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THULIUM-170 OXIDE HEAT SOURCE
EXPERIMENTAL AND ANALYTICAL
RADIATION AND SHIELDING STUDY

Prepared By
Joseph Cyr

Prepared For
Division of Isotopes Development
U.S. Atomic Energy Commission
Contract Number: AT-(40-1)-3693

July 1969

By

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SANDERS NUCLEAR CORPORATION

Nashua, New Hampshire

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ABSTRACT

The results of radiation dose rates measurements performed with three Thulium -170 oxide radioactive sources (initial strengths 20.7, 10.0 and 5.0 thermal watts) through aluminum, stainless steel, lead, tungsten and depleted uranium absorbers at two source-to-dose-point separation distances agreed well with corresponding calculated values, thereby validating the calculation technique.

SECTION 1

INTRODUCTION

Thulium-170 oxide is a potential heat source for various radioisotopic thermal power applications. Shielding engineers performing system studies and lacking specific data as to active impurities in the heat source, often must proceed in their analytic computations on the basis that the heat source contains only Thulium-170 as an active species. Typical heat sources do in fact contain some Thulium-171; the amount is dependent on the activation time and flux. The activated thulium oxide target, in addition, contains typically approximately 0.1% other rare earth oxides, principally erbium and ytterbium, and 50 to 200 ppm of calcium, magnesium, iron and silicon, all to a greater or lesser degree contributors to heat source dose rate.

The objective of this study was to measure⁽¹⁾ the dose rate emitted by a representative, pressed and sintered, thulium oxide heat source of significant thermal power, and to determine, by comparison of measured and predicted dose rates, whether significant shield design error is incurred by application of classical shield calculation methods to Thulium-170 oxide heat sources when it is assumed the source contains only Thulium-170 as the active isotope.

(1) The experimental procedure used included rigorous traceability and calibration standards for the heat sources and for dosimetry so that measured results would in fact reliably evaluate the accuracy of calculated values of dose rate.

SECTION 2

SUMMARY

The Sanders Nuclear Corporation program with respect to Thulium-170 oxide was intended to experimentally determine the radiation dose rates absorbed at given separation distances from various thermal strengths of this radioisotope through successive thickness of selected absorber materials in such a manner as to reliably evaluate calculated values for corresponding configurations.

The radiation sources used in these measurements consisted of stacks of representative, pressed and sintered, Thulium-170 oxide wafers doubly encapsulated in Hastelloy alloy C. Three such sources were used containing respectively: 2225 ± 44.5 , 4450 ± 44.5 and 9211 ± 44.5 curies of this radioisotope.

Radiation dose rate measurements were performed with each of these sources at source-to-dose-point separation distances of 19 and 100 centimeters through each of three thicknesses (0.635, 1.270 and 2.54 cm) of each of five absorber materials: aluminum, stainless steel, lead, tungsten and depleted uranium.

Subsequently, the results obtained from these measurements were analyzed and mathematically normalized in respect to the corresponding calculated values. Comparison of these results revealed that the predicted

dose rates usually exceeded the measured values. This divergence arises as a result of the nature of the build-up factor assumed in the calculations. A further discrepancy was detected with the depleted uranium absorbers and was analyzed as being due to the radioactivity inherent within this material.

SECTION 3

GAMMA RADIATION ATTENUATION DISCUSSION

The reduction of intensity of an electromagnetic radiation (gamma, x-rays) flux as it passes through matter is accomplished by two mechanisms. These are: (1) simple separation distance from radiation source to dose point, and (2) the interactions of the photons with the atoms of the absorber material through which they pass. The first effect is significant for small sources (which approximate "point" sources), the dose rates at given distances from which are inversely proportional to the square of the distance. Consequently, the dose rates from such sources decrease with increasing distance in air, even though the photon attenuation by this medium is negligible.

With the second effect, there are several kinds of interactions by which gamma or x-ray photons are attenuated in matter. Of these, the most significant are:

- Photoelectric absorption
- Compton scattering
- Pair production

In the photoelectric absorption interaction, a gamma photon with energy greater than the binding energy of an extra-nuclear atomic

electron interacts with it so that the latter receives the entire photon energy. This electron is subsequently ejected from the atom, and the photon disappears. The probability (P_{pe}) of occurrence of photoelectric absorption depends upon both the photon energy (E) and the atomic number (Z) of the absorber and varies approximately as follows:

$$P_{pe} \approx \left[(\text{constant}) \left(\frac{Z^n}{E^3} \right) \right]$$

where n varies from 3 for low photon energies to 5 for high energies. Consequently, the probability of photoelectric absorption is greatest with high atomic number absorbers and low energy photons. However, except in the heaviest elements, the incidence of photoelectric absorption usually decreases for photon energies greater than about 1 MeV.

