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TRANSIT ANALYSIS

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ABSTRACT

The helium release from $^{238}\text{PuO}_2$ microsphere fuel was calculated for six different heat-sources that have been proposed for the Transit RTG. The release mechanism was assumed to be classical diffusion, with a two-term, temperature-dependent diffusion coefficient:

$$D = D_1 \exp (-E_1/RT) + D_2 \exp (-E_2/RT) .$$

The temperature history assumed was based upon contractor-supplied information. Two different methods of approach were used and the results were compared. Helium release was calculated both for the normal mission profile and for various possible aborts. Upper and lower limits for the release were also calculated. The origins and nature of the rather sizable uncertainties involved are discussed.

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TABLE OF CONTENTS

	<u>Page</u>
I. Capsules Studied	5
II. Thermal History	6
III. Analysis of Capsules	7
IV. Series A Calculations	8
V. Results of Series A Calculations	12
VI. Series B Calculations	12
VII. Summary and Conclusions	14
VIII. References	15
APPENDIX A - New Diffusion Constant Expressions Corrected for ORNL Temperature Errors	27
APPENDIX B - Steady State Analysis of Gas Release from Spherical Particles	31

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TRANSIT ANALYSIS

Introduction

In connection with design work for Transit space power systems, six capsules were analyzed with regard to helium release before and during the mission. This paper documents and summarizes the problem constraints, the methods of calculation, and the results.

Two somewhat different series of calculations were performed. These are designated as Series A and Series B. Sections I and II apply to both series. In Sections III, IV, and V, the general discussion of the uncertainties in the diffusion coefficient, and the limitations of the simple diffusion model used, apply to both series, while the details of the specific diffusion parameters, the calculational techniques, and the results, apply only to Series A. The latter subjects are discussed for Series B in Section VI. Section VII summarizes the results and attempts to explain the meaning of the differences, such as they are, in the results obtained by the two methods.

I. Capsules Studied

The six capsules studied were:

1. Isotec unvented capsule
2. Isotopes Pioneer capsule (vented)
3. TRW SNAP-19 (vented)
4. RCA AIR-VAC capsule (vented)
5. 3M SNAP-27 capsule (vented)
6. GGA Isotec capsule (vented)

II. Thermal History

The capsules were assumed to undergo a postulated thermal history, which consisted of the following.

A. Prelaunch storage - 1 year

1. Water storage - 9 months

Skin line temperature $\sim 212^{\circ}\text{F}$

Center line temperature $\sim 450^{\circ}\text{F}$

(assuming gradients to be same as under operating conditions)

2. Shipping containers - 3 months

Skin line temperature $\sim 400^{\circ}\text{F}$

Center line temperature $\sim 640^{\circ}\text{F}$

B. Launch pad storage - 1000 hours

Because of air convection cooling, capsule temperatures were estimated as approximately 160°F less than space operating temperatures.

C. Launch, orbit, and short-term reentry. This portion of the thermal history was treated for two possibilities:

1. Launch, orbit, and reentry after one day in orbit; and

2. Launch, orbit, and reentry after 10 days in orbit.

In both cases the reentry configuration was assumed to be side-on stable, which results in severe heating of the capsule. It is, consequently, a "worst" case with regard to helium release.

D. Long-term operation. Assuming no reentry, the capsule was allowed to operate for 5 years. As in reentry calculations, the required temperature profiles were obtained from contractor design information.

III. Analysis of Capsules

Analysis of the various capsules was carried out assuming a simple diffusion model with uniform spherical particles. This analysis is most likely an oversimplification because of at least two factors which modify the diffusion kinetics: (1) the existence of porosity within the microspheres, and (2) the existence of radiation-produced defects which possibly function as trapping sites. Nonetheless, from a phenomenological viewpoint, a simple diffusion model comes close to describing the observed experimental behavior, and in view of engineering and design expediency, the data contained in this report were generated using a simple diffusion model which must suffice until future study yields a more definite description.

