# MODEL FOR RADON DIFFUSION THROUGH THE LUNAR REGOLITH

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**Abstract.** A model for radon diffusion through the lunar regolith is proposed in which the atom migrates by random walk. The regolith is represented by a system of randomly oriented baffles in which the mean distance $\bar{d}$ which the atom travels between two collisions takes on the role of a mean free path. The effective mean time between two collisions depends on two entities: the actual mean time-of-flight and the mean sticking time on grain surfaces for one collision. The latter depends strongly on the temperature and the heat of adsorption of radon on regolith materials. Both $\bar{d}$ (mean free path) as well as $Q$ (heat of adsorption) are either poorly known or unknown for the lunar regolith; hence these quantities are treated as free parameters. Because of the greatly different mean lifetimes against radioactive decay of $^{219}$Rn, $^{220}$Rn, and $^{222}$Rn, the regolith acts as a powerful 'filter' for these species. $^{222}$Rn escape is significant (32%) even for $\bar{d} = 1$ $\mu$, $Q = 7.0$ kcal/mole and a regolith depth of 4 m. Calculations of radon escape from a 4 m thick regolith, using $\bar{d} = 1$, 10 and 80 $\mu$ and $Q = 4.0$, 5.2 and 7.0 kcal/mole show that the $^{222}$Rn/$^{220}$Rn escape ratio can be as small as 7.7 and as large as, or larger than 47. The small value of 7.7 is of particular interest, because it is nearly equal to the escape ratio inferred by Turkevich *et al.* from their Surveyor 5 results.

## 1. Introduction

The formation of an $\alpha$-radioactive deposit on the surface of the Moon involves at least three distinct processes: the emanation of radon out of minerals into the void spaces between regolith fragments, the diffusion of radon through the regolith into the lunar atmosphere, and the trapping of polonium daughters at the top of the regolith (see Kraner *et al.*, 1966). The escape of radon from terrestrial soils is a well-known phenomenon, which has been studied both experimentally as well as theoretically (see Junge, 1963). Turkevich *et al.* (1970), who discovered the radioactive deposit on the lunar surface at the Sea of Tranquillity used a terrestrial model to calculate the ratio of emissivity of $^{222}$Rn to $^{220}$Rn from the regolith. Lindstrom *et al.* (1971) also used a theory developed for the terrestrial environment to discuss their results of $^{210}$Pb equilibrium on the surface of lunar rock and soil. Both groups have concluded that the terrestrial models are probably inadequate, or that their applicability to the lunar case is very questionable. In this communication we should like to present an alternative model for the second of the above mentioned processes and then discuss some numerical results of our calculations.

## 2. Physical Model

Although the first of the three processes, namely the emanation (diffusion, recoil?) of radon from the minerals of regolith particles has important implications for the

### TABLE I

$^{222}$Rn escape probabilities for regolith thickness greater than 4 m

| Thickness (m) | Probability | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| | $Q = 4.0$ $\bar{d} = 1$ | 4.0 10 | 4.0 80 | 5.2 1 | 5.2 10 | 5.2 80 | 7.0 10 | 7.0 80 |
| 4 | 0.188 | 0.383 | 0.454 | 0.046 | 0.352 | 0.453 | 0.058 | 0.377 |
| 5 | 0.147 | 0.356 | 0.444 | 0.025 | 0.323 | 0.443 | 0.034 | 0.351 |
| 6 | 0.115 | 0.333 | 0.433 | 0.014 | 0.295 | 0.431 | 0.021 | 0.327 |
| 7 | 0.090 | 0.321 | 0.426 | 0.008 | 0.271 | 0.420 | 0.011 | 0.305 |
| 8 | 0.071 | 0.291 | 0.413 | -- | 0.248 | 0.410 | 0.007 | 0.284 |
| 9 | 0.056 | 0.272 | 0.403 | -- | 0.227 | 0.400 | -- | 0.265 |
| 10 | 0.043 | 0.254 | 0.394 | -- | 0.208 | 0.390 | -- | 0.247 |

### TABLE II

Average $^{222}$Rn and $^{220}$Rn escape probabilities for a regolith of 4 m depth

