

EARLY HISTORY OF THE MOON: IMPLICATIONS OF U-TH-PB AND RB-SR SYSTEMATICS

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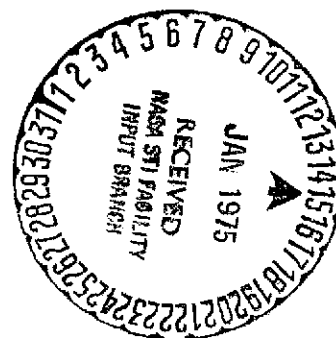
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EARLY HISTORY OF THE MOON:
IMPLICATIONS OF U-TH-PB and RB-SR SYSTEMATICS

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Abstract--Anorthosite 60015 contains the lowest initial $^{87}\text{Sr}/^{86}\text{Sr}$ ratio (0.69884 \pm 0.00004) yet reported for a lunar sample. The initial ratio is equal to that of the achondrite Angra dos Reis and slightly higher than the lowest measured $^{87}\text{Sr}/^{86}\text{Sr}$ ratio for an inclusion in the C3 carbonaceous chondrite Allende. The Pb-Pb ages of both Angra dos Reis and Allende are 4.62×10^9 years (4.62 billion years). Thus, the initial $^{87}\text{Sr}/^{86}\text{Sr}$ ratio found in lunar anorthosite 60015 strongly supports the hypothesis that the age of the moon is about 4.65 b.y. The $^{238}\text{U}/^{204}\text{Pb}$ value estimated for the source of the excess lead in "orange soil" 74220 is ~ 35 and lower than the values estimated for the sources of KREEP (600-1000), high K (300-600) and low K (100-300) basalts. We hypothesize from these and other physical, chemical and petrographic results: (1) that the moon formed ~ 4.65 b.y. ago, (2) that a global-scale gravitational differentiation occurred at the beginning of lunar history, and (3) that the differentiation resulted in a radical chemical and mineralogical zoning in which the $^{238}\text{U}/^{204}\text{Pb}$ ratios increased toward the surface, with the exception of the low $^{238}\text{U}/^{204}\text{Pb}$ surficial anorthositic layer which "floated" at the beginning of the differentiation relative to the denser pyroxene-rich material. All Apollo

and Luna U-Pb data which scatter about a 3.9-4.5 b.y. concordia chord (and its extension above concordia) and Rb-Sr whole rock model ages older than 3.9 b.y. may be readily interpreted as reflecting multiple impact events occurring between ~4.5 b.y. and ~3.9 b.y. ago. We propose that the ~4.47 b.y. age observed for some KREEP material corresponds to the "South Imbrium" impact event. Abundant Rb-Sr internal isochron and $^{40}\text{Ar}/^{39}\text{Ar}$ ages of lunar basalts indicate that a major period of mare basalt flooding occurred from ~3.8 b.y. to ~3.2 b.y. ago.

INTRODUCTION

The purposes of this paper are to present an extremely primitive $^{87}\text{Sr}/^{86}\text{Sr}$ ratio observed in lunar anorthosite 60015, to compare the ratio to similar data obtained from meteorites, and to summarize how U-Th-Pb and Rb-Sr systematics of lunar samples relate to the early history of the moon. Although we have tried to cite most of the important papers relating to this subject, no attempt has been made to complete an exhaustive bibliography of all U-Th-Pb and Rb-Sr reports, owing to space and time limitations.

Enrichment of refractory elements and deficiency in volatile elements in the lunar rocks and their source regions down to a few hundred kilometers depth relative to chondritic and terrestrial abundances have been well documented by many investigations. For example, VINOGRADOV (1971, 1972), GANAPATHY et al. (1970), GANAPATHY and ANDERS (1974), GAST et al. (1970) and HASKIN et al. (1970) clearly demonstrated that volatile chalcophile (Ag, Zn, Cd, In, Tl, and Bi) and siderophile (Au, Ir, Co and Ni) elements are depleted in lunar rocks, whereas refractory elements (Ca, Zr, Hf, Ta, and rare earth elements) are 10- to 100-fold enriched relative to chondritic abundances. LSPET (1969, 1970) and TERA et al. (1970) have

shown that alkali elements are depleted in Apollo 11 rocks and the K/U ratios of lunar material (~ 2500) are low compared to terrestrial (10^4) and chondritic (8×10^4) values. Chronological studies have also shown that the U/Pb ratios are high and the Rb/Sr ratios are low in lunar material (e.g. TATSUMOTO and ROSHOLT, 1970; Silver, 1970; Lunatic Asylum, 1970; Compston et al., 1970). These unique features have been interpreted as being due to the preferential accretion of high-temperature condensates in the later stage of the moon's formation from the solar nebula (GAST et al., 1970; GAST and McCONNELL, 1972). Alternatively, it might be argued that some volatile elements escaped from a dominantly chondritic moon during accretion. Although it is still debatable whether the moon originally accreted heterogeneously or homogeneously, geophysical, geochemical, and petrological studies indicate that the moon is now heterogeneously zoned both chemically and mineralogically.

INITIAL $^{87}\text{Sr}/^{86}\text{Sr}$ FOR METEORITES

If a rock of a planetary body is Rb free and remains closed with respect to Rb and Sr migration, the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio is constant from the time of rock formation. The lower the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio in such a rock, the earlier we infer that it formed and that the host planetary body must have separated from the solar nebula. This reasoning assumes that the solar nebula had a uniform Sr isotopic composition and Rb/Sr ratio at the time of its formation. Achondrites are nearly Rb free and the initial $^{87}\text{Sr}/^{86}\text{Sr}$ ratio can be determined rather accurately. Extensive studies on initial $^{87}\text{Sr}/^{86}\text{Sr}$ values in different planetary bodies have been performed especially by Wasserburg's group. PAPANASTASSIOU and WASSERBURG

(1969) reported a primitive initial $^{87}\text{Sr}/^{86}\text{Sr}$ value of BABI (Basaltic Achondrite Best Initial) for seven basaltic achondrites. Since then, $\text{BABI} = 0.69897 \pm 0.00003$ (standardized) has been used as a reference value to calculate Rb-Sr model ages of planetary bodies.

All laboratories normalize their Sr data to $^{86}\text{Sr}/^{88}\text{Sr} = 0.11940$ to correct for mass fractionation. Measured $^{87}\text{Sr}/^{86}\text{Sr}$ ratios among different laboratories, however, reflect a small bias which must be considered if meaningful comparison of very precise $^{87}\text{Sr}/^{86}\text{Sr}$ ratios measured in the different laboratories is to be made. The National Bureau of Standards' SRM-987 $^{87}\text{Sr}/^{86}\text{Sr}$ ratios (the certified value is 0.71014) obtained in Wasserburg's, Nyquist's and our laboratories are 0.71015, 0.71026, and 0.71018, respectively. In this paper, we standardize all data to the NBS value.

PAPANASTASSIOU (1970) reported a lower initial $^{87}\text{Sr}/^{86}\text{Sr}$ ratio for the augite achondrite Angra dos Reis ($\text{ADOR} = 0.69883 \pm 0.00003$) and suggested the achondrite was formed earlier than other achondrites. Recent $^{207}\text{Pb}/^{206}\text{Pb}$ age determinations of three achondrites--Angra dos Reis, Sioux County, and Nuevo Laredo (TATSUMOTO *et al.*, 1973a)--support PAPANASTASSIOU'S (1970) relative age interpretation. Using the Canyon Diablo troilite Pb as a common initial Pb, TATSUMOTO *et al.* (1973a), calculated a $^{207}\text{Pb}/^{206}\text{Pb}$ age of 4.62×10^9 yr (b.y.) for the angrite Angra dos Reis and ages $\sim 30 \times 10^6$ yr (m.y.) younger than this for the eucrites, Sioux County and Nuevo Laredo.

