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Accumulation and circulation of gaseous radon between lunar fines

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During the lunar night, the temperature of the regolith upper layer is lower than the radon freezing point. Thus radon atoms coming from the interior can be trapped at the surface of the cold lunar fines. The ^{222}Rn daughter products, ^{210}Pb and ^{210}Po, are embedded in a very thin layer at the surface of the grains.

It is therefore possible, by spectrometry, to distinguish between the continuum due to uranium, thorium (and decay products) homogeneously distributed and the narrow peak at 5.3 MeV, due to an excess of ^{210}Po. We have determined a mean day-and-night concentration of about 3.5×10^3 atoms of intergranular ^{222}Rn per gram of superficial fines, corresponding to a continuous flow of 3 atoms min^{-1} cm^{-2} of soil. To account for such a flow of radon atoms moving in a random way from a 6 m source depth, the pore size of the regolith should be 60 μm. On the other hand, the involved changes in the isotopic composition of the radiogenic lead remain less than 1 %.

The study of gaseous ^{222}Rn circulation in the regolith may be a good approach of soil–atmosphere noble gas exchanges in the Moon. Moreover, this study is interesting in itself since ^{222}Rn is an intermediate nuclide between ^{238}U and ^{206}Pb: the mobility of the gaseous Rn has set the problem of the validity of U–Pb datings. Particularly the accumulation of Rn decay products at the surface of the Moon has been considered by several authors and might give there an excess of radiogenic ^{206}Pb.

1. Evidence for a gaseous Rn circulation in the regolith

During the lunar night, the soil temperature drops to about 100 K in a 1.5 cm deep upper layer (figure 1). This temperature being lower than radon freezing point (163 K), the gaseous atoms are adsorbed at the surface of the cold grains. In this layer, the ^{222}Rn density increases throughout the night approaching 6 % of the secular equilibrium value. At sunrise, the Rn frost is immediately re-evaporated and its concentration drops to zero at high day time temperatures.

Throughout the lunar night, the α-disintegrations of those ^{222}Rn atoms trapped at the surface of the grains give rise to recoiling atoms of ^{218}Po and afterwards ^{210}Pb and ^{210}Po in radioactive equilibrium. All of which are embedded in a very thin layer about 20 nm deep on the surface of the grains. Since the ^{210}Pb half-life is about 20 years, even a long time after the lunar samples have been brought back onto Earth, each grain is covered by a superficial layer of ^{210}Pb, which is seen by an α-spectrometer as a quite thin α-source.

Figure 2 shows α-spectra directly obtained by Lambert *et al.* (1975) without any treatment on fines (14163,92 and 15021,110). One can distinguish the continuum due to the thick source

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of ^{238}U, ^{226}Ra, ^{222}Rn and their daughter products, homogeneously distributed within the grains, and a supplementary narrow peak of ^{210}Po just at 5.3 MeV. This peak is hardly visible on the spectrum of an Apollo 14 sample rich in uranium but very clear in a low uranium concentration sample. This peak is also observable on the bottom face of the rock 64475,16 which was, before sampling, in direct contact with the lunar fines.

Taking into account the dates of sampling and measuring, and the 4.6 month half-life of ^{210}Po, this superficial polonium activity cannot be attributed to a direct contamination of the grains surface. Therefore, the activity is obviously due to ^{210}Pb in radioactive equilibrium and consequently to an equal mean activity of intergranular ^{222}Rn which is of the order of 2×10^{-5} disintegration s^{-1} cm^{-2} of sample, i.e. about 10 atoms/cm^2.

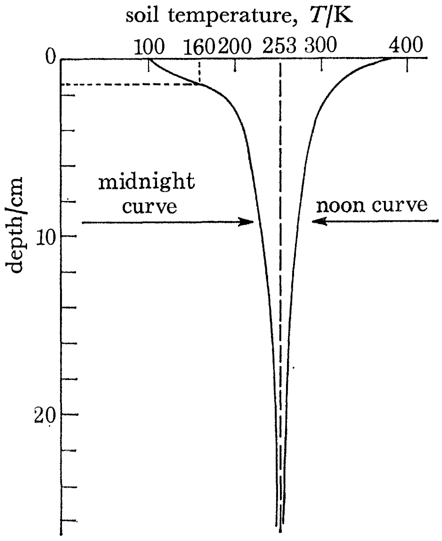

FIGURE 1. Vertical distribution of temperature in lunar regolith.

