

Table 2. Exposure ages of two lunar samples. Sample 10087,59 is lunar fines; sample 10057,20 is fine-grained crystalline rock.

Exposure age type	10084,59 (10 ⁶ years)	10057,20 (10 ⁶ years)
³⁸ Ar	3020	125
⁸² Kr	1600	64
¹³⁰ Xe	720	63

in terms of absolute gas concentration, can perhaps be attributed to fission effects in the Stannern meteorite.

A comprehensive display of the xenon isotope variations in rock 57 is given in Fig. 4, where ratios $M\text{Xe}/^{130}\text{Xe}$ are plotted against $^{126}\text{Xe}/^{130}\text{Xe}$. For M equal to 124, 131, and 132, the points fall reasonably well on lines joining the 1200°C composition and the composition seen at 820°C in the lunar fines. Small corrections have been made for a fission contribution at masses 131 and 132 (see the following paragraph). Otherwise, these isotopes, with ^{126}Xe and ^{130}Xe , appear to belong to a two-component system of trapped and cosmogenic xenon. For M equal to 128 and 129, the values observed between 600° and 900°C fall above the lines containing the other points, as would occur if there were an excess of ^{128}Xe and ^{129}Xe being released at these temperatures. The correlation lines in Fig. 4 pass close to the compositions for cosmogenic xenon in calcium-rich achondrites, except at mass 131, where the correlation line observed has the opposite slope from that observed in meteorites.

Fission xenon in rock 57 was indicated, after subtraction of cosmogenic xenon, by higher values of $^{136}\text{Xe}/^{132}\text{Xe}$ and $^{134}\text{Xe}/^{132}\text{Xe}$ than in the lunar fines. The amount of fission xenon observed is approximately what should accumulate in 4.1×10^9 years (see below) of uranium decay in rock 57 (780 ppb uranium).

Argon from the samples is a mixture of radiogenic ^{40}Ar from ^{40}K decay, cosmogenic argon, and trapped argon, which can doubtless be ascribed to the solar wind. Values of the ratio $^{40}\text{Ar}/^{36}\text{Ar}$ as low as 0.82 were observed in argon fractions from the lunar fines, and provide an upper limit for this ratio in the solar wind. The much higher values of $^{40}\text{Ar}/^{36}\text{Ar}$ in gas fractions from rock 57 have to be attributed to ^{40}Ar from potassium decay. On that basis we compute a K-Ar age of $4.1 \pm 0.3 \times 10^9$ years for rock 57,

using the preliminary value (5) for the potassium content of the rock.

Krypton exhibits a cosmogenic component in all samples, and an excess of ^{80}Kr in rock 57. Radioactive ^{81}Kr was observed in the highly cosmogenic 1200°C fraction of rock 57, permitting calculation of a $^{81}\text{Kr}/^{80,82}\text{Kr}$ age (6) of 37 million years for exposure of the sample to galactic cosmic rays. Crude exposure ages (Table 2) were calculated for rock 57 and for the fines from the cosmogenic contents of ^{38}Ar , ^{82}Kr , and ^{130}Xe . We used the production rates from Munk's (7) study of the calcium-rich achondrite Nuevo Laredo for this purpose, assuming targets of calcium, strontium, and barium, respectively. These ages for rock 57 exceed the $^{81}\text{Kr}/^{80,82}\text{Kr}$ age as they should; the results would agree only if that rock was abruptly shifted to its final location from a completely shielded location. The ^{38}Ar exposure ages are older than the others. For the soil, the ages represent an average for the fine-grained material, but much of the present surface soil material at

Tranquillity Base has been within a meter or so of the surface for 10^9 years or so, according to our data.

Although we have not completely analyzed our results from rock 44, one important fact is immediately evident: in rock 44 the large excess of ^{131}Xe in the cosmogenic component is absent.

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Solar Wind Gases, Cosmic Ray Spallation Products, and the Irradiation History

Abstract. *The isotopic abundances of the rare gases in the fines are found to be similar to those previously reported for gas-rich meteorites. Relative to the heavy gases, neon and helium are depleted by factors of 2.5 and 10 respectively. Accurate krypton-krypton ages for rocks 10017, 10047, 10057, and 10071 were obtained covering a range of from 47 to 509×10^6 years. Varying relative production rates of the krypton and xenon isotopes in these rocks suggest different irradiation conditions and a complex history for at least some of the rocks.*

All 23 stable isotopes of the noble gases He to Xe and Kr^{81} with a half-life of 2.1×10^5 years have been determined in the soil (fines ≤ 1 mm) and in four rocks (10017, 10047, 10057, 10071) from the Apollo 11 mission. The results show that we are dealing with gases of different origin. The soil contains large amounts of trapped solar wind gases which almost completely mask other components. In the crystalline rocks, on the other hand, cosmic ray induced spallation and radiogenic gases are predominant. Small amounts of solar wind gases are found too in chips close to the rock surface. No trapped primordial or "planetary" gases were found in the rocks, which suggests that they were completely out-gassed at the time of their formation.