A more primitive initial $^{87}\text{Sr}/^{86}\text{Sr}$ ratio for Ca-Al-rich chondrules separated from the Allende carbonaceous chondrite was reported by GRAY

et al. (1973, ALL = 0.69876 ± 0.00002) and WETHERILL et al. (1973, 0.69880). Theoretical and petrographic studies suggest that the Ca-Al-rich inclusions in Allende are among the earliest condensates of the solar nebula (CLARKE et al., 1970; MARVIN et al., 1970; KEIL and FUCHS, 1971; and GROSSMAN, 1972, 1973). Although WETHERELL et al. (1973) observed that all their Allende Rb-Sr data points lay between lines corresponding to ages of 4.5 b.y. and 4.7 b.y., GRAY et al. (1973) showed, in their more extensive study, that the Allende inclusions do not form an isochron in a Rb-Sr evolution diagram. Rather, their data scattered between 4.63 and 3.59 b.y. BABI model ages. They concluded that the Rb-Sr isotopic systems in Allende may have been seriously disturbed within the past 3.6 b.y. and they further pointed out that the range of initial $^{87}\text{Sr}/^{86}\text{Sr}$ ratios they found for Allende inclusions could reflect differences of time and possibly places of condensation from the solar nebula prior to agglomeration.

U-PB AGES OF METEORITES

Recently, TATSUMOTO et al. (1973) and TILTON (1973) verified Patterson's (1955) original meteorite age of 4.55 b.y. and reported more precise lead isotopic compositions and Pb-Pb ages of meteorites. TILTON (1973) reported a whole rock $^{207}\text{Pb}/^{206}\text{Pb}$ age of 4.635 b.y. for two carbonaceous and four ordinary chondrites, while TATSUMOTO et al. (1973a) reported that the Pb-Pb age of Angra dos Reis is 4.62 b.y. and that two eucrites are 4.59 b.y. old, using the uranium decay constants $\lambda_{^{238}\text{U}} = 0.15369 \times 10^{-9} \text{ yr}^{-1}$; $\lambda_{^{235}\text{U}} = 0.97216 \times 10^{-9} \text{ yr}^{-1}$).

TATSUMOTO et al. (1973c) reported a Pb-Pb "internal isochron" of the Allende meteorite which will be presented in detail elsewhere later. Their analyses of the matrix material, a magnetic separate, and six inclusions (chondrules and aggregates) yielded a range of $^{206}\text{Pb}/^{204}\text{Pb}$ ratios from 9.9 to 56, and define an isochron with a slope of 0.6188 ± 0.0016 (2σ , YORK, 1969) in a $^{207}\text{Pb}/^{204}\text{Pb}$ versus $^{206}\text{Pb}/^{204}\text{Pb}$ diagram. This line passes within error through the primordial Pb isotopic values (TATSUMOTO et al., 1973a) and corresponds to an age of 4.62 b.y. This age is in good agreement with the Pb-Pb age of Angra dos Reis although the position of one chondrule below this line may reflect later "metamorphic processes," which GRAY et al. (1973) considered to explain the scatter they obtained for a Rb-Sr isochron plot of the various Allende inclusions.

In view of the irregularities displayed by Allende Rb-Sr data, it is perhaps surprising that the Pb isotope data are so linear. Because a Pb-Pb age is not sensitive to very recent U-Pb fractionation and the Allende inclusions do not yield concordant U-Th-Pb ages, it seems likely that the hypothesized time of the Rb-Sr system disturbance is very recent -- possibly related to the break-up of a parent body. Angra dos Reis is a well-differentiated augite-rich achondrite but Allende is an agglomerate of chemically unequilibrated materials. We interpret the Allende 4.62-b.y. internal Pb-Pb isochron age as probably reflecting the time of agglomeration. The significance of the Pb-Pb age agreement and the initial $^{87}\text{Sr}/^{86}\text{Sr}$ difference between the two meteorites are not clear to us. The Pb-Pb age agreement for these two meteorites may indicate that the time span separating accretion of many planetesimal bodies was rather short. A detailed discussion about the inconsistency among Pb-Pb, Rb-Sr, and K-Ar

(FIREMAN et al., 1970) ages is beyond the scope of this paper. The above discussion is presented to show that Allende contains the most primitive $^{87}\text{Sr}/^{86}\text{Sr}$ ratio yet reported and that the Allende Pb-Pb internal isochron age is 4.62 b.y.

INITIAL $^{87}\text{Sr}/^{86}\text{Sr}$ OF THE MOON

If we find a lunar rock with an initial $^{87}\text{Sr}/^{86}\text{Sr}$ ratio as primitive as the meteorites discussed above, it would be indirect evidence for a similar old age of the moon. The lunar anorthosites are excellent candidates for the search for primitive $^{87}\text{Sr}/^{86}\text{Sr}$ ratios because (1) they concentrate Sr and exclude Rb during crystallization and (2) at least some of them may have formed during an early stage of lunar differentiation (WOOD et al., 1970; SMITH et al., 1970). Rather primitive initial $^{87}\text{Sr}/^{86}\text{Sr}$ values as low as BABI have been reported for lunar anorthosites 15415 (0.69896 ± 0.00004 , WASSERBURG and PAPANASTASSIOU, 1971) and 60025 (0.69893 ± 0.00003 , PAPANASTASSIOU and WASSERBURG, 1972).

Anorthosite 60015

The lunar rock 60015 is a football-sized, white, coarse-grained anorthosite which is coated with about a 1-cm-thick rind of dark-brown glass. The glass contained white fine-grained plagioclase clasts. The clasts appeared to be incorporated in the glass from either preexisting anorthosite or anorthositic breccia when the glass formed by meteoroid impacting (Lunar Sample Information Catalogue, 1972; SCLAR and BAUER, 1974).

A coarse-grained interior gray plagioclase sample, a portion of plagioclase about 2 mm from the plagioclase-glass boundary, white plagioclase clasts in the glass, and the dark glass coating itself were all analyzed for Rb, Sr, U, Th, Pb, and K. The analytical results of the U-Th-Pb and the Rb-Sr analyses for

these samples of 60015 are shown in Tables 1 and 2, respectively. We obtained each $^{87}\text{Sr}/^{86}\text{Sr}$ ratio by averaging 10 sets of 20 ratios, measured on an automated mass spectrometer, using programmed magnet switching and on-line digital data acquisition and processing. The rubidium and strontium concentrations were determined by isotope-dilution analysis using ^{87}Rb and ^{84}Sr spikes. Corrections for mass spectrometric fractionation were made by normalization to $^{86}\text{Sr}/^{88}\text{Sr} = 0.11940$. The errors given in Table 2 are twice the standard deviation of the mean. Interlaboratory comparison and reproducibility of our data were checked with replicate ^{84}Sr spiked runs of the National Bureau of Standards' standard SRM-987 which gave an average $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of 0.710184 ± 0.000025 ($2\sigma_m$) for 10 separate Sr isotope analyses during this investigation.

The U-Pb data of various portions of anorthosite 60015 yield insight into times of Pb migration relative to U in the glass rind and interior of anorthosite 60015. In the following section, this age information is briefly considered in order to justify the ages used for the in-situ Rb decay corrections in calculating initial $^{87}\text{Sr}/^{86}\text{Sr}$ ratios.

i) U-Pb age of 60015--As we previously discussed (NUNES et al., 1973), although the Pb in the interior of anorthosite 60015 may have been slightly contaminated with terrestrial Pb, calculated U-Pb model ages of 3.57 and 3.80 b.y., using two- and three-stage U-Pb evolution models respectively, agree well with the $^{40}\text{Ar}/^{39}\text{Ar}$ age of 3.55 ± 0.05 b.y. that SCHAEFFER and HUSAIN (1974) reported for this rock. Indeed, this agreement between the $^{40}\text{Ar}/^{39}\text{Ar}$ age and the U-Pb model ages suggests terrestrial Pb contamination in our sample of 60015 was negligible. In any case, these age data, together with the very primitive $^{87}\text{Sr}/^{86}\text{Sr}$ ratio observed in this anorthosite, indicate

that Pb was probably introduced into the interior of a preexisting anorthosite about 3.5 to 4.0 b.y. ago due to metamorphism likely related to one or more impact events. Introduction of the Pb into anorthosite 60015 at this time was apparently accompanied by Ar loss from the plagioclase as indicated by the $^{40}\text{Ar}/^{39}\text{Ar}$ age and may have been accompanied by gain, or loss of Rb as well.

