RECEIVED BY DTIC FEB 16 1970

3416

SC-DC-69-1892B
November 1969

HELIUM RELEASE DATA FOR $^{238}\text{PuO}_2$
MICROSPHERES

W. A. Stark, Jr., 5321

MASTER

THIS DOCUMENT CONFIRMED AS
UNCLASSIFIED
DIVISION OF CLASSIFICATION
BY J. H. Cahn
DATE 2/24/70

P4234

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

SANDIA LABORATORIES

DISCLAIMER

This report 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.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

LEGAL NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission

A. Makes any warranty or representation, expressed or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or

B. Assumes any liabilities with respect to the use of, or for damage resulting from the use of any information, apparatus, method, or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission, or employee of such contractor, to the extent that such employee or contractor of the Commission, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor.

SC-DC-69-1892B

HELIUM RELEASE DATA FOR $^{238}\text{PuO}_2$ MICROSPHERES

W. A. Stark, Jr., 5321
Sandia Laboratories, Albuquerque

The information in SC-DC-69-1892, dated July 1969, was
presented at the AEC-SNS Helium Release
Information Meeting
August 7, 1969,
Germantown, Pa.

November 1969

ABSTRACT

The results of mass spectrometer analyses of the helium gas released during annealing of production grade, plasma torch fused $^{238}\text{PuO}_2$ microspheres are presented for several fuel samples at varying temperatures. Each data set consists of a temperature-time curve, a release rate-time curve, a fractional total release-time curve, and a diffusion coefficient-time curve. The fractional release curve was obtained by integrating the release rate-time curve, and the diffusion coefficient is calculated from the fractional release curve assuming a simple diffusion model with a time-varying diffusion coefficient.

NOTE: This report updates and supersedes SC-DC-69-1892, dated July 1969, same title, to correct for a computational error and time-adjusted helium inventories. The correction affected many of the computer-produced graphs. Destroy copies of SC-DC-69-1892.

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

ACKNOWLEDGMENTS

The results reported in this work were obtained from experiments performed at Oak Ridge National Laboratory and Mound Laboratory, Monsanto Research Corporation, under the direction of Ray McHenry and Ronald Zielinski, respectively. The author is indebted to these investigators and their co-workers for their hard work, expertise, and clarifying comments.

The author also wishes to acknowledge many enlightening discussions with, and insights from, Read Holland, Clyde Northrup, and David Williams, Sandia Laboratories; and Peter Angelini, John Prados, and Jim Scott, ORNL.

HELIUM RELEASE DATA FOR $^{238}\text{PuO}_2$ MICROSPHERES

Introduction

In the past year and one-half, a large amount of experimental data has been accumulated in an attempt at defining the helium gas release from production grade, plasma-torch fused $^{238}\text{PuO}_2$ microspheres. In the present report, that portion of the data obtained from annealing experiments performed at ORNL and MRC has been collected, manipulated, and partially analyzed assuming a simple diffusion model. The results are presented in the form of graphs summarizing the behavior in each experiment. For more detailed examination of the data, a magnetic tape containing the numerical data is available.

Discussion

Since the initial experiments of Mulford,¹ which showed that large amounts of helium gas can be stored, enough information has been accumulated to indicate that, at least at temperatures less than about 1400°C , the release of helium gas from $^{238}\text{PuO}_2$ microspheres does not occur by a simple diffusion mechanism. Rather, the kinetics of release indicate some modified diffusional mechanism. Nonetheless, from a phenomenological viewpoint, a simple diffusion model comes close to describing the observed behavior, and in view of engineering and design expediency, the data contained in this report were analyzed using a simple diffusion model which must suffice until future study yields a more definite description.

As has been described in detail elsewhere,² the present data were generally obtained by mass spectrometer analysis of the helium release rate as the fuel sample was raised to some elevated temperature and held there. The raw experimental data, then, are temperature-time data

and release rate-time data (corrected to a sample size of 1 gram). The release rate as a function of time is numerically integrated to obtain the total amount of helium released as a function of time. This last quantity is then divided by the total quantity of helium initially in the sample to obtain the variable known as the fractional release $F(t)$. The total inventory of helium used was the greater value of either the amount measured in a separate experiment or the amount obtained in the given experiment; this manipulation eliminates the problem of fractional releases > 1.0 .

Given the fractional release as a function of time, one can calculate an effective diffusion coefficient, D' ($=D/R^2$, where R is the effective radius of the particle). The variation of D' with temperature is accounted for by using the standard analysis for time-dependent diffusion coefficient.³ Then, for a spherical particle of radius R and assuming only radially directed diffusion and no helium generation during the annealing,⁴ one obtains

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

for which the following are reasonably good approximations:⁵

$$F(\tau) \approx \frac{6}{\sqrt{\pi}} \sqrt{\tau} - 3\tau \quad (2)$$

$$(0 < \tau < 0.2)$$

and

$$F(\tau) = 1 - \frac{6}{\pi^2} \exp \left[-\pi^2 \tau \right] \quad (3)$$

$$(\tau \geq 0.2)$$

For the above,

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

Thus, given a value of F at time t , a value of τ may be determined; consequently, a τ -time curve is generated. The derivative at any point,

as can be seen from Eq. (4), is the effective diffusion coefficient. This technique yields the fourth graph in each series.