The relative abundances of the solar wind gases in the soil are given in Table 1. The heavier rare gases Ar to Xe show abundance ratios similar to those found in the Pesyanoe meteorite (1), and agree within factors of 2 to 3 with estimates from cosmic abundance calculations. Relative to the heavy gases, Ne and He in the fines are depleted by factors of about 2.5 and 10 respectively as compared with Pesyanoe, a typical gas-rich meteorite. The concentrations of the heavy gases in Pesyanoe are about 300 times lower and there is no evidence of saturation effects. Therefore the agreement between the abundance pattern of Ar, Kr, and Xe in lunar soil and in Pesyanoe is an important fact in the discussion of the origin of these gases.

Table 1. Relative abundances of the rare gases.

	He ⁴ /Ne ²⁰	Ne ²⁰ /Ar ³⁶	Ar ³⁶ /Kr ⁸⁴	Kr ⁸⁴ /Xe ¹³²
Lunar soil 10084,29	104	6.63	2040	7.8
Pesyanoe (1)	388	16.4	2270	7.5
Atmospheric	0.318	0.522	48.5	27.8
Av. chondritic			~200	1.3
Cosmic				
Aller (7)	355	72	6760	30
Suess-Urey (8)	400	61	4300	27
Cameron (9)	1000	13	18,000	13

Table 2. Krypton-krypton ages.

Sample	Kr ⁸³ /Kr ⁸¹	Kr _{spall} /Kr ⁸¹	P ₈₁ /P ₈₃	Kr ⁸¹ -Kr ⁸³ age (10 ⁶ years)	Kr ⁸¹ (×10 ⁻¹² cm ³ STP per gram)
<i>Rocks</i>					
10017,55	2840 ± 160	2840	0.591	509	0.347
10017,56	2597 ± 125	2520	0.588	449	0.367
10047,40	495 ± 22	470	0.604	86.0	0.343
10057,58	424 ± 18	266	0.580	46.8	0.372
10071,32	1739 ± 175	1700	0.606	312	0.420
<i>Fines</i>					
10084,29	62000 ± 9300	1800	0.60	330	0.40

If we accept trapping and gas retention efficiencies close to unity and a He⁴ solar wind flux of 6.3×10^6 atoms per cm² per sec (2), we calculate an average irradiation time of about 3×10^5 years for a dust layer of 1 g/cm². The isotopic compositions of the solar wind gases are well within the range of values previously reported for some gas-rich meteorites, except for Ar⁴⁰. All gases He to Xe contain a small but clear-cut cosmic ray spallation component and therefore do not represent the pure solar wind composition. The magnitude of the spallation components in the soil can be estimated from results obtained from stepwise heating experiments or from the Kr-Kr age of 330×10^6 years (Table 2). The measured Ne²⁰/Ne²² ratio of 12.80 is little affected by the spallation component, but in the bulk sample the measured ratios of He⁴/He³ = 2540, Ne²²/Ne²¹ = 28.3, and Ar³⁶/Ar³⁸ = 5.25 are all slightly too small. The concentration of

Ar⁴⁰ = 3.6×10^{-4} cm³ (STP) per gram in the dust is unusually high; the measured ratio Ar⁴⁰/Ar³⁶ is 1.09. From nuclear abundance systematics one would expect a much lower relative abundance of the Ar⁴⁰ isotope in the sun. The low Ar⁴⁰/Ar³⁶ ratio of 0.35 in the meteorite Novo Urei (3) also indicates a much smaller value for primordial argon. Less than 30 percent of the Ar⁴⁰ can be accounted for on the basis of the measured K and a K-Ar age of 4.5×10^9 years. A stepwise heating experiment using temperature increments of about 200°C has revealed a variable release of the Ar isotopes. The Ar³⁸/Ar³⁶ ratio, however, only increased at higher temperatures when a spallation component became apparent. The lowest ratio Ar⁴⁰/Ar³⁶ = 0.78 was found in the 800°C fraction, but gives only an upper limit for the occurrence of Ar⁴⁰ in the solar wind. The true value is probably much lower. The excess Ar⁴⁰ over this upper limit com-

bined with the measured potassium yields a K-Ar age of 4.65×10^9 years. These results suggest the presence of a radiogenic Ar component which is not related to the K in the dust. This is supported by the fact that the release of excess Ar⁴⁰ shows two maxima at 600° and 1200°C.