U-Th-Pb data for the 60015 rind, plagioclase clasts within this rind, glassy white material taken from the glass-plagioclase boundary, plagioclase ~ 2 mm from the boundary, and gray plagioclase from the interior of the rock (Table 1) are plotted in Figure 1. Also plotted for reference in Figure 1 is the Apollo 16 South Ray crater soil field (NUNES *et al.*, 1973).

The 60015 plagioclase clast sample and the glassy white boundary material (Figure 1) apparently both underwent an increase of their $^{206}\text{Pb}/^{207}\text{Pb}$ ratios and possibly a net gain of Pb relative to U when the glass rind formed. The glass rind itself lost much Pb relative to U during formation. It appears from Figure 1 that (1) the U-Pb system of the plagioclase 2 mm from the plagioclase-glass boundary was very slightly or not at all affected, by this event, (2) the plagioclase interior was negligibly affected, and (3) the plagioclase clast sample included in the glass was the most strongly affected plagioclase sample measured. If the Pb isotopic compositions of the glass rind and the plagioclase sample from within this glass rind completely equilibrated during the glass formation, and these samples remained closed to the migration of U and Pb after glass formation, the line connecting these 2 samples (line BA, Fig. 1) which corresponds to an age of ~ 200 m.y., dates the time of the glass rind formation.

The U concentrations of the black glass rind (~ 0.4 ppm, Table 1), the South Ray Crater soils (~ 0.6 ppm, NUNES et al., 1973) and the anorthosite interior (~ 0.0015 ppm U, NUNES et al., 1973) suggest that the glass rind is a mixture of about $\sim 65\%$ soil and $\sim 35\%$ anorthosite.

Because evidence of Pb isotope equilibration between the plagioclase clasts and the glass is elusive, the 200 m.y. age may have no meaning at all. In any case, a maximum age of glass formation of ~ 1.1 b.y. may be obtained from the line connecting the 60015 interior sample with the glass rind sample (line EA, Fig. 1) since it is highly unlikely that the initial $^{206}\text{Pb}/^{207}\text{Pb}$ ratio in the glass was any lower than that in the anorthosite. The $^{40}\text{Ar}/^{39}\text{Ar}$ temperature-release pattern for the glass rind was highly irregular and did not yield a reliable age (SCHAEFFER and HUSAIN, oral communication, Mar., 1974) which is compatible with the very young glass age of < 1.1 b.y. we infer from the U-Pb systematics.

ii) The initial $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of 60015--Except for the plagioclase clast sample included in the black glass, the observed $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of 60015 plagioclase (Table 2) are similar to BABI. The observed $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of the plagioclase clasts included in the glass rind is 0.69887 ± 0.00003 --significantly below BABI. Because the clasts may have gained Pb at the time of glass rind formation as discussed above, the Rb we measured in this sample may also have been incorporated at that time. If this is the case, the observed $^{87}\text{Sr}/^{86}\text{Sr}$ ratio would closely approximate the initial $^{87}\text{Sr}/^{86}\text{Sr}$ ratio since a ^{87}Sr correction due to in-situ decay of Rb over the last 1.0 b.y. would lower the ratio by only 0.00002. Of course, some Rb may have been present prior to ~ 1.0 b.y. ago and the observed $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of the clast sample is therefore an upper limit for the true initial value. Assuming a

residence time of 4.0 b.y. for the Rb inside the plagioclase sample collected 2 mm from the glass-plagioclase boundary, we calculate a Rb-corrected $^{87}\text{Sr}/^{86}\text{Sr}$ initial ratio of 0.69886--in good agreement with the observed $^{87}\text{Sr}/^{86}\text{Sr}$ ratio obtained from the plagioclase included in the glass (Fig. 2). Similarly correcting the plagioclase interior sample for in-situ decay of Rb over the past 4.0 b.y. yields an initial $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of 0.69892. We obtain the initial $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of 0.69888 ± 0.00007 , 0.69882 ± 0.00006 , and 0.69881 ± 0.00003 ^(standardized) for the gray plagioclase interior, a portion of plagioclase 2 mm from the glass-plagioclase boundary, and the plagioclase clast sample in the glass, respectively. The averaged initial $^{87}\text{Sr}/^{86}\text{Sr} = 0.69884 \pm 0.00004$ for anorthosite 60015 compares with higher values for lunar anorthosites 60025 (0.69902 ± 0.00006 , this paper; 0.69893 ± 0.00003 , PAPANASTASSIOU and WASSERBURG, 1972), 15415 (0.69896 ± 0.00004 , WASSERBURG and PAPANASTASSIOU, 1972), and BABI (0.69897 ± 0.00003 , PAPANASTASSIOU and WASSERBURG, 1969).

PAPANASTASSIOU and WASSERBURG (1972) reported the initial $^{87}\text{Sr}/^{86}\text{Sr}$ value for anorthosite 60025 as 0.69893 ± 0.00003 and stated that this was the first clear-cut evidence that the moon formed with a slightly more primitive $^{87}\text{Sr}/^{86}\text{Sr}$ ratio than BABI. Subsequently, NYQUIST et al. (1973) reported a still lower initial $^{87}\text{Sr}/^{86}\text{Sr}$ ratio for anorthositic clasts in breccia 61016. Correcting their observed data for in-situ Rb decay over the last 4.6 b.y., they obtained LUNI (lunar initial) = 0.69888. The initial $^{87}\text{Sr}/^{86}\text{Sr}$ ratios reported here for anorthosite 60015 appear to be the lowest yet reported for any lunar sample, although these initial $^{87}\text{Sr}/^{86}\text{Sr}$ ratios are not outside of error below LUNI (NYQUIST et al., 1973). Further efforts to verify the 60015 lowest $^{87}\text{Sr}/^{86}\text{Sr}$ initial ratio are in progress. BIRCK and ALLEGRE (1974) recently reported orally that the initial $^{87}\text{Sr}/^{86}\text{Sr}$ ratio for the anorthosite 60015 is 0.6988.

The above discussion is presented not only to compare small differences in the initial $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of lunar samples, but also to emphasize that the lowest $^{87}\text{Sr}/^{86}\text{Sr}$ ratio yet found for the moon is essentially equal to that found for the achondrite Angra dos Reis (ADOR = 0.69883 ± 0.00003 , PAPANASTASSIOU, 1970) and apparently slightly higher than the lowest measured $^{87}\text{Sr}/^{86}\text{Sr}$ ratio found for the Allende carbonaceous chondrite (ALL = 0.69876 ± 0.00002 , GRAY et al., 1973; 0.69880 , WETHERILL et al., 1973). The agreement between the lowest $^{87}\text{Sr}/^{86}\text{Sr}$ ratio for the moon and Angra dos Reis indicates that the moon formed about the same time or earlier than did Angra dos Reis, since the lowest documented lunar initial $^{87}\text{Sr}/^{86}\text{Sr}$ ratio (this paper) is an upper limit for the absolute lunar initial $^{87}\text{Sr}/^{86}\text{Sr}$ ratio (i.e. we may not have found the lowest lunar $^{87}\text{Sr}/^{86}\text{Sr}$ ratio yet). Because Angra dos Reis has a Pb-Pb age of ~ 4.62 b.y. (TATSUMOTO et al., 1973a), it follows that the moon formed at least ~ 4.62 b.y. ago. This reasoning assumes, of course, that the moon and Angra dos Reis both condensed from the same solar nebula which was homogeneous with respect to its Sr isotopic composition and Rb/Sr ratio.

Recently, Albee et al. (1974) reported an old Rb-Sr two point isochron age of 4.60 ± 0.09 b.y. for Apollo 17 dunite breccia 72417 and concluded that the rock represents a cumulate formed during early lunar differentiation. Although the Pb model age on the leach of the rock is 4.48 b.y. indicating Pb addition at later times (TERA et al., 1974), the Rb-Sr isochron age strongly supports our hypothesis that the moon is ~ 4.65 b.y. old.

