rocks: $^{206}Pb/^{204}Pb$ = 23.6, $^{207}Pb/^{204}Pb$ = 31.5 and $^{208}Pb/^{204}Pb$ = 40.9. These values were calculated from the $^{206}Pb/^{204}Pb$ intercept and on the assumption that lead had evolved from 4.7 to 3.7 billion years prior to crystalline rock formation. The intercepts obtained from 10084 fractions are the same within experimental error as the value obtained from meteorite lead.

The apparent ages are shown in Table 2. The initial leads used for correction are primeval lead of meteorite (*7*) for samples 10061 and 10084 and common lead (given above) for crystalline rocks. The ages indicate that sample 10071 may be younger and 10057 older than the other four crystalline rocks. Thus it might be conceivable that the common lead used for age calculation remains small because the isochrons in Fig. 1 are strongly controlled by the position of sample 10071.

The results corrected for initial lead are plotted in the Pb-U evolution diagram (Fig. 2). The points of the dust and breccia are above the concordia line, but those of crystalline rocks are below the concordia line. The apparent ages of 3.8 to 4.1 billion years for crystallized rocks indicate that they solidified several hundred million years after the formation of the moon. The dust apparently contains only minor amounts of fine fragments of the younger rocks that were analyzed. The most feasible interpretation is that dust and breccia probably were formed at an early stage in the history of the moon.

In Fig. 3, $^{206}Pb/^{204}Pb$ and $^{207}Pb/^{204}Pb$ for lunar dust and breccia are plotted with Patterson's data (*1*) on stone meteorite and Oversby's measurement (*7*) of primordial lead on the Canyon Diablo troilite. These data indicate that the system, of which earth, moon, and meteorites are parts, probably was formed about 4.66 $^{+0.07}_{-0.16}$ billion years ago. We set the upper limit from our experimental error, which amounts to about 1.5 percent, and the lower limit to include Patterson's meteorite data. It might be argued that the obtained age for the moon is from breccia and dust—not the lunar crystalline rock—and there may be doubt that those two samples represent the oldest rock of the moon. However, inasmuch as (i) the observed data fit into the extension of the meteorite lead array, (ii) the data lie near the U-Pb evolution curve, and (iii) the obtained age is very similar to the "older" meteorite ages measured by the Rb-Sr method (*10*), the age may be correct.

Thorium-232, with a 4.0-Mev α particle emission, has been used as a natural yield tracer for the determination of the state of radioactive equilibrium between ^{230}Th and parent ^{238}U in crustal silicate rocks from the earth (*11*). This technique of using ^{232}Th, of known concentration in a sample, as the yield tracer for ^{230}Th was used on several samples from Apollo 11. Although, within experimental error, ^{234}U is in radioactive equilibrium with ^{238}U, preliminary data indicate a value for the $^{230}Th/^{238}U$ activity ratio that is greater than unity in three of the samples analyzed (10017, 10020, 10057). A $^{228}Th/^{232}Th$ activity ratio of unity would indicate that ^{228}Th is in radioactive equilibrium with parent ^{232}Th. In two of the rocks (10020, 10057) the ^{228}Th α activity was measured to be about 10 percent greater than the 4.0-Mev α activity of ^{232}Th. The reasons for these variations are not understood at this time.

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Ages, Irradiation History, and Chemical Composition of Lunar Rocks from the Sea of Tranquillity

Abstract. *The ^{87}Rb-^{87}Sr internal isochrons for five rocks yield an age of 3.65 ± 0.05 × 10^9 years which presumably dates the formation of the Sea of Tranquillity. Potassium-argon ages are consistent with this result. The soil has a model age of 4.5 × 10^9 years, which is best regarded as the time of initial differentiation of the lunar crust. A peculiar rock fragment from the soil gave a model age of 4.44 × 10^9 years. Relative abundances of alkalis do not suggest differential volatilization. The irradiation history of lunar rocks is inferred from isotopic measurements of gadolinium, vanadium, and cosmogenic rare gases. Spallation xenon spectra exhibit a high and variable $^{131}Xe/^{126}Xe$ ratio. No evidence for ^{129}I was found. The isotopic composition of solar-wind xenon is distinct from that of the atmosphere and of the average for carbonaceous chondrites, but the krypton composition appears similar to average carbonaceous chondrite krypton.*