In the Compton scatter interaction, the gamma photon interacts with an absorber atom in such a way that only part of its energy is transferred to an electron. The photon then leaves the interaction, moving in a different direction and with reduced energy. As with the photoelectric effect, the probability (P_{cs}) of occurrence of Compton scattering varies with absorber atomic number and the photon energy. The form of its variation may be stated very approximately as:

$$P_{cs} \approx \left[(\text{constant}) \left(\frac{Z}{E} \right) \right]$$

The significant difference between photoelectric capture and Compton scattering is that, while the former is a true absorption process in which the gamma photon disappears, the latter is not. Thus, each Compton scatter interaction produces a photon of somewhat reduced energy which may then undergo continued (multiple) scattering interactions. If it does not escape the absorber, the photon will eventually be absorbed through photoelectric capture, the probability of which increases as the photon energy decreases. Therefore, Compton scatter interactions within a given quantity of absorber may or may not result in the removal of gamma photons from an incident radiation beam. Thus, several interaction possibilities must be considered when assessing the attenuation of the latter by absorber materials, e.g.,

- Single, large angle scatter out of the radiation beam intercepted by a detector
- Single, small angle scatter within the beam
- Multiple scatter within absorber with ultimate photoelectric absorption of photon
- Multiple scatter within absorber with ultimate scattering of photon out of radiation beam
- Multiple scatter within absorber with ultimate scattering of photon into detected radiation beam.

It is the second and fifth of these interaction modes which necessitates inclusion of a build-up factor (B) when conducting radiation shielding analyses.

The pair production interaction mechanism becomes possible for gamma photons with energies in excess of 1.02 MeV. When such photons pass within the electromagnetic field of the nucleus, they can undergo annihilation accompanied by the formation of an electron-positron pair. The probability (P_{pp}) of occurrence of this interaction is related to the atomic number of the absorber and the photon energy by:

$$P_{pp} \approx \left\{ (\text{constant}) \left[Z^2 (E - 1.02) \right] \right\}$$

so that it increases proportionally with the square of the atomic number of the absorber and with increasing photon energy above 1.02 MeV. Since both photoelectric absorption and Compton scattering decrease with increased photon energy, it is evident that the pair production mechanism becomes the dominant interaction mode at the higher energies. As in photoelectric absorption, pair production results in total absorption of the gamma photon. However, owing to the low photon energies associated with the radioactive decay of Thulium-170 and to a maximum energy less than 1.0 Mev, there is, therefore, no pair production or interaction.

Summarizing, therefore, it is evident from consideration of spectral distribution of photon energies emitted in radioactive decay of Thulium-170 as presented by Arnold⁽¹⁾ that the interaction modes of these radiations with absorber materials will be photoelectric absorption and Compton scattering. In addition, the significance of each mode and its effects upon the residual photon flux will vary with the atomic number of the absorber.

The probability of a photon passing through a given amount of absorber material without undergoing any attenuation interaction is the product of the individual probabilities of survival for each interaction mechanism. The probability of traversing a thickness (x) of absorber without photoelectric absorption is:

$$\exp (-\tau x)$$

where τ is the total linear attenuation coefficient for photoelectric absorption. Similarly:

$$\exp (-\sigma x) = \text{survival probability for Compton scattering}$$

$$\exp (-\kappa x) = \text{survival probability for pair production}$$

(1) Arnold, E. D., Handbook of Shielding Requirements and Radiation Characteristics of Isotopic Power Sources for Terrestrial, Marine, and Space Applications, USAEC Report - ORNL-3576, April 1964.

Thus, a collimated (no extraneous scatter) beam of photons of initial intensity ϕ_0 , after traversing a thickness of absorber material will have a residual intensity $\phi(x)$ given by:

$$\phi(x) = \phi_0 e^{-\mu x} \quad (1)$$

The factor μ in this equation is the total linear attenuation coefficient, and it is numerically equal to the sum of γ , δ , and K . Accordingly, its value will vary primarily with absorber atomic number, Z , and photon energy (E) as discussed above. The magnitudes of μ for various materials and energies are available in White (2) and Rockwell (3).

