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Reentry Thermal Testing of Light-Weight Radioisotope Heater Units

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REENTRY THERMAL TESTING
of
LIGHT-WEIGHT RADIOISOTOPE HEATER UNITS

by
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ABSTRACT

Two Light-Weight Radioisotope Heater Units (LWRHU) were exposed to thermal ramps simulating atmospheric reentry. The helium release rates were measured during each test and modeled after simple diffusion theory. The reentry pulses did not result in swelling of the claddings or degradation of the fuel pellets.

I. INTRODUCTION

The Light-Weight Radioisotope Heater Unit (LWRHU) was designed by the Los Alamos National Laboratory¹ to warm instrumentation on interplanetary satellites. The first application of the unit is on the Galileo Mission to Jupiter. During an accidental reentry into the Earth's atmosphere, a LWRHU would be exposed to a thermal ramp in which a fuel temperature above 1600°C could be attained within a few minutes. To determine the response of a LWRHU to this severe treatment and to provide data useful in theoretical modeling, the reentry simulation tests described in this report were performed.

A schematic of a LWRHU capsule is shown in Fig. 1. The fuel is a 238-plutonium dioxide pellet (weighing about 2 g) encapsulated in iridium. The fuel capsule is contained within a graphite impact shell. Gases released from the fuel pass through a vent assembly consisting of a frit (serving as a particulate filter) and an orifice (0.6-mm diam).

Fig. 1. Schematic of a Light Weight Radioisotopic Heater Unit (LWRHU).

Helium is being continuously generated by alpha decay of the ^{238}Pu isotope. Under storage or operating conditions associated with a LWRHU, most of the helium is retained within the fuel lattice. When a LWRHU is subjected to a simulated atmospheric-reentry thermal pulse, the stored helium is released in a burst that can have several negative effects: (1) The integrity of the fuel body can be degraded, leading to additional fines generation upon impact. (2) The associated pressure buildup within an improperly vented LWRHU capsule could result in swelling of the cladding and compromise the fuel containment. (3) Finally, the released helium can fill the insulator gaps, leading to an unacceptable rise in the clad temperature. To test for these effects and to provide data useful in modeling a LWRHU reentry, we performed the experiments described herein.

The study was conducted by applying a simulated reentry ramp to fuel pellets (aged over seven years) contained in actual LWRHU components. Thermal reentry ramps were applied to two LWRHUs, during which the helium release rates were measured as functions of time and temperature. A semiempirical model, assuming simple diffusion, was used to analyze the release behavior. We measured the extent of swelling of the capsules, determined the fuel integrity, and analyzed the residual helium content of the fuel.

II. THEORY

The helium release rates measured in this study were analyzed based on Fick's laws of simple diffusion, an approach previously discussed by Peterson and Frantz.² Using the fractional release, F , as a function of time, we calculated an effective diffusion constant, D' ($= D/a^2$, where a is the effective radius of the releasing unit). Variations in D' with temperature and therefore time, t , are accounted for by using the parameter, τ , defined as

$$\tau = \int_0^t D' dt \quad (1)$$

Assuming that Fick's laws of diffusion apply, one can derive the expression

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

from which a value of τ may be determined at a time, t , from a corresponding F value. Thus a τ vs time curve is

generated that has a derivative at any point equivalent to the effective diffusion constant.

The temperature dependence of D' was used in Eq. (3) to obtain an activation energy, E_a , a constant for diffusion, D'_0 , and the gas constant, R .

$$D' = D'_0 \exp \{-E_a/RT\} \quad (3)$$

The magnitude of the activation energy provides an indication of the energy barrier for diffusion of the helium atoms within the plutonia lattice.

III. EXPERIMENTAL

The reentry simulations were performed with the apparatus depicted in Fig. 2. The Centorr furnace was heated by a tungsten mesh element and the system was evacuated with an oil diffusion pump containing a liquid nitrogen trap and backed by a forepump. A CEC leak detector was connected to the furnace with a leak valve apportioning the helium released between the detector and the furnace. A picoammeter amplified the signal from the leak detector, which was displayed on an x-y recorder containing an internal time base. The helium release rate was calibrated with a Teledyne-Hastings flowmeter prior to the reentry simulated pulse. Tank

Fig. 2. Apparatus for study of helium release from radioisotopic heater units.

helium was flowed into the furnace through the flowmeter over a range of 9 to 24 cc(STP)/min. This range corresponded to anticipated helium release rates from plutonia pellets based on previous helium release measurements.^{3,4} The flowmeter reading was correlated with the leak detector signal. The leak valve between the furnace and the detector was set so as not to flood the detector at the highest helium release rate.

