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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7. This equation assumes a two-component mixture. However, Ne in lunar samples may well consist of more than just two components: solar wind, solar cosmic rays (both trapped), and spallation components due to galactic cosmic rays and solar cosmic rays.
8. The point for the acetone-washed sample 19 (105 to 250 μm) which is not shown almost coincides with that of sample 4.
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4 January 1970

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 ± 0.3) × 10^{-4}	63 × 10^{-4}
Xe^{132}/Ar^{36}	(1.1 ± 0.2) × 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}	$\leq$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.

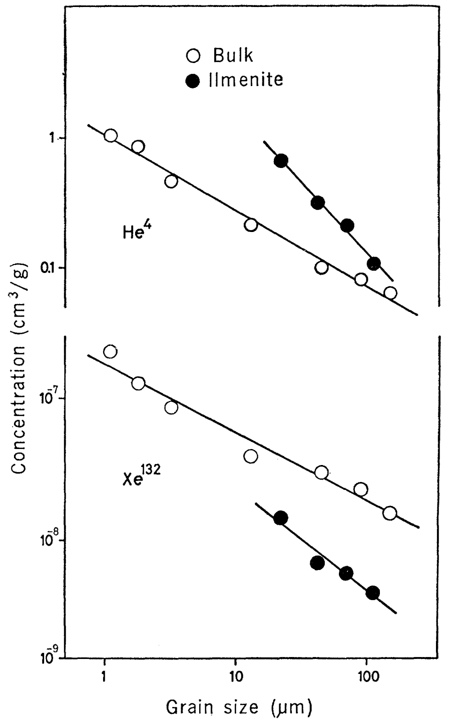
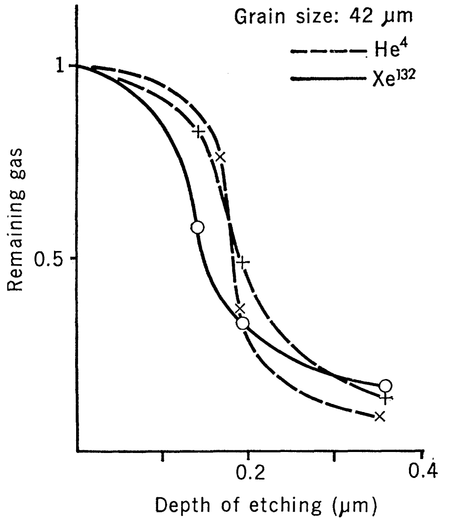


Fig. 1 (left). Grain-size dependency of trapped He and Xe in unseparated Apollo 11 fine material and in ilmenite ($FeTiO_3$) separated from the fine material. Trapped Ne, Ar, and Kr show similar dependencies.

Fig. 2 (right). Trapped He and Xe remaining in the 42-µm ilmenite grain-size fraction after a thin surface layer has been removed from the individual grains by etching with HF. The horizontal axis is equivalent to the thickness of the layer removed from the surface of the individual grains. The two curves for He^4 represent two independent sets of etching experiments. Trapped Ne, Ar, and Kr show similar dependencies.



an energy of 1 kev per nucleon, as observed in the present-day solar wind, is of the order of a few hundred to thousand angstroms. Independent experiments have already established that solar-wind noble gas ions indeed reach the lunar surface (*4*).

For ilmenite, the He^4 concentration is inversely proportional to grain size within 10 percent (Fig. 1), that is, the He^4 concentration per square centimeter of surface area is independent of grain size. This requires that either the grains were exposed to the solar wind for the same time interval, independent of grain size, or the exposure was so long that saturation occurred. The observed surface concentrations of 10^{-3} cm^3 He^4/cm^2 correspond to an integrated solar wind flux of 7 × 10^{17} $atom/cm^2$. Such an irradiation dose would require an exposure time of approximately 300 years at the present level of the solar wind flux. At these levels of irradiation severe saturation effects have to be expected (*5*), which will most likely influence the relative abundance of the trapped solar wind ions. If we assume that the effective trapping efficiency is reduced to about 10 percent because of saturation, a few hundred million years are needed to obtain the observed helium loading of ilmenite surfaces down to a depth of about 60 cm. This figure compares well with a cosmic ray exposure age of the Apollo 11 sample of fine material of 500 × 10^6 years, as estimated from $Xe^{126}_{\text{spallation}}$ and measured Ba concentrations.

