energy of Th^{230} (4.66 ± .06 Mev peak). All the crystalline rocks analyzed have an excess of radioactivity at this α-particle energy. Unusually large excesses are indicated for three of the samples (12009, 12021, 12063) where the expected ratio/measured ratio (Table 2) exceeds unity by 60 to 80 percent. A distribution diagram of the comparison of the expected ratio/measured ratio (Fig. 2) shows the variations of thorium isotopic composition in different rock types. The pattern of variations suggests agreement with the KREEP hypothesis of HUBBARD *et al.* (1971) because the dark portion of rock 12013 has a thorium isotopic composition at one extreme, the basalts have a composition at the other extreme, and the fines have an intermediate composition. We do not interpret the variations in the Th^{232}/Th^{230} ratios as due to disequilibrium in the U^{238}-U^{234}-Th^{230} decay sequence because of (a) the equilibrium conditions demonstrated between U^{234} and U^{238} in all samples measured (Table 1), (b) the Th^{230} equilibrium demonstrated in samples 12013, 12033, 12034 and in some of the Apollo 11 samples (ROSHOLT and TATSUMOTO, 1970), and (c) the concordancy between Pb^{206}/U^{238} ages and Pb^{208}/Th^{232} ages in the two-stage lead evolution model by TATSUMOTO (1970a, b).

Continuing measurements of the α-particle spectra of thorium separates of samples from both Apollo 11 and Apollo 12 indicate that samples with deviations in the Th^{232}/Th^{230} ratios also show deviations in the radioactivity ratios of Th^{228} and its immediate daughter product Ra^{224}. Ra^{224} has a half life of 3.64 days and the radioactivity ratio of Ra^{224}/Th^{228} should be unity after approximately 30 days following the electrodeposition of thorium. An example is illustrated in Fig. 1 for sample 12063, measured 6 months after thorium was electrodeposited on the platinum counting disk. The Ra^{224}/Th^{228} α-radioactivity ratio in this sample at this time of measurement is $1.08 \pm .03$; Th^{228}, with a 1.9-year half life, has decayed to 83 percent of its original activity. The amount of recoil atom loss in production of Ra^{224} from electrodeposited thorium has not been evaluated completely; thus, a quantitative comparison of Ra^{224} excess to 4.6 Mev α-particle excess cannot be made at this time.

A possible interpretation of the α-spectral data, in lunar samples with excess 4.6 Mev α-activity, is that some of the Ra^{224} is produced by a mode of radioactive decay other than direct α-particle emission of Th^{228}. A collateral radioactive series, with the long-lived parent being an isomer of Th^{232} (Th^{232m}), could account for the α-particle data observed. A block diagram of this suggested collateral series (indicated by cross-hatched blocks) and its relation to the Th^{232} decay series is shown in Fig. 3. Such a collateral series would require an isomer of Ra^{228} (Ra^{228m} as daughter of Th^{232m}) and possibly an isomer of Ac^{228}, both of which have significant components of α-particle emission in their decay modes to bypass the Th^{228} branch of the series.

The possibility for rare α-particle emission in Ra^{228} has been considered from the time of classical investigations of natural radioactivity; however, FEATHER et al. (1957) have presented evidence for only very slight amounts of α-emission in Ra^{228} with an upper limit of the α/β branching ratio of about 1.5×10^{-8}. The radioactive properties of the parent isomer, Th^{232m}, as indicated by the lead data in lunar samples, suggest that its half life would be similar to that of Th^{232} ($\sim 1.4 \times 10^{10}$ years) and the energy of its α-particle emission, as suggested by α-spectrometry, would be about 4.7 Mev. Positive identification of its presence is apparently masked by the 4.68 Mev α-particle emitted by Th^{230}. Our best spectral resolution, using 300-sq.-mm. silicon surface barrier detectors, has been 30 Kev, FWHM, and there is only slight evidence of the existence of an α-particle with an energy of 5-10 Kev greater than that emitted by Th^{230}. The suggested decay scheme and α-particle energy for Th^{232m} in relation to the decay properties of Th^{232}, Th^{230}, and Th^{228} are shown in Fig. 4.

