DISCLAIMER

This book was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

Reentry Thermal Testing of a General Purpose Heat Source Fueled Clad

D. E. Peterson

C. E. Frantz

Los Alamos Los Alamos National Laboratory
Los Alamos, New Mexico 87545

REENTRY THERMAL TESTING OF A GENERAL PURPOSE HEAT SOURCE FUELED CLAD

by

D. E. Peterson and C. E. Frantz

ABSTRACT

A General Purpose Heat Source (GPHS) module was exposed to heat treatments simulating an isothermal prelaunch condition, followed by thermal pulses corresponding to atmospheric reentry. Helium release rates were determined during each heating and modeled after simple diffusion theory. Following the tests, the module was examined metallurgically with no evidence of swelling of the cladding nor degradation of the fuel.

I. INTRODUCTION

The General Purpose Heat Source (GPHS) is to be used on the Galileo and Interplanetary Solar-Polar Missions. The GPHS unit uses the nuclear decay of 238-plutonium to provide heat that is converted via thermoelectric elements into electrical power for the mission instrumentation. We performed the simulation tests described in this report to verify that the GPHS heat sources can survive (1) the high-temperature changes associated with an approach to operational temperatures or (2) an accidental reentry into Earth's atmosphere without compromising the fuel containment.

The GPHS is modular in design. An individual GPHS fuel module (Fig. 1) consists of a 238-plutonium dioxide cylinder encapsulated in iridium surrounded by a graphite impact shell, insulation, and an ablator. The iridium capsule contains a vent assembly that allows for the release of helium generated by the plutonium's alpha decay. The helium is retained within the fuel crystal lattice under GPHS storage conditions but is released upon attainment of operational or reentry temperatures. The modular design of the heat source enables testing of individual fuel modules under conditions corresponding to those experienced by an actual flight unit.

All proposed missions involving GPHS are planned to take off from the Shuttle. The minimum time that the GPHS unit will be in the Shuttle prior to launch is 5 days, during which the clad will be at 1113°C. A previous reentry simulation¹ to 1700°C of GPHS

Fig. 1. GPHS module IRG-62.

capsule IRG-58 resulted in deformation of the cladding. Our investigations suggested that the vent operated as designed, but that it was swamped by the high helium release rate from the one-year-old fuel pellet (GP-17). The thermal history of IRG-58 was not typical of an operational heat source, so this second reentry-like exposure was carried out using more realistic conditions.

Our new test sample was IRG-62, a GPHS fueled capsule containing a 21-month-old Los Alamos fuel pellet, GP-19. The capsule was heated to launch pad operating temperature for 5 days and then subjected to a reentry heat pulse. Helium emissions from the fuel were monitored during these heat treatments and modeled by using simple diffusion theory. The diffusion parameters calculated from this analysis should be useful in predicting helium release rates for varying fuel treatment conditions.

The goals of this study were:

- (1) to subject a GPHS capsule to a heat treatment corresponding to a preflight exposure;
- (2) to subject the capsule to a simulated reentry ramp;
- (3) to test for capsule swelling, vapor transport, or degradation of the fuel, vent, or cladding; and
- (4) to model the helium release behavior, to enable future predictions for different fuels or temperatures.

II. THEORY

In spite of intensive studies of helium in UO_2 and PuO_2 over the past 20 years, neither mathematical descriptions proposed nor any acceptable mechanism is available to explain the migration/release of helium in these materials. The helium release behavior of the GPHS fuel was modeled in this study by assuming simple diffusion in which Fick's first law² applies;

$$P = -D \frac{\partial C}{\partial x}, \quad (1)$$

where P = gaseous flux,

D = diffusion coefficient, and

$\frac{\partial C}{\partial x}$ = concentration gradient.