$^{222}$Rn

| $Q$, kcal/mole | | $\bar{d} = 1\,\mu$ | $\bar{d} = 10\,\mu$ | $\bar{d} = 80\,\mu$ |
|---|---|---|---|---|
| | $N$ | | 0.437 (0.77) | |
| 4.0 | $M$ | | 0.434 (0.77) | |
| | $A$ | | 0.436 (0.77) | |
| | $N$ | 0.189 | 0.421 (0.72) | 0.476 (0.91) |
| 5.2 | $M$ | 0.087 | 0.390 (0.67) | 0.472 (0.90) |
| | $A$ | 0.138 | 0.406 (0.695) | 0.474 (0.905) |
| | $N$ | | 0.227 | 0.442 (0.78) |
| 7.0 | $M$ | | 0.025 | 0.337 (0.60) |
| | $A$ | | 0.126 | 0.390 (0.69) |
| | | $^{220}$Rn | | |
| | $N$ | | 0.023 | |
| 4.0 | $M$ | | 0.014 | |
| | $A$ | | 0.019 | |
| | $N$ | 0.0057 | 0.019 | 0.062 |
| 5.2 | $M$ | < 0.001 | < 0.001 | 0.030 |
| | $A$ | 0.0029 | 0.010 | 0.046 |
| | $N$ | | 0.010 | 0.031 |
| 7.0 | $M$ | | < 0.001 | < 0.001 |
| | $A$ | | 0.005 | 0.016 |
| | | $^{222}$Rn/$^{220}$Rn | | |
| | $N$ | | 19 : 1 | |
| 4.0 | $M$ | | 31 : 1 | |
| | $A$ | | 23 : 1 | |
| | $N$ | 33 : 1 | 22 : 1 | 7.7 : 1 |
| 5.2 | $M$ | > 85 : 1 | > 389 : 1 | 16 : 1 |
| | $A$ | 48 : 1 | 41 : 1 | 10 : 1 |
| | $N$ | | 23 : 1 | 14 : 1 |
| 7.0 | $M$ | | > 29 : 1 | > 335 : 1 |
| | $A$ | | 25 : 1 | 24 : 1 |

$N$ = lunar noon, $M$ = lunar midnight, $A$ = arithmetic average. Numbers in parentheses represent the cases of reflection at 4 m depth.

formation of radioactive deposits on the lunar surface we shall restrict ourselves to the second process, the diffusion of radon through the void spaces of the regolith. We shall assume all along that the first process does not fractionate between $^{219}$Rn, $^{220}$Rn, and $^{222}$Rn, i.e. that exactly the same percentages of the numbers of atoms of each of these isotopes formed in the minerals from radium decay escape into the void spaces. This assumption is probably unrealistic, but nevertheless useful because the numerical results in Tables I and II represent emanation rates and fractionation factors for a distinct step in the formation of the deposits.

Let us then consider a radon atom which has just emanated from the mineral in which it was formed (Figure 1). The atom is available for diffusion through the regolith, a very porous and permeable medium (see Birkebak and Dawson, 1971),
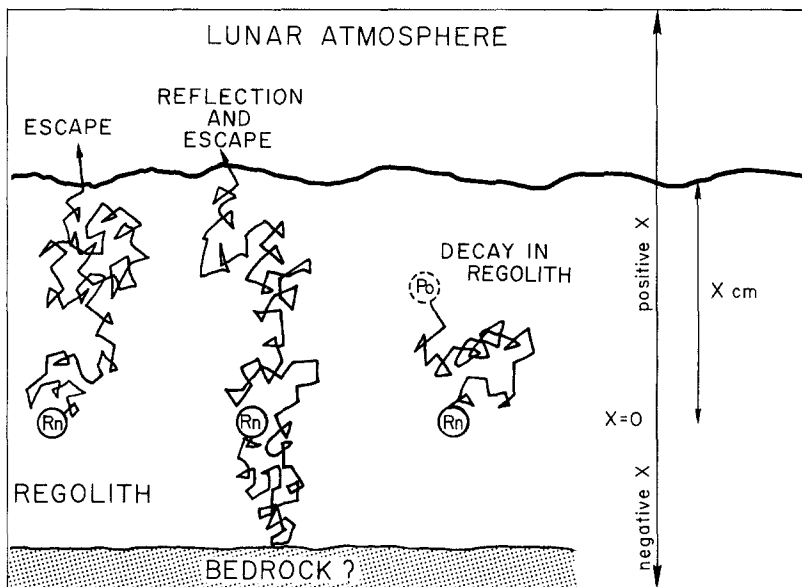