SUMMARY OF U-Th-Pb and Rb-Sr SYSTEMATICS OF LUNAR SAMPLES

Lunar differentiation model

Many researchers have demonstrated from Apollo 11 sample studies that refractory elements--for example, Zr, Hf, and the rare earths--are enriched in lunar surface material with respect to solar abundances and, on the other hand, volatile elements--for example Bi, Zn, Cd, Tl, Pb, Cl, and Br--are significantly depleted with respect to solar abundances. We use the U/Pb ratio as one way to measure relative refractory enrichment and volatile depletion of rocks. The ratio is commonly expressed in the ratio $^{238}\text{U}/^{204}\text{Pb}$ (μ) as measured today. The Pb isotopic compositions of lunar samples are quite radiogenic compared to compositions of carbonaceous chondrites, ordinary chondrites and terrestrial samples. The observed μ 's for low K lunar basalts range from about 300 to 500 and those for high K basalts range from about 600 to 1000, whereas the μ 's of ordinary chondrites (TATSUMOTO et al., 1973a, TILTON, 1973) and terrestrial basalts (e.g. TATSUMOTO, 1966) are about 8 to 30 and those of carbonaceous chondrites are < 1 to 4. Ca-Al-rich inclusions in Allende meteorites (TATSUMOTO et al., 1973c) have μ 's ranging from 15 to 40, while μ 's of Mg-rich inclusions range from 7 to 10. Only the μ 's (about 200) of achondrites approach those of lunar mare basalts (TATSUMOTO et al., 1973a).

U-Th-Pb systematics of the lunar rocks indicate that typical mare basalts and KREEP rocks were derived from sources in which μ ranged from about 100 to 300 and from 600 to 1000, respectively, assuming an age of 4.65 b.y. for

the moon and a simple two-stage U-Pb evolution history. Of course, the μ values will be different if we use a different lunar origin age and/or different evolution models for the moon, although relative relations for the source μ 's are not grossly changed. It is not clear to what extent large multiple impacts may have altered μ values in source rocks, thereby making these observations qualitative rather than quantitative.

Most lunar breccias and soils contain excess Pb relative to U. The excess Pb in these materials was apparently released from other rocks during lunar outgassing due to volcanic activity and meteoroid impact events. In particular, anorthosite 15415 (TATSUMOTO et al., 1972a; TERA et al., 1972) and 60015 (NUNES et al., 1973), a "Rusty Rock" breccia 66095 (NUNES AND TATSUMOTO, 1973), and "Orange glass" 74220 (TATSUMOTO et al., 1973b) contain considerable excess Pb which is not supported by in-situ decay of U and Th. Excess Pb in the anorthosites appears to have been produced in $\mu \sim 300$ source environments equivalent to basaltic sources and that in breccia 66095 appears to have been produced in a $\mu \sim 1000$ which is typical of KREEP sources. All these U-Pb data indicate that the moon was originally extremely depleted in the volatile element Pb relative to cosmic abundances.

A recent U-Th-Pb systematics study of Apollo 17 "Orange Soil" 74220, however, indicates that Pb in this sample was derived from a source with a μ of about 35 (TATSUMOTO et al., 1973b; SILVER, 1973). The origin of the glass spherules in soil 74220 is thought to have resulted from either meteorite impact into a lava lake or lava fountaining (e.g. ROEDDER and WEIBLEN, 1973; PRINZ et al., 1973; MCKAY and HEIKEN, 1973; CADENHEAD, 1973). We prefer the lava fountain hypothesis, because Pb and other volatile element concentrations are too high to be easily derived from a volatile-rich carbonaceous chondrite via some sort of impact transfer.

On the basis of geophysical and geochemical constraints, some investigators have suggested that the moon formed from a mixture of high-temperature condensates (like the Ca-Al-rich inclusions in the Allende meteorite) and CI carbonaceous chondritic material. ANDERSON (1973) postulated that the moon is composed almost entirely of pre-iron high-temperature condensates like the Ca-Al-rich inclusions in the Allende meteorite (GROSSMAN, 1973), whereas WANKE et al. (1973, 1974) suggested that the moon is made of 60% high-temperature condensates and 40% chondrites, and GANAPATHY and ANDERS (1974) suggested a mixture of 25% high-temperature condensate component and 75% chondritic component. The CI carbonaceous chondrites contain about 8 ppb U and the Ca-Al-rich condrules and aggregates in Allende contain about 110 ppb U. If a simple two-component mixing model for the moon is appropriate, the U concentration data suggest that the moon is a 50-50 mixture of the high-temperature condensate and chondritic material. However, GROSSMAN et al. (1974) pointed out that the oxygen isotope data for the Allende high-temperature inclusions are very different from anything yet found on the moon and that consideration of the Allende Ca-Al-rich inclusions as a significant lunar component is therefore not strictly correct.

In any case, the moon did accrete from the solar nebula into a body in which volatile elements were depleted and refractory elements enriched relative to cosmic abundances about 4.65 b.y. ago. We find most attractive the hypothesis that the moon underwent large-scale and possibly total melting by mostly gravitational energy at the beginning of its history (e.g. MIZUTANI et al., 1972). This hypothesized primary accretional global melting would result in global differentiation, with feldspar "floating"

(WOOD et al., 1970; WOOD, 1972) relative to the more dense succession of pyroxene-rich assemblages accumulating in the mantle (RINGWOOD and ESSENE, 1970; PHILPOTS et al., 1972; HASKIN et al., 1973, 1974).

As previously discussed (TATSUMOTO, 1973) a plagioclase-rich noritic layer was likely formed between an anorthositic surface layer and a pyroxenite mantle, as a result of silicate liquid accumulation during the primary differentiation stage. Apparently the lunar crust consists of both anorthositic and noritic layers and is about 60 km thick (e.g. TOKSOZ et al., 1973). The anorthositic surface layer contains only small amounts of K, Rb, Pb, U, and Th. Anorthosite 60015, which has an initial $^{87}\text{Sr}/^{86}\text{Sr}$ ratio as low as that determined for Angra dos Reis, probably formed in this outer anorthositic layer during our hypothesized primary differentiation. Although preliminary data on anorthosite 15415 (e.g. TATSUMOTO et al., 1972a; TERA et al., 1972) suggested ~ 0.2 ppm Pb might be typical of lunar anorthosites, later studies (TERA and WASSERBURG, 1972b; NUNES et al., 1974) revealed that at least some anorthosites could be almost free of Pb as well as of U and Th.

The noritic layer contained a large amount of compatible lithophile elements such as K, REE, P, Rb, U, and Th. This layer was presumably the source of the KREEP component in KREEP-rich rocks. We hypothesize that the crust-upper mantle layering was established ~ 4.63 b.y. ago and that the mantle likely continued to differentiate after this time. The 4.60 b.y. age for dunite 72417 (ALBEE et al., 1974) strongly supports this hypothesis. Meteoroid impact events around 4.47 b.y. ago may well have induced extrusion of KREEP-rich basalts (SCHONFELD and MEYER, 1972). KREEP-rich igneous rock 14310 contains 3.10 ppm U (TATSUMOTO et al., 1972b; TERA and WASSERBURG, 1972a) and KREEP itself was estimated to

contain 4.5 ppm U (SCHONFELD and MEYER, 1972). If KREEP source material contains only ~ 0.1 ppm U as suggested by HUBBARD and GAST (1971) then KREEP is only about a 2% partial melt of the source material if we assume that the partition coefficient of U measured for the terrestrial diopside-liquid system (SEITZ, 1973) is applicable to lunar rocks. In our model, as we discuss below, we assume that the noritic layer contains 0.4 ppm U or more and that the U in KREEP was increased to ~ 4 ppm U by $\sim 10\%$ partial melting.

The lunar pyroxenite mantle--whose μ 's appear to decrease from ~ 300 downward to 100--provided the source material for the mare basalts that were produced ~ 3.2 - 3.9 b.y. ago. The excess Pb in "Orange Soil" 74220, whose calculated first stage μ is about 35, was probably produced from a still deeper part of the lunar mantle than the mare basalt source region. These estimated μ changes suggest that the moon had well differentiated and formed a zonal structure in which μ 's decrease downward.

