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On the question of the ^{40}Ar excess in lunar soils

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The retrapping mechanism proposed to explain the excess ^{40}Ar in lunar soil samples is discussed. Questions arise from the following experimental facts. Gas release in linear heating experiments is similar for excess ^{40}Ar and for solar wind ^{36}Ar, the latter having much higher implantation energy. In feldspar and olivine-pyroxene separates from a lunar soil the ^{40}Ar/^{36}Ar ratio of surface correlated argon is different. In feldspar grain size separates, potassium and ^{36}Ar appear linearly correlated. Possible influences of the Moon's ancient magnetic properties for the efficiency of the retrapping mechanism are discussed. Arguments supporting the notion of a second process, the transient K coating, are presented.

The problem remains unsettled and further studies are needed.

1. Introduction

The covariance of the noble gas concentration with grain size in lunar soils led to the conclusion that a large fraction of the noble gases originates from the solar wind and resides close to the surface of the individual grains. In lunar soils the ratio of ^{40}Ar to ^{36}Ar is around unity, in the incoming solar wind, however, about 10^{-4} (Cameron 1968). Therefore, lunar soils are said to contain parentless, orphan or excess ^{40}Ar. The excess argon was explained by retrapping from the lunar atmosphere (Eberhardt *et al.* 1970; Heymann & Yaniv 1970; Manka & Michel 1970). Gravitationally bound ^{40}Ar atoms in the lunar atmosphere become photoionized and are then accelerated by the solar wind $\boldsymbol{v} \times \boldsymbol{B}$-field. About half of the ions are lost into space, and the other half are driven back onto the lunar surface. Only a few percent of the ions driven back have acquired enough energy to become trapped in the surface matter of the Moon. The rest is recycled into the atmosphere. The small fraction of the atmospheric ^{40}Ar which is trapped is the source of the excess ^{40}Ar.

In this communication we report some experimental findings and present considerations which not only cast some doubt on the retrapping mechanism as being the sole source of the ^{40}Ar but also have some far reaching implications.

2. Experimental incongruities

(*a*) *Energy of implantation and gas release patterns*

The scale height of the lunar argon atmosphere on the day side is about 50 km and the acceleration due to the $\boldsymbol{v} \times \boldsymbol{B}$-field is on the order of volts per kilometre. Therefore, only ions produced at high altitude and low magnetic latitude have trajectories long enough to acquire the energies needed for retrapping. ^{40}Ar can only reach energies above 2000 eV if the solar wind velocity exceeds 400 km s^{-1} or if the magnetic field is stronger than 10 nT (Manka & Michel 1971). On the average the energy of implantation of ^{40}Ar ions originating in the lunar atmosphere is two orders of magnitude lower than that of the solar wind ^{36}Ar.

Baur *et al.* (1972, 1973) expected that the substantial difference in implantation energies of the two argon isotopes should be reflected by different gas retentivities of ^{40}Ar and ^{36}Ar. This can be experimentally evaluated by determining the gas release rates from samples subjected to temperatures increasing linearly with time. The similarity of such gas release patterns led Baur *et al.* (1972) to question the proposed retrapping mechanism as the major source of excess ^{40}Ar.

Frick *et al.* (1973) pursued the question of a correlation between gas release pattern and energy of implantation. To account for the complex host material with its structurally and chemically interactive surface, they implanted argon with energies of 2 keV into a lunar soil glass sample. The artificially implanted ^{40}Ar was indeed released at much lower temperatures than the natural ^{36}Ar. Thermal treatment of their sample prior to the analysis changed the gas release pattern of ^{40}Ar towards that of ^{36}Ar. However, the thermal treatment led to a loss of about 95 % of the artificially implanted ^{40}Ar whereas the natural ^{36}Ar content was practically unaltered. Thus, these experiments indicate that the energy of implantation may correlate with the release patterns and that thermal influences not only affect release patterns but also cause gas loss. With respect to the origin of the ^{40}Ar excess, the results suggest that the yield of trapping and especially retention of ^{40}Ar ions from the lunar atmosphere could be so low that the retrapping mechanism is not able to account for the excess ^{40}Ar.