Fig. 1.   Schematic representation of radon diffusion in the regolith.

consisting principally of fragments smaller than 1 mm (for size distributions see LSPET, 1969, 1970, 1971). The atom, starting at some depth $X$ moves through this medium by random walk, which is imposed upon it by the collisions with the surfaces of the regolith particles. Collisions with gas atoms or molecules in the porespaces of the regolith can be safely neglected because of the very low gas pressures on the surface of the Moon. Although grain-boundary diffusion may become important when the 'passage' between two relatively large void spaces is very narrow, we shall assume that the random walk is governed in the main by the free flight of radon atoms in the porespaces. There is one important exception; however. During each collision the atom 'sticks' for a certain time on the grain surface. This sticking time becomes especially important at low temperatures.

We shall represent the regolith by a system of randomly oriented baffles, in which the mean distance which the atom travels between two collisions, $\bar{d}$, takes on the role of a mean free path. Although a fixed value could be estimated for this parameter, we have decided to treat it as a free parameter. Judging from the median grain sizes of lunar soils as reported by the Preliminary Examination Teams (1969, 1970, 1971) it appears likely that the best value for $\bar{d}$ lies between 1 and 10 $\mu$. In our calculations we have used values for $\bar{d}$ of 1, 10, and 80 $\mu$.

The mean time of flight between two collisions depends on the mean thermal velocity $\bar{v}$, which is a function of temperature, which in turn is a function of depth in the regolith. The diurnal heat wave penetrates only very shallowly (on the order of 10 cm); the temperature profile at greater depth is not accurately known at this writing. We have assumed a constant temperature of 200 K at depths greater than 12 cm. For the temperature distribution in the upper 12 cm of the regolith we have restricted ourselves to two cases: lunar noon and midnight (a few calculations were done for different Sun angles). The temperature vs. depth curves of Winter and Saari (1969) were used.

At low temperatures the diffusion of radon is no longer governed by $\bar{d}$ and $\bar{v}$, but by $\bar{d}$ and $\tau$, the sticking time. Following De Boer (1953) we have assumed that the sticking time can be represented by

$$\tau = \tau_0 e^{Q/RT}, \tag{1}$$

where $Q$ is the (unknown) heat of adsorption of radon on regolith material. Until this quantity is determined experimentally, we can only make qualitative inferences from the potential energies of adsorption of Ne, Ar, Kr, and Xe, which have been well-determined for charcoal (see Ross and Oliver, 1964). Judging from these data we conclude that $Q$ might be about 5–6 kcal/mole, but we suspect that the very clean and perhaps very active surfaces of regolith grains on the Moon could have much greater adsorption potentials than charcoal. In a recent paper Heymann and Yaniv (1971) have suggested that the measurement of 'Rn-pileup' at the lunar terminator by orbiting $\alpha$-spectrometry experiments could in principle be used to determine $Q$. However, since $Q$ is still unknown we shall treat this quantity also as a free parameter, using values in the range of 4–7 kcal/mole.

There has been much discussion in the literature whether or not the regolith is underlain by a contiguous or broken-up bedrock, or even whether there is a bedrock at all at some depth. For our model the presence or absence of a bedrock makes little difference. If there is no bedrock the atom has an equal probability to diffuse upward and possibly out of the regolith or to diffuse downward and decay at some depth. If there is a bedrock (assumed to be impermeable for radon diffusion), it acts as a reflecting plane.

We are dealing with the diffusion of radioactive species: $^{219}$Rn($t_{1/2} = 4$ s), $^{220}$Rn (54.5 s), and $^{222}$Rn(3.825 days). The decay must be taken into account because the model should calculate the probability that a radon atom made available for diffusion at depth $X$ will escape into the lunar atmosphere.

