by the fact that this sample had been stored in air for several weeks prior to measurement. We conclude that the hydrogen abundance of the rock was less than or equal to 10 ppm.

The tritium activity released from the fine material in the various temperature intervals is given in Table 2. Sixty-three percent of the tritium was released at 600°C, closely paralleling the evolution of hydrogen (80 percent). The total tritium activity observed in the fine material was approximately the same as that observed in the rock. All of these samples have approximately the same amount of tritium actvity as that usually observed in stone meteorites (*7*)—namely 200 to 700 dpm/kg. It is clear that the high tritium content that one might expect from solar-flare tritons imbedded in an exposed surface is not observed. Perhaps the samples measured were not truly surface samples. The fines were well mixed, and, if there were a high tritium content on the surface, it could be highly diluted with deeper-lying material. The exterior rock sample did have micrometeorite pits on its surface, but it is not at all certain that this surface had been exposed during the last 20 years or so. The interesting observation of Fireman and his associates (*2*) of a triton component in solar-flare particles should be tested with lunar material known to have been exposed in recent years on the surface.

The release of ^{37}Ar and ^{39}Ar radioactivities from the fine material during the stepwise heating shows that the argon evolved almost completely in the temperature interval 600° to 1200°C. Measurements on the combined sample evolved below 600°C and above 1200°C (see Table 2) showed that less than 4 percent of both of these activities was released outside the 600° to 1200°C interval. This result indicates that the argon produced throughout this silicate material with an average particle size of 100 microns (*4*) is strongly retained. The total ^{37}Ar and ^{39}Ar activity measured in this material is approximately the same as that measured in the rock sample. Since the fine material contains large quantities of solar-wind argon, it is impossible to determine the ^{38}Ar produced by cosmic rays, and therefore an effective exposure age cannot be deduced.

The ^{37}Ar and ^{39}Ar activities produced in the fines and the rock were compared with those expected from cosmic ray bombardment. The magnitude of the activities almost exactly equals those observed in recently fallen meteorites (*7*). The meteorite samples studied are regarded as interior pieces of objects, weighing 10 to 100 kg, that receive an isotropic cosmic ray exposure. The surface lunar material might be expected to contain higher activities produced by low-energy solar and galactic cosmic rays, but on the other hand the lunar surface receives only about half the isotropic flux. Apparently the geometric and shielding effects nearly cancel. The ratio of the ^{37}Ar and ^{39}Ar activities can be compared to the ratio expected from cosmic ray bombardment. The cross section for producing these isotopes in thin targets (1 to 5 g cm^{-2}) of iron, titanium, calcium, and potassium at 600 Mev is given in Table 3, along with the chemical analysis of rock 10057 (*4*). On the basis of these data the calculated ^{37}Ar/^{39}Ar atom ratio, constant cosmic ray bombardment being assumed, is 3.2 for the fine material and 2.2 for rock 10057. The observed ^{37}Ar/^{39}Ar ratio for both of these samples is about 30 percent lower than the estimated cross-section ratios. To obtain an accurate model for the production ratio for these isotopes, a series of bombardments of thick targets of lunar-like material at several energies is required. Experiments along these lines are in progress, but for the present we must use the available information at 600 Mev. To deduce a cosmic ray exposure age based upon the ^{38}Ar content of rock 10057, one must evaluate the relative production rates of ^{38}Ar and ^{39}Ar. Using the cross sections listed in Table 3, we calculate an ^{38}Ar/^{39}Ar cross-section ratio of 2.8. The ^{38}Ar content of rock 10057 was measured, by Funkhouser and his associates (*5*), to be 7.1×10^{-8} cm^3 (STP)/g. Using the measured ^{38}Ar and ^{39}Ar production rates and the calculated ^{38}Ar/^{39}Ar cross-section ratio, we calculate that rock 10057 was exposed for 110 million years on the lunar surface.

Since the lunar samples are high in Sr, Y, and Zr, it is possible that measurable amounts of ^{81}Kr and ^{85}Kr may be present. A search was made for krypton activities in rock 10057, but only an upper limit of less than 0.5 dpm/kg could be set. From the composition of the rock and an estimate of the production cross sections, we conclude that the level of both ^{81}Kr and ^{85}Kr activities is about 0.2 dpm/kg.