Samples investigated were 10017, 10044, 10050, 10057, 10059, 10069, 10071, soil (10084), coarse fines (10085), and core 2 (referred to by Nos. 17, 44, etc.). Contamination levels were negligible. Standards were regularly run to control precision *to the levels reported.*

Rubidium-strontium analyses on total rocks and minerals were obtained. The Sr is concentrated in plagioclase and the K in a fine-grained or glassy interstitial phase, containing up to 12 percent K. Maximum Rb/Sr was obtained in cristobalite and ilmenite separates due to the interstitial high-K phase. Precise internal isochrons on five rocks give the same age of 3.65 × 10^9 years (Fig. 1). All $(^{87}Sr/^{86}Sr)_I$ are distinctly above BABI (*1*). The $(^{87}Sr/^{86}Sr)_I$ for No. 44 is distinct from that of other samples, indicating that the samples represent at least two different rock bodies (Fig. 2). The possibility that the isochrons are two-phase mixing lines cannot be excluded, considering the distribution of alkalis, but the coincidence of ages for rocks of different $(^{87}Sr/Sr^{86})_I$ and texture argues that 3.65 × 10^9 years is the time of crystallization.

The possibility that the age of 3.65 × 10^9 years represents closed-system meta-

morphism of rocks initially formed 4.6 $\times$ 10^9 years ago can be ruled out, as this would require values of $^{87}Sr/^{86}Sr$ at 4.6 $\times$ 10^9 years which are considerably less than BABI for Nos. 17, 57, 69, and 71. Rock textures and chemical heterogeneity in minerals do not support post-crystallization metamorphism.

From

$$\Delta^{44} = (^{87}Sr/^{86}Sr)_I^{44} - (^{87}Sr/^{86}Sr)_{BABI} = 0.00011$$

the time required to evolve this difference in an environment of chondritic Rb/Sr is 11 million years (4 million years for solar). Material which makes up these rocks was therefore physically separated from the protosun at the same time as other planetary objects. This separation was accompanied by a large reduction in Rb/Sr. Thus the moon as a whole cannot have chondritic Rb/Sr unless a rapid development of a lunar crust occurred with enrichment of Sr relative to Rb in contrast to the earth. The rocks studied cannot be the source of basaltic achondrites, as all lunar $(^{87}Sr/^{86}Sr)_I$ exceed the $(^{87}Sr/^{86}Sr)$ of several achondrites. The high $^{87}Sr/^{86}Sr$ in tektites cannot be produced from any of the materials studied except by an elaborate process.

On the basis of soil analyses, lunar Rb/Sr is similar to that of the earth's upper mantle. For modern terrestrial $^{87}Sr/^{86}Sr \geq 0.702$, the value of $(^{87}Sr/^{86}Sr)_I$[44] means that the moon cannot have formed from the earth unless this took place prior to 4.3 $\times$ 10^9 years ago.

Rubidium-strontium analyses of soil give a model age of 4.5 $\times$ 10^9 years, assuming $(^{87}Sr/^{86}Sr)_I$ = BABI. Magnetic separates form a linear array which yields an "age" of 4.07 $\times$ 10^9 years. This is a mixing line whose lower intercept is fixed by Sr-rich feldspar; the resulting "isochron" does not represent a meaningful age. The soil is an aggregate of particles with different ages and complex histories, and, as shown by the glass fragment (5 mg) which falls far off the isochron, individual particles are not necessarily compatible with the model age. The 4.5 $\times$ 10^9 year model age of the soil (which includes fragments of 3.6 $\times$ 10^9 year rocks) is striking and should not be considered accidental. A reasonable interpretation of the data is that the soil represents a nearly valid sampling of lunar crust which was formed about 4.6 $\times$ 10^9 years ago. This is not surprising, since the soil is produced by an impact process which is capable of sampling and averaging large segments of the lunar crust. The younger rocks on the lunar surface were produced by subsequent melting in this ancient crust. The whole crust is considered to behave as a closed system without the injection of large volumes of highly differentiated material from depth within the moon after 4.6 $\times$ 10^9 years ago. A fortuitous coincidence with the age of meteorites and the earth is demanded if the 4.5 $\times$ 10^9 year model age of the soil was produced by Rb loss through volatilization. The general pattern of K, Rb, and Cs abundances does not suggest differential losses of these elements by volatilization.