If such an isomer exists in lunar samples and accounts for the excess α-radioactivity in the 4.6 Mev spectra of thorium, then a calculation of its activity abundance in individual samples can be made using the following equation:

$$\frac{Th^{232m}}{Th^{232} + Th^{232m}} = \frac{R_{measured} - R_{expected}}{R_{measured} + 1}$$

where $R = \frac{4.6 \text{ Mev } \alpha\text{-counts of Th}}{4.0 \text{ Mev } \alpha\text{-counts of Th}}$. The concentrations of thorium, the expected and measured ratios, R, and the calculated activity ratios of $Th^{232m}/(Th^{232} + Th^{232m})$ are given in Table 3. Values of about 0.25 for this ratio, are indicated for the most anomalous samples of crystalline rock. Significant variations exist among the 8 crystalline rocks analyzed, suggesting fractionation between Th^{232} and Th^{232m}. Preliminary measurements (ROSHOLT, unpublished data) indicate that such an isomer may exist in some terrestrial rocks in the amount of about 3 percent of the terrestrial thorium. It should be noted that no long-lived isomers of even proton-even neutron nuclei of heavy elements have been reported in the literature.

Primordial thorium, in r-process nucleosynthesis, would have been produced by beta decay of neutron-rich nuclides and through neutron capture by Th^{231}. If a significant fraction of this thorium was produced as the isomer Th^{232m}, a variable cosmogenic history for Th^{232} and Th^{232m} may exist (ROSHOLT and TATSUMOTO, 1970). WASSERBURG *et al*. (1969) have given evidence for "last-minute" synthesis of heavy elements at about 4.8 billion years ago in their model for nuclear chronology in the galaxy. Thus, radiogenic Th^{232} would have been produced up to 400 million years after the "last-minute" synthesis and after the formation of the solar system, by decay of Pu^{244} progenitor. Different modes for the production of primordial Th^{232m} and radiogenic Th^{232} are suggested in the block diagram shown in Fig. 5.

Acknowledgments--We thank R. J. Knight and D. M. Unruh for laboratory assistance. This study was carried out under NASA Contract T-75445.

REFERENCES

FEATHER N., MILLER N. and PEAT S. W. (1957) Search for a rare α-emission in ^{228}Ra (MsTh 1). Proc. Phys. Soc. [London] 70A, 478-480.

HUBBARD N. J., MEYER C., Jr., GAST P. W. and WIESMANN H. (1971) The composition and derivation of Apollo 12 soils. Earth Planet. Sci. Lett. 10, 341-350.

ROSHOLT J. N., Jr., PETERMAN Z. E. and BARTEL A. J. (1967) Reference sample for determining the isotopic composition of thorium in crustal rocks. U.S. Geol. Surv. Prof. Paper 575-B, B133-B136.

ROSHOLT J. N. and TATSUMOTO M. (1970) Isotopic composition of uranium and thorium in Apollo 11 samples. Proc. Apollo 11 Lunar Sci. Conf., Geochim. et Cosmochim. Acta Suppl. 1, Vol. 2, 1499-1502. Pergamon.

TATSUMOTO M. (1970a) Age of the moon: An isotopic study of U-Th-Pb systematics of Apollo 11 lunar samples--II. Proc. Apollo 11 Lunar Sci. Conf., Geochim. et Cosmochim. Acta Suppl. 1, Vol. 2, 1595-1612. Pergamon.

TATSUMOTO M. (1970b) U-Th-Pb age of Apollo 12 rock 12013. Earth Planet. Sci. Lett. 9, 193-200.

WASSERBURG G. J., SCHRAMM D. N. and HUNEKE J. C. (1969) Nuclear chronologies for the galaxy. Astrophys. J. 157, L91-L96.

Table 1. Concentrations and isotopic composition of uranium

Sample	Rock type	U (ppm)	U^{234}/U^{238} (activity ratio)	U^{238}/U^{235} (atom ratio)
12013,10-09	Breccia	5.67	1.01	137.7
12013,10-42	"	10.80	.99	138.0
12013,10-45	"	5.75	1.02	137.6
12034,16	"	3.58	1.00	137.7
12033,53	Fines	2.67	.99	137.7
12070,56	Fines (contingency)	1.64	1.01	137.8
12009,22	Crystalline rock	.243	1.00	138.0
12021,122	"	.261	1.00	137.8
12022,37	"	.198	.99	137.7
12035,10	"	.199	.99	137.6
12038,42	"	.157	1.01	137.6
12052,66	"	.365	1.00	137.8
12063,49	"	.191	.99	137.7
12064,21	"	.278	1.01	137.9
Error range (percent)			2	0.25

Table 2. Radioactivity ratios of thorium isotopes.