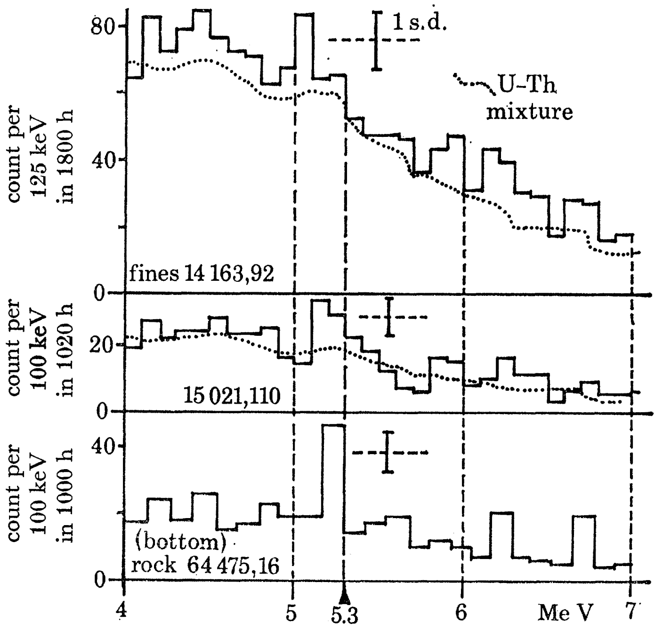

FIGURE 2. Alpha spectra of lunar samples.

The surface of sample to be considered is not the specific surface of the grains, where all possible traps are far from being saturated. In fact, the probability for a grain to catch a recoiling atom of ^{218}Po is proportional to its geometric cross-section. Thus the surface of a grain to be considered is its mean smooth rounded surface (m.s.r.s.). According to the size distributions given by McKay *et al.* (1974) the m.s.r.s. varies from 425 cm^2/g for a mature soil to 325 cm^2/g for an immature one.

Therefore, the concentration of gaseous Rn between the grains in the upper layer is about $\bar{\rho} = 3.5 \times 10^3$ atoms/g of fines. This figure is the mean value averaged over a large number of revolutions of the Moon. A relative density of the order of 1.4 (Houston *et al.* 1974) gives $\bar{\rho} = 5 \times 10^3$ atoms/cm^3. Such a concentration of intergranular radon cannot be accounted for by emanation from the grains themselves.

In fact, lunar fines contain generally between 0.5 and 4 parts/10^6 of uranium, more or less in radioactive equilibrium with all its daughter products. Thus, assuming an average value of 1 part/10^6, we have about 5×10^3 atoms of ^{222}Rn per gram of superficial fines. But most of these atoms remain inside the grains: only a small proportion is able to reach the vacuum space between grains. Table 1 shows the usual values of the emanation-to-production ratios (e.p.r.) found for lunar fines by those authors working in *simulated lunar conditions*. The 5×10^{-3} value characteristic of the ratios in table 1 means that we have about 25 atoms of Rn per gram in a gaseous state between the fines grains. Consequently, in the upper layer, most of the intergranular Rn atoms must come from the deep layers of the regolith.

Taking into account the half-life of ^{222}Rn and the duration of the lunar night, a vertical flow of 3 atoms of radon $\text{min}^{-1}\ \text{cm}^{-2}$ has to reach the upper layer of the soil continuously.

TABLE 1. EMANATION TO PRODUCTION RATIO (e.p.r.)

authors	e.p.r.
Stoenner, R. W. *et al.* (1974)	0.09×10^{-3}–2.7×10^{-3}
Yaniv, A. & Heymann, D. (1972)	2×10^{-3}– 5×10^{-3}
Lambert, G. *et al.* (1973)	1×10^{-3}
Adams, J. A. S. *et al.* (1973)	1.5×10^{-3}–9.5×10^{-3}

2. Rn CIRCULATION IN THE REGOLITH

In lunar conditions of vacuum, we can admit that Rn atoms, in thermal equilibrium with their environment, are moving in a random way between the fines by bouncing from a grain to the next one.