Inspection of the xenon isotopic composition in the soil reveals large excesses of the light isotopes up to Xe¹³¹ which can be attributed to spallation, but the neutron-rich isotopes Xe¹³⁴ and Xe¹³⁶ are deficient as compared to both trapped chondritic and atmospheric xenon. The relative abundances of these isotopes are lower than those reported for bulk analyses of the Pesyanoe meteorite, but are rather similar to those reported for the 1000°C fraction of Pesyanoe. From the stepwise heating experiment (Table 3) more information on the composition of solar wind Xe is obtained. The relative excesses on the light isotopes gradually increase with increasing temperature, clearly indicating a spallation component with a large relative yield of Xe¹³¹. The relative contents of the heavy isotopes are similar in all temperature fractions, although slightly higher values are observed in the 1000° and 1200°C fractions. This is not expected, because a larger relative spallation contribution to Xe¹³² should tend to decrease the relative abundances of Xe¹³⁴ and Xe¹³⁶. Fission Xe from spontaneous fission of U²³⁸ can account for less than 10 percent of the effect. There is no evidence for a large-scale neutron irradiation of solar Xe and Kr such as proposed by Cameron (4). As is mentioned above, the gases in the rocks are predominantly spallogenic and radiogenic. We have determined cosmic ray exposure ages for four rocks (Table 2), using the Kr-Kr dating method, which is based on the ratio

Table 3. Concentrations and isotopic composition of xenon in the lunar soil (10084,29), normalized to Xe¹³² = 100.

Sample fraction	Xe ¹³² (× 10 ⁻⁸ cm ³ STP per gram)	124	126	128	129	130	131	134	136
<i>Bulk samples</i>									
Average	2.05 ± .25	0.632 ± .012	0.725 ± .018	8.75 ± .10	104.5 ± .5	16.66 ± .08	83.5 ± .4	36.93 ± .22	29.90 ± .20
<i>Temperature fractions</i>									
600°C	0.038	0.484 ± .018	0.474 ± .015	8.29 ± .10	104.9 ± .6	16.33 ± .12	82.4 ± .4	36.90 ± .30	29.95 ± .30
800°C	0.454	0.495 ± .015	0.476 ± .012	8.47 ± .08	105.4 ± .5	16.53 ± .10	82.6 ± .3	36.81 ± .20	29.85 ± .20
1000°C	0.647	0.545 ± .012	0.595 ± .012	8.54 ± .08	104.4 ± .4	16.55 ± .10	82.7 ± .3	37.02 ± .22	30.19 ± .20
1200°C	1.168	0.736 ± .012	0.924 ± .013	9.00 ± .09	104.5 ± .4	16.74 ± .09	84.0 ± .3	37.11 ± .20	30.22 ± .20
1400°C	0.017	0.910 ± .018	1.245 ± .018	9.64 ± .11	107.5 ± .6	17.27 ± .13	88.6 ± .7	36.44 ± .30	29.39 ± .30
Total	2.32 ± .30	0.634 ± .015	0.741 ± .014	8.76 ± .09	104.7 ± .6	16.64 ± .10	83.4 ± .4	37.02 ± .25	30.12 ± .25
Pesyanoe (1)	0.0092	0.488	0.504	8.28	103.3	16.40	82.0	37.3	30.7
Atmosphere (10)		0.357	0.335	7.14	98.3	15.17	78.8	38.8	33.0

of a stable Kr isotope and the radioisotope Kr^{81} . This dating method is found to be both accurate and best suited for lunar material for the following reasons: (i) the age determination is based on the isotopic composition of one element only and therefore eliminates errors in concentration measurements; (ii) Sr, Y, and Zr, the main target elements for the production of Kr, are strongly enriched in lunar material, making it possible to measure Kr^{81} in samples as small as 0.1 g; (iii) because of both unusual chemical composition and unusual irradiation conditions the production rates of those stable isotopes generally used for age determinations are not well known.

Kr-Kr ages are obtained from the equation (5)

$$T_r = \frac{\text{Kr}^{83}_{\text{spall}}}{\text{Kr}^{81}} \frac{P_{81}}{P_{83}} \frac{1}{\lambda}$$

where $\lambda = 3.3 \times 10^{-6}$ per year, and $P_{81} = (0.95 \pm 0.05) (P_{80} + P_{82})/2$.