The diffusion constant used was of the form:

$$D(T) = D_1 \exp \left[-E_1/RT \right] + D_2 \exp \left[-E_2/RT \right] , \quad (1)$$

which is in accordance with experimental results on production grade $^{238}\text{PuO}_2$ plasma torch fused (PTF) microspheres. At the time of the Transit analysis*, the values used in the Series A calculations were:

$$\begin{aligned} D_1 &= 2.58 \times 10^{-14} \text{ cm}^2/\text{sec} \\ D_2 &= 3 \times 10^{-3} \text{ cm}^2/\text{sec} \pm 570\% \\ E_1 &= 5 \text{ Kcal/mole} \\ E_2 &= 49.2 \text{ Kcal/mole} \pm 10\% \end{aligned}$$

The E_1 value is approximate, and the D_1 value resulted from a forced fit to the form (1). The error limits on E_2 and D_2 are statistical results based on the scatter of the experimental data. [For a more detailed description of the origin of Eq. (1), see Appendix A.]

*This statement is made because several revisions in these values have been made as the experimental data have been refined. Previous analyses yielded values of E_2 that were lower by a few Kcal/mole and values of D_2 that were lower than those quoted above. Over the limited temperature range for which data exist, these changes essentially compensate for each other; use of the older values would not greatly affect the predicted release behavior.

It should be emphasized that the above values of the parameters in Eq. (1) are mean values derived from experimental data which show a wide range of scatter. It is our opinion that this scatter is "real"; that is, the helium release properties of $^{238}\text{PuO}_2$ microsphere fuel actually may be highly variable from batch to batch, and the scatter in the experimental data may not be merely the result of large experimental error. It was therefore considered necessary to perform some analyses for Transit using "maximum" and "minimum" values of the diffusion coefficient, as well as using the mean values given above.

The maximum and minimum values were obtained by varying D_2 and E_2 within the limits quoted above in such a way that the changes reinforced each other:

Maximum D: $D_2 = 1.8 \times 10^{-2} \text{ cm}^2/\text{sec}$, $E_2 = 44.3 \text{ Kcal/mole}$

Minimum D: $D_2 = 5 \times 10^{-4} \text{ cm}^2/\text{sec}$, $E_2 = 54.1 \text{ Kcal/mole}$.

There is no rigorous statistical justification for this procedure. However, when these values of D and the experimental data are plotted against temperature, about 80 percent of the experimental data points fall between these two extremes, and the remaining data points are only slightly outside these limits (Figure 1). Hence, these values do seem to provide a realistic estimate for the variability of the diffusion coefficient.

IV. Series A Calculations

All capsules were of the general shape of a right circular cylinder with hemispherical end caps. Each was divided into two or more volume segments, the average temperature of which was assumed to be the average of the temperature of the inner and outer boundaries of the segment. End effects were essentially neglected. The helium release was then calculated for each segment; the sum for all segments yielded the total release.

As stated earlier, a simple diffusion model was used as a basis for calculation. The fuel particles were assumed to be uniform spheres of radius $R = 71 \text{ }\mu\text{m}$. An average power density of approximately 2.7

watts/cc and an average mass density of approximately 9.7 gm/cc were assumed.

Some approximations were made in performing the Series A diffusion calculations. In the cases where reentry was assumed to occur after a short time (1 or 10 days) in orbit, most of the helium is generated during the long period of storage at low temperatures, while most of the release occurs during the relatively short period at high temperatures at the end of the fuel history. Under these conditions, one may ignore the fact that the helium is generated continuously over the entire fuel history, and use instead the description valid for a sphere with an initially uniform concentration to calculate the fraction of the helium that is released:

$$F(t) = 1 - \frac{6}{\pi^2} \sum_{n=1}^{\infty} \frac{1}{n^2} \exp \left(-n^2 \pi^2 \tau \right) \quad (2)$$

where

F = fractional release

= $\frac{\text{amount of gas released}}{\text{amount of gas initially present}}$

t = time

$$\tau = \int_0^t \frac{D(t')}{R^2} dt' . \quad (3)$$

For this approximation to be accurate in the case where the helium is generated continuously, most of the contribution to the integral for τ in Eq. (3) must come near the end of the fuel history. This is the case here, and the errors resulting from this approximation in the Transit calculations are no more than a few percent, a trivial value in comparison with the effects due to the uncertainty in the diffusion coefficient.