As mentioned earlier, the occurrence of multiple (Compton) photon scattering within the absorber materials causes the photon flux intensity emerging from an absorber to exceed that which would be calculated using simple exponential attenuation only. Thus, equation (1) above gives the uncollided flux, i.e., the gamma flux which has not undergone any collisions in its passage through the thickness (x) of absorber. Therefore, in order to correlate measured and calculated gamma dose rates, it is necessary to allow for the effects of multiply scattered photons by means of

(2) White, G. R. and Grodstein, N.B.S. Circular 583, 1957.

(3) Rockwell, T., Reactor Shielding Design Manual, 1956.

what is called the build-up factor. This is defined as the ratio of the observed dose rate to that which may be calculated using simple exponential attenuation with the linear absorption coefficient. This factor is a function of the absorber material and thickness, as well as the energy of the incident radiation. Experiments and calculations have been performed to determine build-up factors for various configurations of sources and absorbers; however, the best source for gamma photon build-up factors for point isotropic and planar sources in infinite absorbing media is USAEC Report NYO-3075 (4). These data are also available in Chapter 10 of Rockwell (3). Thus, for a given absorber sample and radiation energy, the residual flux is obtained by modifying equation (1) to take the form:

$$\phi(x) = \phi_0 B(\mu x) e^{-\mu x} \quad (2)$$

where $B(\mu x)$ is the appropriate build-up factor. It is generally expressed as a function of μx , since μ depends upon the absorber material and radiation energy, and x is the absorber thickness.

(4) Goldstein, H. and Wilkins, J. F., Calculations of the Penetration of Gamma Rays, USAEC Report NYO-3075, 1954.

SECTION 4

ABSORBER MATERIAL EFFECTS

The introduction into a beam of gamma radiation of selected absorber materials changes the intensity of the residual flux according to the magnitude of the linear attenuation coefficient for each absorber. Thus, the radiation dose rate induced by this flux also varies by the same factor. Accordingly, it was anticipated that the introduction of aluminum, stainless steel, lead, tungsten and depleted uranium absorbers into a radiation beam emitted by Thulium-170 would provide a broad range of mass densities and linear attenuation coefficients by which to evaluate the severity of radiation hazard presented by this radioisotope. In addition, since certain of these absorbers are also structural materials commonly used in the vicinity of radioisotope heat sources, it was further anticipated that this selection might yield meaningful data regarding their suitability as radiation shielding materials in such applications. Subsequently, after performance of these experiments in various configurations of absorber material and thickness, source strength and dose point separation distance, the measured

radiation dose rates from Thulium-170 were observed to vary exactly as anticipated. Selected results demonstrating this, extracted from the experimental data tabulations of Appendix A, are tabulated below.

Table 4-1 Variation of Measured and Calculated Radiation Dose Rates (1) with Absorber Material

Absorber Material	Material Form	Density (gm/cm ³)	Linear Atten. Coeff ⁽²⁾ (cm ⁻¹)	Dose Rate Measured	(mR/hr) Calculated
Al	alloy-1100	2.71	0.166	5030±16.3	5800
SS	alloy-304	7.90	0.462	2610±7.55	3720
Pb	99.90%	11.34 (pure)	0.797	316±0.34	310
W	99.98%	19.30	1.23	104±0.66	134
U	99.97%	18.97	1.46	38.3±0.11	42

- (1) The results presented here are extracted from the results for the following geometry: source number 3
absorber number 2
separation distance - one meter

- (2) The values tabulated are for monoenergetic (1 MeV) gamma photons only.