The relative production rates of the Kr isotopes are similar for all the investigated rocks, which is indicated by rather constant P_{81}/P_{83} ratios (Table 2). The largest variation in the $\text{Kr}^{78}/\text{Kr}^{83}$ ratio is 15 percent (between rocks 10047 and 10057). Spallation Kr^{83} in rocks containing solar wind Kr were corrected for solar wind gases by using for the spallation component the lowest $\text{Kr}^{86}/\text{Kr}^{83}$ ratio of rock-specimen 10017, 55, an interior sample, taking into account a contribution to Kr^{86} from spontaneous fission of U. The $\text{Kr}^{83}_{\text{spall}}/\text{Kr}^{81}$ ratio of an outside chip of rock 10017 was found to be 10 percent smaller than that of an inside sample, which may be attributed to a slightly higher Kr^{81} content. However, the relative errors in the Kr contents are about 6 percent.

Do the obtained Kr-Kr ages in fact give the actual time interval of exposure to cosmic rays? As has already been mentioned, there are obvious differences in the relative production rates of certain isotopes which cannot be explained by a varying chemical composition but only by differences in the composition and energy distribution of the interacting cosmic rays. Variations in the relative production of He^3 and Ne^{21} may in part be explained by differences in chemical composition or by possible diffusion losses. One is tempted to ascribe the observed variations in the relative production of the Kr isotopes to differences in the relative abundances of the target elements Sr, Y, and Zr. Large-scale variations in the

relative production of the Xe isotopes, however, cannot be explained simply on the basis of chemical differences. Furthermore, the fact that specific relative production rates correlate with each other, such as the $\text{Kr}^{78}/\text{Kr}^{83}$ and $\text{Xe}^{126}/\text{Xe}^{131}$ ratios, leads us to conclude that the irradiation conditions of the four investigated rocks did differ from each other. The measured $\text{Xe}^{131}/\text{Xe}^{126}$ ratios vary by almost a factor of 2 and are on the average also higher than the corresponding ratios found in meteorites. However, Pepin (6) found a $\text{Xe}^{131}/\text{Xe}^{126}$ ratio of ≥ 5.3 in the Estherville meteorite, a value which is within the range covered by our four rocks and the soil sample. The evidence of such large relative Xe^{131} yields, which require a heavily moderated cosmic ray irradiation, suggests that special reactions may be partly responsible for the excess Xe^{131} production. Because of the high capture cross section, Gd is a sensitive indicator for thermal neutrons. We have measured the isotopic compositions of Gd in rock 10017 and in our soil sample. The depletion of Gd^{157} is between 0.1 and 0.4 percent for rock 10017, and for the soil sample we can give an upper limit of 0.2 percent. Because of background interferences in the Gd mass spectrum we can give only limits. This result shows that thermal neutron capture contributes less than 2 percent

to the total Xe^{131} production. No neutron resonances are known for Ba^{130} and it seems unlikely that the neutron resonance integral will contribute much to the total cross section.

If the irradiation history of a rock is complex, if the rock has been partially and variably shielded from cosmic rays, then the reported radiation age represents only a lower limit for the actual time of irradiation. Our results indicate such complex histories at least for rocks 10017 and 10057 and for the soil sample.

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Isotopic Composition of Rare Gases in Lunar Samples

Abstract. Data from total melt and step-by-step heating experiments on the Apollo 11 lunar samples suggest a close affinity between lunar and meteoritic rare gases. Trapped neon-20/neon-22 ratios range from 11.5 to approximately 15, resembling those for the gas-rich meteorites. Trapped krypton and xenon in the lunar fines and in the carbonaceous chondrites are similar except for an interesting underabundance of the heavy isotopes in both lunar gases which suggests that the fission component found in carbonaceous chondrites is depleted in lunar material. Spallation gases are in most cases quite close to meteoritic spallation gases in isotopic composition.

Mass spectrometric analyses were carried out for all rare gas isotopes in six lunar crystalline rocks, a breccia, coarse fines sieved from the lunar soil, and the soil.

Gases in the crystalline rocks are in general dominated by radiogenic and spallogenic isotopes. The ^3He radiation ages, on the assumption of a ^3He production rate of $1 \times 10^{-8} \text{ cm}^3(\text{STP}) \text{ g}^{-1} 10^6 \text{ year}^{-1}$, range from $\sim 30 \times 10^6$ (sample 10069,21) to 210×10^6 years (10071,20). U, Th-He, and K-Ar gas-retention ages, which are consistent

with the values $\sqrt{2} 3.5 \times 10^9$ years reported in the preliminary analysis (1), as well as Ne, Ar, Kr, and Xe radiation ages, will be discussed in a later publication.

Neon isotopic compositions in the rocks and coarse fines are shown in Fig. 1. The pattern indicates a highly variable mixture of a single spallation composition, perhaps represented in virtually pure form in 10020,47[2], and a trapped neon component with $^{20}\text{Ne}/^{22}\text{Ne} = 12.7 \pm 0.4$. Neon in 10020,47[2] is essentially identical to that in the C3