Sample	Th^{232}/U^{238} (atom ratio)	$[Th^{232}/U^{238}]_{atom} \times [\lambda_{232}/\lambda_{238}]$[1] (expected Th^{232}/Th^{230} activity ratio)	Th^{232}/Th^{230} (measured activity ratio)	Expected rat / Measured rati
Breccias and fines				
12013,10-09	3.78	1.20	1.30	0.92
12013,10-42	3.28	1.04	1.03	1.01
12013,10-45	3.42	1.09	1.09	1.00
12034,16	3.75	1.19	1.23	.97
12033,53(a)	3.75	1.19	1.20	.99
12033,53(b)	3.84	1.22	1.18	1.03
12070,56(a)	3.79	1.20	1.18	1.02
12070,56(b)	3.78	1.20	0.99	1.21
Crystalline rocks				
12009,22	3.74	1.18	0.73	1.63
12021,122	3.69	1.17	.69	1.69
12022,37	3.70	1.17	.95	1.24
12035,10	3.54	1.12	.95	1.19
12038,42(a)	3.92	1.24	1.03	1.24
12038,42(b)	3.97	1.26	1.05	1.21
12052,66	3.63	1.15	1.02	1.13
12063,49(a)	3.67	1.17	.73	1.60
12063,49(b)	3.45	1.09	.61	1.80
12064,21	3.64	1.16	1.02	1.13
Error range (percent)		2	2-3	

[1] $\lambda_{232} = 4.88 \times 10^{-11} yr^{-1}$; $\lambda_{238} = 1.537 \times 10^{-10} yr^{-1}$

(a) and (b) indicate different portions of sample

Table 3. Concentrations and alpha radioactivity ratios of thorium isotopes

Sample	Th (ppm)	$\frac{\text{4.6 Mev alpha counts of Th}}{\text{4.0 Mev alpha counts of Th}}$ (expected)[1/]	$\frac{\text{4.6 Mev alpha counts of Th}}{\text{4.0 Mev alpha counts of Th}}$ (measured)	$\frac{Th^{232m}}{Th^{232} + Th^{232m}}$ (calculated activity ratio)
Breccias and fines				
12013,10-09	20.73	0.834	0.772	-0.035 ± .030
12013,10-42	34.29	.960	.968	.004 ± 030
12013,10-45	19.05	.920	.916	-.002 ± .035
12034,16	13.00	.838	.814	-.010 ± .030
12033,53(a)	9.70	.839	.835	-.003 ± .030
12033,53(b)	12.14	.821	.847	.014 ± .035
12070,56(a)	6.02	.831	.847	.009 ± .035
12070,56(b)	7.70	.833	1.01	.090 ± .030
Crystalline rocks				
12009,22	.881	.842	1.37	0.22 ± .03
12021,122	.932	.853	1.44	.24 ± .03
12022,37	.710	.851	1.06	.10 ± .05
12035,10	.682	.890	1.06	.08 ± .04
12038,42(a)	.615	.778	.969	.10 ± .04
12038,42(b)	.816	.794	.964	.09 ± .04
12052,66	1.28	.868	.981	.06 ± .03
12063,49(a)	.679	.858	1.37	.22 ± .02
12063,49(b)	.637	.913	1.65	.28 ± .03
12064,21	.977	.865	.980	.06 ± .04

[1/] $\lambda_{232} = 4.88 \times 10^{-11} yr^{-1}$; $\lambda_{238} = 1.537 \times 10^{-10} yr^{-1}$.

(a) and (b) indicate different portions of sample.

