EMANATION FROM LUNAR SOIL

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ABSTRACT The emanation characteristics of eight bulk lunar fines samples with 0.5 to 10.0 grams plus one small sample of *Kreep* glass were analysed by alpha cintillation counting techniques. The objective was to investigate the problem of radon mobility on the lunar surface and its possible contribution to the lead isotopic fractionation and redistribution, particularly its contribution to the anomalous U^{238}/Pb^{206} isotopic ratios.

The samples were de-emanated at room temperature and a pressure of one atmosphere. An uniformily low radon escape rates and losses were observed in the analysed material. Three terrestrial analogues (obsidian, tektite and W-1) were analysed for comparison purposes. The observed data suggest that very little Rn^{222} is escaping from the soil at the lunar surface and therefore it has a small direct contribution to the excess of isotopic lead.

RESUMO As propriedades emanométricas de oito amostras, variando de 0,5 a 10 g, do solo lunar e uma amostra de vidro *Kreep* foram analisadas com auxílio de técnicas não destrutíveis de cintilações alfa. Esse estudo teve por objetivo investigar a possibilidade de transferência de radônio de um local para outro da superfície da Lua, o que poderia contribuir na distribuição isotópica do chumbo, particularmente nos casos de razões isotópicas U^{238}/Pb^{206} anômalas, já descritas nos primeiros trabalhos de geocronologia da Lua.

As amostras foram de-emanadas a temperatura ambiente e a uma atmosfera de pressão. Uma uniforme e pequena perda de ^{222}Rn foi observada essencialmente de 0 a 0,68% e uma razão de escape 0,04 a 0,2 átomos/h/g. Três amostras de material terrestre de composição mineralógica e concentrações de U e Th análogas (vidro vulcânico, tectitos, W-1) foram analisadas a fim de se comparar os resultados, obtendo-se valores de escape de radônio mais elevado. Os dados obtidos sugerem que muito pouco ^{222}Rn escapa do solo na superfície da Lua e portanto deve-se esperar que ^{222}Rn tenha uma muito pequena contribuição ao excesso do chumbo isotópico observado.

INTRODUCTION The low bulk radon emanation rates, reported in all but one case for the lunar returned samples of the Apollo program the reports of excess ^{210}Po and the parentless lead in dust, do not fit neatly yet into a coherent model. Because it is important to establish, what causal relationships, if any, there are, between radon emanation and venting; and between radon emanation and parentless lead, this study of the bulk radon emanation from returned fines was undertaken.

The objective was to investigate the possibility of radon transfer at the lunar surface which might contribute to the lead isotopic distribution in particular to the "parentless lead" described in earlier studies to lunar age dating (Silver, 1970; Tatsumoto, 1970; Tatsumoto *et al.*, 1971) and at the same time, to compare the lunar results with those from analogous terrestrial materials.

Evidence for lunar isotopic transfer was presented by Tatsumoto *et al.* (1972); systematic enrichments in old leads in the fines compared to local rocks are reported by Silver (1972a) and volatile transfer as a major geochemical process on the lunar surface is discussed by Silver (1972b) with respect to lead.

Earlier, Kramer *et al.* (1966) suggested, based on several physical assumptions, that radon and thoron might be present in the lunar surface and atmosphere in detectable quantities. Using data collected by Explorer 35, Yeh and Van Allen (1969) calculate an

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particle emissivity for the Moon and the lunar surface radioactive concentration ten times less than Kramer's theoretical estimate.

The only indications of ^{222}Rn migration observed on the lunar surface are the indirect detection of an excess of radon daughters by the spectrometer on Surveyor V at Mare Tranquillitatis (Turkevich *et al.*, 1970) and direct detection (although marginally) by the Apollo-15 orbiting spectrometer (Gorenstein and Bjorkholm, 1972 and 1973). The later report on value for the activity of ^{222}Rn at most 10^{-3} dis/cm^2-sec, a factor of 9 smaller than the quantity of radon that would be in equilibrium with the observed amount of ^{210}Pb ($4.6 \pm 1.4 \times 10^{-3}$ dis/cm^2-sec) for an area west of Mare Crisium to the Van Der Graaf-Orlov region. This amount is approximately one-nineth of the value reported by Turkevich and ten times higher than the upper limit reported by Lindstron *et al*, (1971) for an Apollo-11 rock sample. Gorenstein and Bjorkholm concluded that their data implies that higher levels of radon emanation have occurred on the moon in the last 10-100 years.