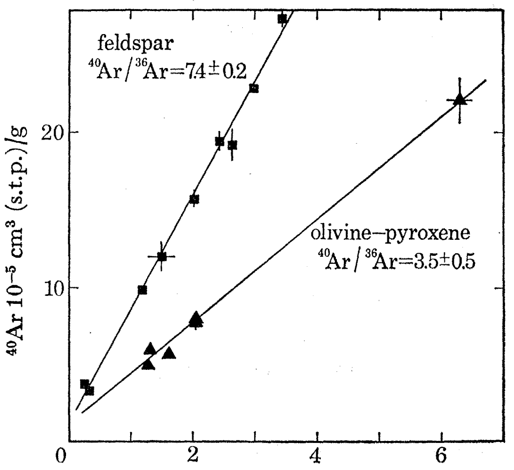

FIGURE 1

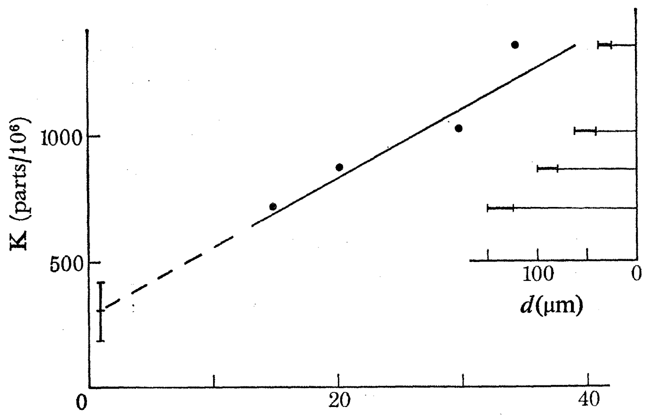

FIGURE 2

FIGURE 1. Isotope correlation of ^{40}Ar against ^{36}Ar for feldspar and olivine-pyroxene separates from the soil 15421. Note the different ^{40}Ar/^{36}Ar ratio of the surface correlated argon. (Data from Frick *et al.* 1975.)

FIGURE 2. Correlation of K against ^{36}Ar in the feldspar separate from soil 15421. (K data from Palme & Wänke, private communications.)

(*b*) ^{40}*Ar*/^{36}*Ar in soil constituents*

Frick *et al.* (1975) investigated grain-size separates of feldspar, olivine-pyroxene and green glass from the soil 15421. The ^{40}Ar-^{36}Ar correlation obtained for feldspar and olivine-pyroxene is shown in figure 1. The difference in the deduced ^{40}Ar/^{36}Ar ratio of the surface-correlated argon is remarkable. Signer *et al.* (1976) argued that this difference is unlikely to be the result of different exposure times of the two minerals and that processes other than retrapping from the lunar surface contribute to the ^{40}Ar excess.

(*c*) *K-^{36}Ar correlation*

On aliquots of some of the feldspar grain size separates shown in figure 1, Palme & Wänke determined the concentration of K and other trace elements. In figure 2 one notes a linear correlation between K concentration and that of ^{36}Ar. This finding concurs with a similar investigation by Basford (1974) and Evensen *et al.* (1974). The covariance of K and other trace elements with grain size could result from surface coatings of compounds originating from the lunar atmosphere or from material mobilized by impacts. K in feldspar grains and to some extent in soils could thus be considered as consisting of a volume correlated component and a surface correlated component.

3. DISCUSSION

(*a*) *Frame of references*

A re-evaluation of the origin of the excess ^{40}Ar in lunar soils and soil clods has to take into account the following evidences and considerations:

(i) Essentially all soils and soil clods contain comparable amounts of ^{40}Ar supposedly retrapped from the lunar atmosphere and ^{36}Ar trapped from the solar wind and, possibly from the solar flares (Frick *et al.* 1975).

(ii) Practically no evidence for the different energies of implantation of the two argon species is seen in the gas release patterns (§2*a*).

(iii) In at least one soil, two different minerals contain different ratios of trapped ^{40}Ar/^{36}Ar with roughly similar argon concentrations, which rules out effects due to differences in histories and/or of diffusive losses (§2*b*).

(iv) Not only noble gases and other constituents (such as N and C) originating from the solar wind but also K and other elements of non-solar origin seem to correlate with grain size (§2*c*).

(v) Lunar samples show remanent magnetization, which may indicate the existence of considerable magnetic fields during the formation of these rocks (cf. Fuller 1974).

(vi) According to Hoffman *et al.* (1973), the ratio of ^{40}Ar to ^{36}Ar in todays lunar atmosphere is around 10.