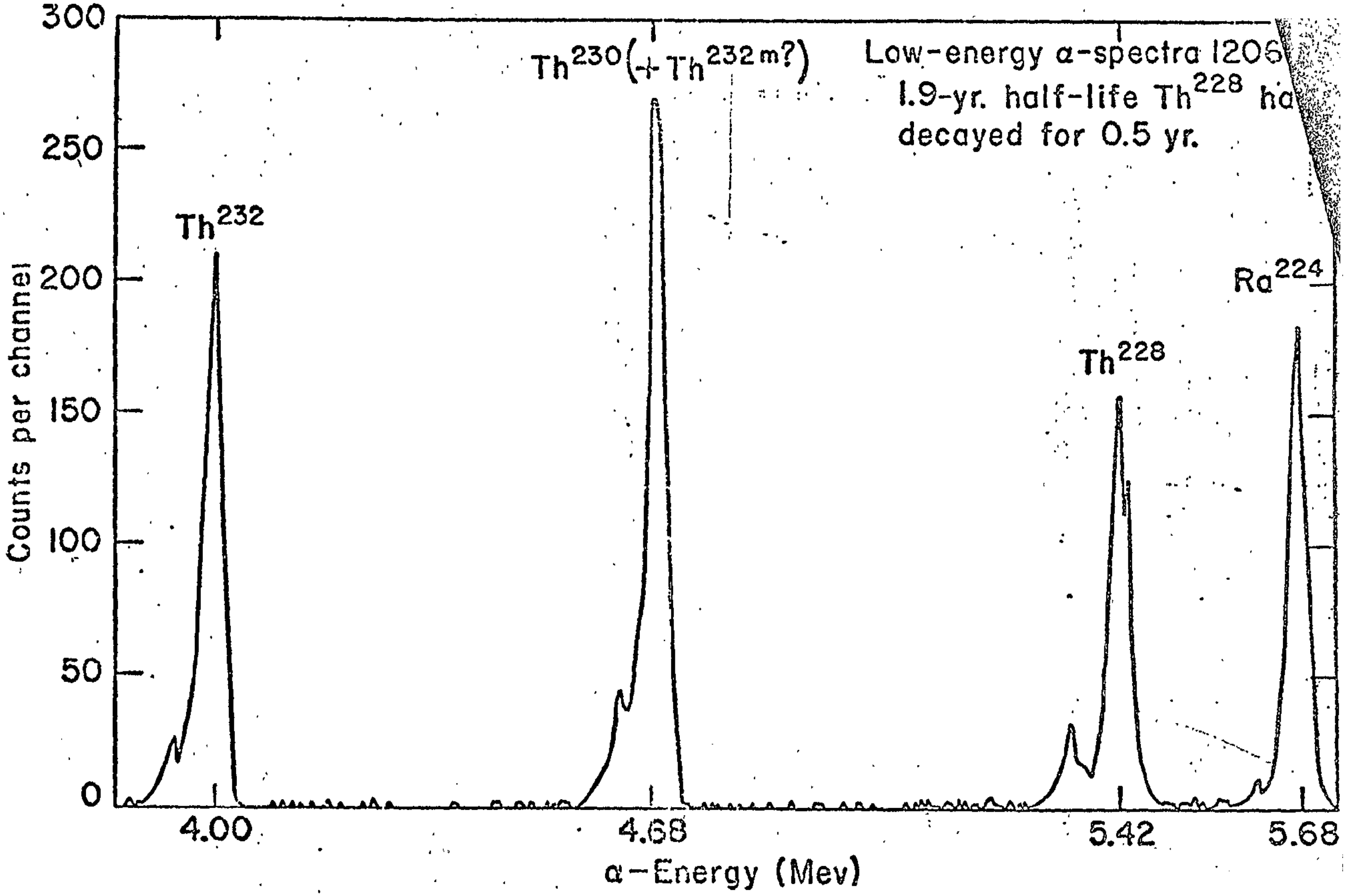

Figure 1. Low-energy α-particle spectra of sample 12063.