SECTION 5

ABSORBER THICKNESS EFFECTS

Owing to the build-up effect due to multiple radiation scatter, the residual photon flux of a given energy from a heavy and/or thick absorber, as expressed by equation (2), does not fall off in a simple exponential manner. Nevertheless, it is possible to retain the exponential description of the flux decline with absorber thickness by writing:

$$\phi(x) = \phi_0 e^{-x/\lambda} \quad (3)$$

where the factor λ is called the relaxation length of the absorber for the specified radiations. Physically, it is the thickness of absorber in which the flux is decreased by a factor of $1/e$. Comparison of equations (1) and (3) shows that the relaxation length λ is formally equivalent to $1/\mu$. Since the quantity x/λ used in the above exponential represents the number of relaxation lengths in a thickness x of absorber, it follows, therefore, that the quantity μx used in connection with the build-up factor is also approximately equivalent to the number of relaxation lengths. This relationship is approximate because, whereas the relaxation length incorporates some allowance for multiple scatter in the absorber, the mean free path ($1/\mu$)

of the same situation does not. Accordingly, it was anticipated that performance of dose rate determinations from Thulium-170 through varying thicknesses of absorber materials would provide information regarding the reduction of the former with increasing number of relaxation lengths. Selected results demonstrating this, extracted from the experimental data tabulations of Appendix A, are tabulated below.

Table 5-1 Variation of Measured and Calculated Radiation
Dose Rates⁽¹⁾ with Absorber Thickness

Absorber Material	Lin Atten. Coeff. (2) (cm ⁻¹)	Absorber Thickness (cm)	No. of Relax. Lengths (μ x)	Dose Rate Measured	(mR/hr) Calculated
Al	0.166	0.635	0.105	5900±32.1	5800
		1.27	0.211	5030±16.3	5800
		2.54	0.422	3630±14.7	5600
SS	0.462	0.635	0.293	4410±18.7	4770
		1.27	0.587	2610±7.55	3720
		2.54	1.17	1140±3.72	2130
Pb	0.797	0.635	0.505	1050±3.96	970
		1.27	1.01	316±0.34	310
		2.54	2.02	36±0.18	47
W	1.23	0.635	0.782	524±3.03	600
		1.27	1.56	104±0.66	134
		2.54	3.12	5.59±0.03	10
U	1.46	0.635	0.927	324±0.66	280
		1.27	1.85	38.30±0.11	42
		2.54	3.71	2.02±0.02	1.8

- (1) The results presented here have been extracted from the
results for this geometry: source number 3
separation distance - one meter
- (2) The values tabulated here are valid for 1 MeV gamma photons
only.

SECTION 6

SOURCE-TO-DOSE-POINT SEPARATION DISTANCE EFFECTS

As discussed in Section 3, the radiation dose rates at given distances from small, compact sources are inversely proportional to the square of the separation distance between the source and the detector. In this experiment two separation distances (19 cm and 100 cm) were measured, and selected results, extracted from the experimental data tabulations of Appendix A, are presented in the following table.

Table 6-1 Variation of Measured and Calculated Radiation Dose Rates⁽¹⁾ with Separation Distance

Absorber Material	Dose Rate (mR/hr)		Inverse Proportionality Factor
	19 cm	100 cm	
Al measured calculated	171,000±1980	5030±16.3	2.94 (10 ⁻²)
	160,000	5800	3.62 (10 ⁻²)
SS measured calculated	73,800±418	2610±7.55	3.53 (10 ⁻²)
	102,000	3720	3.64 (10 ⁻²)
Pb measured calculated	6960±22.4	316±0.34	4.54 (10 ⁻²)
	8720	310	3.55 (10 ⁻²)
W measured calculated	3140±17.0	104±0.66	3.31 (10 ⁻²)
	3700	134	3.62 (10 ⁻²)
U measured calculated	979±8.65	38.3±0.11	3.91 (10 ⁻²)
	1170	42	3.58 (10 ⁻²)

(1) The results presented here have been extracted from the data of this configuration: source number three
absorber number two

SECTION 7

SOURCE STRENGTH EFFECTS

The general equation which describes the collided photon flux detected at a given distance (r) from a point isotopic radiation source through a thickness (x) of absorber material is:

$$\phi(r,x) = S_0 \frac{B(\mu x)}{4\pi r^2} e^{-\mu x} \quad (4)$$

where all terms are as previously described and,

S_0 = source strength, photons/sec

Accordingly, the equation which describes the radiation dose induced at the detector by this flux is:

$$D(r,x) = (K)\phi(r,x)$$

where: K = conversion factor from photon flux-to-dose rate.

Thus, it is evident that radiation dose rates vary linearly with strength of small (point) sources. Despite the perturbing influences of larger source sizes, collimated geometry self-absorption within sources and build-up within absorbers, it was anticipated that the measured radiation dose rates would show this linear variation with strength of Thulium-170 source strengths. Selected measurement results extracted from data of Appendix A are tabulated below.