Assuming spherically symmetrical diffusion and using spherical polar coordinates, the concentration dependence on time as derived from Eq. (1) is expressed as

$$\frac{\partial C}{\partial t} = D \left[\frac{\partial^2 C}{\partial r^2} + \frac{2}{r} \frac{\partial C}{\partial r} \right], \quad (2)$$

where r is the radial position in the sphere. Assuming that the radius of the releasing sphere is a and boundary conditions of (a) $C(r,0) = C_0$, implying initially uniform helium concentration, and (b) $C(a,t) = 0$, implying zero surface concentration, then Eq. (2) may be solved by Laplace transforms as discussed by Crank.³ The resulting equation is

$$F = 1 - \frac{6}{\pi^2} \sum_{n=1}^{\infty} \frac{1}{n^2} \exp[-n^2 \pi^2 D t / a^2], \quad (3)$$

where F is the fractional release, or the helium released at time, t , divided by the total amount initially present. Because the radius of the releasing unit is difficult to obtain, the effective diffusion constant, D' , equivalent to D/a^2 , is used in this study.

For the case where D (or D') varies with time, a parameter, τ , is defined as

$$\tau = \int_0^t D' dt. \quad (4)$$

It can be shown that τ simply replaces Dt/a^2 in Eq. (3). Then use of the approximation derived by Sung and Turnbaugh⁴ gives

$$F = 6 \left(\frac{\tau^n}{\pi} \right)^{1/2} - 3\tau \text{ for } \tau \leq 0.07. \quad (5)$$

For $\tau > 0.07$ the following approximation is applicable

$$F = 1 - \frac{6}{\pi^2} \sum_{n=1}^4 \frac{1}{n^2} \exp[-n^2 \pi^2 \tau]. \quad (6)$$

The measured fraction, released as a function of time and temperature during the initial hours of the experiment, was used in Eqs. (5) and (6) to obtain τ and therefore D' . The Newton-Raphson⁵ method of successive approximations was used to solve Eq. (6) for τ .

The simple diffusion model⁶ involving an interstitial diffusion mechanism would be expected to have a diffusion constant of the form,

$$D' = D'_0 \exp[-E_d/RT], \quad (7)$$

where E_a = activation energy,
 D'_0 = constant for diffusion,
 R = gas constant, and
 T = temperature (K).

The released helium fractions that were measured during the first four hours of the study were used to obtain τ and therefore D' . A plot of $\log D'$ vs $1/T$ yielded an E_a and D'_0 , which were used to calculate the time-and-temperature dependence of the helium released during the entire experiment.

The above gives a mathematical model with three parameters: a , E_a , and D'_0 , where a describes the geometry and E_a and D'_0 give the temperature dependence. This model has been used successfully to analyze much helium release data. However, the fit to the experimental data is not always good, particularly in cases such as reentry-type pulses when the temperature varies drastically with time. One modification that we have applied in the past to the reentry pulse data is the assumption of two mechanisms with different temperature dependence. This gives a second set of D' and E_a , or five total parameters in the model.

In the present case we have chosen another approach, namely, to assume a geometry effect with two different values of a . This means that there are two sizes of spheres in the problem and this can be said to relate to the fuel microstructure. In the GROG-type* fuel there are, in fact, large and small particles. Because a represents a sphere radius in the mathematics, it can be loosely interpreted as the radius of an irregular, approximately equiaxial particle and thus represents a distance the helium has to diffuse through in order to be released (for example, by reaching a surface connected pore or crack). In this two-particle model, the helium releasing units are assumed to be of two sizes with effective radii, a_1 and a_2 . Both releasing units have the same diffusion constant, D , activation energy, E_a , and constant of diffusion D_0 . However, because of the different radii, each has its own effective diffusion constant, D'_1 and D'_2 , respectively. The total fractional release, F_T , at any given time, can be expressed as

$$F_T = W_1 F_1 + W_2 F_2, \quad (8)$$

where F_1 and F_2 are the fractions of helium released from each unit and W_1 and W_2 are the weight fractions of the releasing units. Hence, the two-particle model requires five parameters a_1 , a_2 , E_a , D_0 , and W_1/W_2 —to describe the helium release behavior. By combining Eqs. (3) and (8) and substituting the individual parameters for each

releasing unit, it is possible to obtain values of D'_1 and D'_2 for given measurements of F_T as a function of time and temperature.

