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.

The Lunatic Asylum of the*
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* The inmates are A. L. Albee, D. S. Burnett, A. A. Chodos, O. J. Eugster, J. C. Huneke, D. A. Papanastassiou, F. A. Podosek, G. Price Russ II, H. G. Sanz, F. Tera, G. J. Wasserburg.

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3. We thank our fellow inmates Pai Young and Uwe Derksen for the sophisticated skill and knowledge they have brought to this study, without which this work would not have been accomplished. We also acknowledge the unstinting efforts of J. Brown, H. L. Derksen, L. Ray, and T. Wen. This work was supported by NASA contract 64049.

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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-

Table 1. Argon-40/argon-39 ages of lunar samples. Ages calculated on the basis of $\lambda\beta = 4.72 \times 10^{-10}$ year^{-1}; $\lambda\varepsilon = 0.584 \times 10^{-10}$ year^{-1}; $^{40}K/K = 0.0119$ atom percent; J values are 0.341 ± 0.007 for samples 10003,43 to 10022,46 and 0.319 ± 0.007 for the remainder. The two groups of samples were irradiated at the same time in separate Al canisters. An explanation of the significance of the "errors" in the ratios is given in the text. Quoted errors in ages include also the uncertainties in the J values which arise in part from nonuniformity of the measured neutron flux. Absolute uncertainties in the monitor age (~2 percent) have not been included as all samples are affected equally.

Sample No.	Fractional ^{40}Ar loss	Total argon		High-temperature argon		Low-temperature argon	
		^{40}Ar/^{39}Ar	"Age" × 10^9 years	^{40}Ar/^{39}Ar	"Age" × 10^9 years	^{40}Ar/^{39}Ar	"Age" × 10^9 years
10003,43	0.10	185.1 + 1.4	3.76 + 0.05	206.2 +6.8	3.93 ± 0.07	< 21	< 1.0
10017,49	⩾ 0.48	69.8 ± 0.5	2.30 ± 0.05	132.9 + 3.0	⩾ 3.23 ± 0.06	< 4.2	< 0.25
10022,46	0.19	135.5 ± 0.7	3.26 ± 0.05	166.3 ± 3.8	3.59 ± 0.06	< 14	< 0.75
10024,26	⩾ 0.45	91.5 ± 0.5	2.58 ± 0.05	166.2 ± 1.4	⩾ 3.48 ± 0.05	< 3.7	< 0.2
10044,40	0.07	182.0 ± 0.9	3.62 ± 0.05	195.6 ± 2.5	3.74 ± 0.05	< 16	< 0.8
10062,30	0.22	160.3 ± 1.6	3.42 ± 0.05	206.5 ± 3.7	3.83 ± 0.06	< 22	< 1.0
10072,45	0.14	148.2 ± 0.8	3.30 ± 0.05	172.5 + 2.6	3.53 ± 0.05	< 12	< 0.6

tively retained argon the two isotopes are expected to reside in fixed proportions in equivalent crystal sites since they have both been produced from potassium. Since the two isotopes have nearly identical diffusion coefficients, they will be released in proportionate amounts during a thermal diffusion experiment and the (constant) ^{40}Ar/^{39}Ar ratio will be related in a simple way to the potassium-argon age of the rock.

In contrast, a rock which has lost some of its argon at an intermediate stage of its history will contain easily outgassed crystal sites which are low in ^{40}Ar relative to ^{40}K. These sites may be, for example, minerals of high diffusion coefficient, small mineral grains, surface regions of large mineral grains, or regions surrounding crystal defects. When such a sample is analyzed by the ^{40}Ar-^{39}Ar method the gas released in the early stages of the diffusion experiment is expected to come from the more easily outgassed sites and therefore to be low in ^{40}Ar relative to ^{39}Ar. As the experiment proceeds, the more retentive sites will release their argon and the ^{40}Ar/^{39}Ar ratio will increase. The ^{40}Ar/^{39}Ar ratio of the gas released at high temperatures may be converted to an age which will be closer, though not necessarily equal, to the "true" age of the rock, whereas the initial ^{40}Ar/^{39}Ar ratio may, in suitable circumstances, indicate the most recent time at which argon loss occurred.

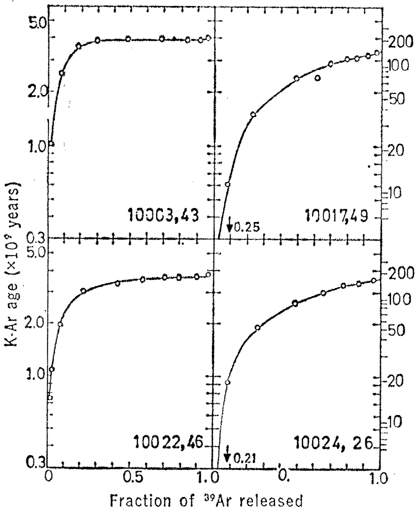

Fig. 1. Argon-40/argon-39 release patterns.