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Inert Gases in Lunar Samples

Abstract. *Sample 10084,40 (fines, less than 1 millimeter) contains substantial amounts of the inert gases. Their concentrations are inversely proportional to particle size; hence the gases appear to be surface-correlated in the soil fragments. The most likely origin of the gas is solar wind or solar cosmic rays. Glass and feldspar are generally poorer in gas than lithic fragments. Ratios of elements in the sample differ significantly from solar values. Ratios of isotopes in the sample are similar to those in meteorites. Argon-40 appears to consist of a radiogenic and a surface-correlated component. An apparent potassium-argon age of* $4.42^{+0.24}_{-0.28}$ *billion years is calculated.*

We have determined (mass-spectrometrically) the concentrations of the inert gases, and in all cases the isotopic compositions of He, Ne, and Ar in the following "splits" of sample 10084,40 ("fines," < 1 mm): 1 and 2, "bulk" of 10084,40 (weights 3.507 and 10.827 mg); 3, lithic fragment, $\sim$800 μm

Table 1. Inert gas contents. Numbers refer to sample description in the text. Units: 3 = 10^{-7}; 4 = 10^{-3}; 20 = 10^{-5}; 21 = 10^{-8}; 22 = 10^{-6}; 36 = 10^{-6}; 38 = 10^{-7}; 40 = 10^{-5}; 84 = 10^{-8}; 132 = 10^{-9}, all cm^3/g at standard temperature and pressure. Numbers in parentheses are cumulative percent errors of the values listed. Only errors which could be experimentally determined are included: weighing error, standard deviation of calibrations, standard deviation of dilutions, standard deviation of peak heights, and errors in blank corrections. We believe that the accuracy of the data is within twice the listed errors. Errors of element ratios such as ^{4}He/^{20}Ne can be computed from this table, but errors in isotope ratios such as ^{20}Ne/^{22}Ne cannot.

Sample	Helium		Neon			Argon			Krypton	Xenon
	3	4	20	21	22	36	38	40	84	132
1	923 (1.9)	246 (2.1)	240 (2.1)	640 (3.1)	180 (1.9)	403 (1.8)	754 (1.7)	49.4 (2.0)	18.2 (4.7)	40.3 (9.0)
2	818 (2.4)	225 (2.8)	236 (2.4)	633 (2.7)	180 (2.5)	390 (2.0)	726 (2.0)	48.6 (2.0)	16.4 (3.1)	34.3 (2.6)
3	101 (1.9)	11.7 (1.9)	10.1 (1.7)	150 (1.9)	8.55 (1.7)	17.2 (1.7)	41.2 (1.7)	14.3 (12)	2.3 (10)	10.7 (10)
4	74.6 (1.7)	21.2 (1.9)	28.0 (1.7)	93.5 (3.4)	21.8 (1.7)	86.5 (1.7)	164 (1.7)	15.9 (2.5)	4.66 (8.4)	16.0 (17)
5	80.8 (1.7)	19.1 (1.9)	18.6 (1.7)	93.4 (4.3)	14.6 (1.7)	26.9 (1.7)	56.2 (1.7)	8.72 (5.1)	1.50 (20)	6.3 (20)
6	119 (2.6)	26.1 (1.9)	26.4 (1.7)	143 (3.3)	21.0 (1.7)	50.0 (1.7)	101 (1.7)	13.6 (1.9)	2.71 (15)	7.0 (20)
7	297 (1.8)	75.0 (1.9)	79.3 (1.7)	264 (2.6)	61.1 (1.7)	145 (1.7)	275 (1.7)	24.8 (1.9)	7.4 (10)	19 (20)
8	693 (1.9)	182 (1.9)	202 (1.7)	564 (2.5)	151 (1.7)	333 (1.7)	623 (1.7)	41.2 (1.9)	13.7 (7)	34 (15)
9	368 (1.9)	97.8 (1.9)	129 (1.7)	380 (1.9)	98.8 (1.7)	277 (1.9)	518 (1.7)	34.9 (2.1)	11.9 (10)	34 (15)
10	107 (2.0)	24.2 (1.9)	29.2 (1.7)	107 (6.2)	22.3 (1.9)	36.5 (2.0)	70.7 (1.7)	7.74 (2.6)	2.1 (20)	8.6 (50)
11	7.8 (16)	0.9 (50)	3.34 (5.3)	34 (50)	2.85 (8.6)	5.87 (5.5)	12.0 (6.8)	19.9 (13)		
12	46.2 (4.4)	11.5 (2.2)	26.0 (2.0)	140 (5.3)	20.8 (2.7)	76.9 (1.9)	149 (2.1)	36.4 (2.9)	4.8 (20)	7.6 (50)
13	15.9 (5.6)	3.59 (4.4)	13.2 (2.2)	66 (11)	10.4 (2.8)	14.4 (2.3)	28.7 (2.8)	37.6 (2.8)		
14	11.4 (11)	1.95 (3.6)	1.76 (3.7)	13.7 (15)	1.33 (4.5)	3.31 (3.4)	9.9 (3.7)	2.44 (14)		
15	9.1 (2.6)	2.24 (1.9)	3.02 (1.9)	7.6 (12)	2.24 (2.6)	6.96 (1.9)	13.1 (1.9)	4.19 (5.3)		
16	8.0 (5.2)	2.03 (1.9)	2.16 (1.9)	33.9 (6.2)	1.92 (4.3)	4.45 (4.4)	12.6 (2.7)	7.18 (8.1)		
17	23.5 (2.7)	2.85 (2.6)	3.33 (1.9)	46 (10)	3.02 (4.8)	2.02 (2.8)	3.8 (2.8)	3.15 (6.2)		
18	8.8 (5.3)	2.44 (2.6)	7.59 (1.7)	68.9 (5.2)	6.46 (2.6)	9.19 (1.7)	19.0 (2.6)	6.05 (3.4)		