The working hypothesis for the present investigation is that, although on a much smaller scale, consequences similar to lead diffusion or volatilization (separation of the stable radiogenic lead from the site of parent uranium and thorium) can be achieved by cumulative radon migration.

As demonstrated for the zircon and sphenes concentrates (Barretto, 1972) samples which exhibit more lead loss (discordant) correspond to those with higher radon loss. Thus, if the moon is a closed system for some of the radiogenic lead isotopes and radon, depletion from one site would correspond to some isotopic enrichment at another.

RESULTS Eight bulk lunar soil samples, weigthing 1 to 10 grams, plus a small sample of Kreep glass, were analysed for ^{222}Rn emanation using an α-scintillation counting technique. Samples were analysed as they arrived without any fractionation sieving or other treatment. They were analysed at room temperature and atmospheric pressure. To disturb as little as possible the emanation characteristics, a volume share technique

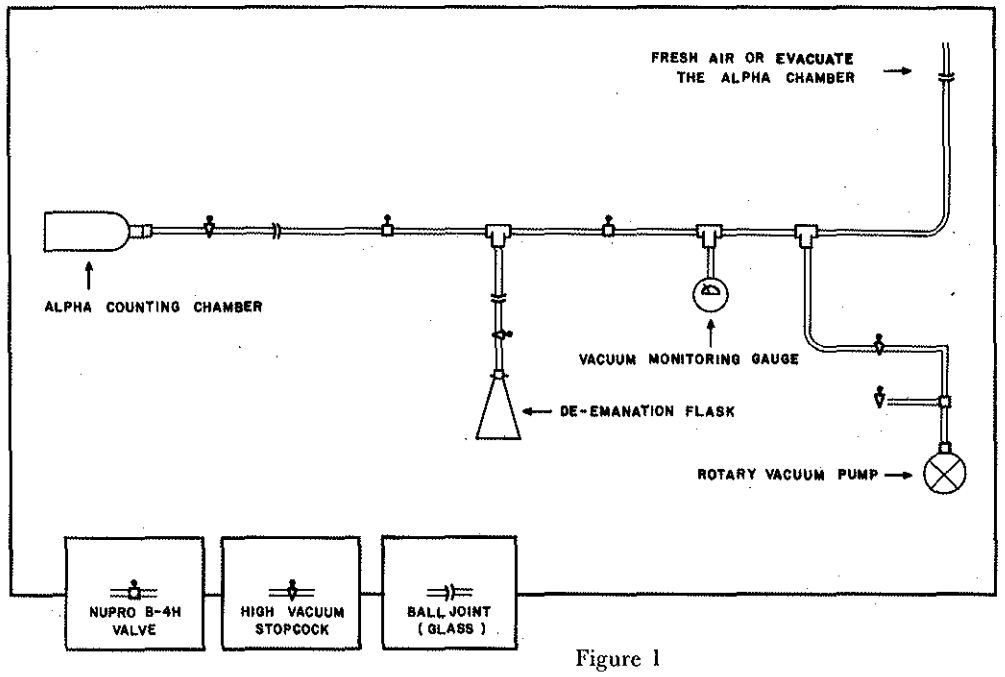

Figure 1

of gas transfer was used (Fig. 1). The volumes were such that, approximately 62% of the gas was transferred to the counter.

The results for the nine samples analysed are given in Table I. The samples were also de-emanated at room temperature and a pressure of one atmosphere. Background collection and sample counting was for periods longer than 15 hours. As Table I shows, a uniformly low radon losses were observed, from essentially zero to 0.68% assuming the uranium concentrations reported for the bulk samples of which the analysed material are aliquots, or when that was not available, the uranium value for the closest or average site material.