Luny rock 1 (No. 85) is a fine-grained rock fragment (shocked) consisting of low-Ca pyroxene [$(Ca_{0.04}Mg_{0.65}Fe_{0.31})SiO_3$], isotropic "plagioclase" ($An_{90}Ab_{10}$) and "K-feldspar," ilmenite, troilite, chlorofluorapatite, and whitlockite. It contains 0.56 percent K and 16.7 ppm Rb, has $(^{87}Sr/^{86}Sr)$ of 0.71399 $\pm$ 0.00011, and yields a model age (T_{BABI}) of 4.44 $\times$ 10^9 years. An assumed age of 3.65 $\times$ 10^9 years would require $(^{87}Sr/^{86}Sr)_I$ = 0.70174. This peculiar rock may represent a nonmare sample.

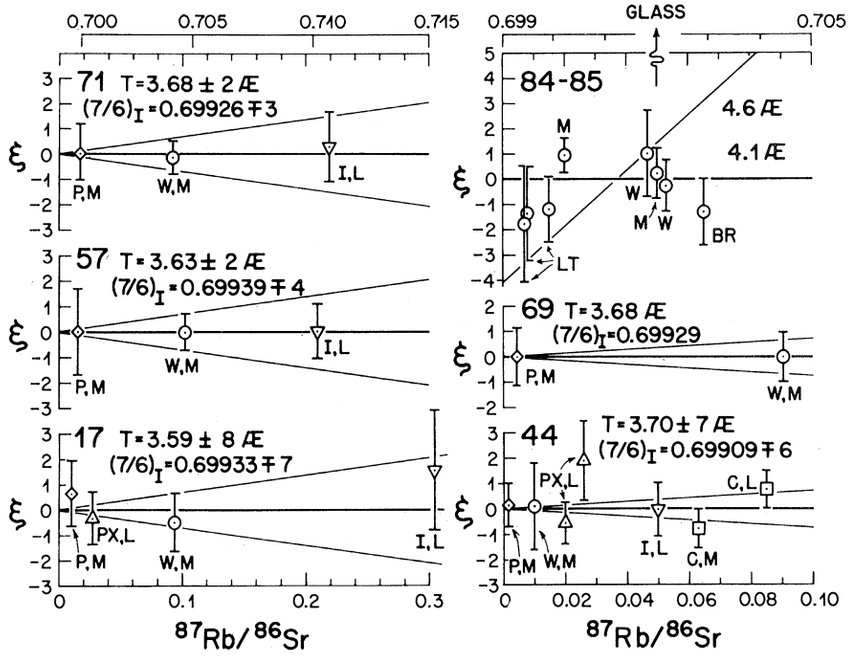

Fig. 1. Displacement of measured samples (m) from best fit (BF) (York) isochron

$$\left(\xi = \frac{(^{87}Sr/^{86}Sr)_m - (^{87}Sr/^{86}Sr)_{BF}}{(^{87}Sr/^{86}Sr)_{BF}} \times 10^4 \right)$$

vs. $(^{87}Rb/^{86}Sr)_m$. Errors on data points are 2σ (mean). Ages T ($\lambda = 1.39 \times 10^{-11}$ yr^{-1}) and initial $(^{87}Sr/^{86}Sr)_I$ are given (errors correspond to last digit). Lines on either side of $\xi = 0$ correspond to $\pm$ 50 million years. Symbols are whole rock (W), plagioclase (P), pyroxene (PX), cristobalite (C), and ilmenite (I). For dust (Nos. 84, 85) $\xi = 0$ corresponds to $T = 4.1 \pm 0.1$ AE and $^{87}Sr/^{86}Sr)_I = 0.69927 \mp 11$ calculated from lithic fragments (LT), dust (W,M), and No. 59 breccia (BR). A glass fragment lies off scale. Mechanical separate (M), heavy liquids (L).