(0.879); 4, lithic fragment with black fusion crust, ~800 μm (0.704); 5, eight lithic fragments, ~500 μm (0.644); 6, twenty-nine lithic fragments, ~250 μm (0.637); 7, sieve fraction 70 to 105 μm (0.574); 8, sieve fraction < 70 μm (0.409); 9, strongly magnetic portion 70 to 105 μm (0.499); 10, weakly magnetic portion 70 to 105 μm (0.552); 11, brown sphere and spindle, ~250 μm (0.051); 12, three black spheres, ~400 μm (0.175); 13, dark brown glass, ~400 μm (0.171); 14, white "sugary" fragment (feldspar ?), ~550 μm (0.243); 15, lithic fragment with glass and ilmenite, ~800 μm (1.050); 16, four transparent colorless fragments, ~350 μm (0.519); 17, yellow glass, ~600 μm (0.420); 18, green glass, ~500 μm (0.467). The samples were melted in a vacuum furnace, using induction heating. The gases were then cleaned on hot Ti-Zr getters. Absolute amounts were obtained by peak comparison, with carefully metered standards to calibrate the sensitivity of the spectrometer. Preliminary results are listed in Table 1. These results confirm one finding of the preliminary examination team (*1*), namely that the Apollo 11 "fines" contain vast quantities of the inert gases. However, glassy fragments (samples 11–18) are nearly always poorer in gas than lithic fragments of the same size. The same difference is shown by the strongly and weakly magnetic portions of the 70 to 105-μm fraction (samples 9 and 10): the latter contains a substantially greater proportion of clear glassy fragments. The difference could in part be explained by diffusion loss of gas, because the lowest ^{4}He/^{20}Ne ratios occur among samples 11–18. Closer inspection of the data reveals that this cannot be the sole cause—some of the highest ^{20}Ne/^{36}Ar ratios are found among the same samples. Perhaps the glassy fragments were formed more recently than the lithic fragments; hence they were exposed to solar wind and solar cosmic rays for a shorter time. Alternatively, the diffusivity of the inert gases in the glasses is so high that gas implanted by solar wind was nearly wholly lost from them, leaving the more firmly implanted solar cosmic-ray ions behind, which might explain the increased ^{20}Ne/^{36}Ar ratios. Even so, the ^{4}He diffusivity in the glasses must be exceptionally high to account for the low ^{4}He/^{20}Ne ratios.

For samples 3–10 (we restrict this discussion to lithic fragments of similar composition and mineralogy), the amount of ^{36}Ar per gram increases in the smaller fragments (Fig. 1), which suggests that the gas is surface-correlated. There is, of course, a possibility that part of the gas is really contained in the very fine, micrometer sized (or less) dust which adheres loosely to surfaces of larger fragments, thus simulating the correlation of Fig. 1. However, an acetone-cleaned sample of fragments in the 105 to 250 μm range agrees closely with the correlation (Fig. 1, point 19). This shows that the trend is real.