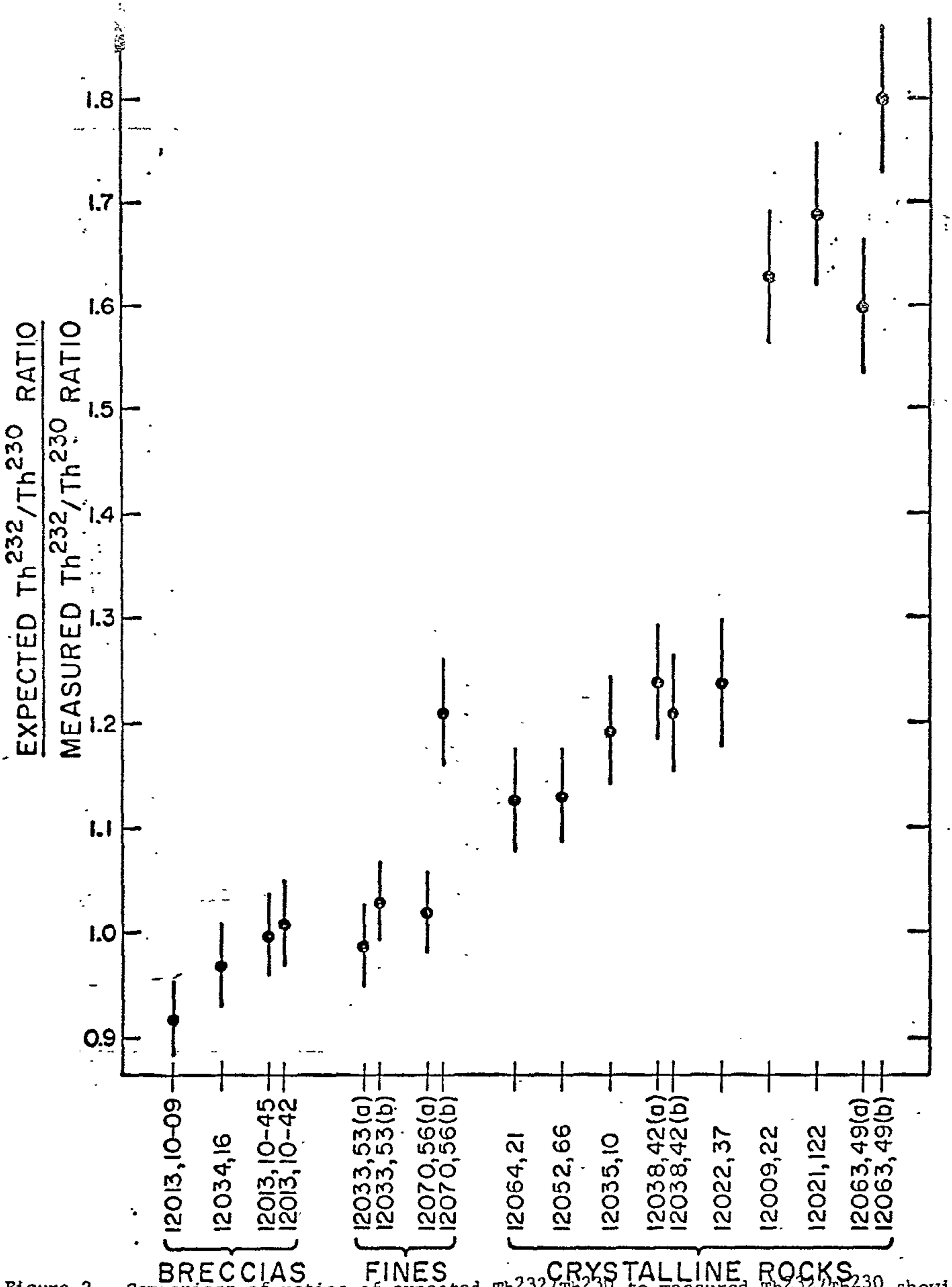

Figure 2. Comparison of ratios of expected Th^{232}/Th^{230} to measured Th^{232}/Th^{230} showing variations of thorium isotopic composition in different rock types. Vertical bars represent uncertainties in the expected ratio/measured ratio values calculated from standard deviations based on counting statistics and a 2 percent uncertainty in the Th^{232}/U^{238} ratio.