Table 7-1 Variation of Measured and Calculated Radiation
Dose Rates (1) With Source Strength

Absorber Material	Source Number	Dose Rates (mR/hr)	
		Measured	Calculated
Aluminum	3	178,000±2140	161,000
	2	141,000±2420	119,000
	1	94,700±986	81,600
Stainless Steel	3	131,000±1492	132,000
	2	87,400±406	95,400
	1	57,400±363	63,600
Lead	3	25,300±172	27,300
	2	15,900±241	17,200
	1	9920±111	9970
Tungsten	3	15,100±180	16,600
	2	9380±104	10,200
	1	5060±50	5850
Uranium	3	5780±59.8	7710
	2	3540±21.0	4650
	1	2200±33.8	2620

(1) The results presented here have been extracted from the
data for this configuration: absorber one
source separation - 19 cm

SECTION 8

MEASURED AND PREDICTED DOSE RATES COMPARISONS

The objective of this study was to obtain measured and calculated values of radiation dose rates for representative Thulium-170 oxide sources, and to determine from their comparison whether significant error is introduced into shield design calculations by the assumptions of isotopic purity of such sources.

Tabular comparisons of the measured vs. predicted Thulium-170 dose rate data for the five absorber materials are presented in Appendix A. These same results are presented graphically below in Figures 8-1 through 8-5. Cursory examination of these results indicates that, while the agreement between predicted vs. measured values for the heavier absorbers (i. e., lead, tungsten) seems acceptable, the results for aluminum, stainless steel, and depleted uranium appear to show marked variances between corresponding curves. It is thought that the reasons for these discrepancies with aluminum and stainless steel lie in the nature and behaviour of the build-up factors assumed for these materials for purposes of calculation (see Appendix C). The divergence in the results for depleted uranium absorbers, on the other hand, is thought to arise due to the radioactivity inherent with this material. Further examination of the curves shows that for aluminum and stainless steel absorbers, the predicted and measured values appear in reasonable agreement for the thin (1/4 inch) absorber specimens, but that the results diverge significantly with increasing thickness.

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Figure 8-1 Variation of Calculated and Measured Thulium-170 Dose Rates with Thickness of Aluminum Absorber.

Figure 8-2 Variation of Calculated and Measured Thulium-170 Dose Rates with Thickness of Stainless Steel Absorber.

Figure 8-3 Variation of Calculated and Measured Thulium-170 Dose Rates with Thickness of Lead Absorber.

69-H65987-004

Figure 8-4 Variation of Calculated and Measured Thulium-170 Dose Rates with Thickness of Tungsten Absorber.

69-H65987-005

Figure 8-5 Variation of Calculated and Measured Thulium-170 Dose Rates with Thickness of Uranium Absorber.

As noted in the discussion of Section 5, as absorber thickness becomes increased, there is a corresponding increase in the number of relaxation lengths within the absorber for each particular photon energy. Therefore, the interaction probability for each energy photon likewise increases; and as a result, it would normally be anticipated that the residual photon flux would decline with increased absorber thickness. The experimental results in each case appear to substantiate this. However, the predicted results show no such decrease, and it is felt that the explanation for this may be found in further examination of the infinite build-up factors assumed for these calculations.

The use of this factor means an assumption of both source and dose point contained within an infinite homogeneous absorbing medium where the source emission, dose point radiation absorption, and all radiation interactions with the attenuating medium occur isotropically. Thus, for materials such as aluminum and stainless steel, with very low attenuation coefficients, each Compton scatter interaction could be expected to lead to an extremely large number of subsequent interactions, because the secondary photons would be only slightly less penetrating than the original in each interaction event. Consequently, a flux of photons passing through an infinite quantity of such materials survive many scattering interactions and a significant number would become incident upon the dose point. This is in contrast with the heavier ($Z > 40$) absorber materials which have very high absorption coefficients for the secondary photons produced from Compton scattering.

Therefore, for dose rate calculations which assume infinite build-up factors in light absorbers, the radiation flux about the source should remain fairly constant with increasing separation distances out to very large numbers of relaxation lengths. This behaviour appears in the calculated dose rate results presented herein.

