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**RADIATION MEASUREMENTS OF PLUTONIUM-238 ENDOGENOUS HEAT
STUDY SOURCES FOR THE ARTIFICIAL HEART PROGRAM**

M. E. Anderson, R. A. Neff, A. R. Campbell and G. R. Hagee

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MONSANTO RESEARCH CORPORATION

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M. E. Anderson
R. A. Neff
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SUMMARY

Detailed radiation measurements were performed on 16-W and 24-W encapsulated plutonium-238 heat sources for use in endogenous heat studies as part of the Artificial Heart Program. The neutron energy spectra, neutron emission rates, gamma energy spectra and gamma dose rates were measured, and the neutron dose rates were calculated. In general, the results did not deviate significantly from results obtained previously from many other encapsulated plutonium-238 heat sources in which the plutonium is present as the metal.

INTRODUCTION

Radiation measurements were performed on two plutonium-238 heat sources to be used for endogenous heat studies for the Artificial Heart Program.¹ Figure 1 shows the type of source used. The plutonium within these sources is present as the metal and has the following nominal isotopic composition: plutonium-238 - 80%, plutonium-239 - 16%, plutonium-240 - 3%, and plutonium-241 - 1%. The radioactive decay of plutonium-238 is responsible for 99.9% of the thermal power of these sources. The larger of the two sources being reported herein had a thermal power of 24.038 ± 0.096 W (May 5, 1967) and the smaller had a thermal output of 16.126 ± 0.048 W (May 6, 1967). The mass of plutonium-238 in each source was 42.55 g and 28.34 g, respectively.

During the endogenous heat studies the sources are to be located within a stainless steel and aluminum heat exchanger covered with Silastic (a silicone rubber). The heat exchanger is shown in Figure 2. Radiation measurements were made in air with the sources both in and out of the heat exchanger. Results are reported for each source as follows:

	<u>Bare Source</u>	<u>Source in Heat Exchanger</u>
Total Neutron Emission Rate	X	X
Neutron Energy Spectrum	X	
Neutron Dose Rate	X	X
Gamma Energy Spectrum	X	
Gamma Dose Rate	X	X

The principal effect of the heat exchanger on the neutrons is to scatter some of them and thus change the anisotropy of the neutron flux. Emission rate measurements were made of the sources both in and out of the exchanger. Within the comparison accuracy of $\pm 2\%$ the total emission rates were the same in both cases, confirming that few neutrons are absorbed by the exchanger when the source and exchanger are located in air. The neutron energy spectrum would be changed only insignificantly by the heat

exchanger and therefore no neutron spectra measurements were made of the source in the exchanger.

Gamma spectra measurements were made principally to identify the gamma energies present from the sources, and the presence of a heat exchanger would have essentially no effect on these results. The presence of the heat exchanger does affect the dose rates due to gamma and neutron radiation and these results, both with and without the heat exchanger, are reported.

NEUTRON EMISSION MEASUREMENTS

The total neutron emission rates of these sources were determined with the use of a long counter and a $^{239}\text{PuBe}(\alpha, n)$ standard source (M-591) calibrated at the National Bureau of Standards. A geometric integration was done for each source to account for the anisotropy of emission. A correction was made to account for the difference between the long counter response to these heat sources and the standard source. Another correction was made to take into account the change of yield with time for the standard since the calibration at the National Bureau of Standards (this change is due to the ingrowth of americium-241 from the decay of plutonium-241 present in the standard source). The total neutron emission rates for each source on May 18, 1967, were as follows:

16-W source - 1.02×10^5 n/sec $\pm 5\%$

24-W source - 1.60×10^5 n/sec $\pm 5\%$

The $\pm 5\%$ is an estimate of the absolute uncertainty. The specific yield of the 16-W source is 3.58×10^3 n/sec/g of plutonium-238; for the 24-W source the yield is 3.76×10^3 n/sec/g.

The neutron flux anisotropy $\epsilon(\theta)$ (defined as the ratio of the flux at a given angle relative to the source axis of symmetry to the average of the spatially integrated flux) was measured for several angles and these results are summarized in Table 1.

NEUTRON ENERGY MEASUREMENTS

Neutron energy spectra for these sources were obtained with the stilbene crystal, fast neutron spectrometer.² For these measurements the detector was situated on the source axis. No energy measurements were made at other locations relative to the source because variations in the spectrum as a function of location would be insignificant. The results of these energy measurements for both sources are shown on semi-logarithmic plots in Figures 3 and 4 and on linear plots in Figures 5 and 6. In each case the ordinates are the specific yields for each source (neutron emission rate per gram of plutonium-238).