## 3. Mathematical Model

The coordinates for the random walk are shown in Figure 2. At $t=0$ the atom is at depth $X(x=0)$. Positive values of $x$ are upward, negative downward. We have considered the case of an 'infinite' plane, i.e. Rn atoms are made available from time-to-time at different locations in this plane. This allows us to consider diffusion in the positive and negative $x$-direction only (i.e. by the projection of the random walk on the $x$-coordinate). The solution of the diffusion equation is readily available from diffusion theories. By considering the diffusion of only *one* atom instead of a large number, say $n$, number densities in the solutions of diffusion equations become probabilities. For instance the probability that the radon atom is located at a distance $x$ above or below plane $X$ at time $t$ is simply

$$p(x, t) = 1/\sqrt{\pi D t} \, \exp\left(-x^2/4Dt\right);$$ (2)

and when we account for radioactive decay, the equation changes to

$$p(x, t) = 1/\sqrt{\pi D t} \, \exp\left(-x^2/4Dt\right) \exp\left(t/t_1\right)$$ (3)

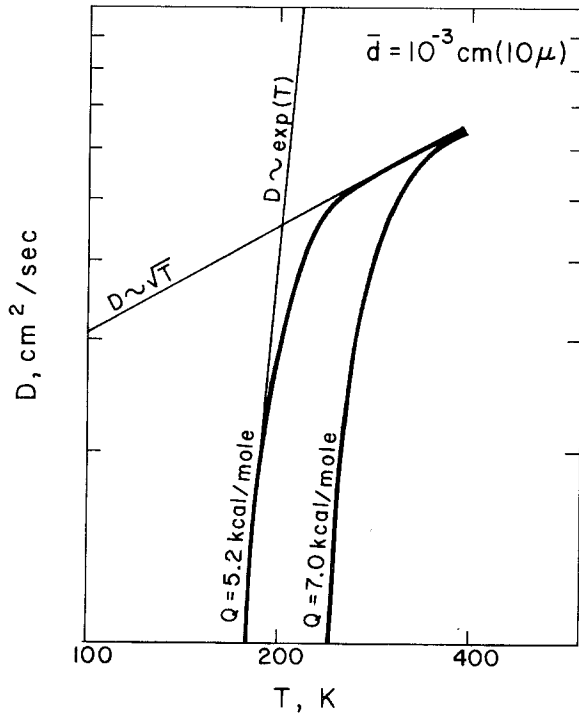$t_1$ = mean lifetime of radon atom = $t_{1/2}/0.693$

$t_{1/2}$ = half-life.



Fig. 2. Diffusion coefficients for $^{222}$Rn calculated with equations (1), (6), and (7) for $\bar{d} = 10 \, \mu$ and two values of $Q$: 5.2 and 7.0 kcal/mole. Note that $D$ has a $1/\sqrt{T}$ dependence at high temperatures and an $\exp(T)$ dependence at low temperatures.

Fick's equation for particle flux in a concentration gradient can be used to represent the probability that the atom moves through the plane $x$ at time $t$ in the positive $x$-direction in the form

$$p(x, t) = -D \frac{\partial p(x, t)}{\partial x} = \frac{x}{2t\sqrt{\pi Dt}} \exp(-x^2/4Dt) \exp(-t/t_1). \qquad (4)$$