The ^{40}K-^{40}Ar ages are for No. 17: whole rock, 2.45 $\times$ 10^9 years; the ^4He age, 2.5 $\times$ 10^9 years [U-Th from (2)]; plagioclase, 3.2 $\times$ 10^9 years. For No. 44: whole rock, 3.45 $\times$ 10^9 years; pyroxene, 3.6 $\times$ 10^9 years. For No. 69: whole rock, 2.9 $\times$ 10^9 years. For soil: feldspar glass, 4.9 $\pm$ 0.4 $\times$ 10^9 years; brown glass, 1.6 $\times$ 10^9 years.

Comparison of mineral and rock data demonstrates gas loss. The plagioclase for No. 17 yields a much higher age than the total rock, indicating Ar loss from the fine-grained, K-rich, interstitial phases. The concordance of He and Ar ages must be fortuitous. The maximum age is equal to the Rb-Sr age, and the general pattern is compatible with the Sr results. Assuming no inheritance of Ar, the age of the brown glass fragment shows that the soil contains particles produced by events of intermediate age ($\sim$ 10^9 years).

A widespread 3.6 $\times$ 10^9-year event due either to impact or to internal melting would cause major crustal differentiation and melting of a significant part

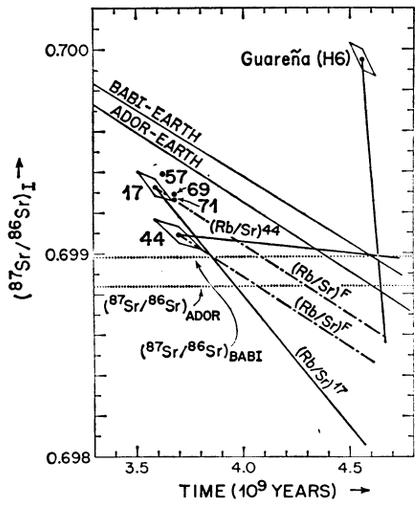

Fig. 2. Age-$(^{87}Sr/^{86}Sr)_I$ anticorrelation plot. Distinct error envelopes of Nos. 17 and 44 correspond to 2σ errors. Similar error envelopes for Nos. 57, 69, and 71 are omitted for clarity. The Guareña chondrite is shown for comparison. BABI-EARTH and ADOR-EARTH lines represent evolution of terrestrial Sr for corresponding $(^{87}Sr/^{86}Sr)_I$. Extrapolation of Nos. 17 and 44 to times before 3.6 AE are shown assuming Rb/Sr is either (i) as measured in the whole rock or (ii) as for the fines (F).

of the moon. The high U and Th content (*2*) of surface rocks must have been derived from material of much lower U and Th if the moon is not extensively molten today (U < 10 ppb). This requires that mare materials be differentiated from a layer at least 100 km thick. If the 3.6×10^9-year event was due to internal melting, the moon should be differentiated to a depth greater than 100 km, it should be partially molten today, and volcanic events should occur at the present time. If the event was the result of widespread impacts, the earth was also subjected to severe bombardment at that time. This may explain the absence of a geologic record before 3.6×10^9 years ago.

Microprobe analyses of phosphates showed fluorapatite in No. 44 and in lithic fragment 85-1-11 (F ~ 3.4 percent; Y and rare earths (REE) ~ 2 percent). Sufficient F and Cl are present so that no OH is required. Whitlockite in No. 69 contains no REE. Luny rock 1 (No. 85) contains chlorofluorapatite (F ~ 2.4 percent; Cl ~ 1.2 percent) with no REE and co-existing whitlockite with ~ 9 percent REE.