We prefer an extensive lunar differentiation model such as that proposed by NAKAMURA et al. (1974) and TAYLOR and JAKES (1974). If such a model is appropriate and the lunar asthenosphere deeper than 1000 km (or more) consists of pyroxenite (ANDERSON, 1973) or Fe-FeS (BRETT, 1972; TAKSOZ et al., 1974), then the noritic layer must contain 0.4 ppm or more uranium, assuming the present-day average U concentration is 0.06 ppm for the moon (LANGSETH et al., 1972). In this calculation, we followed TAYLOR and JAKES' (1974) geochemical zoning model and we assumed U concentrations in the mantle decrease downward from 70 to 20 ppb. The depth assignment for the lunar basalt sources of the TAYLOR-JAKES model might be too deep, considering possible U-Pb clock resetting of the sources by planetesimal impacts (NUNES

et al., 1974). If this is the case, the U concentration of the noritic layer probably is higher than 0.4 ppm.

Early chronology of the moon

Well-documented Rb-Sr internal isochron and $^{40}\text{Ar}/^{39}\text{Ar}$ ages of lunar mare basalts range from 3.2 b.y. to 3.8 b.y. (e.g. PAPANASTASSIOU et al., 1970; PAPANASTASSIOU and WASSERBURG, 1971; COMPSTON et al., 1970; TURNER, 1970; HUNEKE et al., 1973; HUSAIN et al., 1972; STETTLER et al., 1973). In this section we consider the moon's early history prior to the 4.0 b.y. "cataclysm" (TERA et al., 1973).

The first Apollo 11 lunar breccia and soil U-Pb analyses (Fig. 3) yielded nearly concordant U-Pb ages of ~ 4.65 b.y. These data together with the slightly discordant U-Pb Apollo 11 mare basalt analyses and ~ 3.8 b.y. Rb-Sr isochron age (Lunatic Asylum, 1970) prompted TATSUMOTO (1970a) to explain the mare basalt data with a simple two-stage U-Pb evolution model, which would have 4.60-4.66 b.y.-old source material produce mare basalt at ~ 3.8 b.y. ago. Using this model, TATSUMOTO (1970a) postulated that $\sim 1/3$ of the Pb which had accumulated in the basalt source material by in-situ decay of U and Th was transferred to the soil by volatilization during eruption ~ 3.8 b.y. ago. Acid leaching (SILVER, 1970) and size fraction (TATSUMOTO et al., 1971) studies provided evidence for lead mobility on the lunar surface. However, the U-Pb data of the acid leaches and of the acetone float fraction of soil 10081 do not exactly lie on the upper extension of the 3.8-4.65 b.y. discordia line, but rather on an extension of the 3.8-4.4 b.y. chord (fig. 4). U-Pb analyses of silicic brecciated rock 12013 (TATSUMOTO, 1971) and of mineral separates of basalt 12064 (TATSUMOTO et al., 1971) also yielded upper concordia intercept ages at ~ 4.4 b.y. This event became unambiguously clear when TERA and

WASSERBURG (1972a) obtained U-Pb internal isochrons for KREEP-rich basalts 14310 and 14053 that yielded concordia intercept ages of ~ 4.47 and ~ 3.95 b.y. Rb-Sr whole rock data of some KREEP-rich basalts and breccias also suggested a Rb-Sr equilibration event occurred approximately 4.45 b.y. ago (SCHONFELD and MEYER, 1972; NYQUIST *et al.*, 1972, 1973). In addition whole rock U-Pb data for "igneous rock" 68415 are concordant at ~ 4.47 b.y. (NUNES *et al.*, 1973), and TERA *et al.* (1973) also obtained an upper concordia intercept age of ~ 4.47 b.y. from a 2-point whole-rock-plagioclase isochron for this rock. Chemical and petrographic studies suggest that 14310, 14053, and 68415 (e.g. GREEN *et al.*, 1972; JAMES, 1973; HELZ and APPLEMAN, 1973; TATSUMOTO *et al.*, 1972b) are probably of regolith (impact melt) origin. We suggest that the upper concordia ages of about 4.47 b.y. for these rocks represent a period of impact melting and regional differentiation rather than a primary global differentiation resulting from lunar accretionary heating. The ~ 4.0 b.y. "Imbrium event" is then envisioned as another much later impact event. U-Pb data of almost all terra cataclastic anorthosites, breccias and soil samples from Apollo 14, 15, and 16 and Luna 16 and 20 sites plot on or near an extension of the 4.0 - 4.47 b.y. concordia chord on a U-Pb evolution diagram (TERA *et al.*, 1974)(Fig. 4). The distinct non-linear scatter of these data, however, requires at least three stages of U-Pb evolution for lunar highland rocks, and suggests that the 4.47 b.y. age may be of regional rather than global scale (NUNES and TATSUMOTO, 1973).

Whole rock U-Pb analyses of the Apollo 17 mare basalts 74235, 72155, 71569 and 75035 (NUNES *et al.*, 1974) are within error concordant at 4.57 b.y., 4.54 b.y., 4.53 b.y. and 4.50 b.y., respectively (Fig. 4). The 4.57 ± 0.01 b.y. age of mare basalt 74235 is the oldest lunar concordant whole-rock U-Pb

age yet determined, and may be considered as a minimum age of the moon. ALBEE et al.'s (1974) Rb-Sr two-point isochron age of 4.60 ± 0.09 b.y. for a lunar dunite is compatible with this interpretation.

A suggestion that the moon is older than 4.85 b.y. was made by SILVER (1970). Later, SILVER (1973) reported a soil from Apollo 16 with U-Pb concordant ages in excess of 5 b.y., and pointed out that the determination of the age of the moon by a concordia treatment of soils and breccias is not yet feasible. Although most breccia and soils have apparent $^{207}\text{Pb}/^{206}\text{Pb}$ ages > 4.65 b.y. and plot above concordia which is indicative of Pb gain relative to U, none of the breccia samples plot below the extension of the 4.0-4.65 b.y. chord above concordia in a U-Pb evolution diagram. U-Pb data of some soil samples which plot below this chord can be easily explained by "third event(s)" which occurred at a younger time; e.g. ~ 0.8 b.y. for Apollo 12 soils (TATSUMOTO et al., 1971) and ~ 2 b.y. for Apollo 14 (TATSUMOTO et al., 1972b) and Luna 20 soil (TATSUMOTO, 1973). Soils lost Pb relative to U by the "third event(s)." These "event" may be only apparent, due to an integrated bombardment history and associated Pb loss throughout the last ~ 4.0 b.y. (TATSUMOTO, 1973; CHURCH and TILTON, in press).

As TATSUMOTO and ROSHOLT (1970) originally pointed out, accepting the age of the moon as ~ 4.65 b.y. from their U-Pb data of Apollo 11 breccia and soil samples involved some speculation and these samples may have been only coincidentally nearly concordant at ~ 4.65 b.y. Regardless of whether or not this is the case, the very primitive $^{87}\text{Sr}/^{86}\text{Sr}$ ratios reported in this paper (anorthosite 60015) and by NYQUIST et al., (1973, for 61016 anorthosite clast) strongly support an age of ~ 4.65 b.y. for the origin of

the moon. As discussed above, all U-Pb and Rb-Sr data obtained from lunar samples are compatible with this primary age of ~ 4.65 b.y.

