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Fission Product Decay Heat Studies as of December 15, 1975

by

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FISSION PRODUCT DECAY HEAT STUDIES

(December 15, 1975)

J. L. Yarnell and P. J. Bendt

Abstract

This report describes the status of the Los Alamos Fission Product Decay Heat Studies program as of December 15, 1975.

Introduction

The purpose of this project is to study fission product decay heating rates, with emphasis on short decay times. Isothermal calorimetry is used to perform benchmark experiments for decay times between 20 seconds and 2000 seconds with an absolute accuracy of better than 5%. Experiments are being done with ^{235}U (Task 1) and will be done eventually with ^{239}Pu (Task 2). Thermal neutron spectra are used for the irradiations.

The project was initiated in July 1974. Final results for ^{235}U are expected by 6-30-76, and for ^{239}Pu by 1-1-77. Final reports for each task will follow the final data by ~ 3 months.

This report describes the work done to date, together with the status of the final experimental configuration. We note that in an experiment of this type, the most efficient approach is to devote a majority of the available time to optimization and perfection of the experimental apparatus and technique, and that when this has been done, the final results are obtained rather quickly. We have adopted this approach in our work on this project.

Experimental Method

The experimental principle is to use decay heat to evaporate liquid helium and to measure the rate at which the boil-off gas is evolved. Since the heat of vaporization of helium is known, the amount of decay heat may be calculated from the boil-off rate. Well-established radiochemical techniques are used to determine the number of fissions in the sample.

Operation of a boil-off calorimeter with liquid helium has a number of advantages. The heat of vaporization of helium is small (~ 21 joules/gram) so that high sensitivity is achieved. (For comparison, the heat of vaporization of water is 2261 joules/gram.)

At the temperature of liquid helium (~ 4°K), the heat capacity of most solids is very much less than it is at room temperature (by a factor of ~ 45 for Pb and ~ 1000 for Cu). This lowers the thermal inertia of the massive absorbers required, and leads to thermal time constants shorter than those usually obtained in calorimetry.

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A third possibility available with liquid helium is to operate in the superfluid regime. This results in a very high thermal conductivity in the liquid, but requires operation at reduced pressure, with its attendant complications in pressure and temperature control.

Initial Measurements

A dual concentric calorimeter was constructed, and numerous tests were carried out on it, using electrical heating, radioactive decay of ^{224}Ac , and ^{235}U fission products.

The inner, "beta", calorimeter was filled with liquid nitrogen, and the absorptive material along a radius was $\sim 2 \text{ g/cm}^2$. The outer, "gamma", calorimeter contained lead shot in liquid helium and had a material density along a radius of $\sim 47 \text{ g/cm}^2$. The gamma section was tested using several sizes of lead shot, and with both normal and super-fluid helium.

The following conclusions were reached as a result of our studies on this first version of the calorimeter.

1. It would be preferable to make the "beta" measurements with liquid helium rather than liquid nitrogen, since under the experimental conditions, nitrogen has a tendency to super-heat, leading to erratic flow variations.
2. Operation in the super-fluid regime, while feasible, requires sufficiently complex control procedures and data analysis to take away the inherent simplicity and "cleanness" of the experiment. We believe that these features are important in securing general acceptance of the experimental results, and therefore wish to retain them.
3. The thermal time constants ($\sim 20 \text{ sec}$) obtained with lead shot and normal liquid helium were longer than desired, even with optimum size of lead shot. A shorter and less restricted convection path for the liquid appeared to be desirable.
4. The gamma escape correction was larger than desired, due to the low packing density of the lead shot (64% of theoretical density) and the space occupied by the beta calorimeter. We concluded that since the quantity of most interest was the total decay heat, this quantity should be measured directly using a single calorimeter, and that the "beta" (or easily absorbed) portion of the energy should be determined in an auxiliary measurement. The total decay heat measurement will be carried out first. The "beta" measurement can be carried out in the same apparatus if the absorber is replaced by a much thinner one.