Glasses (balls and crusts) commonly contain Fe-Ni spherules with 10 percent Ni, troilite, and schreibersite. By contrast Ni-free Fe blebs within troilite in crystalline rocks could not have been in equilibrium with a silicate melt containing significant Ni. The Ni-free metal must have crystallized from a melt already depleted in Ni or have been produced by secondary reduction of Fe in Ni-free phases already separated from the melt. The Fe-Ni spherules in glasses, as well as Fe-Ni fragments in soil, are evidence of meteorite impact, since the crystalline rocks contain almost no Ni.

Chemical abundance measurements (Fig. 3) show the alkaline earths, Gd, and Li strongly enhanced relative to chondrites. Soil, breccia, and fine-grained rocks are not highly depleted in K-Rb-Cs, but the coarse-grained rock has K-Rb-Cs abundances similar to those found in Ca-rich achondrites. Concentrations of alkalis (except Na) and Ba are highly correlated. Strontium has a definite anticorrelation with the alkalis, whereas Ca is almost constant. Gross abundance features such as the alkaline earth, Gd, and Li enhancement probably reflect the composition of the source material, whereas the "internal correlations" (for example, Ba vs. alkalis) are a result of magmatic processes. It is not possible to form the soil precisely from a mixture of the rocks analyzed. In general, a component with high Rb, Cs, and Ba relative to K is required. Except for Na and Ca, the chemical composition of the breccia (No. 59) can be described as a mixture of local soil and fine-grained rocks. No evidence for fractionation of volatile alkalis can be seen in comparing soil and rocks.

Integrated thermal neutron fluxes were determined by measuring the depletion of ^{157}Gd and ^{155}Gd and the corresponding enrichment of ^{158}Gd and ^{156}Gd (Fig. 4). Taking a cosmic ray exposure age of 5.5×10^8 years for No. 17 and 7×10^8 years for soil, we obtain fluxes of 1.1 and 0.4 n/cm² sec. The neutron exposure of the top and bottom of core 2 is the same. If the flux at the core bottom is twice that at the top, the soil has been mixed to 13 cm at least once in the last 10^8 years. Assuming the soil to be well mixed over 4.5×10^9 years, the total depth of this zone must be less than about 25 times the thickness of the neutron production zone (~ 2 to 4 m) using an average flux of 1.5 n/cm² sec.

Vanadium isotopic composition measurements on whole rock samples of Nos. 17 and 84 showed that $(^{50}V/^{51}V)$ has the terrestrial value to within 3 percent and 2 percent. Vanadium appears to be concentrated in ilmenite

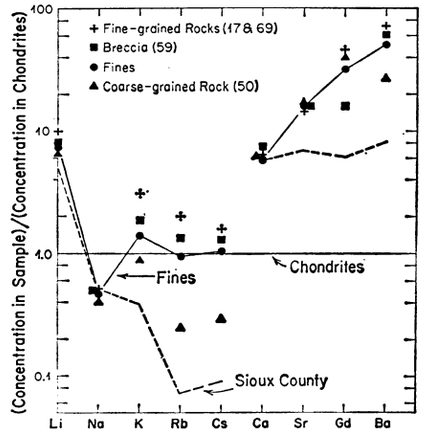

Fig. 3. Chemical abundances in lunar samples relative to chondrites. Assumed chondritic abundances in ppm: Li = 1.72, Na = 6600, K = 786, Rb = 2.81, Cs = 0.099, Ca = 12,200, Sr = 9.96, Ba = 3.37, Gd = 0.40. Typical Ca-rich achondrite (Sioux County) shown for reference.

(~ 100 ppm). A limit of $\sim 4 \times 10^{17}$ particles/cm² ($E \gtrsim 50$ Mev) for a model of uniform differential exposure of terrestrial, meteoritic, and lunar material can be set.