Given the temperature history, one may evaluate the integral (3) by means of the empirical diffusion coefficient (1). τ may then be substituted into Eq. (2) to yield the fractional release. In this regard the approximations:

$$F(t) = \frac{6}{\sqrt{\pi}} \tau^{\frac{1}{2}} - 3\tau \quad (4)$$

for $\tau \leq 0.2$

and

$$F(t) = 1 - \frac{6}{\pi^2} \exp(-\pi^2 \tau), \text{ for } t > 0.2 \quad (5)$$

were often used.

For the initial 1-year storage period, calculations (Series A) based on the highest temperature encountered by the capsule indicate an overall release of approximately 5 percent or less. Experimental work on relatively new fuel at higher temperatures indicates that even this value is probably high; however, fuel approximately 2 years old has been observed to retain only 70 percent of the helium generated. Thus, in the Transit analysis, estimates of the helium release during the first 1-year storage range from 0 to about 30 percent. It was assumed that the helium remaining in the fuel was uniformly distributed.

For the 1000-hour storage in the converter on the launch pad, average temperatures were estimated for each segment as approximately 160°F less than the average space operating temperature. The integral (3) was evaluated and the fraction F calculated for $t = 200, 400, 600, 800,$ and 1000 hours on the launch pad. The amount of helium generated during these periods was added to that remaining after 1-year storage for the calculation of the total helium release.

The description of helium release during the launch, orbit, and reentry required estimates of the temperature profiles of the capsules both during normal operation in space and during reentry. These data were obtained from rough interpolations of calculations supplied by Isotopes and TRW.

For long-term operation, without reentry, equilibrium between generation and release is reached in a time short compared with 5 years for most cases. At equilibrium, the amount retained is given by the steady-state analysis of diffusional release from a sphere with a constant generation A_0 (see Appendix B):

$$C_R = C_{AVG} - \frac{A_0 R^2}{15D} \quad (6)$$

where

C_{AVG} = average concentration of helium in the sphere

C_R = helium concentration at the surface of the sphere.

For the vented capsule systems, it was assumed that $C_R = 0$, so that

$$C_{AVG} = \frac{A_o}{15(D/R^2)}$$

at equilibrium. The amount released is, of course, the difference between the total generated and the amount retained in the microspheres. In the case of the unvented capsule, it was necessary to approximate C_R using Henry's law,

$$C = kP \quad (7)$$

where

P = pressure

k = proportionality constant.

No experimental values of k are available for the helium solubility in PuO_2 ; however, data obtained for the structural isomorph UO_2 were used in the calculation.¹ Using this value of $k = 3.60 \times 10^{-5}$ cc(STP)/gm atm with the results of some very approximate calculations on capsule pressure enabled one to estimate C_R , and hence the average concentration C_{AVG} of helium in the fuel.

The criterion for the validity of the equilibrium assumption is that

$$\frac{Dt}{R^2} \gtrsim 0.2 \quad .$$

This criterion was not met for the Isotopes-Pioneer and TRW SNAP-19 vented capsules when the minimum value of D was used. Hence, the following procedure was employed in Series A.

The helium generated prior to launch and that generated after launch were treated separately. Eq. (2) was used to evaluate the release of the helium generated prior to launch, as described previously. For the helium generated after launch, the helium generation rate was assumed to be constant with time, i.e., the slight decline due to radioactive

decay was neglected. In addition, (D) was assumed to be constant after launch, giving $\tau = Dt/R^2$. Eq. (2) then gives, not the fractional release, but rather the ratio (release rate)/(generation rate). Integration over time, starting with the time of launch, gives the amount of this component released, and addition of the contribution from the helium generated prior to launch gives the total released.