Figure 3 shows the observed range of abundances of trapped gas relative to trapped Ar^{36}. The overall abundance pattern is quite similar to the predicted cosmic abundances (*6*) and to the abundances of trapped gas observed in some gas-rich aubrites (*7–9*). The Ar-Kr-Xe abundances in the bulk fine material and the separated ilmenite are identical. However, the He^4/Ar^{36} and Ne^{20}/Ar^{36} ratios are considerably higher in the ilmenite than in the bulk material. This is best explained by diffusion loss of He and Ne in the bulk material leading to a relative enrichment of trapped Ar, Kr, and Xe. The abundances and isotopic compositions of the trapped gases in the Apollo 11 fine material are not necessarily identical with those of the solar wind. They may have been altered by diffusion losses, systematic differences in trapping probabilities and saturation effects. Diffusion loss and the mass dependency of trapping probabilities should always lead to a depletion of the light gases and isotopes. The highest observed He^4/Ar^{36} and Ne^{20}/Ar^{36} ratios should thus be most representative of the true solar-wind abundances. Since no experimental evidence on the influence of saturation effects on the elemental and isotopic composition is presently available we have to disregard this effect.

The four ilmenite fractions show the least evidence for diffusion loss, and the abundance ratios for the trapped component determined from these mineral concentrates are given in Table 1. The errors correspond to the range of values observed in the four different ilmenite grain-size fractions (except Ar isotope ratios). The necessary corrections for spallation-produced He^3, Ne^{21}, and Ar^{38} were obtained from correlation diagrams and the HF-etched ilmenite fractions. The Ar^{40}/Ar^{36} ratio given is the lowest we observed. A contribution of radiogenic argon from the decay of K^{40} cannot be excluded, and the ratio Ar^{40}/Ar^{36} of 0.7 must be considered as upper limit for the Ar^{40}/Ar^{36} ratio of the trapped gas. All grain-size fractions of the bulk material give Ar^{40}/Ar^{36} ratios higher than 0.7. In the finer grain-size fractions the measured K concentration is not high enough to account for this Ar^{40} excess in terms of *in situ* decay.

All ilmenite fractions have a sizable spallation Kr content, and the very precise evaluation of the trapped Kr isotopic composition which is required to distinguish between atmospheric Kr and AVCC Kr (*10*) is difficult. The bulk grain-size fractions contain relatively little spallation Kr, and correlation diagrams indicate that the trapped Kr in these fractions is more similar to atmospheric Kr than to AVCC Kr.

All bulk and ilmenite grain-size fractions show spallation Xe. From suitable correlation diagrams we can show that the trapped Xe cannot have atmospheric composition. The correlation lines for the light Xe isotopes $Xe^{124,\,126,\,128,\,130}$ virtually pass through the points corresponding to AVCC Xe, and we conclude that the relative abundances of these isotopes in the trapped gas can be compatible with AVCC Xe within 2 percent. We shall use the AVCC isotopic composition for these isotopes for the further discussion. The abundance of the heavy isotopes $Xe^{131,\,132,\,134,\,136}$ relative to Xe^{130} is significantly lower than in atmospheric or AVCC Xe (spallation correc-

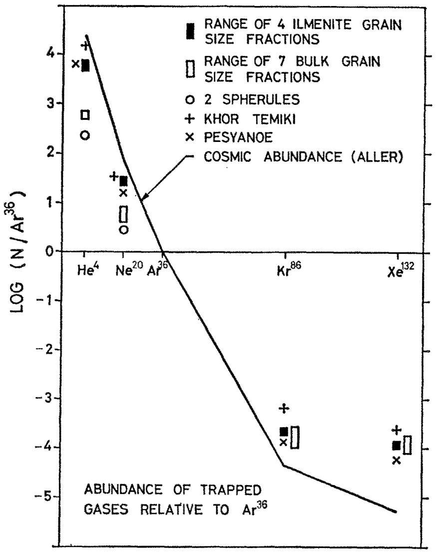


Fig. 3. Trapped gas abundances relative to Ar^{36} in bulk grain size (1 to 150 μm) and ilmenite grain size (20 to 110 μm) fractions separated from Apollo 11 fine lunar material. The two spherules separated from the fine lunar material had diameters of 0.2 mm. Other data from (*6–9*).

tion applied). From six fractions with the smallest spallation we obtain the δ values given in Fig. 4.