Thus, we conclude that most of the inert gases were implanted in the fragments from the solar wind, and from solar cosmic rays of greater energy. We estimate that the average "hemispherical" exposure of a 250-μm fragment to solar wind of present-day intensity was $\sim 10^3$ years (adopted $H^+ = 10^8/\text{cm}^2$ sec; $H/^{36}Ar = 10^5$). However, a calculation based on ^{84}Kr gives $\sim 10^4$ years (adopted $H/^{84}Kr = 4 \times 10^9$). These times become much longer if solar cosmic rays were the source of the gas. In addition to direct implantation of solar wind and solar cosmic rays, gas could have been acquired from gas-rich cosmic dust, firmly bonded to the surface of the soil fragments.

Among the principal elemental ratios, $^4He/^{20}Ne$ ranges from 26 to 116, with 13 of 18 ratios >75. These values are substantially below those normally found in gas-rich chondrites or carbonaceous chondrites (*2*), but are similar to the ratios in the howardites (*3*).

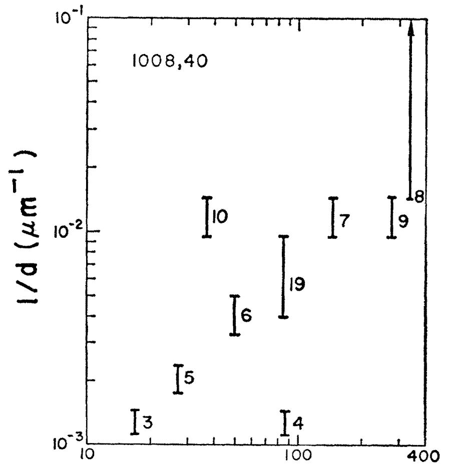

Fig. 1. The ^{36}Ar plotted against inverse of particle size (1/d) for lithic fragments. Glass fragments and feldspar, like the weakly magnetic portion 70 to 105 μm (sample 10) nearly always contain less ^{36}Ar than the lithic fragments.

The $^{20}Ne/^{36}Ar$ ranges from $\sim 3 \simeq 17$, lower than in gas-rich chondrites, but greater than in most carbonaceous chondrites (*2*). The $^{36}Ar/^{84}Kr$ ranges from $\sim 700 \simeq 2400$, similar to the value in the dark portion of the Fayetteville chondrite (*4*), but considerably above the values in carbonaceous chondrites (*2*). Finally, $^{132}Xe/^{84}Kr = 2 \simeq 6$, somewhat above chondritic values (*2*). Certainly, all these ratios are strikingly less than any "solar" or "cosmic" ratios that have been reported. The reasons for this apparent fractionation are not wholly clear; there may be several: preferential diffusion loss of the lighter gases, admixture of meteoritic (chondritic?) material which is known to contain a fractionated component, perhaps a "saturation" effect in the surface for the more abundant, lighter elements, and preferential removal of the lighter gases by sputtering.

A cosmic-ray produced component is present in a number of the samples. We have estimated cosmogenic ^3He_c, $^{21}Ne_c$, and $^{38}Ar_c$ with the following assumptions: "solar" $(^3He/^4He)_s = 3.73 \times 10^{-4}$; $(^{20}Ne/^{21}Ne)_s = 370$; $(^{36}Ar/^{38}Ar)_s = 5.36$; and $(^{36}Ar/^{38}Ar)_c = 0.6$. The amounts of ^3He_c range from ~ 100 to 600×10^{-8} cm^3/g at standard temperature and pressure; $^{21}Ne_c \sim 15 \simeq 120 \times 10^{-8}$ cm^3/g at standard temperature and pressure; and $^{38}Ar \sim 10 \simeq 100 \times 10^{-8}$ cm^3/g at standard temperature and pressure. With one-half of the production rates in Ca-rich achondrites (*5*), the corresponding "exposure ages" are estimated as ranging from tens to hundreds of millions of years. However, the lunar samples may have been exposed to solar cosmic rays as well as to galactic cosmic rays (the effects of solar cosmic rays cannot normally be detected in meteorites because of ablation) such that these "exposure ages" must be considered maximum values. More information on the radioactive species is needed to settle this question more firmly.

