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April 19, 1954

FINAL REPORT ON PRODUCTION TEST NO. 105-551-SR
THE FISSION GAS EXPERIMENT, KAPL-M-108

INTRODUCTION

This experiment was designed to furnish data on the quantitative relationship between exposure of enriched uranium metal to thermal neutron bombardment and the subsequent release of gaseous fission fragments from the metal.

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SUMMARY

Studies of data obtained from the in-pile phase of the KAPL Fission Gas Experiment indicate, neglecting startup and shutdown transients, that gaseous fission fragments generated within a 6-mil foil of uranium metal maintained at an operating temperature of about 450 C are released from the foil nearly as fast as they are produced.

DETAILS OF THE EXPERIMENT

Figure I exhibits the essential details of the in-pile experimental assembly. It is composed of two primary components: namely, a vessel in which is sealed the enriched uranium sample, and a pressure sensing diaphragm coupled to the sealed vessel. The void space around the uranium sample is referred to as the "hot section"; it is connected to the void space around the upper side of the diaphragm by a capillary tube. The void space on the upper side of the diaphragm is referred to as the "cold section." The hot and cold sections of the fission gas chamber plus the connecting tubing constitute a closed system of known volume and temperature.

Due to small temperature gradients, the fission gas temperature in the cold section can be measured more accurately than the fission gas temperature in the hot section. To reduce the uncertainties in the determination of the overall fission gas temperature, therefore, the volume of the cold section was made large compared to the volume of the hot section.

Fabricated in the shape of a thin washer, the 93 per cent enriched uranium foil is 0.006 inches thick, 0.250 inches OD, 0.125 inches ID, and weighs 0.085 grams. For heat removal, the uranium washer was bonded to molybdenum which in turn was bonded to an isothermal block containing a heater. To provide a temperature high enough to assure adequate thermal diffusion, yet low enough to avoid changes in the crystalline structure of the uranium, the hot section was designed to operate at a temperature of approximately 450 C (uranium changes from the alpha to the beta phase at about 665 C). Heat from the fissioning uranium foil plus an automatically controlled electric heater in the isothermal block maintain the 450 C temperature in the hot section. The temperature at the cold section is approximately 50 C under operating conditions.

Pressure in the fission gas chamber is measured by a null system using a pressure transmitter consisting of a thin metal diaphragm. This pressure transmitter is subjected to the fission gas pressure on one side and an equal, externally measured and controlled pressure of helium gas on the other side. Electrical contact points, one fixed and the other attached to the diaphragm, are used to indicate the null position of the diaphragm. In operation the helium gas is slowly bled from the external system until the electrical points make contact; this event actuates a solenoidal valve, causing the helium pressure to increase about one centimeter of mercury in the external gas system and separating the contact points. This process continues in a cyclic fashion with a period of approximately three minutes. Low point in the recorded pressure cycle is reached when the electrical points just make contact. This event

serves as a reference point in the pressure cycle and thus it is this value which is used to indicate the pressure in the fission gas chamber.

During the DR Pile shutdown of January 23, 1953, the test assembly was charged into tube 1078-DR, an annulus tube. Location of the assembly was adjusted to the one-half flux region by positioning the test unit 104 inches from the center of the tube. This was done to avoid overheating the foil by fission heat alone. At maximum tube power and with the fissioning uranium as the only heat source, the hot section of the fission chamber attained a temperature of 330 C.

The fission gas assembly was operated in DR Pile from January 23, 1953, to October 12, 1953. During the irradiation, DR Pile was shutdown for an extended period from March 2, 1953, to April 6, 1953, as indicated in Figure II. Other shutdowns during the irradiation were short--of the charge-discharge type requiring a maximum of about three days--and occurred infrequently, possibly once or twice per month. No significant changes in the nominal pile operating level were made during the irradiation.

EXPERIMENTAL DATA

Curve A, Figure II, is a freehand fit to the observed pressure data corrected for temperature. The observed pressure was corrected by the formula

$$P_c = P_o \left(\frac{71.5}{T_h} + \frac{291.2}{T_d} \right)^*$$

where: P_c is the corrected pressure,
 P_o is the observed pressure,
 T_h is the absolute temperature of the hot assembly, and
 T_d is the absolute temperature of the diaphragm assembly.