Philosophically, we endeavor to invent a mathematical model that will fit observed data with as few parameters as possible. A good model will fit a wide range of data. It is desirable that the parameters be derivable from some attributes of the fuel to enable predictive power without having to perform helium release measurements on each different kind of fuel. For example, if we could hold D'_0 and E_a constant for all fuel and correlate two or more a values with a particular microstructure, predictions would be facilitated.

We used the following equation, based on the decay rate and pellet weight to calculate the helium level generated by alpha decay in the fuel:

$$V_{He}(cc[STP]) = 0.8296(1 - \exp^{-0.0079t}) (G) (E), \quad (9)$$

where t = time (yr),

G = pellet weight (152.46 g), and

E = isotope enrichment factor (80% ^{238}Pu).

II. EXPERIMENTAL

A. Pre-reentry Heat Treatment

The fuel pellet, GP-19, was composed of a GROG-type mixture of shards (fired at high temperatures) and unfired powder and was hot-pressed at Los Alamos on March 14, 1979 and sintered on March 19, 1979. A description of the fabrication conditions has been previously published by Kent⁷. After sintering, the pellet had a density of 84.4%, weighed 152.462 g, with a 27.69-mm diam and 27.69-mm height. The capsule assembly, IRG-62, was fueled with GP-19 and welded on October 22, 1980.

The pre-reentry heat treatment was conducted at a clad temperature of 1133°C from December 1 through 5, 1980. The apparatus used a closed system equipped to collect evolved gases and to measure the pressure as a function of time (Fig. 2). The test assembly was a graphite (ATJ)** container enclosed in a welded tantalum can connected by a tantalum tube to a manifold external to the furnace tank. The manifold consisted of a Wallace-Tiernan gauge, a ballast volume, a gas-sampling

*A mixture of high-fired and unfired plutonia powders.

**ATJ graphite is a Union Carbide brand name.

Fig. 2. Gas collection apparatus.

bulb, and associated valves and fittings. A clad Pt/Pt-10Rh thermocouple, with the junction in contact with the iridium capsule, measured the temperature. Initial argon pressure in the system was 0.204 atm. Over a period of nearly four hours, the capsule temperature was increased to 1133°C test temperature to simulate the rise of the heat source after removal from its storage container. This temperature was held over five days, with periodic pressure monitoring. At the end of the study, we sampled the gas in the system at temperature with the gas-tap bulb and determined its composition with a mass spectrometer.

B. Simulation of GPHS Reentry Thermal Pulse

Simulation of the reentry ramp of GPHS module, IRG-62, was performed with the apparatus depicted in Fig. 3. The impact shell was replaced with two graphite caps. A Pt-Pt10Rh thermocouple was positioned between the caps onto the exterior of the iridium shell. The thermocouple reading on a digital voltmeter was recorded as a function of time by simultaneously videotaping the voltmeter and a clock. This approach was

Fig. 3. Diagram of apparatus used in the reentry simulation of IRG-62.

required because the voltmeter was at 2000 V relative to ground.

A CEC leak detector, attached to the furnace housing, measured helium levels. A picoammeter amplified the signal from the leak detector, which was displayed as a function of time on an X-Y recorder. We calibrated the detection system prior to the reentry study by flowing helium into the furnace over the range, (0.2-11 cc(STP)/min, and measuring the corresponding signal levels on the recorder. A Hastings-Teledyne flowmeter (Model NALL-100P), with a calibration factor of 1.44×10^{-10} amp/(cc[STP]/min), measured helium flow rates into the furnace.

After the system was evacuated to 10^{-9} amp and the initial temperature stabilized at 1258°C, the reentry ramp was performed by electron bombardment of the module. The power level was manually correlated with the time and temperature. The experimental thermal pulse was based on the theoretical minimum-gamma reentry ramp calculated at General Electric.⁸ This ramp is the most severe case of possible pulses with the steepest temperature dependence on time. The excellent agreement between the experimental and theoretical ramps is shown in Fig. 4.

C. Post Mortem

The capsule was radiographed and its dimensions measured with a micrometer before and after both the pre-reentry and reentry phases of this study. Radiograph

showed fuel cracking to be somewhat more extensive following the heat treatment but unchanged by the reentry exposure. The changes were relatively minor and probably had no relationship to helium release; capsule dimensions did not change during the study.