I have applied the ^{40}Ar-^{39}Ar technique to seven lunar rock samples; each sample weighed approximately 100 mg. The samples were broken into small chips 1 to 3 mm in diameter for analysis. They came into contact with the atmosphere during this operation, and also briefly when being loaded into the gas extraction system, but during the neutron irradiation they were vacuum-encapsulated in quartz ampules to minimize contamination with atmospheric argon. The samples were irradiated for 2 days in the core of the Herald reactor at the Atomic Weapons Research Establishment (AWRE), Aldermaston, and received an integrated fast neutron flux of 6.0×10^{18} neutron cm^{-2}. The irradiation was monitored by the inclusion of two terrestrial hornblende samples of known age. After the irradiation, samples were heated for 1-hour periods at successively higher temperatures from 400°C up to the melting point and the gas released at each stage was analyzed.

Corrections for atmospheric argon released from the gas extraction system were applied by measuring system blanks before and after each series of extractions. In general, these corrections were small (~ 1 to 2 percent) except for the first and the last extraction for each sample. A small correction was made for trapped ^{40}Ar on the assumption that the ^{40}Ar/^{36}Ar ratio of the trapped gas is unity (*3*). It was also necessary to apply a correction for ^{39}Ar produced by the ^{42}Ca(n,α)^{39}Ar reaction. This was very conveniently done for each extraction by measuring the amount of ^{37}Ar produced by the reaction ^{40}Ca(n,α)^{37}Ar. The release patterns of ^{37}Ar and ^{39}Ar are quite distinct, the ^{37}Ar not being released until the very highest temperatures. From a consideration of the ^{40}Ar/^{39}Ar and ^{37}Ar/^{39}Ar ratios in two ^{37}Ar-rich fractions of sample 10003,43, it is possible to show that the ^{37}Ar/^{39}Ar ratio of the calcium-derived argon is 1250. All ^{39}Ar abundances have therefore been corrected by subtracting 1/1250 of the ^{37}Ar abundance. In most fractions where significant radiogenic argon is released, this correction is also small.

One may convert the corrected ^{40}Ar/^{39}Ar ratio to a potassium-argon age t by using the expression

$$t = T \ln [1 + J\,(^{40}\mathrm{Ar}/^{39}\mathrm{Ar})]$$

where

$$J = (1 + \exp\,(t_s/T) / (^{40}\mathrm{Ar}/^{39}\mathrm{Ar})_s$$

T is the mean life of ^{40}K, and the subscript s refers to measurements on the monitor.

All the samples analyzed have been crystalline igneous rocks of either type A or type B (*3*). Breccias and soil

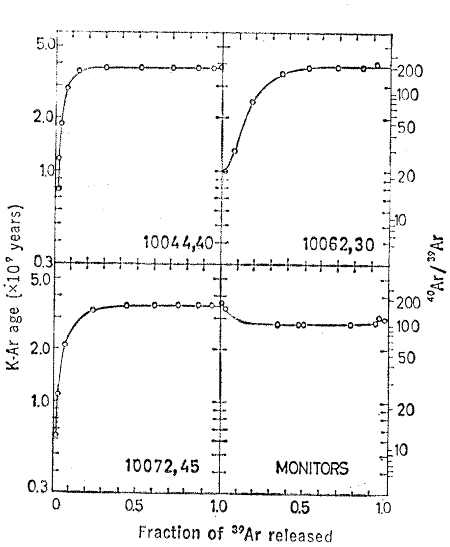

Fig. 2. Argon-40/argon-39 release patterns. The monitor release pattern given is a composite of three experiments and is characteristic of the monitor used (*2*).

samples have not so far been analyzed because of their complex history and their extremely high solar wind content. Figures 1 and 2 summarize the results obtained so far. Without exception, the release patterns are consistent with a recent episode of argon loss having occurred in relatively old rocks, after the fashion described above.

The evidence, in the low-temperature argon release, of regions within each of the samples that are low in radiogenic ^{40}Ar, relative to potassium, makes it possible in principle for one to estimate the time of the event which produced the loss of argon. In practice, with the present samples, it has been possible to place only upper limits on the time of outgassing because in most cases the gas loss was relatively small. These times are included in Table 1 and indicate that the events concerned, possibly the impact events which transported the rocks across the lunar surface, occurred relatively recently. It will be necessary to examine cosmic-ray exposure ages for details of this part of the rocks' history.