Modifications to the original design

A calorimeter with a single solid absorber was considered. The transient thermal behavior may be calculated from the heat conduction equation:

$$\frac{\partial U}{\partial t} = \alpha \nabla^2 U,$$

where U is the deviation from a reference temperature, t is time, and α diffusivity, defined by

$$\alpha = \frac{k}{\rho C_p} \frac{\text{cm}^2}{\text{sec}}.$$

The density is ρ , heat capacity at constant pressure is C_p , and the thermal conductivity is k . Diffusivities of several materials considered are given in the table.

Element	Z	Density	Thermal Diffusivity in cm^2/sec	
			300 K	4 K
Cu	(29)	8.96	1.14	$\sim 3000^*$
Ag	(47)	10.96	1.70	$\sim 2000^*$
W	(74)	19.3	0.62	~ 900
Pb	(82)	11.36	0.24	~ 80

* Uncertainty due to thermal conductivity, which is very sensitive to impurities. Diffusivities up to an order of magnitude higher may be obtained with some specimens.

The diffusivity of lead was too low to permit the use of a solid lead absorber of the size required, so it was rejected.

Tungsten appeared to be promising because of its high density and relatively high diffusivity. However, difficulties in fabrication of pure tungsten (and low diffusivity of its more easily fabricated alloys) lead to its eventual rejection.

Silver would be acceptable, but was rejected because of cost.

Copper was finally selected on the basis of its high diffusivity, ease of fabrication, and cost. A copper absorber which will fit into the existing apparatus was designed. It provides $\sim 50\%$ more material density along a radius than the original design using Pb shot. Calculations (described later) indicate that with the copper absorber the gamma escape correction can be made with acceptable accuracy.

Current status

The remainder of this report provides a description of the experiment and its status as of December 15, 1975.

Samples

Samples consist of approximately 60 milligrams of uranium enriched to 93.08% ^{235}U . The fission rate for such a sample in the irradiation position is 2.3×10^{12} fissions per second. The power generated in the sample during irradiation is approximately 75 watts. The standard irradiation time is 2×10^4 seconds, and the decay heat after 20 seconds cooling is calculated to be ~ 2.3 watts.

Assurance that no fission products are lost from the sample

The samples are sealed in 0.13-mm (0.005-inch) thick aluminum foil by electron beam welding. After welding, the samples are alpha-counted to detect possible leaks and/or external contamination with ^{235}U . During irradiation, a stream of helium gas flows over the sample and then through a charcoal trap adjacent to a gamma counter. During the counting period, the sample is held at 4°K, at which temperature all fission products are solid and diffusion rates are exceedingly slow. Finally, the samples are checked for the presence of external fission products before the radiochemical analysis, to determine the number of fissions, is started. So far, no evidence for the escape of fission products has been found. Any sample in which fission product leakage is detected will be rejected.

Assurance of constant neutron flux during sample irradiation

The sample irradiation facility contains a miniature fission chamber adjacent to the sample position. The signal from this chamber is used to control the reactor power during irradiations. By this means, the neutron flux at the sample position may be held constant within $\pm 0.25\%$ during an irradiation.

Determination of the number of fissions in the samples

After the calorimetric measurements have been completed, the sample is dissolved, and the activities due to specific fission products are separated, following standard radiochemical techniques. The separated activities, normally ^{99}Mo , ^{140}Ba , and ^{147}Nd , are counted in a standard beta proportional counter. Repeated checks have shown the precision (reproducibility) of these determinations to be better than 2%.

The overall procedure is calibrated by fission counting, using a technique in which the mass of the standard comparison foil cancels out. These calibrations have been carried out yearly for the past twenty years; the maximum spread in this series of determinations was less than 2%. The maximum bias in the calibration is considered to be less than 1%.

Sample transfer

A pneumatically driven sample carrier with an inertially operated release mechanism is used to transfer the samples from the irradiation position to the calorimeter. The transfer time is approximately 2 seconds. An additional 4 seconds is required for sample cooldown. Data may then be obtained as soon as the initial transient following sample introduction has died away to a sufficiently low level. We expect that the total delay time before data can be obtained to be less or equal to 20 seconds.