V. Results of Series A Calculations

The results of the Series A calculations using the mean value of the diffusion coefficient are shown in Figures 2 through 7, in which the cumulative helium release is plotted as a function of time. In addition, release calculations for the Isotopes Pioneer capsule and the TRW SNAP-19 capsule were repeated using the "minimum" diffusion coefficient. The results for these two cases are shown in Figures 8 and 9.

Table I summarizes the results for the cases where reentry is assumed to occur after 1 or 10 days in orbit. The table gives the fractional release at launch, after 1 or 10 days in orbit, and after reentry following 1 or 10 days in orbit. Results are given for the "minimum" and "maximum" values of the diffusion coefficients as well as for the mean value.

VI. Series B Calculations

A computer program, HELIUM, which employs an implicit numerical solution to solve the problem of classical diffusion of helium in alpha-emitting fuels, was used in the Series B calculations.² Because of the speed of the HELIUM solution, it could be supplied with a number of diffusion coefficients and activation energies to indicate the variability in the predicted helium release. In addition, values of the fuel activity, nuclear decay constant, mean fuel radius, etc., were used as input to the program. HELIUM does not employ the approximations involved in the use of Eqs. (2) and (3) in the Series A calculations; however, as mentioned previously, those approximations are expected to be of little significance in the Transit calculations.

The diffusion coefficients employed in Series B were obtained from essentially the same body of experimental data as those employed in Series A (Figure 1). The chief difference between the two sets of calculations lies in the way these coefficients were derived from the experimental data and used in the calculations.

In Series B, the program HECON2 was used together with another program, PULSE, to extract diffusion coefficients.¹ Again, these coefficients showed a wide range of variability, but they also fell within an envelope qualitatively consistent with a two-term diffusion coefficient such as that described by Eq. (1). The envelope is shown in Figure 10 in which $D' = D/R^2$ is plotted.

Many (but not all) of the data were derived from experiments in which the helium release rate was measured while fuel samples were heated to some elevated temperature which was then held constant for some period of time. During the "transient" or heating-up period, there was often a temperature range over which the diffusion coefficient extracted by PULSE increased more rapidly with increasing temperature than do the bounding curves of the envelope shown in Figure 10, though they remained within that envelope. The extent to which this occurred, and the temperature range over which it occurred, were quite variable. After the peak temperature was reached, the diffusion coefficients extracted by PULSE typically declined by one to two orders of magnitude even though the temperature remained constant.* This decline may be due to a time-dependence of the diffusion coefficient in addition to that resulting from changes in temperature; on the other hand, it must be noted that the fuel is composed of particles with a considerable range of surface/volume ratios, which is expected to lead to qualitatively similar effects. None of our calculations take into account this size spectrum of the microspheres.

In the Series B calculations, the parameters of Eq. (1) were chosen to best fit the behavior observed for the individual fuel samples, instead of choosing the mean value or the limiting values

*The diffusion coefficients used in the Series A calculations were extracted by a different method. They also sometimes showed this behavior, but less generally than did those obtained from "PULSE."

of the envelope of Figure 10. The variations observed between the different fuel samples were matched with corresponding variations in the diffusion parameters, giving a range of predicted releases.

The procedure used gives higher weight to the transient phenomena, as HELIUM cannot take into account the frequently observed decline in the diffusion coefficient with time at constant temperature. Parameters that best fit the diffusion coefficients extracted by PULSE tend to give higher values of the diffusion coefficient than do parameters describing the behavior after a considerable time at constant temperature. However, both the "transient" and "long-term" diffusion coefficients show wide variability and overlap considerably.

Results of the Series B calculations are shown in Figures 11 through 16.