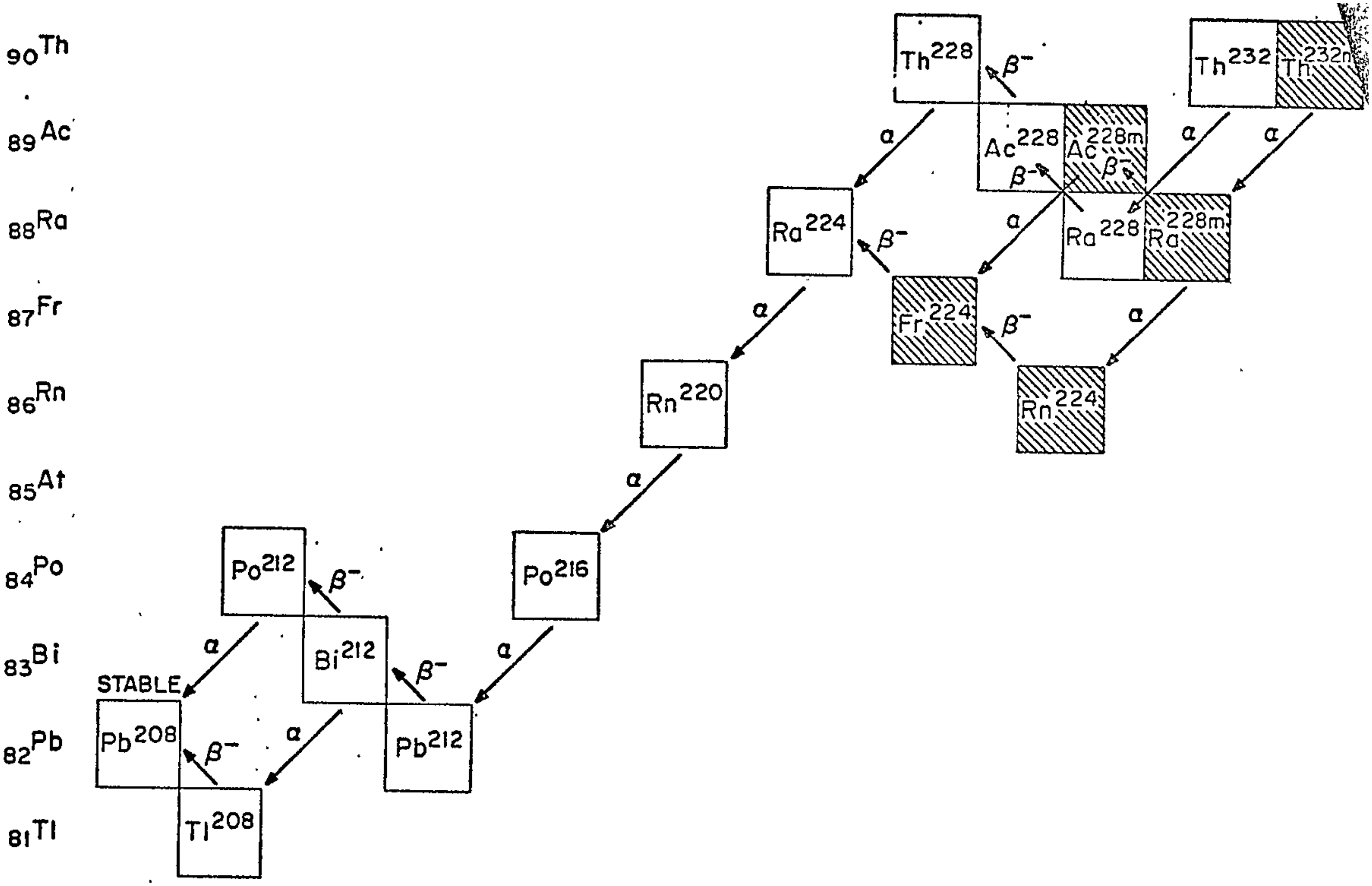

Figure 3. Block diagram of suggested decay series (cross-hatched) of Th^{232m} collateral to regular decay series of Th^{232}.