The LWRHU temperature was determined with a Pt-Pt/10Rh thermocouple touching the capsule base. The temperature readings were continuously monitored with a Brown chart recorder, the range of which was calibrated with a millivolt power source. The thermocouple readings were correlated with temperatures measured simultaneously with a Leeds and Northrup automatic pyrometer sighted onto the LWRHU wall. The desired temperature ramp was achieved by manually adjusting the power level to the heating element. An LWRHU fueled with a plutonia pellet was used to optimize the temperature ramp. The reentry ramps actually applied were significantly more severe than the reentry pulse (AP2737) calculated at Applied Physics Laboratory.

Two sets of ²³⁸PuO₂ pellets (14367), weighing 1.90 and 2.10 g, were selected from a batch that had been aged for 7.83 and 7.93 yrs, respectively. These pellets were encapsulated in LWRHUs designated 004 and 027. Prior to encapsulation the vent assemblies had flow rates of 12.4 and 6.57 mole/sec/atm, respectively, as measured at Mound Facility. Each LWRHU was placed on a Pt-10Rh stand in the center of the furnace and the system evacuated to 1×10^{-4} torr. The temperature was increased to 400°C and allowed to stabilize at this temperature for one hour. Each LWRHU was then subjected to the simulated reentry pulse.

IV. RESULTS

Exposure of the LWRHU 004 to the reentry ramp resulted in flooding of the leak detector. The temperature and detector levels are shown as a function of time in Table I. A large helium signal was noted at 850°C, and detector flooding occurred at about 1270°C.

The leak valve was partially closed and the furnace recalibrated. The time dependence of the helium release from LWRHU 027 is shown in Fig. 3. Also displayed are the measured and calculated temperature ramps. The first detectable helium signal was noted at 910°C with the release rate maximizing at 1288°C. The values

Table I. Temperatures and Leak Rates Measured During Exposure of RHU 004 to a Thermal Reentry Pulse.

T(°C)	Leak Rate
436	0.13
659	0.36
846	---
1023	120.0
1266	160.0
1567	120.0
1580	120.0
1515	50.0

Fig. 3. Time dependence of helium release and temperature in reentry simulation of RHU 027.

corresponding to the measured temperatures and release rates are given at 1-sec intervals in Table II.

The helium inventory contained in each set of pellets was calculated from the alpha decay rate as given by the equation,

$$n = 3.701 \times 10^{-3} (1 - e^{-0.0079t}) (G)(P) , \quad (4)$$

where n = helium amount (moles),

t = time (yrs),

G = mass PuO_2 (g), and

P = per cent ^{238}Pu enrichment/100 = 0.809.

The total helium levels calculated for LWRHUs 004 and 027 were 337 and 378 μmole , respectively. The residual helium levels analyzed following reentry were 17 and 49 μmole , respectively, correlating with a total release of 95 and 87% of the helium inventories. Integration of the total helium released from LWRHU 027 gave 330 μmole , or 88% of the calculated inventory.

The values of the fractional helium released, the logarithm of the effective diffusion constant, and the

reciprocal temperature used in modeling the release behavior of LWRHU 027 are given in Table III. A plot of the $\log D'$ vs the reciprocal temperature is shown in Fig. 4. There appears to be a definite break at about 1100°C. Least-squares analysis performed on the two respective data sets yielded the results shown in Table IV, where the slope and intercept of each line is given along with the corresponding activation energy, E_a , and constant for diffusion, D'_0 .

The external dimensions of LWRHUs 004 and 027 measured before and after the reentry pulses are given in Table V. No swelling of either capsule from the reentry pulse was evident. Each capsule top was sawed off and the vent and fuel pellets examined. The vent assemblies showed no degradation nor any evidence of vapor transport in the frits or vent holes. Fuel-pellet integrity appeared to be excellent following the reentry.

Based on the alpha-decay rate, approximately 8.2 and 8.46 cc(STP) of helium were calculated to have been resident in the fuel of LWRHUs 004 and 027. Following reentry, residual helium levels of 0.4 and 1.1 cc remained

Table II. Times, Temperatures, and Helium Release Rates Associated with Thermal Reentry Pulse Given LWRHU 027.