Integration of Equation (4) from $t = 0$ to $t = \infty$ represents the probability that the radon atom has moved *at least* a distance $x$ from its initial position *without having*
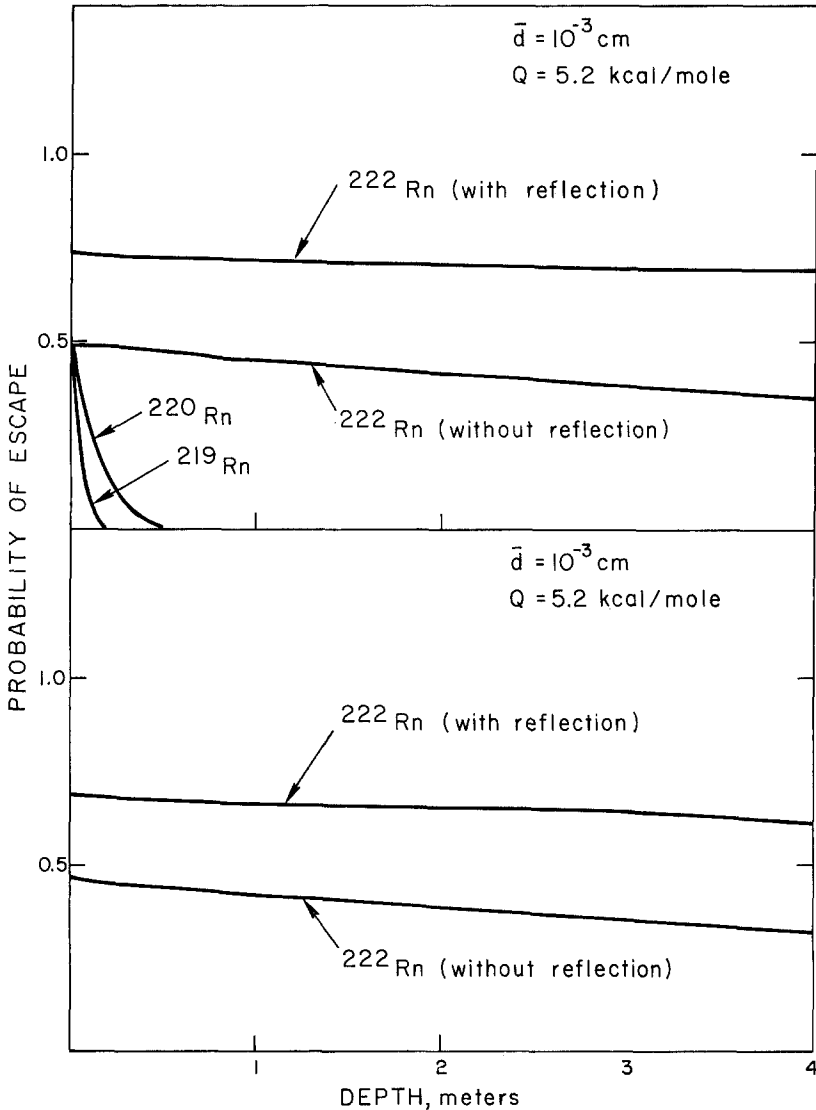


Fig. 3.   Radon escape probabilities for a regolith of 4 m thickness; $\bar{d} = 10\,\mu$, $Q = 5.2$ kcal/mole.
Top: lunar noon; bottom: lunar midnight.

*decayed* namely,

$$P(x) = \exp\left(- x \sqrt{0.693/Dt_{1/2}}\right).\tag{5}$$

The integration to $t = \infty$ seems irrational because the conditions of lunar day prevail for only 14 (Earth) days, to be followed by 14 days of lunar night. However, $P(x)$ can be obtained in a closed form such as in Equation (5) only for the integration between the chosen limits. Any other choice would have required very lengthy numerical integrations of Equation (4). The consequences of integrating to $t = \infty$ are insignificant for the $^{219}$Rn and $^{220}$Rn results. These species escape only during the lunar day, if at all (see Figures 3, 4, and 5). At sunset the escape rate falls rapidly to zero, or to very low values (Table II). During the lunar night practically all of $^{219}$Rn and $^{220}$Rn decays in the regolith proper. At sunrise, when the top of regolith warms up quite rapidly, the steady-state conditions for the escape of these species will be established on time scales comparable to the half-lives, i.e. very short compared to the length of the lunar day.

The case of $^{222}$Rn is more complicated, because the temperature variations in the top of the regolith are now reflected in the escape probability with a relaxation time of
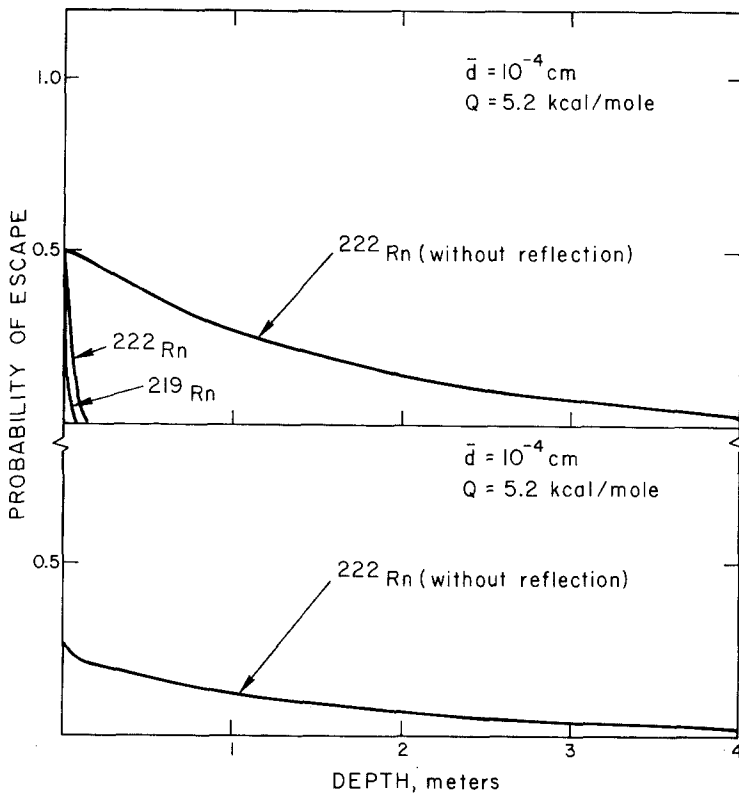