Cosmic ray exposure ages from ^{21}Ne, ^{36}Ar, and ^{126}Xe for each sample are essentially self-consistent, giving 550, 100, and 40 million years for Nos. 17, 44, and 69, respectively. Production rates are estimated from meteorite data, making allowance for chemical composition and geometry. Solar flare contributions are ignored. These ages may be in error because of erosion, variable shielding, and greater shielding than in meteorites.

Xe and Kr concentrations [ ] are

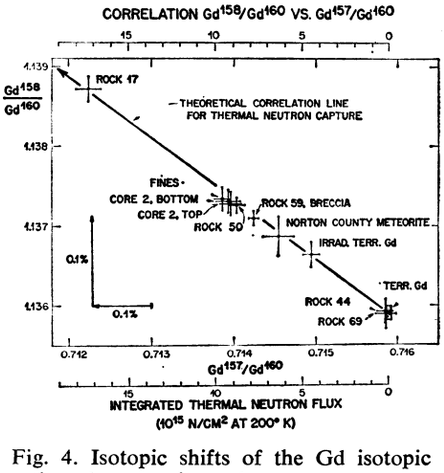

Fig. 4. Isotopic shifts of the Gd isotopic ratios due to thermal neutron capture. Mass fractionation is corrected by normalizing ratios to $^{156}Gd/^{160}Gd = 0.9361$. Errors correspond to 2σ.

given in units of 10^{-12} cm^3 (STP)/g. Cosmogenic (c) Xe and Kr spectra were derived for No. 17, giving

Xe 124/126/128/129/130/131/132/134/136 = 0.55/≡ 1/1.58/1.67/1.10/7.90/0.91/0.099 ≡ 0;
$^{126}Xe_c$ = [170];
$^{136}Xe_{tr}$ = [30] and
Kr 80/82/83/84/86 = 0.484/0.743/≡ 1/0.384/≡ 0;
$^{83}Kr_c$ = [1400]
$^{86}Kr_{tr}$ = [70].

We assumed ^{136}Xe and ^{86}Kr represented trapped (tr) gas of atmospheric composition after correcting for ^{238}U spontaneous fission ($^{136}Xe_f/^{136}Xe_{tr} \approx 0.1$). For No. 69 ($^{126}Xe_c$ = [14]; $^{136}Xe_{tr}$ = [13]), cosmogenic spectra derived with these assumptions agree well with those for No. 17. Using soil Xe for Xe_{tr} leaves the Xe_c spectra for No. 17 relatively unaffected but destroys good agreement between Nos. 69 and 17, suggesting that either large amounts of atmospheric Xe contamination are present or that Xe_{tr} in lunar rocks has atmospheric composition.

Xe_c and Kr_c spectra for No. 17 are compatible with meteoritic spallation spectra except at masses 129 and 131. The 129 and 131 excesses appear cosmogenic in that, for chemically similar Nos. 17 and 69, $(^{129}Xe/^{126}Xe)_c$ and $(^{131}Xe/^{126}Xe)_c$ are the same to 10 percent, although the exposure ages differ by a factor of 14. The ratio $(^{131}Xe/^{126}Xe)_c$ = 3.51 in No. 44 is reasonable for meteoritic spallation, but the $(^{129}Xe/^{126}Xe)_c$ = 1.47 is still anomalous and similar to that in No. 17. Measured neutron exposures and estimated target concentrations are too low to account for excesses in ^{131}Xe and ^{129}Xe by capture on Ba or Te. Solar proton reactions (15 to 30 Mev) on Ba would predominantly produce ^{129}Xe and ^{131}Xe (from ^{130}Ba and ^{132}Ba); however, excess ^{83,84}Kr from similar reactions on ^{84}Sr and ^{85}Rb are not observed. Excess ^{129}Xe may indicate that meteoritic $(^{129}Xe/^{126}Xe)_c$ is too low. Excess lunar ^{129}Xe is not attributable to ^{129}I.

A fraction (< 37 μm) of soil (No. 85) was used for rare gas analysis to enhance solar-wind Xe relative to spallation. Measured spectra are

Xe 124/126/128/129/130/131/132/134/136 = 0.590/0.658/8.68/105.0/16.68/83.08/ ≡ 100/37.13/30.24

with

^{132}Xe = [54,000]

and

Kr 80/82/83/84/86 = 12.87/65.00/65.51/323.6/≡100
^{86}Kr = [80,000].