The formula corrects the observed pressure to temperatures of 50 C and 450 C at the diaphragm and hot assemblies respectively. These values were chosen because they are close to the actual operating temperatures and make the correction factors small for variations in operating temperatures. Observation has shown that the corrections for values of T_h below approximately 350 C cause the data to scatter greatly; consequently, such data have been eliminated from Curve A. This is probably due to the fact that the two temperature regions in the assembly are not well defined.

Curve B, Figure II, giving the accumulated exposure at the experimental assembly as a function of time, was determined from measurements of heat energy release in the uranium near the experimental assembly in the surrounding process tubes. The integrated thermal neutron exposure at the assembly is merely the product of the volume-average exposure in the adjacent uranium (related to the accumulated exposure by $nvt = MWD/AT \times 3.0 \times 10^{-17}$) and the "rise factor" for a nearly neutron-transparent process channel. This "rise factor," the ratio of the thermal flux in an empty process channel to the volume-average thermal flux in an adjacent uranium-filled channel, has been determined theoretically from diffusion theory

* See the appendix.

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and by experiment. The rise factor for this assembly is approximately 3.0.

To obtain the effective or volume-average exposure of the foil to thermal neutrons, it is necessary to correct for neutron self shielding of the foil. This correction factor, 0.51, will be mentioned later. Thus, the effective exposure of the foil to thermal neutrons is $nvt_{eff} = MWD/AT \times 3.0 \times 10^{17} \times 0.51 \times 3.0 = 4.6 \times 10^{17} \times MWD/AT$. Per cent U-235 burnup may be expressed as: $\% \text{ U-235 burnup} \approx (\sigma_f + \sigma_a) \times nvt_{eff} \times 100 = 6.50 \times 10^{-20} \times nvt_{eff} = 3.0 \times 10^{-2} \times MWD/AT$.

Curve D, Figure III, gives P_c as a function of accumulated exposure, MWD/AT. The points for the curve were obtained from Figure II, using pressures corresponding to integral values of exposure. Line E, Figure III, is a straight line "best-fit" drawn through the linear portion of Curve D. The first five datum points and the four points subsequent to the extended shutdown were neglected to exclude transient phenomena; the remaining data were used in a least squares analysis for a straight line. The formula for Line E is $P_c = 84.6 + 0.090 \times MWD/AT$. Although the processes whereby fission gases escape from the uranium metal are not well known, (1) it is reasonable to expect that with a constant rate of exposure and constant metal temperature the amount of fission gases released from the uranium metal will eventually become a linear function of exposure. Since this appeared to be the situation, Line E was drawn. The deviation of the linear portion of Curve D from a straight line is probably due to variations in pile power level, assembly operating temperatures, shutdowns, and the method used for determining the assembly exposure, which is possibly responsible for a five to ten per cent variation. It is evident that Curve D exhibits a definite discontinuity at the extended shutdown. This discontinuity could be explained either by a change in instrument calibration during the extended shutdown, which is unlikely, or by an actual release of fission gases during this period, a questionable phenomenon at the low temperatures prevalent during the period. Post irradiation measurements made to measure additional evolution of fission gases from the uranium were inconclusive. Further studies to be made on the irradiated assembly by KAPL may resolve this question.

The theoretical potential pressure build-up as a function of accumulated exposure, Line C, Figure III, was calculated as follows:

1. Constants of the system: (2)

- V, volume of the fission gas system = 0.473 cc.,
- P_g' , the pressure sealed into the assembly = 0.993 atm.,
- T, the absolute temperature at which helium was originally sealed into the assembly = $273 + 22 = 295^\circ K$ (assumed),
- W, the amount of U-235 in the foil = $0.085 \times 93\% = 0.079$ gm.

- (1) KAPL-402, "The Escape of Fission Gases from Uranium," C. E. Weber, April 12, 1950.
- (2) GEH-20,373, "Fission Gas Release Experiment," V. Ropitsky to T. F. Fisher, January 20, 1953.

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2. Moles of helium sealed into system:

If M_s = moles of helium initially sealed into the system,
 $R = 82.07$ cc - atm.;

$$\text{then, } M_s = \frac{P_s' V}{RT} = \frac{0.993 \times 0.473}{82.07 \times 295},$$

$$\text{and } M_s = \underline{1.94 \times 10^{-5} \text{ moles.}}$$

3. Moles of fission gases generated with exposure, nvt:

Assumptions:

- a. 100% binary fission,
- b. All of the fission gases are monatomic. This is a valid assumption, since the noble gases xenon and krypton constitute most of the fission gases.