Fig. 4.   Radon escape probabilities for a regolith of 4 m thickness; $\bar{d} = 1\ \mu$, $Q = 5.2$ kcal/mole
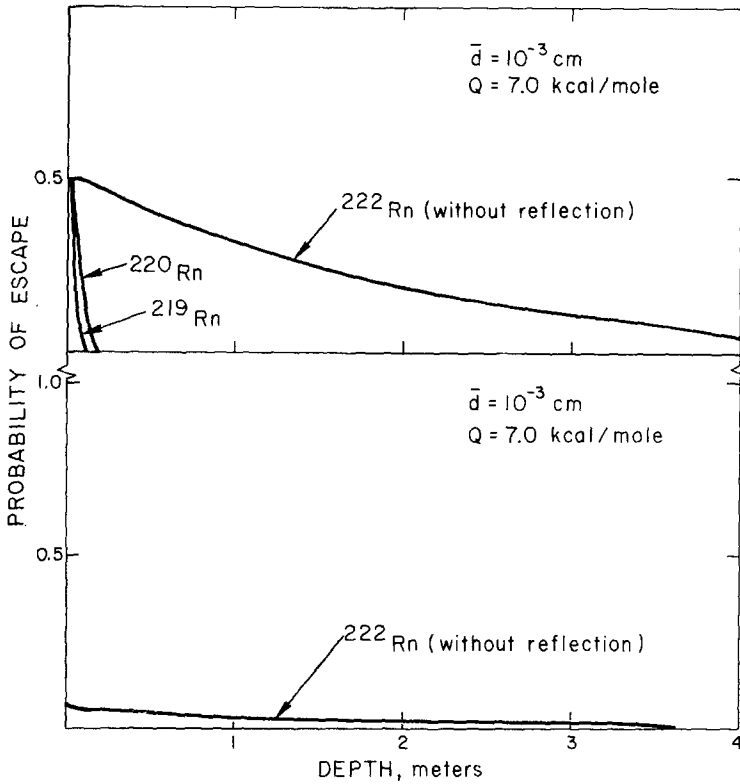Top: lunar noon; bottom: lunar midnight.

Fig. 5. Radon escape probabilities for a regolith of 4 m thickness: $\bar{d} = 10\,\mu$; $Q = 7.0$ kcl/mole. Top: lunar noon; bottom: lunar midnight.

several days. Let us consider a $^{222}$Rn atom made available at 4 m depth at lunar noon (Figure 4). If the conditions of lunar noon prevailed forever, the atom would have a 4% chance of escaping into the lunar atmosphere. But after only 7 days the conditions change rapidly (cooling of the top of the regolith) such that if the atom is not yet actually in the atmosphere, its probabilty of reaching the atmosphere is considerably reduced during the next 14 days, or for more than three $^{222}$Rn half-lives! However, this loss of escape probability will be in part, if not wholly offset by the nighttime diffusion. Consider a $^{222}$Rn atom released at 4 m depth at midnight. It has a probability of only 1.5% to reach the lunar atmosphere. However, from about 12 cm down to 4 m the temperature conditions during lunar night are essentially the same as during the day. What happens is that the atom diffuses as if it were daytime, until it gets into the top 12 cm of the regolith, where it becomes slowed down for 7 days. Soon after sunrise, the undecayed $^{222}$Rn still present in the top layers of the regolith will be released under daytime conditions. It is because of these arguments that we consider Equation (5) a fair representation for both lunar day and nighttime conditions.