For each source, about 70% or more of the neutrons result from spontaneous fission of plutonium-238. The rest of the neutrons are from (α, n) reactions with impurities having low atomic numbers. Above about 5 MeV essentially all of the spectrum is due to spontaneous fission neutrons. The data for energies greater than 5 MeV have been fitted to a Maxwellian distribution, a relationship frequently used to describe fission spectra. The Maxwellian distribution is shown in each figure as a solid line and approximately represents the spontaneous fission portion of each spectrum. The difference between the solid line and the measured spectrum is due to neutrons from (α, n) impurities. The lefthand data point in each spectrum was obtained by a difference calculation using data from two different detectors. The total neutron flux (all energies) incident on the crystal was determined from measurements with a long counter. The flux above the lower limit of detection for the spectrometer was obtained from the stilbene data. The difference between these two measurements is then this lefthand data point. The relatively large uncertainty for this point is a result of taking the difference between two numbers of approximately the same size. The error bars for all of the other data points represent plus or minus one standard deviation and were calculated from counting statistics only. Where no error bar is shown, it is smaller than the printed data point.

The two spectra have the same general features. There are small differences between the spectra. Some differences were expected since the two sources have slightly different specific yields.

NEUTRON DOSE CALCULATION

A factor for converting from neutron fluence to dose equivalent for these two sources was obtained by using the measured spectra and the recommended maximum permissible exposures to neutrons of different energies as given in Table 2 of NBS Handbook 63.³ The factor so obtained for the 16-W source is $(3.16 \pm 0.16) \times 10^{-5}$ mrem/n/cm² and for the 24-W source it is $(3.11 \pm 0.13) \times 10^{-5}$ mrem/n/cm².

The dose equivalent rate (DE) was calculated at several points on the source axis of symmetry by using the following equation which assumes a planar neutron source:

$$DE = \frac{Q \cdot C}{2\pi R^2} \left[A \ln \frac{\sqrt{R^2 + H^2}}{H} + B \left(1 - \frac{H}{\sqrt{R^2 + H^2}} \right) \right] \quad (1)$$

To obtain the above relationship, the following approximate relationship was used for the anisotropy factor:

$$c(\theta) \approx A + B |\cos \theta| \quad (2)$$

In Equation 1, R is the radius of the plutonium, H is the distance from the midplane of the plutonium, Q is the total neutron emission rate of the source, and C is the factor given above for converting from fluence to dose. This formula assumes that all scattering takes place within the plutonium volume, even if it may actually have occurred within the source container or the heat exchanger. The effect of this simplification should be slight and, if anything, may make the calculated values slightly high. In Equations 1 and 2, A and B are simply constants.

The dose at the curved side surface of the heat exchanger was calculated at two reference positions (90° and 270°). These calculations used the actual source dimensions, but the results obtained were within 2% of the result obtained when a point source is assumed. Because it is much simpler, the point source expression is shown:

$$DE = Q \cdot e(\theta) \cdot C / 4\pi H^2 \quad (3)$$

In the latter case, H is the distance between the axis of symmetry and the edge of the heat exchanger and is 4.3 cm. The other factors are as defined above.

Table 2 is a summary of the calculated dose equivalent rates due to neutrons for both sources. The uncertainty for all values, except for those in contact with a surface of the source, is estimated to be about $\pm 20\%$. The uncertainty for those calculations at a source surface is higher and may be $\pm 40\%$.

GAMMA DOSE MEASUREMENTS

Gamma dose equivalent rate measurements were conducted for the two sources using several sizes of Condenser-R meters*. The sources and detectors were held approximately 4 ft (1.2 m) from the floor and 6 ft (1.8 m) from the closest wall by an aluminum rod framework.

The measured values were plotted as a function of distance from the center of the plutonium fuel. Each dose curve was then extrapolated to the surface of the source. This extrapolation was made by using the theoretical variation of dose rate as a function of distance along the source axis. This relationship is shown in Figure 7 for a thin disc source of diameter 3.47 cm. Figures 8 and 9 show these dose measurements and the extrapolations to the surface of the source using the theoretical curve. The theoretical curve and the measured dose rates were normalized at 10 cm. (Measurements, using gamma dosimetry film and Condenser-R meters, were made on a second 16-W source for this program in order to check the agreement with the theoretical curve at distances closer to the source. These measurements and the theoretical curve agree with each other to within at least 1 cm of the fuel center).

*Victoreen Instrument Company, Cleveland, Ohio.

The dose at the side of each source was obtained by extrapolation from the measured values assuming the dose varied inversely as the square of the distance. Figures 10 and 11 show the dose measurements at the side of the sources.

Table 3 presents the dose values for each source. All dose values have an estimated uncertainty of $\pm 20\%$ except those at the surface of the source or the heat exchanger where the estimated uncertainty is about $\pm 40\%$. This large uncertainty at the surface is due mainly to the uncertainty in the extrapolation from the measured values.