SCHAEFFER and HUSAIN (1974) interpreted their $^{40}\text{Ar}/^{39}\text{Ar}$ ages obtained from Apollo 16 and 17 samples, as being related to the McGETCHIN et al. (1973) basin excavation stratigraphy model, and suggested that their $^{40}\text{Ar}/^{39}\text{Ar}$ ages of 4.20 ± 0.05 , $4.05 - 4.20$, and 4.13 ± 0.05 b.y. were related to the Nectaris, Humorum, and Crisium excavations respectively. NUNES et al. (1974) recently attempted to correlate U-Pb lunar whole rock data with the McGETCHIN et al. (1973) basin excavation stratigraphy model. They suggested that the U-Pb and Rb-Sr whole rock documented ages of ~ 4.0 to ~ 4.5 b.y. correspond to multiple major basin excavation events. The preponderance of KREEP material in the Apollo 12 and 14 samples and high radiation just south of the Imbrian area led us (NUNES et al. 1974) to suggest KREEP was produced by a "South Imbrian" excavation event that occurred ~ 4.47 b.y. ago (Fig. 5). The unofficial term "South Imbrian" refers to the large basin whose center lies just east of Copernicus. SCHONFELD and MEYER (1973) have argued, primarily from the distribution of KREEP on the lunar surface, that the Imbrian event must predate the formation of KREEP some 4.4 b.y. ago. If the NUNES et al. (1974) explanation is correct, however, KREEP may have been confined to an area just south of the Imbrian basin and the Imbrian impact event may not have caused as much KREEP redistribution as SCHONFELD and MEYER (1973) thought it should have. Therefore, it appears that the "South Imbrian" basin (rather than the Imbrian basin) formed immediately prior to KREEP extrusion in this region. This explanation allows us to continue to accept the well-documented

4.0 b.y. event as the time of the Imbrian excavation. The basin-excavation chronology, developed at length by NUNES et al. (1974), is summarized in Figure 5, which is a modified version of WILSHIRE and JACKSON's (1972) figure. The original geologic map showing the main outer mountain rings of major lunar basins and the approximate extent of their continuous ejecta blankets was published by WILHELMS and McCAULEY (1971). The basin excavation ages presented in Figure 5 were suggested by SCHAEFFER and HUSAIN (1974) for Nectaris and younger basins and NUNES et al. (1974) for Nectaris and older basins. The relative age assignments primarily follow those of STUART-ALEXANDER and HOWARD (1970).

Numerous Rb-Sr internal isochron ages and $^{40}\text{Ar}/^{39}\text{Ar}$ ages indicate mare basalts were extruded from ~ 3.8 to ~ 3.2 b.y. ago. Thus, the older whole rock Rb-Sr and U-Th-Pb ages discussed above must represent still earlier periods of equilibration--possibly related to earlier episodes of melting and crystallization resulting from impacting planetesimals (NUNES et al., 1974).

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Table 1. U-Th-Pb concentrations and Pb isotopic compositions of anorthosite 60015 samples.

Description	Comp.	Weight (mg)	Concentrations (ppm)			Observed ratios*			Ratios Corrected Analytical Blank ‡						
			U	Th	Pb	$\frac{^{206}\text{Pb}}{^{204}\text{Pb}}$	$\frac{^{207}\text{Pb}}{^{204}\text{Pb}}$	$\frac{^{208}\text{Pb}}{^{204}\text{Pb}}$	$\frac{^{232}\text{Th}}{^{238}\text{U}}$	$\frac{^{238}\text{U}}{^{204}\text{Pb}}$	$\frac{^{206}\text{Pb}}{^{204}\text{Pb}}$	$\frac{^{207}\text{Pb}}{^{204}\text{Pb}}$	$\frac{^{208}\text{Pb}}{^{204}\text{Pb}}$	$\frac{^{207}\text{Pb}}{^{206}\text{Pb}}$	$\frac{^{208}\text{Pb}}{^{206}\text{Pb}}$
						Conc.	Conc.	Conc.	Conc.	Conc.	Conc.	Conc.	Conc.	Conc.	Conc.
Black glass rind	P	165.5				219.9	205.5	220.9			251.8	236.0	245.2	0.9373	0.9738
	C1	108.0	0.409	1.539	0.566	231.9	217.0	—	3.89	628.4	345.6	324.1	—	0.9376	—
	C2	92.4	0.398	1.466	0.621	189.2	179.3	—	3.81	364.1	217.9	206.8	—	0.9491	—
Plagioclase clast in the glass rind	P	117.7				29.26	27.17	47.27			33.15	31.34	50.82	0.9454	1.533
	C	87.7	0.0077	0.0248	0.1486	38.43	38.10	—	3.33	6.29	41.14	40.74	—	0.9902	—
"Glassy" white - plagioclase boundary	P	60.0				63.17	67.28	75.61			94.89	104.0	102.7	1.096	1.082
	C	80.0	0.0421	0.1567	0.3881	71.27	77.47	—	3.85	24.36	81.65	89.48	—	1.096	—
Plagioclase 2 mm from the glass - plagioclase boundary	P	126.4				50.66	62.23	63.19			71.20	91.60	79.66	1.287	1.119
	C	115.2	0.0023	0.0060	0.171	73.82	95.66	—	2.70	4.32	107.9	144.5	—	1.339	—
Plagioclase interior †	P	375				70.27	92.52	79.03			75.45	100.27	83.35	1.329	1.105
	C	355	0.0009	0.0026	0.198	69.12	91.36	—	3.45	1.00	73.90	98.98	—	1.339	—

* ^{208}Pb spike contribution has been subtracted from concentration data.

† From NUNES *et al.* (1973).

‡ Analytical total Pb blanks ranged from 1.1 ng to 3.8 ng.

Table 2. K, Rb, and Sr data from football-sized anorthosite sample 60015 and anorthosite 60025. All data have been corrected for interlaboratory bias relative to NBS-SRM 987 standard $^{87}\text{Sr}/^{86}\text{Sr} = 0.71014$.

Sample	Weight (mg)	Concentration (ppm)			$^{87}\text{Rb}/^{86}\text{Sr}$	$^{87}\text{Sr}/^{86}\text{Sr}$				
		K	Rb	Sr		Observed*	Corrected for in-situ Rb decay			
						t = 1.0 b.y.	t = 4.0 b.y.	t = 4.65 b.y.		
60015										
Black glass rind	15.8	694	1.910	156.8	0.03520	0.70116 <u>+0.00006</u>	0.70067	0.69915	0.69882	
Plagioclase, 2 mm from rind	30.3	68	0.1353	163.0	0.00240	0.69896 <u>+0.00007</u>		0.69882	0.69880	
Plagioclase interior	2.6	54	0.1221	166.2	0.00212	0.69900 <u>+0.00006</u>		0.69888	0.69886	
Plagioclase clast in glass	28.3	79	0.0810	182.9	0.00128	0.69883 <u>+0.00003</u>	0.69881	0.69876	0.69874	
60025										
Anorthosite whole rock	20.0	60	0.02017	213.6	0.000276	0.69904 <u>+0.00006</u>		0.69902	0.69902	

* All errors are $2\sigma_m$. $\lambda_{^{87}\text{Rb}} = 1.39 \times 10^{-11} \text{ yr}^{-1}$. The estimated errors for concentrations are $\pm 2\%$ for K and $\pm 1\%$ for Rb and Sr.

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Table 3. Observed $^{238}\text{U}/^{204}\text{Pb}$ ratios and first-stage estimated $^{238}\text{U}/^{204}\text{Pb}$ ratios assuming a two-stage U-Pb evolution model with an age of 4.65 b.y. for the origin of the moon.

Rock type	$^{238}\text{U}/^{204}\text{Pb}$ (observed)	$^{238}\text{U}/^{204}\text{Pb}$ (estimated for source)
Anorthosites	< 10	indeterminate
KREEP	1000 - 3000	600 - 1000
High-K basalts	600 - 1000	~ 300
Low-K basalts	300 - 600	~ 100
Excess Pb in "orange soil"	--	35

FIGURE CAPTIONS

Fig. 1. $^{206}\text{Pb}/^{207}\text{Pb}$ vs. $^{238}\text{U}/^{207}\text{Pb}$ plot for anorthosite 60015. Data are corrected only for blank. A, black glass rind; B, plagioclase clasts in the glass rind; C, "glassy" white-plagioclase boundary; D, plagioclase 2mm inside the boundary; E, plagioclase interior; M.T., modern terrestrial lead.