Absorber

The absorber section of the calorimeter consists of a right circular cylinder of high-conductivity oxygen-free copper 177.8 mm (7 inches) in diameter and 215.9 mm (8.5 inches) high. A 12.7 mm (0.5 inch) diameter hole along the axis 127 mm (5 inches) deep provides a path for insertion and removal of the sample. This hole is filled with liquid helium, and a reservoir containing additional liquid helium is provided at the top of the absorber section. The reservoir is made shallow to provide a short convection path for the liquid. The copper absorber is suspended in a vacuum chamber whose walls are maintained a few millikelvins warmer than the absorber. This arrangement minimizes heat leaks into the absorber while avoiding heat transfer to the outer bath by reflux action of the helium in the connecting tube. See Fig. 1.

At 4 K, the thermal diffusivity of the copper is expected to be of the order of $3000 \text{ cm}^2/\text{sec}$, and possibly as much as an order of magnitude higher, depending on the impurity content. The thermal time constant is of the order of L^2/α , where α is the diffusivity in cm^2/sec and L is a characteristic length in centimeters. For $L = 21.6 \text{ cm}$, the time constant is $\sim 0.16 \text{ sec}$. For contrast, the room temperature diffusivity is $1.14 \text{ cm}^2/\text{sec}$ and the corresponding time constant is more than 400 seconds.

Temperature measurements in the calorimeter

In order to verify the behavior of the calorimeter and to provide data for any corrections required by small departures from true isothermal behavior, it is necessary to make very accurate temperature measurements in the neighborhood of 4 K. Calibrated germanium resistance thermometers are used for this purpose. These thermometers provide an absolute accuracy of 0.005 K, and can detect temperature changes as small as 0.001 K.

For measurements at higher temperatures, platinum resistance thermometers and thermocouples are used.

Design of the boiloff gas line and time constant of the system

The line which transports the vaporized helium from the region at 4 K to the flowmeter at room temperature poses several problems in the optimization of the system response. The volume of gas in the line should be as small as possible consistent with an acceptable pressure drop at maximum flow (< 1 torr). The heat necessary to raise the temperature of the gas from 4 K to room temperature should be supplied in a controlled manner, and variations in the temperature gradient along the tube as a function of gas flow should be minimized.

Since both the density and viscosity of the helium gas vary strongly with temperature, which in turn depends on the manner in which the required heat is supplied, the calculations were by no means simple, and experimental verification of the design was considered desirable. Accordingly, a mockup of the system was constructed and is being used to study the system response as a function of various design parameters. Heat pulses are introduced electrically by means of a heating element immersed in a liquid helium bath, and the flowmeter response is recorded. While the tests are not yet complete, a response of 90% in 2 seconds and 100% in 5 seconds has been achieved. It is expected that similar or better response will be obtained in the final system.

Measurement of the boiloff gas flow rate

The mass flow of helium evolved from the calorimeter is measured by means of a quartz-coated thin-film platinum sensor on a quartz support, rigidly mounted in the throat of a venturi in the flow stream. There is a temperature compensating resistor on the downstream side of the venturi, and on the upstream side is a straight section of tubing with temperature-controlled walls, which serves to smooth the flow and to reduce temperature variations in the gas stream.

The sensor temperature is maintained at a constant number of degrees above that of the gas stream by a feedback circuit which senses resistance changes in the sensor. Under these conditions, the mass flow is a known function of the power required to maintain the constant temperature difference.

The mass flowmeter has a time constant of less than 10^{-3} seconds, and a dynamic range of 100-1 in mass flow. (For an irradiation of 2×10^4 seconds, the decay heat falls by a factor of ~ 7.1 between 10 and 4000 seconds.) At maximum flow, the pressure drop across the flowmeter assembly is ~ 1 torr, which corresponds to a temperature change of 0.002 K in the helium bath. The associated circuitry provides a linearized output voltage that is proportional to mass flow, as well as the voltage being supplied to the bridge containing the sensor and temperature compensating resistor. See Fig. 2.

The flowmeter temperature compensation, calibration, and linearization were set up for measurements of dry helium at room temperature. In addition, the calibration is checked by means of standard dry-test integrating

gas-flow meters. The overall calibration of the system is also checked by the introduction of a known amount of electrical heating directly into the calorimeter.