Table I — Radon-222 emanation from lunar soil

Sample	Weight gm.	eU (ppm)	Activity (DPH)	Escape rate (atoms/h/g)	Escape-to-production ratios (%)	pCi/h/g
15021,83 (# 2)	5.1	1.5	0.34×10^3	0.20 ± 0.12	0.31 ± 0.19	0.11×10^{-4}
12033,73 (# 3) Kreep	0.23	3.0	0.30×10^2	1.21 ± 0.83	0.95 ± 0.75	1.12×10^{-4}
68501,13 (# 2)	10.05	0.65	0.29×10^3	0.1 ± 0.03	0.38 ± 0.11	0.62×10^{-5}
10084,107 (# 2)	4.37	0.54	0.10×10^3	0.04 ± 0.04	0.15 ± 0.15	0.23×10^{-5}
10084,83 (# 3)	5.01	0.54	0.12×10^3	0.15 ± 0.05	0.62 ± 0.15	0.83×10^{-5}
12070,14,845 (# 3)	1.0	1.5	0.67×10^2	0.16 ± 0.11	0.25 ± 0.15	0.95×10^{-5}
10084,257 (# 2)	6.73	0.54	0.16×10^3	0.12 ± 0.04	0.51 ± 0.14	0.68×10^{-5}
10084,557 (# 2)	10.0	0.54	0.24×10^3	0.16 ± 0.04	0.65 ± 0.16	0.87×10^{-5}
64501,8 (# 1)	7.0	0.65	0.20×10^3	0.13 ± 0.05	0.47 ± 0.18	0.77×10^{-5}
w-1 Basalt (# 2)	5.0	0.52	1.11×10^3	2.10 ± 0.45	8.90 ± 1.60	1.10×10^{-4}
Vol. Glass (# 2)	16.5	9.5	0.69×10^4	19.0 ± 1.85	0.49 ± 0.09	0.12×10^{-3}
Tektite (# 2)	15.5	1.6	0.11×10^4	0.56 ± 0.08	0.77 ± 0.10	0.15×10^{-4}

Number of analysis. Volcanic glass < 60 > 115 mesh.
Tektite < 97 > 150. Sample 12033,73 = Kreep.

For comparative purposes the geochemical rock standard W-1, a volcanic glass and tektite are shown in the table, as they represent the terrestrial materials closest in composition and radioactive concentrations to the lunar rocks. W-1 was measured under precisely identical conditions including using the type of plastic vial in which the lunar materials were delivered. It shows 9% radon loss, and it should be noted that this pulverized rock probably does not have the large surface area of the bulk lunar dust. The emanation characteristics for the volcanic glass (obsidian from Lipari, Italy) and the tektite (Texas bediasites) are closer to that of the lunar soil. They present much lower percentage of radon loss.

The low emanation reported for the lunar material agrees with values for the activity and emanation rate observed by Stoenner *et al.* (1971) and the percentage of radon loss given by Yaniv and Heymann (1972). They do not agree, however, with a previous preliminary result obtained by our group (Adams *et al.*, 1971) from a 0.47 gr sample of Apollo-12 soil.

DISCUSSION Due to the low concentrations (0.04-4 ppm U and 0.1-13 ppm Th) and assuming an age of 4.5 b.y. for its formation, the natural radiation damage in the lunar material will be small, never approaching the dose which could affect the radon escape characteristics as demonstrated for the terrestrial materials (Barretto, 1972). It could be argued, however, that the particles on the top of the lunar soil surface will show considerably

greater damage than the earth's surface material because the latter were exposed to cosmic radiation, solar wind and solar flares effects for hundreds of millions of years. But according to the radiation damage mechanism proposed in our earlier studies of radon emanation, the effective types of damage that would cause radon mobility within the crystal are those occuring at the sites of the uranium and its daughter. Thus, the damage caused to the material exposed to various types of bombardment will no necessarily affect the radon emanation.