If δ is the mean free path and τ the mean time between two successive impingements, the time t to pass through the thickness $Z = n(\delta/\sqrt{3})$ is $t = n^2\tau$, so

$$t = 3(Z/\delta)^2.$$

According to Friesen & Heymann (1972), the time τ is the sum of two elements: the flight time t_f and the desorption time t_d:

$$\tau = t_f + t_d.$$

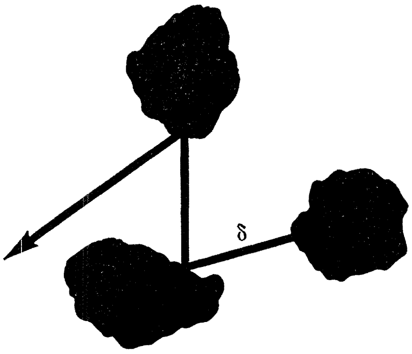

FIGURE 3. Random movement in lunar regolith; $t_f = \delta\sqrt{(m/3\,kT)}$; $t_d = t_0\,e^{Q/RT}$.

Flight time

The first term $t_f = \delta\sqrt{(m/3kT)}$ is the flight time across a mean free path δ for an atom of mass m at temperature T.

In the case of ^{222}Rn we have:

$$t_f \simeq 9.4 \times 10^{-8}(\delta(\mu\text{m})/\sqrt{(T(\text{K}))}).$$

(second)

At the lunar surface T is between 100 and 400 K. Taking into account the size distribution of the grains, δ could not be out of the range 2–50 μm; so $t_{\rm f}$ is between 1×10^{-8} and 50×10^{-8} s.

Desorption time

The second term $t_{\rm d}$ is the desorption time and is given by de Boer (1968),

$$t_{\rm d} = t_0 \exp\,(Q/RT).$$

In this experience, the characteristic time t_0 is the period of vibration of the adsorbed atom and depends only on soil characteristics, by the relation:

$$t_0 = 4.75\times10^{13}\sqrt{\{(MV)^{\frac{2}{3}}/T_{\rm m}\}}$$

where M is the mean molecular mass, V the molar volume, and $T_{\rm m}$ the melting temperature. de Boer (1953) gives for Al_2O_3, $t_0 = 0.75\times10^{-13}$ s, SiO_2, $t_0 = 0.95\times10^{-13}$ s, MgO, $t_0 = 0.67\times10^{-13}$ s.

On the other hand, using the constants M and V, calculated from Morrison's (1971) compilation, for 12070, and the melting temperature taken from Green *et al.* (1971), Stoenner *et al.* (1972) give for t_0 1.1 and 1.2×10^{-13} s and use as a fair average $t_0 = 0.9\times10^{-13}$ s.

The heat of adsorption Q of radon on charcoal has been measured to be 31.4 kJ (7.51 kcal)/mol in the air (Bübeli & Söri 1954) and estimated to be 56.5 ± 5.8 kK (13.5 ± 1.4 kcal) at low surface coverage in vacuum (Chackett & Tuck 1957). In view of the low surface energy observed by Fuller *et al.* (1971) in adsorption of N_2 and CO on soil sample 10087, however, Q in lunar soil may not be so high. But the lower limit is the heat of vaporization of liquid Rn, which is 18 kJ (4.3 kcal), and therefore Stoenner *et al.* (1972) use finally a mean value $Q = 42 \pm 12.5$ kJ (10 ± 3 kcal).

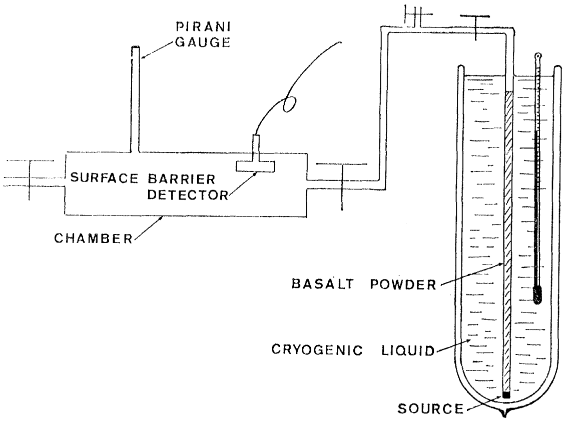

FIGURE 4. Device for the measurement of the heat of desorption.