(*b*) *Efficiency of retrapping and retention*

The similarity of the release patterns of the two argon isotopes indicates that a major redistribution of at least the low energy implanted ^{40}Ar must have taken place. Hodges *et al.* (1974) explain the high ^{36}Ar concentration in the present lunar atmosphere by solar wind saturation of grains on the lunar surface. If this is the case, ^{40}Ar must also be saturated and a large fraction of the initially retrapped ^{40}Ar would have been lost. In fact the loss of ^{40}Ar should be much more severe than the ^{36}Ar loss. According to the theoretical considerations of diffusion processes by Huneke (1974), the equilibrium between implantation and diffusive loss is connected to fractionation of two isotopes according to the inverse ratio of their respective penetration depths (or implantation energies). Even though it is not clear what processes cause the similarity of the gas release pattern, it appears inevitable that much larger losses of ^{40}Ar than ^{36}Ar must have occurred. If the postulate of large ^{40}Ar losses holds – which is equivalent to a comparatively low retentivity of ^{40}Ar – the estimated yields of atmospheric retrapping are invalid. It is then questionable whether the retrapping mechanism can really account for all ^{40}Ar retained today in the regolith.

In this context it is of interest to note that on the basis of the density of todays lunar argon atmosphere, solar wind velocity, field conditions and trapping efficiences, Geiss (1974) expected the ^{40}Ar/^{36}Ar ratio of trapped argon to be less than 0.2. On the other hand, Hodges *et al.* (1974) conclude that this ratio should be about 6, but their conclusion neglects the fact that only a small fraction of the ions driven back at the lunar surface is really trapped.

(*c*) *Retrapping in ancient times and the history of the lunar magnetic field*

Yaniv & Heymann (1972) suggested that the ^{40}Ar/^{36}Ar ratio of the surface correlated argon is also correlated to the age of the regolith at the individual collection sites. This, they argued, is the consequence of the expected decrease in density of the argon atmosphere with time. This postulate has important implications.

If soil samples have indeed kept a memory of the lunar atmosphere in ancient times, by means of the retrapped ^{40}Ar then the Moon could not have had a global magnetic field much stronger than a few hundred nT. A global lunar magnetic field strong enough to cause a standoff of the solar magnetic field by a few hundred kilometres above the lunar surface would severely affect the efficiency of the acceleration mechanism of photoions by the $\boldsymbol{v} \times \boldsymbol{B}$-field. To reconcile this with the omni-presence of excess ^{40}Ar in all returned soil samples and soil clods, one must assume that either

(i) all excess ^{40}Ar detected was retrapped after decay of the global lunar magnetic field, or

(ii) some of the ^{40}Ar excess dates back as much as 3–4 Ga, and the moon never had a global magnetic field strong enough to cause a solar wind standoff, or

(iii) not all of the excess ^{40}Ar is due to the $\boldsymbol{v} \times \boldsymbol{B}$ retrapping mechanism.

The omni-presence of the solar gases and their comparatively constant element abundance ratios in mineralogically similar soils and soil clods do not preclude an ancient global magnetic field of considerable strength. Due to the magnetic rigidity of the solar wind ions they still could penetrate to the lunar surface. Furthermore, the fractionation of solar wind ions in a magnetic field is small because of similar velocities of all species and the high state of ionization.

(*d*) *Other causes for ^{40}Ar excess?*

Baur *et al.* (1972), Frick *et al.* (1973) and Baur *et al.* (1973) have suggested that part of the excess ^{40}Ar in lunar soils could be due to K which was present at the grain surface some time during its history. This postulate was met with criticism (Yaniv & Heymann 1972). However, several new observations can be taken as supporting evidence for the transient K hypothesis:

(i) In lunar soils, potassium (Barnes *et al.* 1973; Garner *et al.* 1975) and other elements are enriched in heavy isotopes (cf. Clayton *et al.* 1974).

(ii) K enrichment on the surface of glass spheres has been detected by Podosek & Huneke (1973) and by Stettler.

(iii) Indication for radiogenic ^{87}Sr excess in a mature soil was found by Nyquist.

(iv) K, Rb, Ba and other elements are relatively enriched not only in the small grains of bulk soils (Evensen *et al.* 1974), but also in small grains of the feldspar separates (Basford 1974; Palme & Wänke).