GAMMA SPECTRA MEASUREMENTS

Gamma spectra measurements were made on the two sources using a 3-in. (7.6-cm) high by 3-in. (7.6-cm) diameter NaI(Tl) scintillation detector. The sources were each mounted with their flat surface parallel to the incident face of the detector. The distance from source to detector was adjusted to produce about 10% or less dead-time in the pulse height analyzer. The spectra were measured in two energy ranges: one range covers approximately 0 to 0.9 MeV and the other 0.2 to 2.7 MeV. Results for the 24-W source are shown in Figures 12 and 13. The spectrum for the 16-W source is quite similar to that for the 24-W source and has not been shown.

The photopeak energies noted on these spectra are listed below:

<u>Energy (keV)</u>	<u>Probable Origin</u>
70	Plutonium-239
100	Plutonium-238
153	Plutonium-238
203	Plutonium-238
336	Uranium-237
767	Plutonium-238
2614	Thallium-208

The gamma rays seen in these spectra are typical of those seen in NaI(Tl) spectra of plutonium-238 metal sources measured previously.

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1. Major Activities in the Atomic Energy Programs: January-December 1967, U. S. Atomic Energy Commission, U. S. G. P. O., Washington, D. C., January 1968, p. 181.
2. M. E. Anderson, Trans. Am. Nucl. Soc., 9, 600 (1966).
3. Protection Against Neutron Radiation up to 30 Million Electron Volts, U. S. Department of Commerce, National Bureau of Standards Handbook 63, April 1967.

Table 1

ANISOTROPY FACTORS, $\epsilon(\theta)$, FOR NEUTRON FLUX CALCULATIONS

<u>Location</u>	<u>16-W Source</u>		<u>24-W Source</u>	
	<u>Without Heat Exchanger</u>	<u>With Heat Exchanger</u>	<u>Without Heat Exchanger</u>	<u>With Heat Exchanger</u>
0°	1.10	1.23	1.19	1.27
90°	0.89	0.69	0.88	0.70
180°	1.14	1.26	1.21	1.29
270°	0.89	0.83	0.88	0.83

Table 2
DOSE EQUIVALENT RATE DUE TO NEUTRONS
(mrem/hr)

<u>Location</u>	<u>16-W Source</u>		<u>24-W Source</u>	
	<u>Without Heat Exchanger</u>	<u>With Heat Exchanger</u>	<u>Without Heat Exchanger</u>	<u>With Heat Exchanger</u>
On top surface, $\theta = 0^\circ$	420	440	710	740
On bottom surface, $\theta = 180^\circ$	850	870	1320	1330
On axis, 3 cm from top of surface	55	61	94	100
On axis, 10 cm from top of surface	8	9	14	14
On axis, 100 cm from top of source	0.10	0.11	0.16	0.17
On surface of heat exchanger at $\theta = 90^\circ$	-	37	-	59
On surface of heat exchanger at $\theta = 270^\circ$	-	44	-	70

Table 3
DOSE EQUIVALENT RATE DUE TO GAMMAS
(mrem/hr)

<u>Location</u>	<u>16-W Source</u>		<u>24-W Source</u>	
	<u>Without Heat Exchanger</u>	<u>With Heat Exchanger</u>	<u>Without Heat Exchanger</u>	<u>With Heat Exchanger</u>
<u>At Surface:</u>				
Top	230	235	360	365
Bottom	355	360	550	560
Side	135	14	145	20.5
<u>10 cm from Surface:</u>				
Top	5.1	-	7.6	-
Bottom	5.6	-	8.5	-
Side	2.9	1.4	3.6	1.9

FIGURE 1 - Photographic and cross-sectional illustrations of the endogenous heat study source.

FIGURE 2 - Photographic and cross-sectional illustrations of the heat exchanger for use with an endogenous heat study source.

FIGURE 3 - Semi-logarithmic plot of the neutron energy spectrum for the 16-W source.

FIGURE 4 - Semi-logarithmic plot of the neutron energy spectrum for the 24-W source.

FIGURE 5 - Linear plot of the neutron energy spectrum for the 16-W source.

FIGURE 6 - Linear plot of the neutron energy spectrum for the 24-W source.

FIGURE 7 - Variation of gamma dose equivalent rate as a function of distance along the source axis.

FIGURE 8 - Measured gamma dose equivalent as a function of distance on the axis of the 16-W source.

FIGURE 9 - Measured gamma dose equivalent as a function of distance on the axis of the 24-W source.

FIGURE 10 - Measured gamma dose equivalent at the side surface of the 16-W source and at the side surface of the heat exchanger containing the source.

FIGURE 11 - Measured gamma dose equivalent at the side surface of the 24-W source and at the side surface of the heat exchanger containing the source.

FIGURE 12 - Gamma energy spectrum for the 24-W source for the range 0-0.9 MeV.

FIGURE 13 - Gamma energy spectrum for the 24-W source for the range 0.2-2.7 MeV.