Time (sec)	Temp (°C)	Rate ($\mu\text{mole/sec}$)	Time (sec)	Temp (°C)	Rate ($\mu\text{mole/sec}$)	Time (sec)	Temp (°C)	Rate ($\mu\text{mole/sec}$)
0	528	---	37	865	2.3387	57	1235	18.410
5	535	---	38	883	2.5786	58	1254	21.408
10	552	---	39	901	2.7585	59	1272	26.805
15	578	---	40	919	2.9983	60	1291	25.905
20	620	---	41	938	3.2982	61	1311	22.607
22	637	---	42	956	3.5980	62	1330	19.909
23	663	0.00090	43	974	3.7779	63	1350	15.411
24	674	0.17990	44	992	4.0177	64	1369	11.813
25	685	0.26985	45	1010	4.4506	65	1389	8.8149
26	695	0.41976	46	1029	4.7973	66	1408	6.7162
27	706	0.53970	47	1048	5.0971	67	1428	5.5169
28	717	0.68961	48	1066	5.4840	68	1447	3.7179
29	727	0.83953	49	1085	6.4164	69	1463	1.9189
30	738	0.98952	50	1104	7.4355	70	1482	---
31	756	1.1394	51	1122	9.4146	75	1551	---
32	774	1.3193	52	1141	10.014	80	1586	---
33	792	1.5291	53	1160	13.838	85	1606	---
34	811	1.7390	54	1179	13.312	90	1610	---
35	829	1.9189	55	1197	11.813	95	1605	---
36	847	2.1588	56	1216	14.212	100	1596	---

Table III. Release Integral and Diffusion Constant Calculated for Reentry of LWRHU 027 as a Function of Time and Reciprocal Temperature.

Time (sec)	Release Integral	-log D' (sec ⁻¹)	10 ⁴ /T (K ⁻¹)	Time (sec)	Release Integral	-log D' (sec ⁻¹)	10 ⁴ /T (K ⁻¹)
22	0	0	10.99	49	0.17326	3.135	7.36
30	0.01064	5.261	9.89	50	0.19295	3.017	7.26
31	0.01366	5.083	9.72	51	0.21788	2.853	7.17
32	0.01715	4.916	9.56	52	0.24440	2.762	7.07
33	0.02120	4.755	9.39	53	0.31629	2.490	6.89
34	0.02580	4.609	9.23	54	0.31629	2.490	6.89
35	0.03089	4.482	9.08	55	0.34757	2.477	6.80
36	0.03660	4.354	8.93	56	0.38521	2.332	6.72
37	0.04279	4.246	8.79	57	0.43395	2.142	6.63
38	0.04962	4.135	8.65	58	0.49064	1.984	6.55
39	0.05693	4.041	8.52	59	0.56162	1.774	6.47
40	0.06487	3.944	8.39	60	0.63021	1.661	6.39
41	0.07360	3.843	8.26	61	0.69007	1.590	6.32
42	0.08313	3.748	8.14	62	0.74279	1.507	6.23
43	0.09313	3.672	8.02	63	0.78360	1.471	6.16
44	0.10377	3.592	7.91	64	0.81488	1.432	6.09
45	0.11555	3.496	7.79	65	0.83822	1.392	6.02
46	0.12703	3.459	7.68	66	0.85600	1.321	5.95
47	0.14175	3.300	7.57	67	0.87062	1.160	5.88
48	0.15627	3.255	7.47	68	0.88046	0.962	5.81

Fig. 4. Plot of log D' vs the reciprocal temperature determined from analysis of RHU 027.

Table IV. Slopes and Intercepts of Log D' vs $1/T$ Plots for Low- and High-Temperature Regions. Corresponding Activation Energies, E_a , and Constants of Diffusion, D'_0 , are Also Listed.

Parameter	Low Temperature	High Temperature
No. pts.	19	20
T range($^{\circ}$ C)	727 - 1104	1066 - 1447
Slope	-8073.8 $\pm$ 120.0	-13667.1 $\pm$ 386.7
Intercept (log D'_0)	2.8096 $\pm$ 0.1033	6.9355 $\pm$ 0.2535
E_a (KJ)	154.6 $\pm$ 2.3	261.7 $\pm$ 7.4

in the fuel pellets of the respective LWRHUs as determined by high-temperature collection and mass spectrometry. These levels correlate to releases of 95 and 87% of the total helium inventories, the latter value agreeing well with the 88.5% determined from the total release measured during the ramp of LWRHU 027.