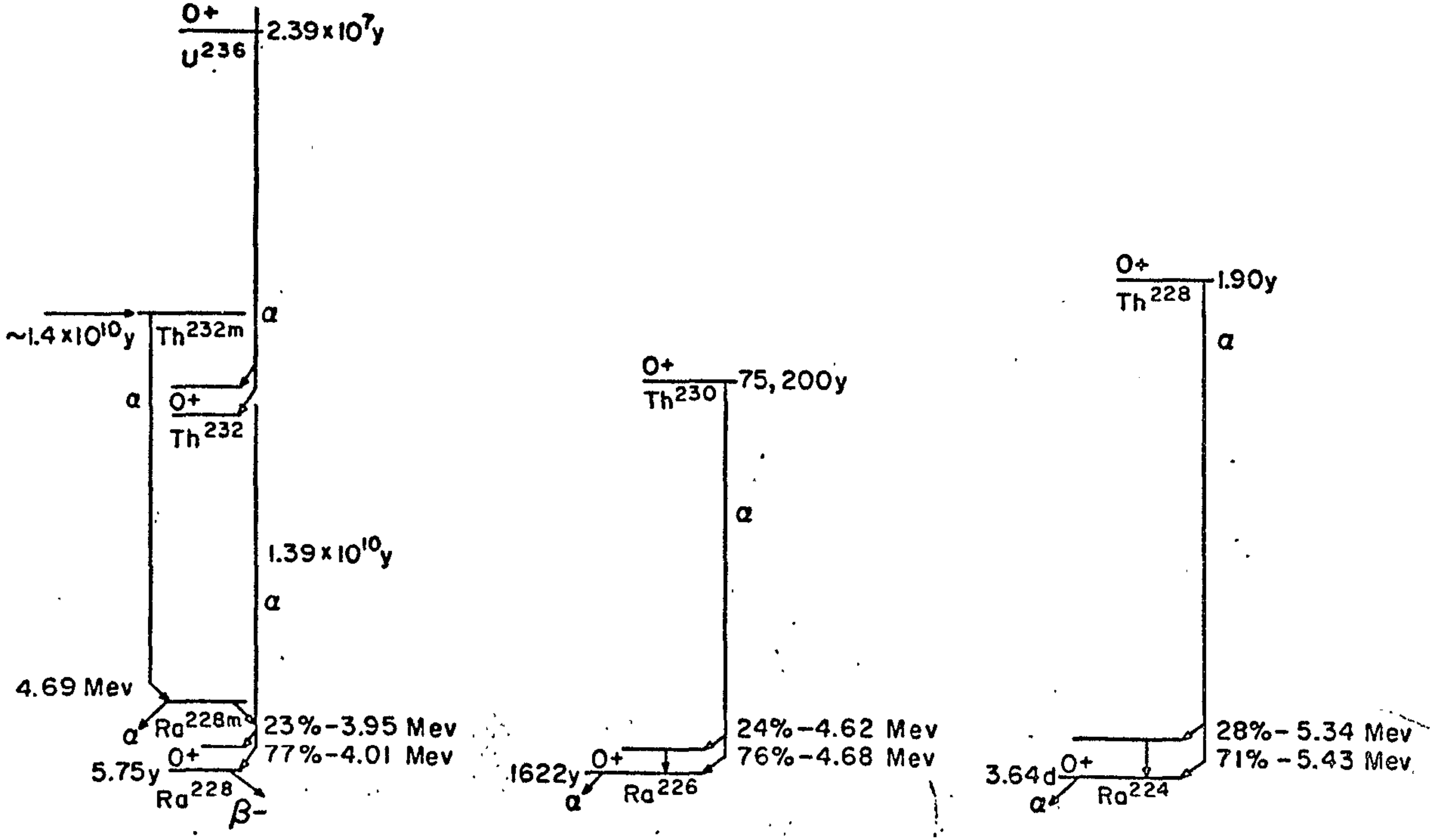

Figure 4. Suggested decay scheme and α-particle energy of Th^{232m} and its relations to Th^{232}, Th^{230}, and Th^{228}.