Unfortunately Equation (5) cannot be easily integrated from depth $X$ to the top of the regolith because of the complicated dependence of the diffusion coefficient $D$

on temperature. $D$ is related to $\bar{d}$ and the effective mean velocity $\bar{v}_{\text{eff}}$ by

$$D = \tfrac{1}{3}\bar{d}\bar{v}_{\text{eff}}. \tag{6}$$

The effective mean velocity reflects the mean time spent by the atom to travel one mean free path, but this time includes both the sticking time as well as the actual mean time of flight between collisions namely,

$$\bar{v}_{\text{eff}} = \frac{\bar{d}}{\tau + \bar{d}/\bar{v}} \tag{7}$$

Two examples of the temperature dependence of $D$ are shown in Figure 2 for $\bar{d}=$ $=10~\mu$ and two values of $Q$: 5.2 and 7.0 kcal/mole. The curve for 5.2 kcal/mole shows clearly that $D$ is governed by the mean time of flight at temperatures greater than about 250 K, but by the sticking time at lower temperatures.

In order to make Equation (5) useful to calculate the escape probability from depth $X$, we have divided the regolith into strips of 1 cm thickness. The temperature in each strip is assumed constant. An adjustment, when necessary, is made at the boundary between two strips. The probability that a radon atom, starting at depth $X$ cm, escapes into the atmosphere is then the product of the probability that the atom 'survives' diffusion through each of the $X$ layers: i.e.,

$$P(X) = \tfrac{1}{2} \prod_{i=1}^{x} P_i. \tag{8}$$

(The factor $\tfrac{1}{2}$ arises from the fact that only one-half of the atoms diffuses in the positive $x$-directions.)

Most of the calculations were done by computer. In a few cases ($X = 4 - 10$ m) we decided to neglect the influence of the top 10–20 cm and solve Equation (4) directly using $T = 200$ K. Hand calculations were also made for the case of reflection, because

$$P'(X) = P(X) + P^2(Y)/P(X),$$

where $P(X)$ is the probabilty without reflection; $Y$ the depth of the reflecting plane.

## 4. Results and Discussion

Figures 3, 4, and 5 show typical results of escape probabilities for different values of $\bar{d}$ and $Q$ at noon and midnight for a regolith depth of 4 m. Results of calculations for greater regolith depth (up to 10 m) are given in Table I.

Let us briefly consider the case for $\bar{d} = 10~\mu$ and $Q = 5.2$ kcal/mole (the value of 5.2 kcal/mole was obtained by extrapolation of the charcoal data for Ne, Ar, Kr, and Xe). For the noontime case it is seen that the escape probability of $^{222}$Rn remains fairly large throughout the entire 4 m depth; in fact at 4 m the probability is still 0.35. Table I shows that the probability is 0.21 at a depth of 10 m! At midnight the probability of escape from the first 10 cm is somewhat less compared to the noontime curve, but is still fairly high at about 0.45. Again the decrease of the escape probability is slow down to 4 m, where the value is now 0.32. With reflection at 4 m all the pro-

babilities increase by some 40–50%. The inferences are that a regolith of this kind will nearly always suffer substantial $^{222}$Rn losses both on the sunlit as well as on the dark side. No great 'pileup' of $^{222}$Rn will occur at midnight in the uppermost 10–20 cm; however, the lowest temperatures are not reached until shortly before sunrise. Our calculations show that some 'pileup' (or retention) of $^{222}$Rn occurs during the last few (Earth) days before sunrise, but since this adsorbed radon can begin to diffuse almost immediately after sunrise when the temperature of the surface increases rapidly, the pileup has only a small effect on the total $^{222}$Rn escape during a full lunar day. At noon both $^{219}$Rn and $^{220}$Rn escape from the regolith, but their probabilities fall rapidly with depth: that of $^{219}$Rn is practically zero at 20 cm depth; that of $^{220}$Rn at 55 cm depth. At midnight neither of these isotopes can escape; in fact it is safe to say that for this case *all* of the $^{219}$Rn and $^{220}$Rn decay *in* the regolith proper during lunar night. In summary: a regolith of this kind acts as a very powerful 'filter' for the three radon isotopes.