VII. Summary and Conclusions

Comparison of Figures 3 through 10 and Table I with Figures 11 through 16 shows that the Series A calculations tend to give somewhat lower values for the helium release than do the Series B calculations, as might be expected from the fact that the diffusion parameters employed in Series A are largely derived from measurements made after the transient variation of the diffusion coefficients had largely died out, while those used in Series B weight more heavily the transient heat-up phenomena. (Actual fuel capsule history will, of course, include both transients and lengthy periods at some relatively constant temperature.) The difference between the two may be regarded as a measure of the magnitude of the uncertainties resulting from the approximate nature of the basic model used in both cases, in which the release mechanism is described, phenomenologically at least, in terms of simple diffusion from homogeneous spheres of a single size.

In general, the differences are not large. There is little difference in the upper limits of the helium release predicted, if only because both methods give upper limits approaching 100 percent. There is more difference in the lower limits (e.g., compare Figures 8 and 9 with Figures 12 and 13, respectively).

In view of the origins of the diffusion coefficients employed in the two series, there may be some preference for Series A when long-term operation is considered. On the other hand, it should be remembered that there are no release data on microsphere fuel much older than about 2 years. Any predictions as to the release behavior for times much greater than 2 years are on somewhat shaky ground.

VIII. References

1. Sung, P., Equilibrium Solubility and Diffusivity of Helium in Single Crystal Uranium Dioxide, University of Washington, Seattle, Washington; PhD thesis, University Microfilms No. 68-9339, 1967.
2. SC-RR-69-103, HECON Computations for the Diffusion of Helium in Alpha-emitting Isotopes, C. J. M. Northrup, Jr., and D. E. Amos, Sandia Laboratories, Albuquerque, New Mexico, May 1969.

TABLE I

Helium Release Summary
(Series A)

Capsule	Mission Stage	Helium Released			STP Volume (cc) "Mean"	Retained (cc)
		"Mean"	Fraction Max	Min		
ISOTEC, unvented	Launch	0.720	1.0	0.17	1410	540
	E.O.*, 1 day	0.746	1.0	0.18	1460	500
	Reentry ⁺ , 1 day	0.780	1.0	0.20	1530	430
	E.O.*, 10 day	0.857	1.0	0.24	1730	290
	Reentry ⁺ , 10 day	0.875	1.0	0.25	1770	250
ISOTEC, vented	Launch	0.599	1.0	0.10	810	540
	E.O., 1 day	0.647	1.0	0.11	875	475
	Reentry	0.670	1.0	0.12	905	445
	E.O., 10 day	0.791	1.0	0.15	1090	290
	Reentry	0.803	1.0	0.16	1110	270
AIR VAC, vented	Launch	~1.0	1.0	0.563	1140	≤10
	E.O., 1 day	~1.0	1.0	0.580	1140	≤10
	Reentry	~1.0	1.0	0.583	1140	≤10
	E.O., 10 day	~1.0	1.0	0.699	1200	≤10
	Reentry	~1.0	1.0	0.701	1200	≤10
SNAP-27, vented	Launch	0.940	1.0	0.49	545**	35
	E.O., 1 day	0.949	1.0	0.51	550	30
	Reentry	0.973	1.0	0.55	564	13
	E.O., 10 day	0.977	1.0	0.65	586	14
	Reentry	0.991	1.0	0.68	594	6
TRW SNAP-19, vented	Launch	0.307	0.96	0.065	360	820
	E.O., 1 day	0.327	0.965	0.07	385	795
	Reentry	0.411	0.98	0.09	485	695
	E.O., 10 day	0.461	0.99	0.095	555	655
	Reentry	0.496	1.0	0.11	600	610
ISOTOPE- PIONEER	Launch	0.373	0.86	0.08	350	590
	E.O., 1 day	0.383	0.87	0.085	360	580
	Reentry	0.560	0.96	0.125	525	415
	E.O., 10 day	0.506	0.92	0.105	490	475
	Reentry	0.634	0.97	0.154	610	355