The capsule was sectioned circumferentially and the vent end removed. Helium was applied externally to the vent at 13.8-kPa differential pressure, while submerging the inside vent area in *Leak-Tek*. The formation of numerous bubbles on the inside of the vent was evidence that the vent remained open during the thermal exposures.

Examination of the fuel, following removal of the capsule vent end, showed a cracked and broken pellet. The recovered fuel weighed 153 g, of which 2.9 g was submitted for helium content analysis. The residual helium was determined by the combined techniques of high-temperature vacuum extraction and mass spectrometry. A fraction of the fuel weighing 144 g was sieved through 4 screens (with mesh sizes ranging from 6000 to 420 microns) to perform a particle size analysis. The remaining fuel (6.1 g), as well as the vent assembly and cladding, were metallographically analyzed. Representative samples from each region were extracted and mounted in an epoxy resin and then polished, etched, and microscopically examined.

IV. RESULTS

A. Experiment

1. Pre-reentry Heat Treatment. The times, total pressures, and capsule temperatures measured during the aging of IRG-62 are summarized in Table I, and variations in the temperature and pressure with time are plotted in Fig. 5. The pressure varied from the initial 0.204 atm of argon to a maximum of 0.642 atm following a 32.83-h exposure. The apparent upward break after 9 h is believed to be from the onset of the generation of carbon monoxide. Release of helium leveled off at that point, enabling the CO/CO₂ cyclic mechanism⁹ to begin. The pressure gradually declined in the last 90 h of exposure, reaching 0.622 atm at 120 h. This pressure decrease can be attributed to gettering of the carbon monoxide by the tantalum container.

Void volumes and temperatures estimated for three regions of the apparatus were as follows: that portion at an average temperature between the capsule and furnace

Fig. 4. GPHS reentry pulse applied to IRG-62 compared with theoretical pulse.

temperatures was 82.5 cc; that at an average between the capsule and room temperatures, 7.3 cc; and that at room temperature, 438.8 cc. The sample bulk had a void volume of 104.3 cc. The pressure drop, from taking the gas tap, was calculated using the estimated volume and temperature of each region. The calculated pressure of 0.506 atm compared favorably with the measured 0.509 atm. Table II lists the measured composition and partial pressures of the gas tap taken at the end of the aging. Combining these results with the previously discussed volume/temperature regions yielded a total helium volume of 115 cc(STP) released during the aging. Substituting into the decay the time since sintering (1.712 y), together with the other appropriate parameters, Eq. (9) yielded a total helium level of 136.4 cc(STP) generated since the pellet sintering. Out of this total 85.4% was released during the pre-reentry heat treatment at 1133°C.

The dimensions of the iridium cladding were essentially unchanged by the aging treatment. The inner surface of the graphite container opposite the capsule's vent contained a single pit, evidence that an oxidizing species had vented during the exposure. The interior of the tantalum container had a gold appearance that could have been from reaction with the carbon monoxide (since tantalum carbide has a gold color).

2. Thermal Reentry Test. Exposure of IRG-62 to a reentry ramp was conducted at times and temperatures listed in Table III. Each measured shows good agreement with the corresponding value calculated from the

TABLE I. Times, pressures, and temperatures measured during the pre-reentry heat treatment of the GPHS capsule IRG-62.