If Y , the fission gas yield = 24.6%, (3)

M_g = the moles of fission gas generated,

N_0 , Avagadro number = 6.023×10^{23} ,

σ_f , the U-235 fission cross section = $549 \times 10^{-24} \text{ cm}^2$,

A , the atomic weight of U-235 = 235,

f , the self shielding factor = 0.51, (4)

nvt = the exposure in neutrons/cm²;

$$\text{then, } M_g \frac{\text{fissions} \times \text{yield}}{N_0} = \frac{W \times N_0}{A} \times \frac{\sigma_f \times f \times Y}{N_0} \times \text{nvt},$$

$$\text{and } M_g = \underline{2.32 \times 10^{-26} \times \text{nvt.}}$$

4. Pressure build-up with exposure:

Let P_s = the observed operating pressure corrected for temperature at the start of the irradiation.

If $P_s = 86.7$ cm Hg (see Curve A, Figure II),

then, the pressure increment, ΔP , due to the build-up of fission gases is:

$$\begin{aligned} \Delta P &= P_c - P_s \left(\frac{M_s + M_g}{M_s} - 1 \right) = P_s \times \frac{M_g}{M_s} \\ &= \frac{86.7 \times 2.32 \times 10^{-26}}{1.94 \times 10^{-5}} \times \text{nvt} \\ &= 1.04 \times 10^{-19} \times \text{nvt} \\ &= 1.04 \times 10^{-19} \times 3.0 \times 10^{17} \times \text{MWDT/AT} \times 3.0 \\ &= \underline{0.4 \times 10^{-2} \times \text{MWDT/AT (cm Hg).}} \end{aligned}$$

(3) ORNL-1153, Quarterly Progress Report, Chemical Division, September 1951.

(4) KAPL-239, "Activation Experiments in the KAPL Preliminary Pile Assemblies,"
 H. B. Stewart and G. B. Gavin, September 20, 1950.

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The prime sources of error in the above data are in the accumulated exposure. MWD/MT, and "rise factor," 3.0. The "rise factor" is in error by possibly fifteen or twenty per cent; the exposure, as was mentioned before, by five to ten per cent. The self shielding factor, since the problem is one which lends itself to an exact calculation, should be quite good, possibly accurate to within a few per cent.

Data presented here supersede any preliminary data published in document HW-29124, entitled "Fission Gas Experiment, KAPL-M-108," August 25, 1953.

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APPENDIX

The reduction of observed pressure data to standard operating conditions:

Equations of state:

$$P_1 V_1 = N_1 R T_1 \quad (1)$$

$$P_2 V_2 = N_2 R T_2 \quad (2)$$

$$N_1 + N_2 = N \quad (3)$$

$$P_1 = P_2 = P \quad (4)$$

Formulae: Solving (1) and (2) for N_1 and N_2 , respectively, and applying (3) we have:

$$\frac{P_0 V_1}{R T_1} + \frac{P_0 V_2}{R T_2} = N = \frac{P_c N_1}{R T_{1S}} + \frac{P_c V_2}{R T_{2S}} \quad (5)$$

where P_0 = the observed pressure, and P_c = the pressure correction to the standard operating temperatures. Standard operation temperatures are $T_{1S} = 723^\circ\text{K}$ (450°C) and $T_{2S} = 323^\circ\text{K}$ (50°C).

Substituting: $V_1 = 0.093$, $V_2 = 0.380^*$, $T_{1S} = 723^\circ\text{K}$, $T_{2S} = 323^\circ\text{K}$, and solving (5) for P_c , we have

$$P_c = P_0 \left(\frac{71.3}{T_1} + \frac{291.2}{T_2} \right)$$

* Obtained from reference (1).

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— KAPL 108 FISSION GAS ASSEMBLY —

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Pg. 6

K.P. MWD/AT

EXP. MWD/AT

FIGURE II (cont)

- A. Observed pressure buildup as a function of time (temperature corrected).
 B. Unperturbed exposure as a function of time.

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