Abundances of heavier Xe isotopes are distinct from both the average for carbonaceous chondrites (AVCC) and atmosphere. Enrichments in lighter Xe isotopes are presumably due to spallation. The $^{130-136}$Xe data can be accounted for by a mass fractionation of 1.5 percent per mass unit relative to AVCC. However, subtraction of this fractionated AVCC component from $^{124-8}$Xe yields residuals

Xe 124/126/128 = 0.37 ± 0.04/1.0/0.5 ± 0.2

which are not compatible with known Xe spallation spectra. The quoted errors arise from uncertainties in AVCC. A similar calculation relative to atmosphere gives a more unreasonable "spallation" spectrum. The Kr spectrum can be accounted for by a mixture of AVCC Kr plus spallation. It is not possible from our data to establish a genetic relationship between solar-wind Kr and Xe and AVCC (or atmosphere).

The average depth to which the soil is well mixed is about 6 m, assuming that the integrated ^{126}Xe production occurred over 3.6 × 10^9 years.

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1. BABI is the initial $(^{87}Sr/^{86}Sr)_I$ in basaltic achondrites; ADOR is the more primitive value found in Angra dos Reis. D. A. Papanastassiou and G. J. Wasserburg, *Earth Planet. Sci. Lett.* **5**, 361 (1969); D. A. Papanastassiou, thesis, California Inst. of Technology (1970).
2. Preliminary Examination of Lunar Samples from Apollo 11, *Science* **165**, 1211 (1969).
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Argon-40/Argon-39 Dating of Lunar Rock Samples

Abstract. *Seven crystalline rock samples returned by Apollo 11 have been analyzed in detail by means of the ^{40}Ar-^{39}Ar dating technique. The extent of radiogenic argon loss in these samples ranges from 7 percent to ⩾ 48 percent. Potassium-argon ages, corrected for the effects of this loss, cluster relatively closely around the value of 3.7 × 10^9 years. Most of the vulcanism associated with the formation of the Mare Tranquillitatis presumably occurred around 3.7 × 10^9 years ago. A major cause of the escape of gas from lunar rock is probably the impact event which ejected the rock from its place of origin to its place of discovery. Upper limits for the times at which these impact events occurred have been estimated.*

The determination of accurate and meaningful ages of rocks and soil from many parts of the lunar surface will be of prime importance in unraveling the sequence of events which have occurred there since the moon formed as an independent object in space. In this paper I report on the measurement and interpretation of potassium-argon ages of crystalline rocks returned by Apollo 11.

In its simplest form the determination of a potassium-argon age involves the measurement of the total amount of potassium and the total amount of radiogenic ^{40}Ar in the sample. The assumptions are made that the rock was free of argon when formed and that it has quantitatively retained ^{40}Ar, from the decay of ^{40}K, since that time. The assumption of quantitative argon retention is particularly *inappropriate* for the lunar rocks. The rocks returned to earth have been picked up loose from the surface of the moon, presumably at some distance from their place of origin. The presence of shock effects in some, if not all, of the crystalline rocks indicates that high-energy events, possibly meteorite impacts, may have transported the rocks from their place of origin to their place of discovery and it is very probable that argon loss occurred at the time of transfer. In an attempt to estimate the extent of gas loss and to apply a suitable correction to the potassium-argon age, an activation technique, the ^{40}Ar-^{39}Ar method, has been applied to seven of the crystalline lunar rocks.

The ^{40}Ar-^{39}Ar method has been described more fully elsewhere (*1, 2*). The technique consists of converting a measured fraction of ^{39}K in the rock to ^{39}Ar by neutron activation, and then heating the sample in stages to release this ^{39}Ar, together with radiogenic ^{40}Ar, by thermal diffusion. The argon is subsequently analyzed in a mass spectrometer. In a sample which has quantita-