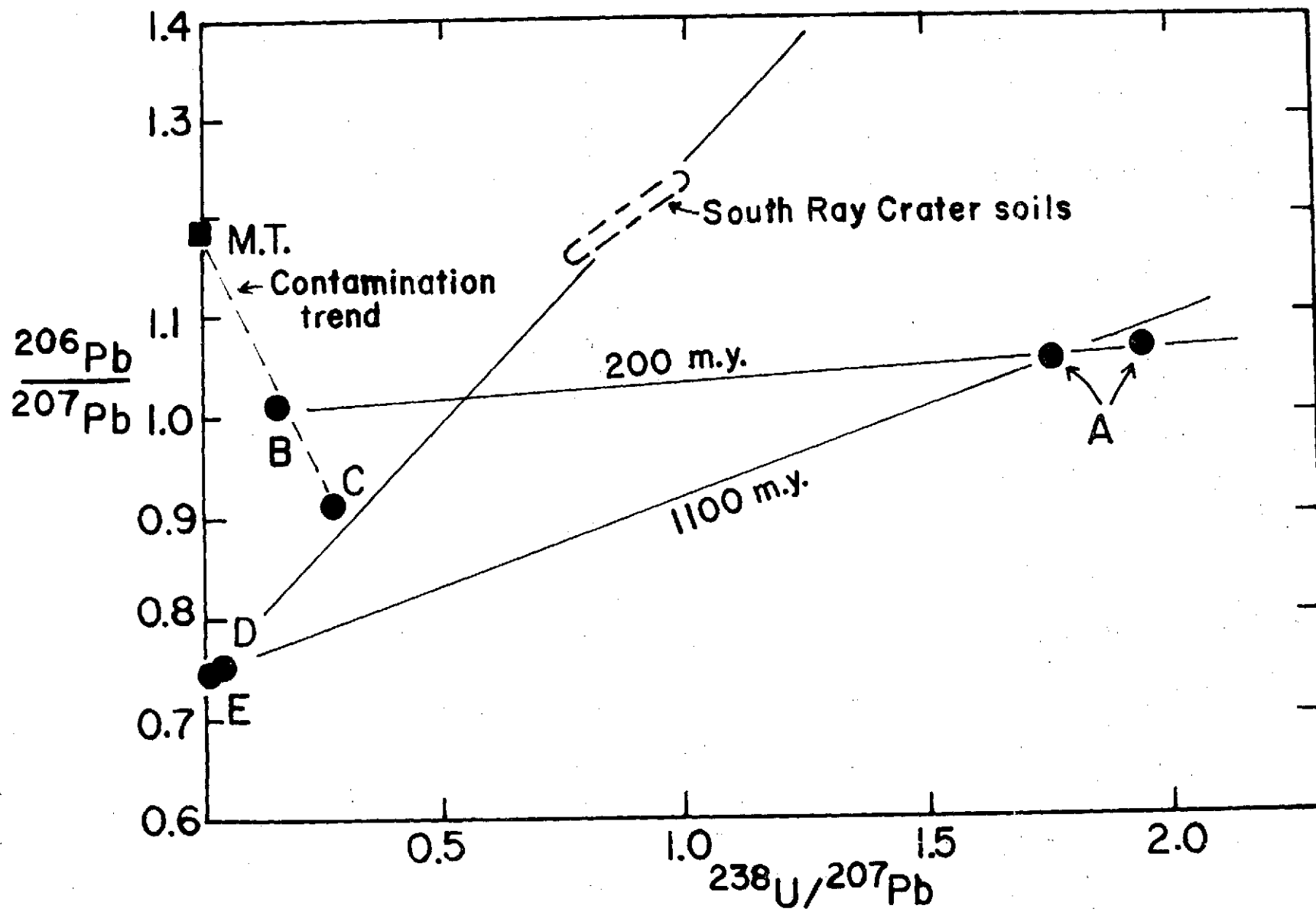
Fig. 2. Rb-Sr evolution diagram. For the initial $^{87}\text{Sr}/^{86}\text{Sr}$ ratio, ages 1.0 b.y. and 4.0 b.y. are used for plagioclase and clasts in glass rind of 60015 (open circles). Anorthosites 15415 and 60025, dunite 72417, BABI, LUNI, ADOR, and ALL are shown by solid dots for comparison. For anorthosite 60025, P & W denotes the data of Papanastassiou and Wasserburg (1972) and T, N & U for data by Tatsumoto et al. (this paper).

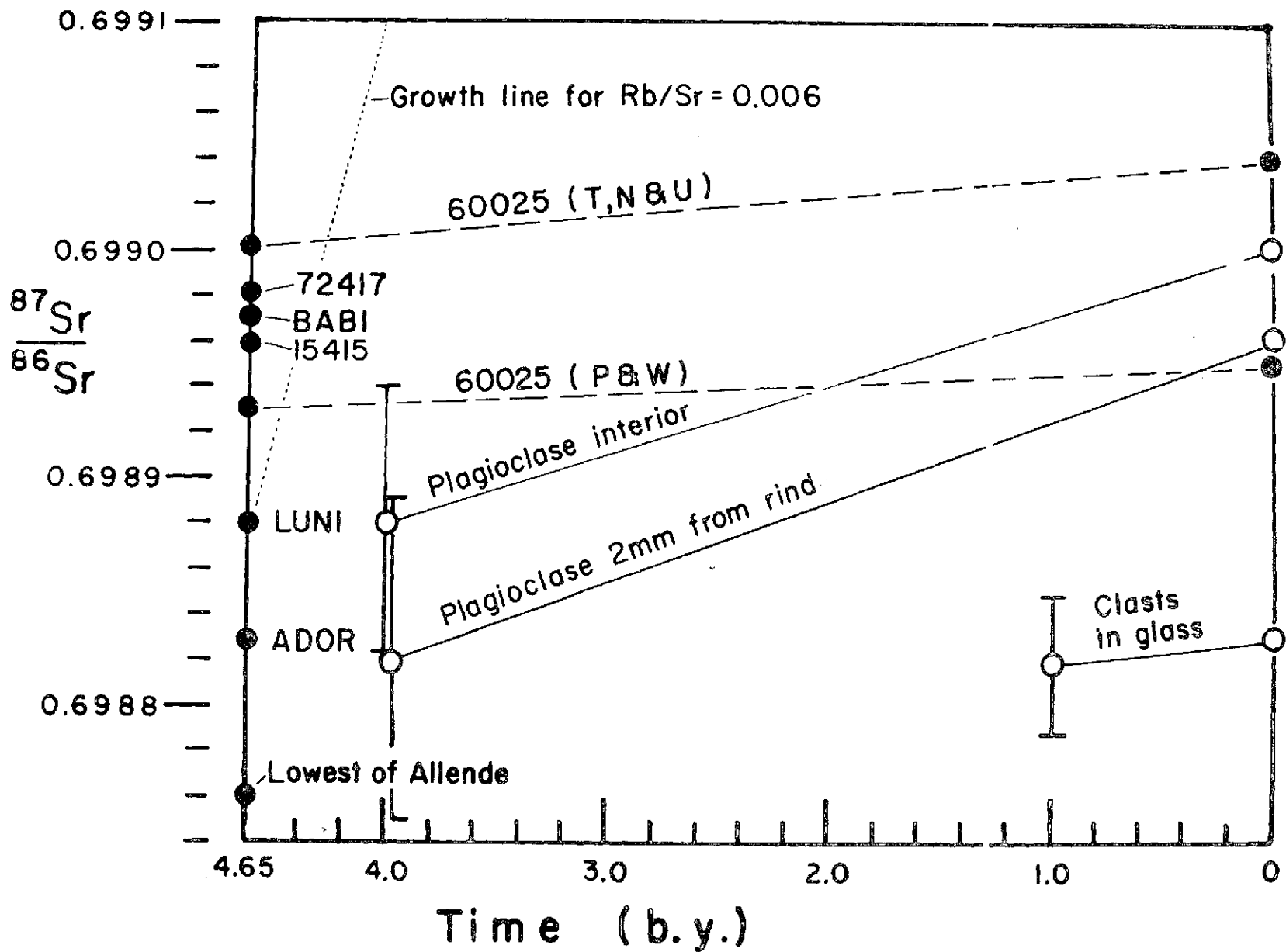
Fig. 3. U-Pb concordia diagram. Data points are for whole rock analyses of Apollo 11 and 12 rocks and soils, density separates of basalt 12064 (numbers 1-5 are from lower density) and portions of rock 12013.

Fig. 4. U-Pb concordia diagram. Data points are for Apollo 14, 15, 16, and 17 samples, and Luna 16 and 20 samples. B, S, and TW denote data reported by Barnes, et al. (1973), Silver (1973), and Tera and Wasserburg (1972a, b) respectively. The 4.0-4.47 b.y. concordia chord (dashed line) and the 3.8-4.65 b.y. concordia chord (solid line) are shown. Acid leach (denoted by L, Silver, 1970) and acetone^e float (AF) are also plotted.