The mass dependence of the absolute abundances of noble gases in the trapped gases is less steep than the predicted cosmic abundances (*6*) (Fig. 3). We cannot yet decide whether this is due to saturation effects in the trapping of the solar wind or to mass discrimination in the solar wind or whether this reflects the true chemical composition of the solar wind reservoir.

For the discussion of the solar-wind noble gas isotopic compositions we will assume that these ratios represent to a good approximation the true isotopic compositions prevailing at the time of formation of the solar system. The lower terrestrial Ne^{20}/Ne^{22} and Ne^{20}/Ne^{21} ratios are in excellent agreement with a mass dependent fractionation process (for example gravitational escape), terrestrial Ne representing the remaining few per mil of the original gas. If we calculate the overall fractionation factor from the Ne^{20}/Ne^{22} ratio, then the terrestrial Ne^{20}/Ne^{21} ratio predicted from the solar wind Ne^{20}/Ne^{21} ratio is rather high compared with the observed terrestrial value. This may indicate that the terrestrial atmosphere contains some Ne^{21} from spallation reactions or (α,n) reactions on O^{18} (*11*). Terrestrial and solar Ar^{36}/Ar^{38} ratios agree within the present experimental uncertainty.

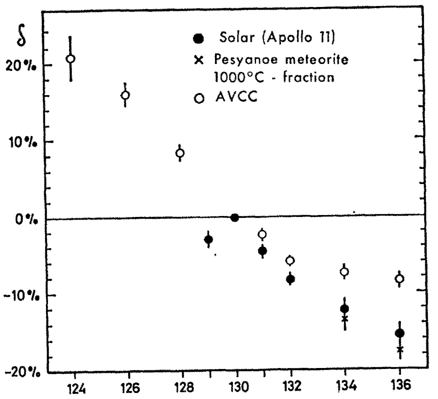


Fig. 4. Isotopic composition of trapped solar wind Xe in Apollo 11 fine material, of AVCC Xe (*10*) and of trapped Xe in the Pesyanoe meteorite (*9*).
$\delta = [(Xe^{M}/Xe^{130})/(Xe^{M}/Xe^{130})_{atm} - 1]$ is the relative deviation from the atmospheric composition.

The difference between the abundance of the heavy isotopes in AVCC Xe and solar Xe could be explained by a fission component in AVCC Xe. The required fission spectrum would be $Xe^{131} : Xe^{132} : Xe^{134} : Xe^{136} = 0.74 : 1.03 : 0.81 : 1.0$ with $Xe^{136}_{fission}/Xe^{136} = 0.08$ in carbonaceous chondrites. A possible long-lived isotope from the "island of stability" near $N = 184$ might be the progenitor (*12*). However, if fission Kr is indeed present in AVCC Kr (*10*), we would have $(Xe^{136}/Kr^{86}{}_{fission}) \sim 10$, a ratio much too low for the fission of super heavy nuclides. The large differences observed between solar wind and atmospheric Xe are difficult to explain as the mass fractionation factors proposed are very hard to realize, especially because virtually no fractionation is observed in Kr and Ar. The assumption of a completely different origin for terrestrial Xe might be required to explain the Xe isotope anomalies, with AVCC Xe and terrestrial Xe originating from different nucleosyntheses.

The noble gases extracted from lunar rocks 10017 and 10071 are essentially mixtures of pure spallation noble gases and radiogenic He^4 and Ar^{40}. Compared to the fine material the trapped gas content is very low. The abundance patterns of the spallation noble gases are similar to the well known spallation yield spectra observed in meteorites. The K/Ar ages and Kr^{81}/Kr exposure ages (*13*) are given in Table 2.

The K/Ar ages of the two feldspar fractions are distinctly higher than the age of the whole rock, reflecting the limited significance of total rock K/Ar ages. The feldspar fractions show He^3 diffusion loss and we cannot yet decide on the basis of this data alone, whether their K/Ar ages are significantly affected by prolonged diffusion losses or shock, or whether they represent the time of crystallization and rapid cooling.

The exposure ages are an order of magnitude lower than the K/Ar ages. Hence crystallization and ejection of the rocks must have occurred at different times. It cannot be decided yet whether the difference in exposure age of the two rocks reflects variable shielding conditions during exposure, or whether the two rocks have been ejected at different times.

From rock 10071 we calculate the following spallation production rates per million years in units of 10^{-12}cm^3 STP/g: $He^3 = 7200$; $Ne^{21} = 1000$; $Ar^{38} = 1300$; $Kr^{83} = 2.7$; and $Xe^{126} = 0.44$.

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