* E.O. = End of period in orbit

** In each of two capsules

+ Following reentry after (1,10) days orbit

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Figure 1

Figure 2

Figure 3

Figure 4

Figure 5

Figure 6

Figure 7

Figure 8

Figure 9

Figure 10

Figure 11

Figure 12

Figure 13

Figure 14

Figure 15

Figure 16

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APPENDIX A

New Diffusion Constant Expressions Corrected for ORNL Temperature Errors

For $T > 800^{\circ}\text{C}$

$$E_u = 49.2 \text{ Kcal/mole} \pm 10\%$$

$$D_u = D_{uo} \exp \left[-E_u/RT \right]$$

$$D_{uo} = 3 \times 10^{-3} \frac{\text{cm}^2}{\text{sec}} \pm 570\%$$

At low temperatures, break is seen.

Approximate activation energy is

$$E_L \approx 5 \text{ Kcal/mole} \pm 100\% .$$

The break temperature is $600^{\circ}\text{C} \pm 200^{\circ}$.

Assume the two activation energies are additive:

$$D_{\text{TOTAL}} = D_{Lo} e^{-E_L/RT} + D_{uo} e^{-E_u/RT} .$$

At 600°C the upper equation gives

$$\begin{aligned} \frac{1}{2}D &= D_{uo} \exp - \frac{49.2 \times 10^3}{1.987 \times 873} \\ &= D_{uo} \exp [-28.363] \\ &= D_{uo} \exp [-28.363] 10^{10} \cdot 10^{-10} \\ &= D_{uo} \exp [-28.363] \exp [23.0258] \times 10^{-10} \\ &= D_{uo} \times 10^{-10} \exp [-5.3372] \\ &= D_{uo} \times 10^{-10} \times 4.81 \times 10^{-3} \\ &= 3 \times 10^{-3} \times 10^{-10} \times 4.81 \times 10^{-3} \\ &= 3 \times 4.81 \times 10^{-16} \\ &= 1.443 \times 10^{-15} \longleftarrow . \end{aligned}$$

The lower equation gives, using the result of the last calculation:

$$\begin{aligned}
 1.443 \times 10^{-15} &= D_{Lo} \exp \left[- \frac{5 \text{ Kcal}}{RT} \right] \\
 &= D_{Lo} \exp \left[- \frac{5 \times 10^3}{1.987 \times 0.873 \times 10^3} \right] \\
 &= D_{Lo} \exp [-2.8824] \\
 &= D_{Lo} 5.6 \times 10^{-2} = D_{Lo} 0.56 \times 10^{-1}
 \end{aligned}$$

$$\therefore D_{Lo} = 2.576 \times 10^{-14} = \frac{1.443 \times 10^{-15}}{0.56 \times 10^{-1}}$$

$$\begin{aligned}
 \therefore D &= D_{Lo} \exp \left[- \frac{E_L}{RT} \right] + D_{Ho} \exp \left[- \frac{E_H}{RT} \right] \\
 &= 2.576 \times 10^{-14} \exp \left[- \frac{5 \times 10^3}{RT} \right] \\
 &\quad + 3 \times 10^{-3} \exp \left[- \frac{49.2 \times 10^3}{RT} \right] \\
 &= 2.576 \times 10^{-14} \exp \left[- \frac{2.516356316 \times 10^3}{T} \right] \\
 &\quad + 3 \times 10^{-3} \exp \left[- \frac{24.76094615 \times 10^3}{T} \right] .
 \end{aligned}$$

Dividing by 3×10^{-3}

$$\begin{aligned}
 D &= \left(8.586667 \times 10^{-12} \exp \left[- \frac{2.516 \times 10^3}{T} \right] \right. \\
 &\quad \left. + \exp \left[- \frac{24.76 \times 10^3}{T} \right] \right) (3 \times 10^{-3}) .
 \end{aligned}$$