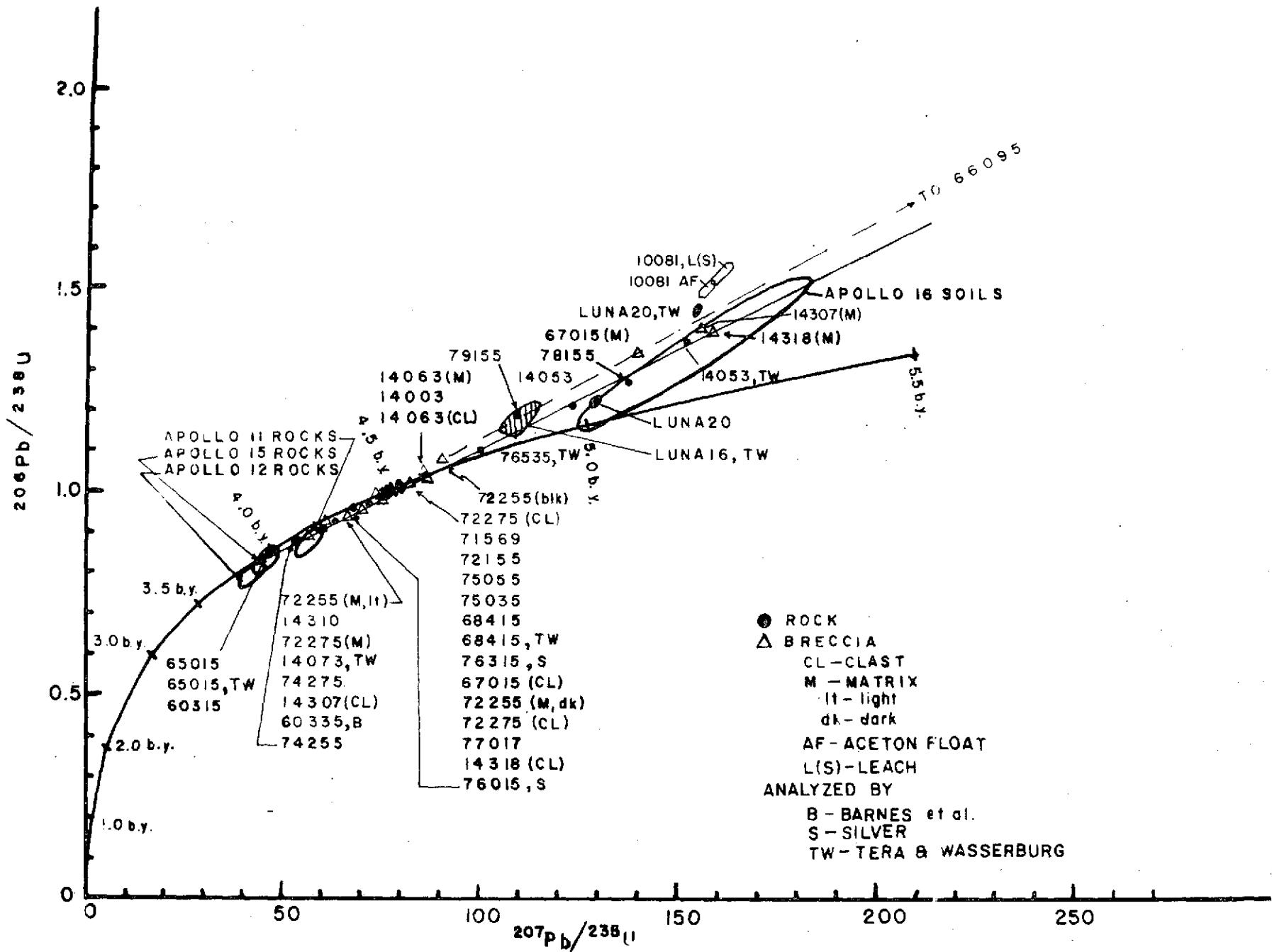
Fig. 5. Map showing main outer mountain rings of major lunar basins (drawn by Wilhelms and McCauley, 1971), ejecta blankets, and relative ages. Numbered in order of decreasing relative age. 1. Nubium, 2. Tranquillitatis, 3. Fecunditatis, 4. Procellarum, 5. "South Imbrium", 6. Serenitatis, 7. Nectaris, 8. Humorum, 9. Crisium, 10. Imbrium, and 11. Orientale.

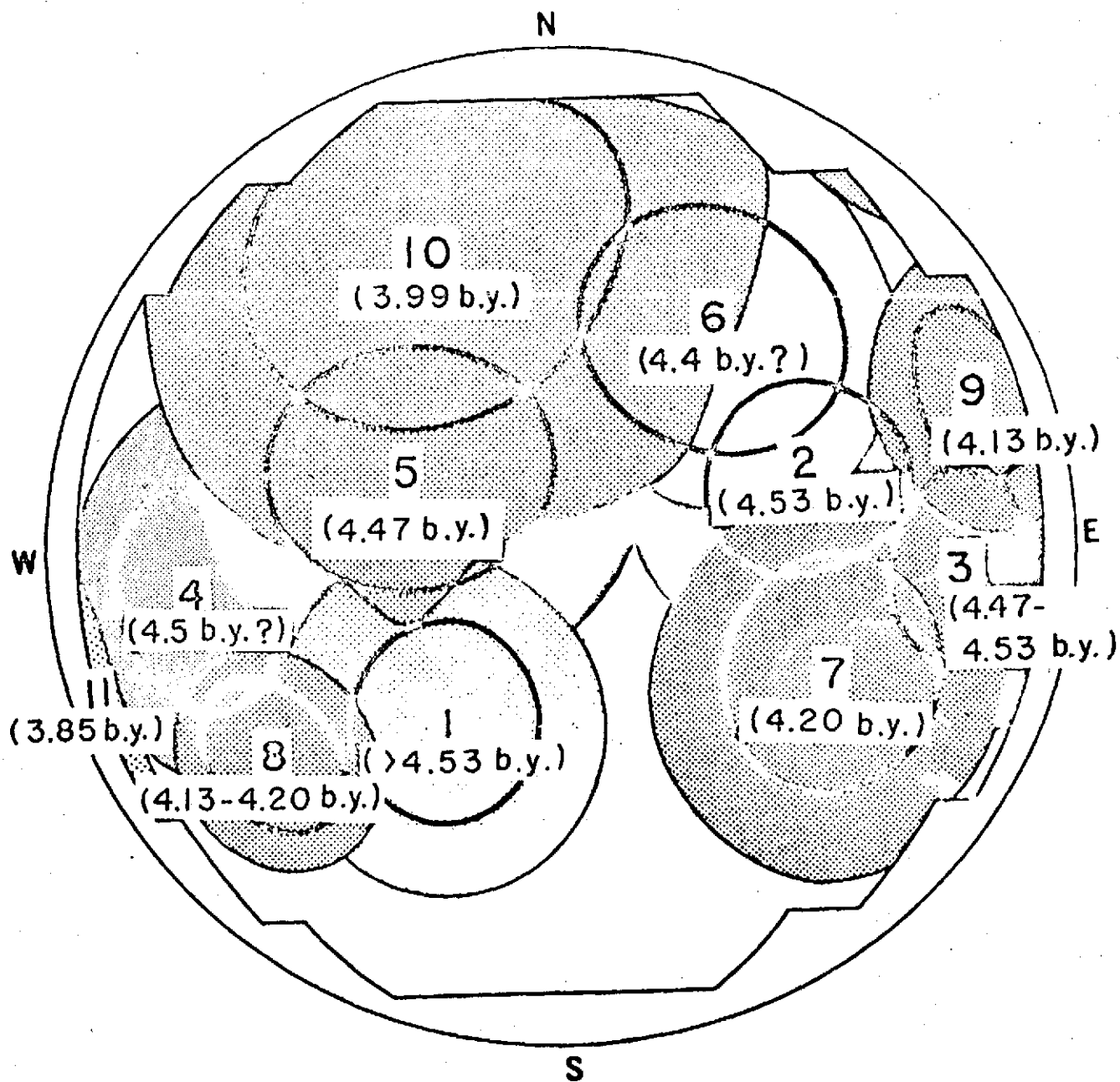
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Fig. 5