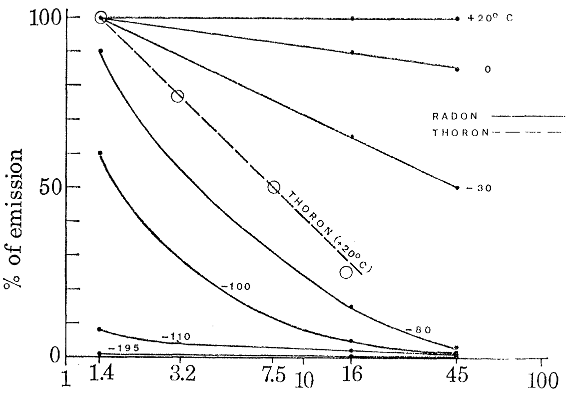

FIGURE 5. Depth decrease of radon.

The temperature of the regolith being of the order of 253 K, the desorption time of radon should be then comprised between two very different figures of 1×10^{-7} and 1.5×10^{-2}. However, all of these figures are not compatible with simulation experiments.

Lambert *et al.* (1973) have simulated lunar fines by a mixture of powdered basalt with a grain size less than 50 μm. The source of Rn has been made with an U–Th ore. The experiments have been performed in a good primary vacuum at different temperatures (figure 4).

The measures have been made by α-spectrometry to distinguish between the isotopes 220 and 222 of radon.

Figure 5 shows the transmission power at different temperatures. It may be seen that ^{222}Rn atoms are able to pass through 45 cm in 1 radioactive half-life of 3.8 days at $T = -30$ °C. Now Q is a logarithmic function of the mean free path. Thus acceptable values of δ comprised between 2 and 30 µm enable to calculate: $Q = 39.7 \pm 5$ kJ (9.5 ± 1.2 kcal). At the mean temperature of the regolith, the desorption time of radon is therefore comprised between 1×10^{-4} and 1×10^{-6} s and consequently, except for the high day time temperatures, the flight time is completely negligible and the time necessary to pass through the thickness Z becomes:

$$t(Z) \approx 3(Z/\delta)^2 t_0 \exp(Q/RT),$$

with the preceding values of t_0, Q and T, Le Roulley (1975) finds

$$t(Z) \approx 4.3 \times 10^{-5}(Z/\delta)^2.$$

Source depth, for radon reaching the upper layer

Whatever figures are not acceptable for Z and δ. We have to take into account a production rate of intergranular gaseous radon of the order of 25 atoms/cm^3. Therefore, the 5000 atoms/cm^3 of the upper layer could be accounted for by a 2 m depth source with a 100 % efficiency. But for the deepest layers and a reasonable value $\delta = 3$ µm, the transit time necessary should be of the order of 200 days, which is impossible owing to the 3.8 days half life of ^{222}Rn.

A much more likely source depth of 3×2 m should be more acceptable provided that the corresponding transit time was of the order of 4 days. This corresponds to a mean free

$$\delta = Z\sqrt{\{(4.3 \times 10^{-5})/t\}} \simeq 60 \text{ µm}.$$

3. Conclusion

The striking feature is that the measured regolith pore size is found to be much larger than the value inferred from the number median diameter (about 3 µm, Görz *et al.* 1971). We must conclude that the finer grains are not isotropically distributed among the bigger ones. Following V. Troitsky (1967), we do think that the pores are not closed but consist of gaps between sintered grains. Such a model agrees with the very low thermal conductivities measured by Dul'nev *et al.* (1966).

On the other hand, the change produced in the isotopic composition of the radiogenic lead seems to be rather small. First, the 0.5 % losses due to Rn emanation are generally negligible. Next, the accumulation of exogenic ^{206}Pb in the upper layer is limited by the turnover process. According to Arnold (1975), a residence time of 10 million years seems to be reasonable in the 1.5 cm layer. Thus the amount of ^{206}Pb due to the intergranular Rn should be only 0.1 % of the total radiogenic ^{206}Pb.

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