The effect of a shorter mean free path, 1 $\mu$, is shown in Figure 4. The principal difference with the preceding case is at depth greater than 20 cm where the escape probability decreases now much faster and reaches the value of only 0.05 at 4 m at noon (0.015 at midnight). Consequently reflection at 4 m has only little influence on the escape probabilities. Also the depth from which $^{219}$Rn and $^{220}$Rn can escape is reduced by about a factor three. The escape of $^{222}$Rn at midnight is considerably smaller than for the preceding case, hence one would suspect a substantially increased pileup from midnight to sunrise in the upper 20 cm.

Figure 5 shows the effect of increasing $Q$ to 7.0 kcal/mole. Qualitatively the effect is the same as that of decreasing $\bar{d}$.

A quantity of interest is the *average escape probability* for a regolith of given depth; averaged both over depth and an entire lunar day, or certain periods of the lunar day. In order to test the influence of changing sunangles we did make a few calculations for angles different from lunar noon or midnight. When it turned out that the more refined calculations gave results not too greatly different from the simplest assumption, namely noontime conditions during all of the lunar day, and midnight conditions during all of lunar night, we continued to use the latter assumption.

Table II shows average escape probabilities for a regolith of 4 m depth. The following assumptions were made. First that the regolith is grossly uniform as far as U and Th concentrations are concerned (i.e. homogeneity on a 1 cm$^3$ scale). Second that there is no difference in the emanation-to-production ratio for $^{219}$Rn, $^{220}$Rn, and $^{222}$Rn, i.e. that the same percentage of each of these isotopes escapes (from the minerals in which they are formed) into the porespaces of the regolith. The table shows the average escape probabilities for three values of $Q$ (4.0, 5.2, and 7.0 kcal/mole), three values of $\bar{d}$ (1, 10, and 80 $\mu$); for lunar noon and midnight (i.e. lunar day and lunar night); and the arithmetic average of day and night.

Table II shows quantitatively the expected trends: radon escape is considerably larger from a coarse ($\bar{d}=80$ $\mu$) than it is from a fine regolith ($\bar{d}=1$ $\mu$); the cooling of the uppermost 12 cm of the regolith during lunar night results in a significant, if not

substantial decrease of the escape rates, and large values of the heat of adsorption also slow the escape of radon down.

The trends of $^{222}$Rn/$^{220}$Rn escape ratios are very interesting. Let us first consider the case for $\bar{d} = 10 \mu$, $Q = 5.2$ kcal/mole. At lunar noon the ratio is 22:1; at midnight $> 389:1$; the average is 41:1. An increase in $\bar{d}$ to 80 $\mu$ causes a decrease in all three of the ratios. The most probable reason is that the depth from which $^{220}$Rn escapes has increased more than the already considerable depth from which $^{222}$Rn has a large escape probability. For a finer regolith ($\bar{d} = 1 \mu$); all the ratios increase. The effects of changing $Q$-values on the other hand appear to be more complicated. For a smaller valne (4.0 kcal/mole), as well as for a larger value (7.0 kcal/mole) all three ratios are seen to decrease. Obviously the smaller $Q$-value increases the $^{220}$Rn escape rate more than that of $^{222}$Rn, whereas the larger $Q$-value inhibits $^{222}$Rn diffusion more strongly than it slows down the $^{220}$Rn diffusion.

The $^{222}$Rn/$^{220}$Rn escape ratios vary from a low of 7.7 to a high of $> 389$. The smallest value of 7.7 is of considerable interest, because it is nearly equal to the value of about 7 required by the Surveyor 5 results of Turkevich *et al.* (1970). The good agreement does not necessarily imply that the $^{220}$Rn and $^{222}$Rn in the Sea of Tranquillity have escaped from a relatively coarse regolith ($\bar{d} = 80 \mu$), because the observed ratio could in part be due to a fractionation of the two species in the first step, emanation from the minerals. However, it is gratifying to note that our model produces ratios which are in general considerably smaller than the value of 75 calculated by Turkevich *et al.* (1970) from the terrestrial model.

## Acknowledgment

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