On the other hand, the low radioactive concentrations in the lunar dust and breccias seems to be distributed among all mineral phases with relatively few high uranium concentrations as in zircons. It was observed that terrestrial plagioclases and pyroxenes, less damage retentive minerals, are very low emanators. Moreover, zircon itself is a poor emanator (Barretto, 1972). Adding to these considerations the fact that chemical weathering is absent in the moon, it is apparent that with the lunar material we are dealing with different conditions.

The hypothesis that the mobility of radon in the moon is inhibited by implantation of the recoiled atom due to the lack of interstitial air or water does not apply to the present measurements as atmospheric air and ambient humidity were present during all analyses, and yet the radon escape observed was minimal.

However, the effects of hydration of the lunar soil in contact with earth atmosphere as an incipient weathering is not ruled out as one possibility to alter the emanation characteristics.

The results obtained in the analysis of Texas tektites and the Italian Lipari volcanic glass (Tables II and III) shed some light on the interpretation of these low emanation

Table II – Particle size influence on radon escape
Texas tektite (bediasite)

Particle size mesh	Weight gm	Concentrations* eU ppm	eTh ppm	Escape-to-production rate (%): Average
+ 23 (3)	14.6	1.4	5.7	0.120 ± 0.053
−38 + 48 (2)	11.4	1.5	5.2	0.228 ± 0.072
−74 + 97 (3)	22.4	1.8	4.5	0.386 ± 0.075
−97 + 150 (2)	15.5	1.6	5.9	0.774 ± 0.104

(3) Number of analyses of each fraction.
*Concentrations determined by γ-ray spectrometry.

Table III – Particle size influence on radon escape
Lipari volcanic glass

Particle size mesh	Weight gm	Concentrations* eU ppm	eTh ppm	Escape-to-production rate (%): Average
+ 10 (2)	46.5	8.3	37.0	0.052 ± 0.015
−10 + 20 (2)	25.4	10.5	35.4	0.087 ± 0.018
−20 + 60 (3)	56.3	7.8	40.2	0.16 ± 0.022
−60 + 115 (2)	16.5	7.5	37.2	0.48 ± 0.035
−115 + 250 (1)	6.7	8.2	34.0	0.66 ± 0.051

(2) Number of analyses of each fraction.
*Concentrations determined by γ-ray spectrometry.

rates. The tektites contain 1.6 ppm uranium and 5.7 ppm thorium and autoradiography studies of these bediasite (Balacek, 1966) indicated that the radioactivity is homogenously distributed through the sample and of a low flux. The Lipari glass with 8.5 ppm uranium and 37 ppm thorium is also believed to have this concentration uniformily distributed. Here again we do not have natural dosage large enough to produce significant damage and both the tektite and the glass have low percentage of escape. The observed radon loss for different size fractions are shown in Figs. 2 and 3.