V. DISCUSSION

The helium release rates measured from both LWRHUs were higher than anticipated, based on previous measurements on microspheres or pellets.^{3,4} The detector flooding in the LWRHU 004 reentry was unexpected and shows the very fast release of the stored helium during exposure to thermal pulse. This experiment yielded approximate temperatures at which the helium release began and maximized, as well as an estimate of the maximum release rate.

The leak valve between the detector and furnace was set to detect the onset of helium release while still not flooding the detector at the maximum release rate. The x-y recorder went slightly off scale during the LWRHU 027 reentry simulation but fortunately made a fast recovery. During this period of flooding the helium release was determined by extrapolation of the rate vs time curve.

The tests performed in this study were overtests of the actual reentry of an LWRHU. The simulated reentry pulses were significantly more severe than the worst case reentry ramp calculated at the Applied Physics Laboratory, the temperature rise being significantly steeper and the maximum temperature over 100° higher. The vent rates extended down to the minimum specification of $0.45 \mu\text{mole/sec.}^1$

The maximum storage period anticipated for LWRHU fuel is 4.5 yrs whereas the fuel used in this study was over 7 yrs old. Therefore, the internal pressure

Table V. Dimensions (mm) of LWRHU Capsules Given Thermal Ramps.^a

Parameter	LWRHU-004		LWRHU-027	
	Before (mm)	After (mm)	Before (mm)	After (mm)
Length	12.475	12.460	12.500	12.500
Upper diam	8.300	8.310	8.290	8.305
Upper Land diam	8.515	8.540	8.530	8.535
Lower Land diam	8.575	8.575	8.570	8.565
Lower diam	8.350	8.355	8.330	8.335

^a ± 0.010 mm.

build-up of helium must be greater in our test cases. The absence of any observable swelling of the LWRHU capsules indicates that the vent assemblies can handle the helium released during the atmospheric reentry conditions.

Any pumping rate effects from the helium release during the ramp would have been offset by the calibration. Most background gases emitted by the system or LWRHU would have been pumped off during the initial heating period at 400°C. Very little change was noted in the furnace pressure while optimizing the reentry ramp, which supports the lack of background gas effects. The helium flow calibration performed at ambient temperature was related to the release rates measured at temperatures associated with the thermal pulse. The effect of these temperature differences on the helium release rates determined in this study are believed to be negligible because of the low pressures involved.

Almost the entire helium inventory was released during the experimental ramps. The difference in the total amounts released during the two reentries was most likely the result of a more severe ramp in the LWRHU 004 exposure. The temperature during this ramp was not controlled and monitored as carefully as in the subsequent pulse given LWRHU 027.

It is interesting to note that the fuel pellets survived the reentry pulses intact, even after being subjected to severe temperature excursions and burdened with a relatively large helium inventory. The break observed in the log D' vs $1/T$ curve at about 1100°C is not unusual; Mulford and Mueller³ observed similar discontinuities in the temperature dependence of the diffusion coefficient, as did Angelini and coworkers.⁴ Diffusion of the helium through the grains, with entrapment at the grain boundaries, probably predominates at temperatures below 1100°C, but at higher temperatures, the helium trapped

in the grain boundaries would be released, accounting for the break. The activation energy of 261 kJ agrees well with the value of 282 kJ determined during the heat treatment² of the General Purpose Heat Source capsule, IRG-62.

REFERENCES

1. R. E. Tate, "The Light Weight Radioisotope Heater Unit (LWRHU). A Technical Description of the Reference Design," Los Alamos National Laboratory report LA-9078-MS (December 1981).
2. D. E. Peterson and C. E. Frantz, "Reentry Thermal Testing of a General Purpose Heat Source Fueled Clad," Los Alamos National Laboratory report LA-9227 (March 1982).
3. R. N. R. Mulford and B. A. Mueller, "Measurements of Helium Release from Materials Containing $^{238}\text{PuO}_2$," Los Alamos National Laboratory report LA-5215 (July 1973).
4. P. Angelini, R. E. McHenry, J. L. Scott, W. S. Ernest, and J. W. Prados, "Helium Release from $^{238}\text{PuO}_2$ Microspheres," Oak Ridge National Laboratory report ORNL-4507 (March 1970).

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