The $^3He/^4He$ ranges from $\sim 3.5 \simeq 9 \times 10^{-8}$; the increase is due to ^3He_c as can be seen from concomitant changes in the Ne and Ar isotopic ratios. The $^{20}Ne/^{22}Ne$ ranges from 13.5 to 11.0; $^{21}Ne/^{22}Ne$ from 0.034 to 0.176. On a three-isotope plot (*6*) the points lie near a straight line: $(^{20}Ne/^{22}Ne) = 13.65$ to $14.47\ (^{21}Ne/^{22}Ne)$ (*7*). The isotopic composition of Xe measured in sample 2 is similar, but not identical to that in carbonaceous chondrites. On the basis of $^{132}Xe = 100$, the composition is: 124 (0.48), 126 (0.54), 128 (7.93), 129 (103), 130 (15.7), 131 (81.78), 134 (37.36), 136 (30.86).

In Fig. 2 we have plotted ^{36}Ar against ^{40}Ar in 9 samples (*8*) (most of the glasses and feldspar and samples 3 and 10 do not fall on the correlation, but most lithic fragments do). The points lie close to $^{40}Ar = 1.056\ ^{36}Ar + (7.120 \pm 1.086) \times 10^{-5}$ cm^3/g at standard temperature and pressure. We interpret the correlation as follows: all the samples contain approximately the same amount of K, have about the same apparent K-Ar age, and contain $(7.120 \pm 1.086) \times 10^{-5}$ cm^3/g at standard temperature and pressure radiogenic ^{40}Ar. With K = 0.1 percent by weight (*1*) and terrestrial abundance for ^{40}K, the corresponding K-Ar age is $4.42^{+0.24}_{-0.28}$ billion years. In addition all the samples contain surface-correlated ^{40}Ar with $^{40}Ar/^{36}Ar = 1.056$. This explanation would alleviate the difficulty of embarrassingly high (>7 billion years) K-Ar ages for samples 1 and 2 but it introduces a new puzzle—where does this ^{40}Ar come from? A solar wind origin as postulated for ^{36}Ar seems very unlikely (*9*). The moon itself provides an environment where ^{40}Ar could have been produced by K-decay. Escape of an ^{40}Ar-rich gas from the interior of the moon would make ^{40}Ar available

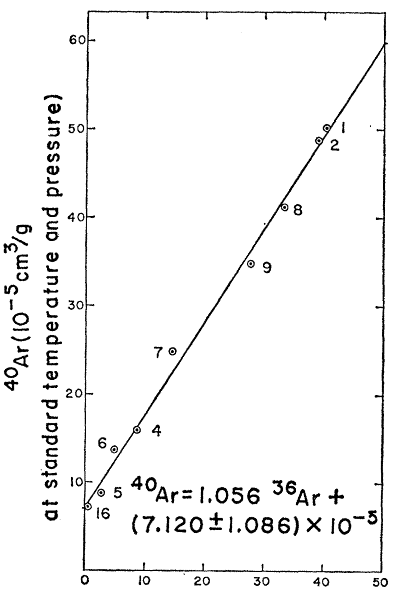

Fig. 2. The ^{36}Ar plotted against ^{40}Ar in nine samples. We conclude that all the samples contain $(7.120 \pm 1.086) \times 10^{-5}$ cm^3/g at standard temperature and pressure of radiogenic ^{40}Ar. With K = 0.1 percent the K-Ar age is $4.42^{+0.24}_{-0.28}$ billion years. The samples also contain surface correlated ^{40}Ar with $^{40}Ar/^{36}Ar = 1.056$.

at the surface (the fraction of ^{40}Ar liberated in the interior need not be large). At the moon's surface this gas must become entrapped in the surfaces of particles. Several mechanisms are possible: shock implantation *(10)*, simple dissolution, or "pumping" by elastic collisions with solar wind or solar cosmic-ray ions. We prefer the third possibility because it does not seem to require substantial gas pressures in the moon's atmosphere at any time in the past. An alternative explanation is that ^{40}Ar was acquired from impacting cosmic dust. This requires that the dust is considerably older than the moon, perhaps some 7 to 8 billion years old *(11)*.