In all but two samples (10017,49 and 10024,26) the ^{40}Ar/^{39}Ar ratio attains a constant value in the last five or so high-temperature extractions. The average value of these high-temperature ratios is presented in Table 1 together with the root-mean-square deviation of the individual ratios. The ages calculated from these ratios are also included in the table and are a truer indication of the time of origin of the rocks than ages based on the total gas release. The ages based on total gas release are included for comparison and also included is an estimate of the extent of argon loss. This is simply the fractional difference between the high-temperature ^{40}Ar/^{39}Ar ratio and the total gas ^{40}Ar/^{39}Ar ratio.

Samples 10017,49 and 10024,26 do not show plateaus in the argon release pattern, and it is probable that the "high-temperature" age, calculated on the basis of the last significant argon extraction, represents only a lower limit on the true age of the rock. Likewise the fractional argon losses calculated (48 percent and 45 percent, respectively) can only be regarded as lower limits.

The effect of applying the correction for argon loss is impressive in that the spread of ages is considerably reduced. The ages based on total argon cover a range of 1.4×10^9 years, from 2.30×10^9 years to 3.76×10^9 years. If we take account of the fact that the "high-temperature" ages of samples 10017,49 and 10024,26 are now to be regarded only as lower limits, the significant spread in ages is reduced to 0.4×10^9 years (from 3.53×10^9 years to 3.93×10^9 years).

Once one has been able to largely eliminate the effects of argon loss, the problem of the interpretation of these ages is considerably eased, provided some estimate can be made of the area of the lunar surface represented by the samples. It is probable that most of the regolith is of comparatively local origin. This viewpoint is supported by the relative uniformity of composition of many of the Apollo 11 rocks (*3*), the existence of well-defined color boundaries associated with differences in underlying rock type (*4*), and the relationship of regolith thickness to local cratering density (*5*). Despite the fact that only a limited number of age determinations have been made, we can therefore now say with a fair degree of confidence that much of the vulcanism associated with this region of Mare Tranquillitatis occurred around 3.7×10^9 years ago.

It is impossible at the present time to reach any firm conclusions about the significance of the remaining spread in the measured ages. It is difficult to escape the conclusion that the spread is real and represents different rock-forming events which have occurred in the vicinity of the Apollo 11 site. However, before reading too much into these differences, it will be useful to seek corroboration by a comparison of the ^{40}Ar-^{39}Ar ages with either Rb-Sr or U-Pb ages.

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Uranium-Thorium-Lead Isotope Relations in Lunar Materials

Abstract. *The lead isotopic compositions and uranium, thorium, and lead concentrations have been measured on six samples of material from the Sea of Tranquillity. The leads are moderately to very radiogenic; the initial lead concentrations are very low; the uranium and thorium levels are 0.26 to 0.88 and 0.87 to 3.35 parts per million, respectively. The Th/U ratios cluster about a 3.6 value. Apparent ages calculated for four rocks are 4.1 to 4.2* $\times 10^9$ *years. Dust and breccia yield apparent ages of 4.60 to 4.63* $\times 10^9$ *years. The uranium-lead ages are concordant, or nearly so, in all cases. The lunar surface is an ancient region with an extended record of events in the early history of the solar system. The discrepancy between the rock ages and dust ages poses a fundamental question about rock genesis on the moon.*

Elemental and isotopic abundances of uranium, thorium, and lead have been studied in six samples of lunar materials collected from the Sea of Tranquillity. The initial analytical results contain some fundamental information bearing upon the early history of the moon in matters of genesis, chemical differentiation, and timing. They raise some new questions concerning lunar evolution.

The samples include lunar dust (fines < 1 mm, sample 10084,35) a fine-grained dark breccia, apparently a sample of indurated lunar regolith (10060,-15), and chips of four rocks fairly representative of the textural and compositional ranges encountered in the Apollo 11 collections (10017,34; 10045,30; 10047,34; and 10072,39). The general character and lunar setting of these materials have been described previously (*1*).

The rocks are basaltic or gabbroic in composition and texture. Rocks 10072 and 10045 are fine-grained, vesicular to vuggy, and olivine-bearing. Rock 10017 has not been definitely identified as containing olivine and is a microgabbro in texture, with striking spherical vesicles. Rock 10047 is a medium-grained, cristobalite-bearing gabbro. The microbreccia, 10060, is very dark and fine-grained with a few scattered glass spherules suggesting its affinity to the dust sample.

The analytical techniques consisted of chemical decomposition with HF and $HClO_4$, extraction of Pb, U, and Th from the residue with concentrated HNO_3 and various procedures of coprecipitation and organic solvent extraction