For each experiment there are thus four graphs presented: (1) a temperature-time curve, (2) a release rate-time curve, (3) a fractional release-time curve, and (4) a diffusion coefficient-time curve. The last should be used with due regard to the basic assumptions (namely, of simple diffusion and neglect of the helium generation during the experiment) and to the approximate method of calculation.

The fuels examined were:

370 - Fuel from a vented capsule which underwent a rather complicated thermal history described in MRC literature.⁶ The fuel was about 13 months old.

DART - Fuel from an unvented capsule which was held at 900° to 1100°C for about 13 months. Measurements of the helium release on opening the capsule indicated that 90 percent of the helium generated since the fuel was made had been released. At the time of the test, the fuel was approximately 15 months old.

106 - A fuel sample stored at relatively low (~150°C) temperatures for about 12 months before the test.

The fuel samples were generally sieved into two size ranges, the so-called high cut 177 to 210 μ m diameter and the low cut 74 to 88 μ m diameter. One sample, 370R (R for random), was used without sieving, as was the 106 fuel sample.

BIBLIOGRAPHY

1. Private communication, R. N. R. Mulford, LASL, to the author.
2. See for example:
 ORNL-4285
 MLM-1527
3. Crank, J., The Mathematics of Diffusion, p. 147, Oxford, 1967.
4. Ibid., p. 84 ff.
5. Sung, P. and Turnbaugh, J. E., J. Appl. Phys., 39, 346 (1968).
6. A description of the thermal histories for all fuels tested can be found on the magnetic tape mentioned in the text.

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.

Figure 17.

Figure 18.

Figure 19.

Figure 20.

Figure 21.

Figure 22.

Figure 23.

Figure 24.

Figure 25.

Figure 26.

Figure 27.

Figure 28.

Figure 29.

Figure 30.

Figure 31.

Figure 32.

Figure 33.

Figure 34.

Figure 35.

Figure 36.

Figure 37.

Figure 38.

Figure 39.

Figure 40.

Figure 41.

Figure 42.

Figure 43.

Figure 44.

Figure 45.

Figure 46.

Figure 47.

Figure 48.

Figure 49.

Figure 50.

Figure 51.

Figure 52.

Figure 53.

Figure 54.

Figure 55.

97

Figure 56.

Figure 57.

Figure 58.

Figure 59.

Figure 60.

Figure 61.

Figure 62.

Figure 63.

Figure 64.

Figure 65.

Figure 66.

Figure 67.

Figure 68.

Figure 69.

Figure 70.

Figure 71.

Figure 72.

Figure 73.

Figure 74.

Figure 75.

Figure 76.

Figure 77.

Figure 78.

Figure 79.

Figure 80.

Figure 81.

Figure 82.

Figure 83.

Figure 84.

Figure 85.

Figure 86.

Figure 87.

Figure 88.

Figure 89.

Figure 90.

Figure 91.

Figure 92.

Figure 93.

Figure 94.

Figure 95.

Figure 96.

Figure 97.

Figure 98.

Figure 99.

Figure 100.

Figure 101.

Figure 102.

Figure 103.

Figure 104.

Figure 105.

Figure 106.

Figure 107.

Figure 108.

Figure 109.

Figure 110.

Figure 111.

Figure 112.

Figure 113.

Figure 114.

Figure 115.

Figure 116.

Figure 117.

Figure 118.

Figure 119.

Figure 120.

Figure 121.

Figure 122.

Figure 123.

Figure 124.

Figure 125.

Figure 126.

Figure 127.

Figure 128.

Figure 129.

Figure 130.

Figure 131.

Figure 132.

Figure 133.

Figure 134.

Figure 135.

Figure 136.

Figure 137.

Figure 138.

Figure 139.

Figure 140.

Figure 141.

Figure 142.

Figure 143.

Figure 144.

Figure 145.

Figure 146.

Figure 147.

Figure 148.

Figure 149.

Figure 150.

Figure 151.

Figure 152.

Figure 153.

Figure 154.

Figure 155.

Figure 156.

Figure 157.

Figure 158.

Figure 159.

Figure 160.

Figure 161.

Figure 162.

Figure 163.

Figure 164.

DISTRIBUTION:

J. E. McDonald, 5300 (2)
W. A. Stark, 5321 (35)
A. J. Clark, Jr., acting, 7511
ARPIC, 9512
B. F. Hefley, 8232 (5)
B. R. Allen, 3422
C. H. Sproul, 3428-2 (15)
L. C. Baldwin, 3412
L. L. Alpaugh, 3412
G. C. McDonald, 3416 (2)
Attn: M. S. Goldstein