Time (hrs)	Pressure (atm)	Capsule T (°C)	Time (hrs)	Pressure (atm)	Capsule T (°C)
0	0.204	472	6.00	0.420	1132
0.17	0.205	519	6.67	0.429	1132
0.33	0.208	577	7.67	0.442	1132
0.50	0.209	632	8.72	0.458	1132
0.67	0.210	683	14.00	0.580	1130
0.83	0.212	728	15.09	0.596	1130
1.00	0.213	769	22.83	0.642	1129
1.17	0.214	810	23.83	0.646	1118
1.33	0.216	854	24.83	0.647	1128
1.67	0.217	874	25.83	0.650	1130
1.83	0.218	904	26.83	0.650	1130
2.00	0.221	910	28.83	0.653	1130
2.17	0.222	945	29.83	0.654	1129
2.33	0.225	952	32.83	0.655	1128
2.50	0.229	990	47.00	0.650	1127
2.67	0.237	997	51.00	0.649	1127
2.83	0.247	1037	56.92	0.646	1128
3.00	0.263	1043	71.00	0.639	1127
3.17	0.282	1081	75.92	0.638	1127
3.33	0.303	1087	79.83	0.638	1127
3.50	0.322	1113	95.17	0.630	1126
3.67	0.342	1118	98.75	0.630	1126
3.83	0.356	1133	104.75	0.629	1125
4.00	0.362	1133	119.83	0.622	1123
5.00	0.403	1132

Fig. 5. The total pressure over IRG-62 increased to a maximum at 32 h.

TABLE II. Composition and partial pressures of gas tap drawn at 1133°C after 5 days during the pre-reentry heat treatment of IRG-62.

Species	mol%	Pressure (mm)
Hydrogen	<0.1	---
Helium	44.0	208
Carbon monoxide	21.0	99
Nitrogen	<0.1	---
Methane	<0.1	---
Oxygen	<0.1	---
Argon	35.0	165
Carbon dioxide	<0.1	---

TABLE III. Times and temperatures measured for the reentry simulation of IRG-62. (Temperatures calculated from a minimum gamma-reentry ramp are also listed.)

Time (min)	Meas. T (°C)	Calc. T (°C)	Time (min)	Meas. T (°C)	Calc. T (°C)
0	1258	1258	5.25	1549	1547
0.25	1259	1258	5.50	1537	1533
0.50	1262	1260	5.75	1517	1517
0.75	1266	1263	6.00	1493	1496
1.00	1272	1270	6.25	1469	1471
1.25	1280	1284	6.50	1450	1442
1.50	1325	1310	6.75	1429	1391
1.75	1347	1346	7.00	1382	1351
2.00	1397	1388	7.25	1325	1316
2.25	1437	1432	7.50	1271	1280
2.50	1477	1468	7.75	1232	1241
2.75	1499	1500	8.00	1204	1209
3.00	1523	1523	8.25	1175	1174
3.25	1544	1541	8.50	1147	1141
3.50	1551	1553	8.75	1121	1106
3.75	1563	1561	9.00	1095	1075
4.00	1567	1564	9.25	1072	1047
4.25	1570	1567	9.50	1050	1018
4.50	1570	1565	9.75	1030	991
4.75	1560	1563	10.00	1015	962
5.00	1553	1556			

minimum gamma reentry pulse; therefore, the desired reentry ramp appears to have been followed satisfactorily throughout the experiment.

The maximum helium release rate observed during the reentry pulse was 7×10^{-3} cc(STP)/min, and the total helium released during the pulse was 0.5 cc(STP). The helium level generated between the end of the heat treatment and the reentry exposure, as calculated using Eq. (9), was about 1 cc(STP). Hence, it appears that no additional helium was released during the reentry pulse beyond that normally generated after the pre-entry heat treatment.

The residual helium content in the fuel following the reentry exposure, as measured mass spectrometrically, was 0.03 cc(STP)/g fuel, or a total of 4.5 cc(STP). Based on the helium generation rate as expressed in Eq. (9), a total helium level of 4.4 cc(STP) should have been generated between the end of the reentry ramp and the analysis, hence there was no helium remaining in the fuel following the thermal reentry simulation. Because there was no helium released during the reentry ramp (other than that generated since the heat treatment), the 84.6% of the calculated inventory released during the pre-entry heat treatment appears to have exhausted the helium content. It is assumed that the difference of 15.4% was released during the fuel-storage and capsule-welding stages.