Dividing by $\exp \left[- \frac{35.76 \times 10^3}{T} \right]$,

$$\begin{aligned}
 D &= \left(8.586667 \times 10^{-12} \exp \left[\frac{24.76 \times 10^3 - 2.516 \times 10^3}{T} \right] + 1 \right) \\
 &\quad \left(3 \times 10^{-3} \exp \left[- \frac{24.76 \times 10^3}{T} \right] \right) \\
 &= \left(1 + 8.586667 \times 10^{-12} \exp \left[\frac{22.24458983 \times 10^3}{T} \right] \right) \\
 &\quad \left(3 \times 10^{-3} \exp \left[- \frac{24.76 \times 10^3}{T} \right] \right) .
 \end{aligned}$$

Diffusion Constant For PuO₂ Microspheres

<u>T(°F)</u>	<u>T(°C)</u>	<u>D(cm²/sec)</u>	<u>D/R² (sec⁻¹)</u>
800	427	7.1 x 10 ⁻¹⁶	1.4 x 10 ⁻¹¹
1000	538	1.3 x 10 ⁻¹⁵	2.6 x 10 ⁻¹¹
1200	649	8.3 x 10 ⁻¹⁵	1.6 x 10 ⁻¹⁰
1400	760	1.2 x 10 ⁻¹³	2.4 x 10 ⁻⁹
1600	871	1.2 x 10 ⁻¹²	2.4 x 10 ⁻⁸
1800	982	8.1 x 10 ⁻¹²	1.6 x 10 ⁻⁷
2000	1093	4.1 x 10 ⁻¹¹	8.1 x 10 ⁻⁷
2200	1204	1.6 x 10 ⁻¹⁰	3.2 x 10 ⁻⁶
2400	1316	5.1 x 10 ⁻¹⁰	1.0 x 10 ⁻⁵
2600	1427	1.4 x 10 ⁻⁹	2.8 x 10 ⁻⁵
2800	1538	3.5 x 10 ⁻⁹	6.9 x 10 ⁻⁵
3000	1649	7.7 x 10 ⁻⁹	1.5 x 10 ⁻⁴

Assumes that R_{avg} = 71 μm.

Minimum Diffusion Coefficient

$$E_H = 54.1 \text{ Kcal/mole (10\% high)}$$

$$D_{HO} = 5 \times 10^{-4} \text{ cm}^2/\text{sec (600\% low)}$$

Retaining the low temperature values of

$$D_{LO} = 2.576 \times 10^{-14} \text{ cm}^2/\text{sec}$$

$$E_L = 5 \text{ Kcal/mole,}$$

we have

$$\begin{aligned} D &= D_{LO} \exp \left[-\frac{E_L}{RT} \right] + D_{HO} \exp \left[-\frac{E_H}{RT} \right] \\ &= 2.576 \times 10^{-14} \exp \left[-\frac{5 \times 10^3}{RT} \right] + 5 \times 10^{-4} \exp \left[-\frac{54.1 \times 10^3}{RT} \right] \\ &= 2.57 \times 10^{-14} \exp \left[-\frac{2.51635 \times 10^3}{T} \right] + 5 \times 10^{-4} \\ &\quad \exp \left[-\frac{27.22698 \times 10^3}{T} \right] . \end{aligned}$$

Dividing by 5×10^{-4}

$$\begin{aligned} D &= 5 \times 10^{-4} \left\{ 5.152 \times 10^{-11} \exp \left[-\frac{2.51635 \times 10^3}{T} \right] \right. \\ &\quad \left. + \exp \left[-\frac{27.22698 \times 10^3}{T} \right] \right\} . \end{aligned}$$

Dividing by $\exp \left[-\frac{27.22698 \times 10^3}{T} \right]$

$$\begin{aligned} D &= 5 \times 10^{-4} \exp \left[-\frac{27.22698 \times 10^3}{T} \right] \times \left\{ 1 + 5.152 \times 10^{-11} \right. \\ &\quad \left. \exp \left[\frac{24.7106 \times 10^3}{T} \right] \right\} . \end{aligned}$$