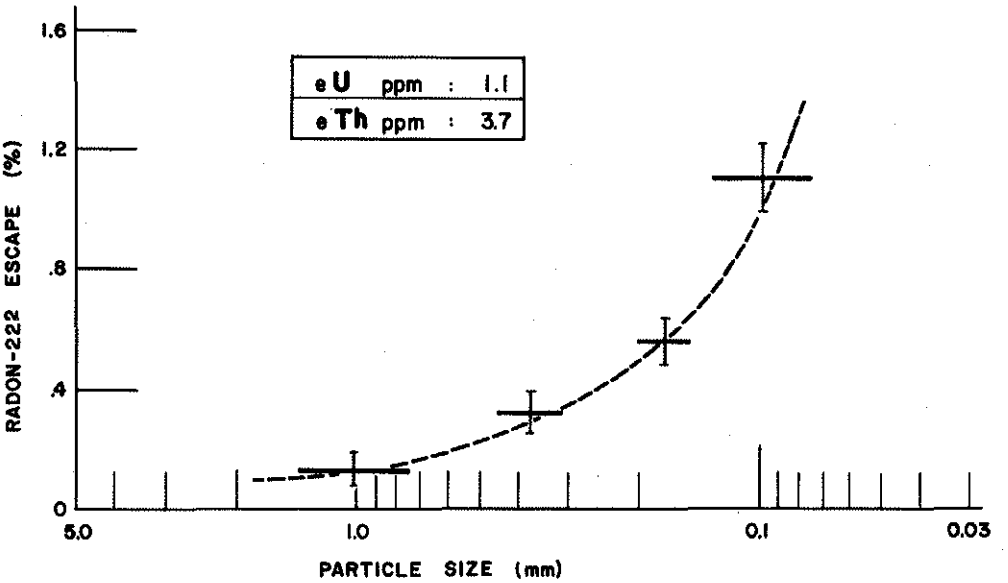

Figure 2 — Particle size influence on radon escape, Texas tektite (bediasite)

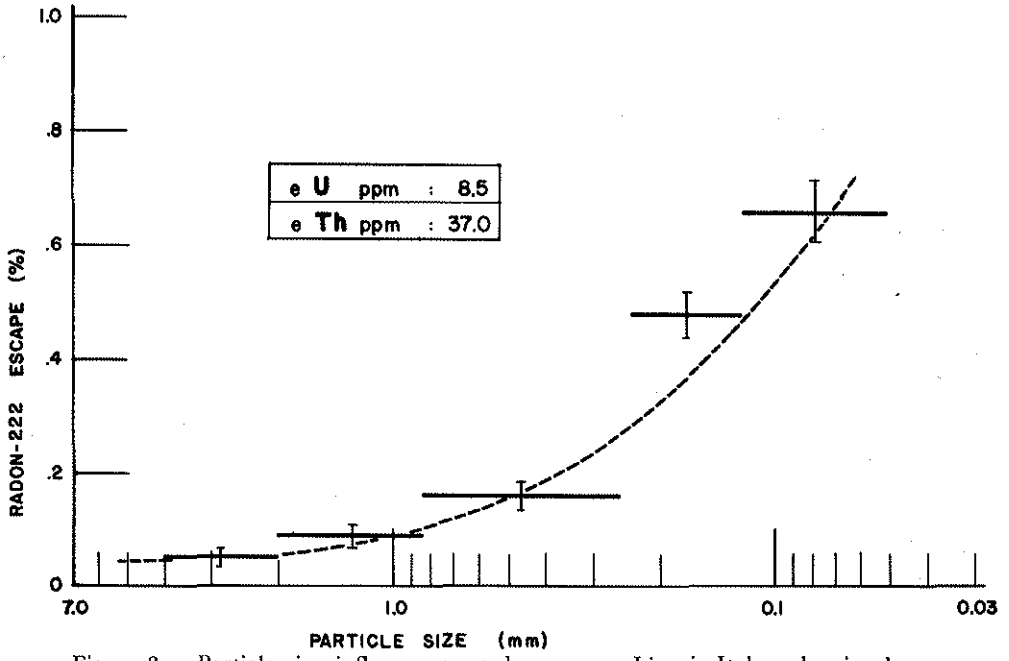

Figure 3 — Particle size influence on radon escape, Lipari, Italy volcanic glass

Thus, in conclusion, the data shown suggest that very little ^{222}Rn is escaping from dust at the lunar surface and therefore one should expect that radon would make a small contribution to the excess isotopic lead. The strong adsorption of ^{210}Po on layers of basalt powder observed by Lambert *et al.* (1973) is very interesting and adds another parameter to be investigated. However the measurements reported herein are confined to surface dust material and the radon behavior in breccias and rocks (specially high uranium material like rock 12 013) must be investigated and experiments be performed at temperatures and condition approximating the lunar day before any further conclusions are reached regarding the radon emanation of the lunar material. Only with more emanation data, more information on the diffusion coefficients of radon for the lunar regolith conditions, studies of other radon properties as effective attachment coefficients (Ikebe and Kawano, 1970, Ikebe *et al.*, 1970) and applying mathematical models (Friesen, 1972) will it be possible to evaluate the radon mobility in the lunar surface.

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