3. Theoretical Analysis. We used the single- and two-particle models to analyze the pre-reentry heat treatment data. An initial 15.4% helium inventory loss, prior to the treatment, was assumed, based upon the experimental observations discussed in the previous section. Using the helium released as a function of time and temperature in Eqs. (4) and (5), an activation energy, E_a , and an effective diffusion coefficient, D'_0 , were calculated. Values of E_a and D'_0 calculated from the single-particle model were 281.5 kJ/mol and $1.023 \times 10^8 \text{ sec}^{-1}\text{cm}^{-2}$, respectively. When two sizes of releasing units, with a radius ratio of 5 and a weight-fraction ratio of 0.42, were used in the two-particle model, an E_a of 321.8 kJ/mol and D'_0 values of the large and small particles of 2.387×10^8 and $5.968 \times 10^9 \text{ sec}^{-1}\text{cm}^{-2}$, respectively, were obtained. These values were then used to calculate the time dependence of the helium pressure for the pre-reentry treatment. The results are plotted in Fig. 6 and indicate that the two-particle model appears more consistent with the experimental measurements.

Fig. 6. Pressure vs time predicted from single- and two-particle models and measured during pre-reentry heat treatment of IRG-62.

4. **Post Mortem.** Table IV lists the dimensions of the iridium capsule before and after the heat treatment and reentry exposures. Ranges in the three readings performed on each set are also given, the slightly larger dimensions shown in the last column being within the error of the measurements. There is no pattern in the dimensional differences. If the internal pressure had caused swelling, the center height measurement would have shown the greatest growth.

The capsule is shown in Fig. 7 after removal from the reentry furnace. Other than the normal marks that result from handling, the capsule appeared unaffected by its thermal exposures. The vent showed no discoloration nor other evidence of any vapor-transported condensate. Metallographic examination of the vent structure showed it was unaffected by the thermal tests (Fig. 8).

A typical microstructure of the iridium capsule from IRG-62 is shown in Fig. 9. The grain structure and size are as expected for the low-temperature aging and short-term reentry exposures. Approximately 23 and 29 grains were measured over the 0.025-in. wall thickness for the vented and blind end caps, respectively.

Inspection of the fuel following removal of the capsule vent end, showed the pellet to be cracked and broken (Fig. 10). This observation agrees with the radiographic analysis.

The microstructure of the fuel is shown in Fig. 11. The mixture of large and small fragments is always observed in fuel made by the GROG process. However, its character is more pronounced in this pellet and a particularly open microstructure is very evident.

The separation of the fuel according to particle size yielded the results shown in Table V. Each fraction is expressed as the weight per cent remaining after sieving through each mesh size.

V. DISCUSSION

This test showed that we can expect most of the stored helium to be released from the fuel without damage to the capsule during its residence in the Shuttle prior to launch. The data showed that no helium remained in the fuel after the reentry pulse, and that little or no helium, above the amounts generated between the heat treatments, was released during the reentry pulse.

A total of 84.6% of the helium generated in the GP-19 pellet during the 21-month storage period was released during the aging or reentry exposures although no helium remained in the fuel following the tests. Initially it was proposed that the additional helium was released during storage of the fuel pellet. However, the storage

TABLE IV. Dimensions of IRG-62 capsule before and after heat treatment and reentry pulse.

Diameter	Pre-aging (mm)	Post-aging (mm)	Post-reentry (mm)
Vent end	29.72 ± 0.05	29.72 ± 0.05	29.77 ± 0.08
Weld	29.84 ± 0.02	29.84 ± 0.08	29.90 ± 0.05
Blind end	29.69 ± 0.02	29.69 ± 0.02	29.74 ± 0.05
Height-center	30.43	30.40	30.45
Height-edge	30.30 ± 0.02	30.30 ± 0.02	30.38 ± 0.05

(a)

(b)

Fig. 7. Capsule IRG-62 appeared to have been unaffected by the aging and reentry thermal exposures.

(a)

(b)

Fig. 8. The vent in IRG-62 was unaffected by the thermal exposures (50X).

Fig. 9. The microstructure of the iridium capsule in IRG-62 was Typical (100X).

temperature of a typical GPHS pellet stored under the same conditions as GP-19 was measured and found to be too low to have released significant helium. Another possible factor could be loss of helium due to alpha recoil, which occurs during decay of ^{238}Pu . The alpha-

particle has an energy of 5.5 MeV and can move a distance of about 12 microns in the fuel. A pellet with small grains, such as GP-19, would allow more alphas to leave the grain surfaces. Some fraction of the alpha-particles would not be readsorbed into other grains and would be ultimately released from the pellet. Additional helium losses probably occurred due to localized heating of the fuel in the weld region.