APPENDIX B

Steady State Analysis of Gas Release from Spherical Particles

The partial differential equation for radial diffusion in a sphere with constant diffusion coefficient D and constant generation rate A_o per unit volume is

$$\frac{\partial N}{\partial t} = A_o + D \left(\frac{\partial^2 N}{\partial r^2} + \frac{2}{r} \frac{\partial N}{\partial r} \right) , \quad (B-1)$$

at steady state $\frac{\partial N}{\partial t} = 0$, and

$$\frac{d^2 N}{dr^2} + \frac{2}{r} \frac{dN}{dr} = \frac{-A_o}{D} . \quad (B-2)$$

Multiplying by r^2 , we find

$$\frac{d}{dr} \left(r^2 \frac{dN}{dr} \right) = \frac{-A_o r^2}{D} . \quad (B-3)$$

The first integration gives

$$r^2 \frac{dN}{dr} = \frac{-A_o}{3D} r^3 + K_1 \quad (B-4)$$

where K_1 is a constant, determined by boundary conditions. A second integration gives

$$N = \frac{-A_o r^2}{6D} - \frac{K_1}{r} + K_2 \quad (B-5)$$

where K_2 is another constant.

The boundary conditions are two

- (1) At steady state, the total helium released is equal to the generation rate

$$J_{r=a} \cdot 4\pi a^2 = A_o \cdot \frac{4}{3} \pi a^3$$

$$J_{r=a} = \frac{A_o a}{3} \quad . \quad (B-6)$$

By Ficks law, $J = -D \frac{\partial N}{\partial r}$, so from Eq. (B-4)

$$J_{r=a} = -D \left. \frac{\partial N}{\partial r} \right|_{r=a}$$

$$= -D \left(\frac{-A_o a}{3D} + \frac{K_1}{a^2} \right)$$

or

$$J_{r=a} = \frac{A_o a}{3} - \frac{K_1 D}{a^2} \quad (B-7)$$

$\therefore K_1 = 0$, by comparison to (B-6).

- (2) The total amount of gas within the sample at steady state is X.

$$X = \int_0^a 4\pi r^2 \cdot N \cdot dr$$

$$= 4\pi \int_0^a \left(K_2 - \frac{A_o r^2}{6D} \right) r^2 dr$$

$$= 4\pi K_2 \int_0^a r^2 dr - \frac{4\pi A_o}{6D} \int_0^a r^4 dr$$

$$= \frac{4\pi K_2}{3} a^3 - \frac{4\pi A_o}{6D} \cdot \frac{a^5}{5}$$

$$= \frac{4\pi}{3} a^3 \left(K_2 - \frac{A_o}{10D} a^2 \right) , \quad (B-8)$$

or by rearrangement

$$K_2 = \frac{X}{\frac{4}{3}\pi a^3} + \frac{A_o a^2}{10D} ; \quad (B-9)$$

hence

$$\begin{aligned} N &= \frac{-A_o r^2}{6D} + \frac{A_o a^2}{10D} + \frac{X}{\frac{4}{3}\pi a^3} \\ &= \frac{X}{\frac{4}{3}\pi a^3} + \frac{A_o}{2D} \left(\frac{a^2}{5} - \frac{r^2}{3} \right) \\ &= N_{av} + \frac{A_o}{2D} \left(\frac{3a^2}{15} - \frac{5r^2}{15} \right) \\ &= N_{av} + \frac{A_o}{30D} \left(3a^2 - 5r^2 \right) \\ &= N_{av} + \frac{A_o a^2}{30D} \left(3 - \frac{5r^2}{a^2} \right) . \end{aligned} \quad (B-10)$$

If one assumes that at the surface of the sphere $N_{r=a} = 0$ for an experiment performed in a vacuum chamber, then it is possible to determine a steady state diffusion coefficient:

$$\begin{aligned} N_{r=a} &= 0 = N_{avg} + \frac{A_o a^2}{30D} \left(3 - 5 \right) \\ N_{avg} &= \frac{A_o a^2}{15D} \\ D/a^2 &= \frac{A_o}{15} \cdot \frac{1}{N_{avg}} . \end{aligned} \quad (B-11)$$

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