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Trapped Solar Wind Noble Gases, Kr^{81}/Kr Exposure Ages and K/Ar Ages in Apollo 11 Lunar Material

Abstract. *Grain size and etching experiments show that the fine lunar material contains large amounts of trapped solar wind particles. Elemental and isotopic compositions of the noble gases in solar material and in the terrestrial atmosphere are significantly different, except for the Ar^{36}/Ar^{38} and the Kr isotope ratios. Exposure ages of two rocks and of the fine material are between 380 and 510 × 10^6 years. Feldspar concentrates give K/Ar ages of 3220 and 3300 × 10^6 years, significantly higher than the unseparated rock.*

The concentrations and isotopic composition of the noble gases He, Ne, Ar, Kr, and Xe were measured in (i) seven grain-size fractions of fine material (sample No. 10084) (grain sizes between 1 and 150 μm); (ii) four grain-size fractions of ilmenite separated from the fine lunar material (grain sizes between 20 and 110 μm); (iii) four etched samples from an ilmenite grain-size fraction (42 μm); (iv) a number of individual crystals, spherules, and other structural elements separated from fine lunar material; (v) three rock samples from rocks No. 10017 and 10071; and (vi) two feldspar concentrates from rock No. 10071. In addition K and Ba were determined in several of these samples.

Table 1. Comparison of trapped solar gas in lunar ilmenite and terrestrial atmosphere.

Ratio	Trapped solar gas	Terrestrial atmosphere
He^4/Ar^{36}	5300 ± 800;	
Ne^{20}/Ar^{36}	24 ± 3	0.5
Kr^{86}/Ar^{36}	$(2 \pm 0.3) \times 10^{-4}$	63×10^{-4}
Xe^{132}/Ar^{36}	$(1.1 \pm 0.2) \times 10^{-4}$	7×10^{-4}
He^4/He^3	2700 ± 100	
Ne^{20}/Ne^{22}	12.90 ± 0.07	9.8
Ne^{20}/Ne^{21}	410 ± 12	338
Ar^{36}/Ar^{38}	5.30 ± 0.05	5.35
Ar^{40}/Ar^{36}	≤0.7.	
Kr isotopes, atmospheric, see text		
Xe isotopes, see text and Fig. 4		

From these results the elemental and isotopic abundances of the trapped noble gases were calculated, and limits on the elemental and isotopic abundances in the solar wind can be inferred. Exposure ages were obtained from Kr^{81}/Kr measurements, and K/Ar ages were derived for the rock samples.

The preliminary examination of the fine lunar material *(1)* already revealed the presence of large amounts of noble gases, presumably of solar wind origin. Other possible sources of these trapped gases *(2)* are energetic solar particles, an ambient atmosphere early in the history of lunar material, or gases released during the impact of comet nuclei. To establish the origin of the trapped gases we measured the noble gas concentration in different grain-size fractions of the bulk material *(3)*. The He^4 and Xe^{132} results are given in Fig. 1. A distinct anticorrelation between grain size and trapped noble gas content is evident. Trapped Ne, Ar, and Kr show a similar grain-size dependence.

The experiment was repeated on ilmenite grain-size fractions separated from the fine material. An even stronger anticorrelation between grain size and trapped gas content was observed (Fig. 1 for He^4 and Xe^{132}; Ne, Ar, and Kr show similar dependence). Such a grain size dependence results if the trapped gases are located at the surface of the individual grains and if their concentration per unit of surface area is roughly independent of the grain size.

In a further study of the location of the trapped gases we etched aliquots of the 42 μm ilmenite grain-size fraction with dilute HF to remove a thin layer from the surface of the individual grains. The mean thickness of the etched layer was determined from the amount of Fe and Ti dissolved. The noble gases remaining in the etched grain-size fractions were then measured. While the concentration of the noble gas spallation fraction was essentially unchanged by the HF treatment, the concentration of all trapped gases decreased strongly after removing a surface layer with an approximate average thickness of 2000 Å (Fig. 2). We conclude that the trapped noble gases are located within a depth of 2000 Å below the surface of the individual grains. This is in agreement with the observed grain size dependence of the trapped gas content.

This experimental evidence strongly supports a solar-wind origin of the trapped gases. The range of ions with

Table 2. The K/Ar ages and Kr^{81}/Kr exposure ages of Apollo 11 lunar rocks 10017 and 10071.

Rock No. and fraction	K/Ar ages (× 10^6 years)	Kr^{81}/Kr exposure age (× 10^6 years)
10017 Unseparated	2200 *	510 ± 50
10071 Unseparated	2820 ± 80	380 ± 25
10071 Feldspar CA4	3300 ± 80	
10071 Feldspar CA6	3220 ± 80	

* Ar^{40} concentration without isotopic dilution.