The presence of carbon monoxide complicated the analysis of the helium released during the heat treatment. Most of the CO was assumed to have been generated following the first four hours of the analysis. This assumption appears reasonable because the movement of helium out of the vents would have restricted any back-diffusion of CO into the capsule. After this helium release rate decreased, the CO cyclic mechanism could take place.

The fuel showed no evidence of severe cracking or other microstructural degradation due to helium release during the pre-reentry heat treatment. Neither the pre-reentry nor reentry exposures resulted in any serious degradation of the fuel microstructure, as was evident from the radiographic examination before and after each treatment.

The absence of any vapor-deposited material in the vent or on the exterior of the capsule indicated that the fuel was relatively free of impurities. *We emphasize that*

(a)

(b)

Fig. 10. Fuel pellet GP-19 was found to be broken following thermal exposures: (a) with capsule top removed and (b) after removal from capsule.

(a)

(b)

(c)

(d)

Fig. 11. Microstructure of GP-19 fuel from capsule IRG-62 showing duplex structure characteristic of the GROG process:
 (a) outer region (50X); (b) outer region (250X); (c) center region (50X); (d) center region (250X).

TABLE V. Particle-size distribution of IRG-62 following re-entry exposure.^a

Screen (microns)	Fraction (wt%)
6000	94.11
2000	3.89
841	0.98
420	0.38
>420	0.64

^aEach fraction represents the amount remaining after sieving through the mesh size listed.

this purity must be maintained in the fuel used in the actual flight units. Any condensate collecting in the vent structure would restrict the helium released during the pre-reentry treatment and during any reentry thermal ramps, resulting in swelling of the capsule.

Application of the two-particle model to the helium release, which occurred during the pre-reentry heat treatment, appeared to fit the data better than the single-particle model. This new approach may be helpful in the future in developing models that include the effect of particle-size differences.

ACKNOWLEDGMENTS

We wish to recognize the contributions of D. Pavone for the pre-reentry treatment, F. Schonfeld for the metallographic analysis, J. S. Starzynski and R.N.R. Mulford for the theoretical analyses, D. Garinger for the particle sizing, G. Langner of group M-1 for the radiographic examinations, and D. Vance of group CMB-1 for the residual helium analysis. We also appreciate the support of the Department of Energy, office of Coordination and Special Projects.

REFERENCES

1. W. J. Maraman, "General-Purpose Heat Source Project and Space Nuclear Safety and Fuels Program," Los Alamos Scientific Laboratory report LA-8713-PR (August 1980).
2. R. M. Barrer, *Diffusion in and Through Solids* (Cambridge University Press, New York, 1951).
3. J. Crank, *The Mathematics of Diffusion* (Oxford University Press, Inc., New York, 1967).
4. P. Sung and J. Turnbaugh, "Calculation of Diffusivity and Solubility with Error Analysis for Diffusion in a Sphere," *J. Appl. Phys.*, **39**, 346 (1968).
5. L. G. Kelley, *Handbook of Numerical Methods and Applications* (Addison-Wesley Publishing Co. Inc., Reading, Mass., 1967), p. 90.
6. P. Shewmon, "Atomic Theory of Diffusion," in *Diffusion in Solids* (McGraw-Hill Book Co., New York, 1963), Chap. 2.
7. R. A. Kent, "LASL Fabrication Flow Sheet for GPHS Fuel Pellets," Los Alamos National Laboratory report LA-7972-MS (August 1979).
8. R. Hartman, Letter, 3 September 1980, General Electric Co., Philadelphia, PA 19101.
9. D. E. Peterson and D. Pavone, "A Study of Carbon Monoxide Production from a Multi-Hundred Watt Fuel Sphere Assembly," Los Alamos National Laboratory report LA-8414-MS (June 1980).