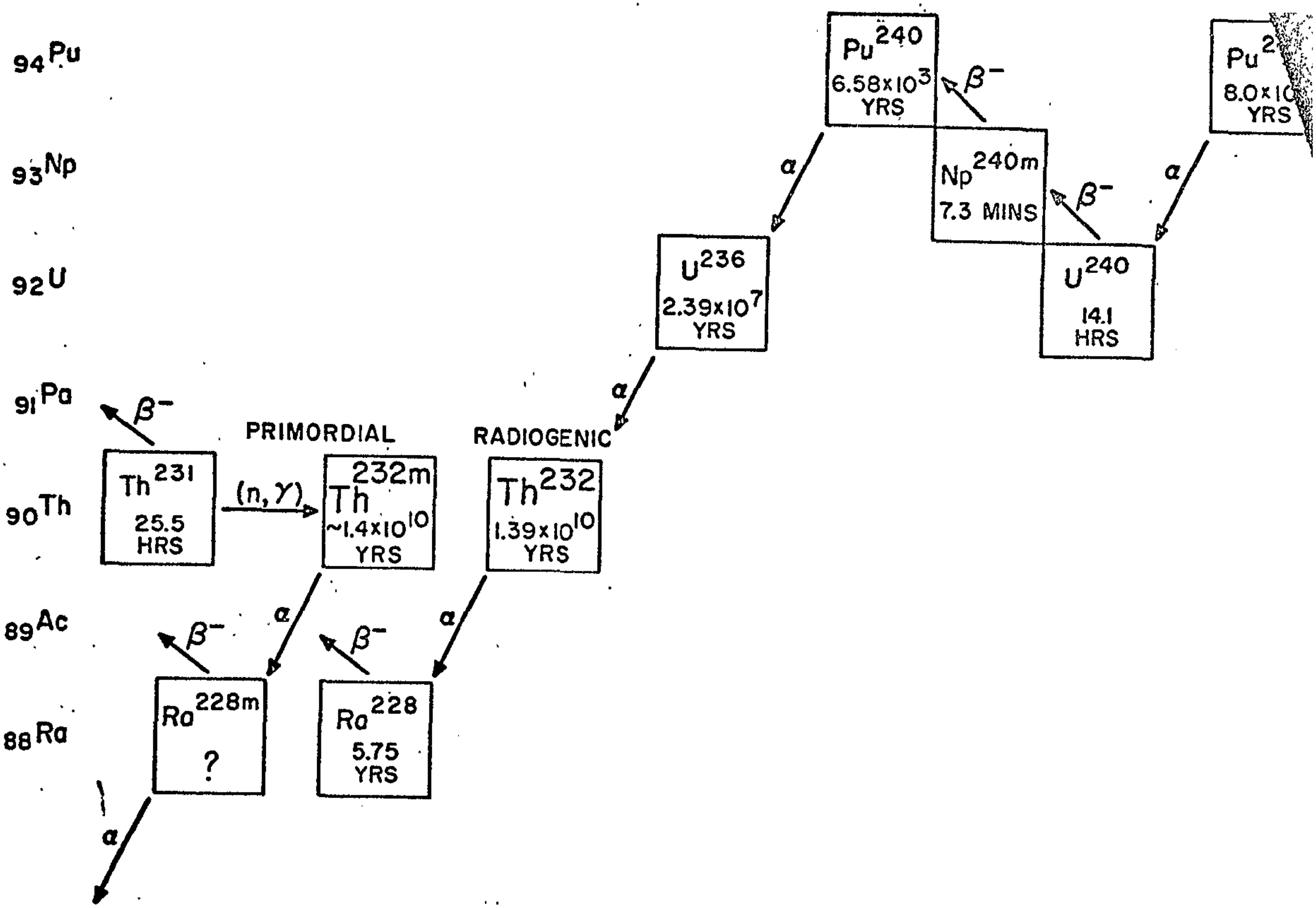

Figure 5. Block diagram of different modes suggested for the production of primordial Th^{232m} component of thorium and radiogenic Th^{232}.

mposition of lead of Apollo 12 samples

Atomic Ratio				
	Corrected for blank			
204	Pb^{206}/Pb^{204}	Pb^{207}/Pb^{204}	Pb^{208}/Pb^{204}	U^{238}/Pb^{204}
	396.6	168.9	403.3	505.6
	672.2	270.3	671.9	871.9
	410.7	172.3	402.3	487.5
	293.4	97.85	304.3	447.0
	391.2	158.8	428.5	473.0
	225.5	96.5	258.6	-
	796.7	331.3	789.2	985.6
	720.2	299.9	710.5	863.3
	739.9	306.9	724.8	972.9
	449.6	234.9	444.6	510.6
	1388	660.3	1217	2485
	924.2	462.9	863.0	1765
	1134	577.6	1028	1581
	433.4	251.7	415.6	495.5
	1201.7	641.6	1203	1612
	(415)**	(230)**	(423)**	(435)**
	2059	1050	1773	2773
	1438	768.1	1266	1510
	(1329)***	(707)***	(1149)***	(1398)***

ned igneous rock; C, breccia; D, soil.
sition was calculated using Pb^{208}/Pb^{206} ratio in #09A.
tamination was 14.56 ppm for sawing.

Table 3. Isotopic co

		Raw data		
Sample No.	Type*	Pb^{206}/Pb^{204}	Pb^{207}/Pb^{204}	Pb^{208}/Pb
12009,22	A	375.7	160.4	383.1
12021,122	B	606.3	244.6	608.0
12022,37	B	377.0	162.1	378.6
12035,10	B	247.0	83.96	259.4
12038,42,a	A	338.9	139.6	376.4
12038,42,b	A	207.8	89.6	239.8
12052,66	A	714.9	298.1	710.3
12063,49,a	A	645.1	269.4	638.6
12063,49,b	A	551.6	230.9	545.6
12064,21	B	397.8	208.5	395.7
12034,16,a	C	926.4	459.7	850.1
12034,16,b	C	555.7	280.05	524.7
12033,53	D	985.5	502.8	896.2
12070,56	D	400.9	233.2	387.7
12013,10	A			
#09A		898.8	481.4	904.8
#09B				
#42		1892	965.6	1631
#45		1202	643.2	1062
sawdust		39.63	26.77	56.4

*Type: A, fine-grained igneous rock; B, medium-grai

**Corrected for Pb^{208} spike. The lead isotopic compo

***A reasonable value was calculated assuming lead con

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