Factory specifications indicate a precision of 0.1% for repetitive measurements at constant temperature and pressure. The absolute accuracy is stated to be 1.5% of reading or 0.5% of full scale, whichever is less, if the inlet temperature is held constant to within ± 3 K. Tests using a mockup of the calorimeter indicate that the temperature of the effluent gas can be maintained constant to within ± 0.6 K.

Pre-test predictions

The decay heat expected for a 2×10^4 -sec irradiation of ^{235}U in a thermal flux of 3×10^{13} n/cm²-sec has been calculated at the Los Alamos Scientific Laboratory (LASL) using CINDER-10 with neutron capture in fission products, CINDER-10 without capture, and RIBD. Similar calculations were made at Oregon State University (OSU) using a modification of an earlier version of CINDER, without capture. The LASL calculations included all of the currently known corrections to ENDF/B-IV.

The LASL calculations agreed within $\pm 0.3\%$ for cooling times through 10^4 seconds. The OSU calculations were higher than those of LASL by up to 8% for the same range of cooling times. The source of this discrepancy is under investigation. The CINDER-10 results are shown in Fig. 3, and a comparison with RIBD and OSU is shown in Fig. 4.

The calculations were also used to estimate the fraction of the total energy emitted in the form of gamma rays. This fraction is needed in the correction for gamma leakage from the calorimeter.

Gamma escape correction

A portion of the energy emitted by the sample in the form of gamma rays will escape from the finite absorber used in the calorimeter. Let

$$F = \frac{E_{\gamma}}{E_{\beta} + E_{\gamma}}$$

be the fraction of the total energy that is in the form of gamma rays, and let G be the fraction of the emitted gamma energy which escapes from the absorber. Then

$$E_{\text{total}} = \frac{E_{\text{calorimeter}}}{1 - FG}$$

The gamma fraction F was calculated, from ENDF/B-IV data and the codes described above. For a 2×10^4 -sec irradiation and cooling times between 10 and 4000 sec, the CINDER-10 results are shown in Fig. 5. We note that under these conditions F varies between 0.50 and 0.60.

The gamma escape fraction G depends on the gamma energy spectrum. For our initial estimates of the spectrum we have assumed the Oak Ridge National Laboratory (ORNL) preliminary data of Dickens et al. for $T_{\text{irrad}} = 100$ sec, $T_{\text{cool}} = 100$ sec, $T_{\text{count}} = 50$ sec. (We have measurements of the spectra at various times following a 2×10^4 -sec irradiation in progress.)

The ORNL spectrum was used as input to a LASL Monte Carlo gamma transport calculation for the gamma leakage from the copper absorber section. The resulting value of G was 0.0654 ± 0.0015 . If F is given its average value of 0.55, the total correction for gamma escape amounts to 3.6%.

A sensitivity analysis was carried out to estimate the accuracy with which this correction is known.

1. Errors in G :

- a. The statistics of the Monte Carlo calculation introduce a 2% error.

- b. The effect of uncertainties or variations in the assumed gamma spectrum was estimated by repeating the calculation with all energies increased by 10%. The increase in leakage was 4%, which we take to be the uncertainty due to the spectrum.

- c. The effects of the albedo of the surroundings on the gamma escape was investigated by performing the Monte Carlo calculation with and without a thick copper cylinder surrounding the absorber. This variation caused a 3% change in G . The overall rms error in G from these sources is then $\sqrt{2^2 + 4^2 + 3^2} \cong 5\%$.

2. Taking the average value of F leads to an $\sim 10\%$ uncertainty in K . Combining errors, we have a total uncertainty in the correction of $\sim 11\%$. Multiplying by the magnitude of the correction, 3.6%, gives an overall error in the decay heat of $\sim 0.4\%$ due to uncertainties in the gamma escape correction. This uncertainty can be further reduced by taking better estimates of F and of the gamma spectrum, and this will be done in the final analysis. However, the present analysis indicates that the errors in estimating the gamma escape fraction will not be an important contribution to the overall error in the measurement of decay heat.

Figure 1

MASS FLOWMETER

Figure 2

Figure 3

Figure 4

Figure 5