The first observation clearly indicates that transport processes causing isotopic fractionation occur on the lunar surface. Observations (ii) and (iii) point to a parallelism between excess amounts of the decay product (i.e. 'parentless' components) of the K-Ar and Rb-Sr systems, systems with volatile parents. The K enrichment near the surface and in the smaller grains supports the transient K hypothesis. The grain-size correlation of other volatile and non-volatile elements (e.g. Rb, Ba) indicates, however, that the K coating is not solely due to the vapour deposition of volatile components. Besides possible vapour deposition, the grain surfaces appear also to be contaminated with matter rich in K and non-volatile elements. Such matter could be produced by small impacts on low melting materials.

Concerning ionization and subsequent removal of K (and Rb) from the Moon, the following processes which favour removal of alkali metals may be considered:

The low ionization potential of alkalis causes a high yield for thermo-ionization of these elements from metal surfaces, but little is known about thermo-ionization from radiation damaged silicate surfaces. Another process for efficient ionization of alkalis is photo-ionization. Because of the low ionization potential of K, the number of solar photons capable of alkali ionization is some 10^5 times higher than that of Ar ionization. The first atomic monolayer of surface matter with a few parts per 10^3 of K contains about 10^3 more K atoms than the lunar atmosphere contains Ar atoms per unit area. The probability for ions to leave the surface layer is difficult to estimate. However, once an ion has escaped the grain surface, it may become accelerated in the surface-near electric field. Local magnetic fields may still be able to retain such ions bound to the Moon; but trajectories of the order of several hundred kilometres carry ions sufficiently high to expose them to the $\boldsymbol{v} \times \boldsymbol{B}$-field. This accelerates half of the ions into space.

Experimental verification of the ionization processes in the laboratory seems extremely difficult, particularly, because the pertinent surface properties of lunar matter available on Earth are severely altered by their exposure to the terrestrial environment. Of interest is, however, the comparison of observations by the suprathermal ion detector experiment (s.i.d.e.), which is basically a lunar surface ion detector, and the lunar atmosphere composition experiment (l.a.c.e.) measuring concentrations of neutral gases in the lunar atmosphere. Vondrak *et al.* (1974) note: 'The differences in the l.a.c.e. and s.i.d.e. values (for the lunar loss rates) can be reconciled if there exists a component(s) of the lunar atmosphere in addition to the observed noble gases'. Thus, one may speculate that alkali ions could be part of such an 'atmospheric component' not detected by the l.a.c.e.

It remains to be noted that the Ar data obtained by Frick *et al.* (1975) on the 15421 feldspar separates and the K determinations by Palme & Wänke (figure 1) can be used to evaluate the surface concentrations of K required to produce the observed ^{40}Ar excess. Signer *et al.* (1976) conclude that with a depth of the ^{40}Ar and K bearing layer of 10 μm thickness, K concentrations around 1 % are required.

An inevitable consequence of the postulate that some of the excess ^{40}Ar is produced by a transient K coating is the requirement of a higher diffusion constant for K than for ^{40}Ar. In terrestrial K/Ar dating, especially of feldspars, just the opposite has been observed. Thus, if transient K is a viable source for some of the excess ^{40}Ar, the apparent inversion of the diffusion behaviour of K and Ar has to be the result of peculiar conditions on the Moon causing severe alterations of the surface layers of the host and loss of K from surface layers by ionization. With respect to the influence of ancient magnetic fields to the source mechanism of the ^{40}Ar excess we noted in §3*c* that the retrapping mechanism is expected to be severely impared. Similarly, loss of K into space would also be reduced by whatever fraction the $\boldsymbol{v} \times \boldsymbol{B}$-field would contribute without lunar magnetic field. This reduction increases the residence time of the transient K and eases the level of K required to account for the ^{40}Ar excess.

4. Conclusion

The above considerations indicate that several experimental observations concerning the ^{40}Ar/^{36}Ar ratio in lunar soils apparently lead to incongruities with the postulate that the ^{40}Ar-excess is due to a retrapping of ^{40}Ar from the lunar atmosphere. Although none of these observations speaks firmly and conclusively against retrapping as the sole source of the surface correlated ^{40}Ar, the sum of all considerations seems to imply that one or more additional sources are needed to explain the observed concentrations. No final ruling is possible, but, in view of the implications of the ^{40}Ar excess to questions such as the lunar regolith evolution, the past of the solar wind, the thermal history of the Moon and the ancient magnetic field of the Moon, a further experimental pursuit of the ^{40}Ar excess seems an important domain of noble gas investigations of lunar soils.

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