APPENDIX A
SUMMARY DATA TABULATIONS

TABLE A-1
DATA SUMMARY FOR ALUMINUM ABSORBERS

Source Strength (watts $\pm \sigma$)	Absorber Thickness (cm)	Exposure Duration (hours)	Average Measured Dose (mR $\pm \sigma$)	Average Zero Dose (mR $\pm \sigma$)	Average Net Dose (mR $\pm \sigma$)	Average Measured Dose Rate (mR/hr)	Normalized Measured Dose Rate (mR/hr)	Predicted Dose Rate (mR/hr)	Ratio: Predicted to Measured
1. 19 cm.									
10.11 $\pm$ 0.05	0.635	0.25	21,720.0 $\pm$ 261.0	6.26 $\pm$ 0.11	21,713.74 $\pm$ 261.0	86,854.96 $\pm$ 104.0	178,000 $\pm$ 2,140.0	161,000	0.90
10.11 $\pm$ 0.05	1.27	0.25	20,900.0 $\pm$ 242.0	6.26 $\pm$ 0.11	20,893.74 $\pm$ 242.0	83,574.96 $\pm$ 968.0	171,000 $\pm$ 1,980.0	160,000	0.94
10.11 $\pm$ 0.05	2.54	0.25	16,193.0 $\pm$ 201.0	6.26 $\pm$ 0.11	16,186.74 $\pm$ 201.0	64,746.96 $\pm$ 804.0	133,000 $\pm$ 1,650.0	156,000	1.17
5.05 $\pm$ 0.05	0.635	0.25	17,786.0 $\pm$ 305.0	9.18 $\pm$ 0.26	17,776.82 $\pm$ 305.0	46,883.56 $\pm$ 488.0	141,000 $\pm$ 2,420.0	119,000	0.84
5.05 $\pm$ 0.05	1.27	0.25	14,266.0 $\pm$ 128.0	9.18 $\pm$ 0.26	14,256.82 $\pm$ 128.0	36,331.56 $\pm$ 128.0	113,000 $\pm$ 1,010.0	118,000	1.04
5.05 $\pm$ 0.05	2.54	0.25	12,453.0 $\pm$ 110.0	9.18 $\pm$ 0.26	12,443.82 $\pm$ 110.0	25,935.56 $\pm$ 148.0	98,600 $\pm$ 871.0	115,000	1.17
2.48 $\pm$ 0.05	0.635	0.25	11,733.0 $\pm$ 122.0	12.11 $\pm$ 0.09	11,720.89 $\pm$ 122.0	46,883.56 $\pm$ 488.0	94,700 $\pm$ 986.0	81,600	0.86
2.48 $\pm$ 0.05	1.27	0.25	9,095.0 $\pm$ 32.0	12.11 $\pm$ 0.09	9,082.89 $\pm$ 32.0	36,331.56 $\pm$ 128.0	73,400 $\pm$ 259.0	81,100	1.11
2.48 $\pm$ 0.05	2.54	0.25	6,496.0 $\pm$ 37.0	12.11 $\pm$ 0.09	6,483.89 $\pm$ 37.0	25,935.56 $\pm$ 148.0	52,400 $\pm$ 300.0	79,000	1.51
2. 100 cm.									
14.96 $\pm$ 0.07	0.635	0.5	2,175.3 $\pm$ 11.6	41.11 $\pm$ 0.21	2,134.19 $\pm$ 11.6	4,268.38 $\pm$ 23.20	5,900 $\pm$ 32.1	5,800	0.98
14.96 $\pm$ 0.07	1.27	0.5	1,858.6 $\pm$ 5.90	41.11 $\pm$ 0.21	1,817.49 $\pm$ 5.90	3,634.98 $\pm$ 11.80	5,030 $\pm$ 16.3	5,800	1.15
14.96 $\pm$ 0.07	2.54	0.5	1,352.6 $\pm$ 5.30	41.11 $\pm$ 0.21	1,311.49 $\pm$ 5.30	2,622.98 $\pm$ 10.60	3,630 $\pm$ 14.7	5,600	1.54
5.05 $\pm$ 0.05	0.635	0.5	1,215.3 $\pm$ 7.08	9.52 $\pm$ 0.25	1,205.78 $\pm$ 7.05	2,411.56 $\pm$ 14.10	4,780 $\pm$ 27.9	4,300	0.90
5.05 $\pm$ 0.05	1.27	0.5	1,056.0 $\pm$ 3.38	9.52 $\pm$ 0.25	1,046.48 $\pm$ 3.39	2,092.96 $\pm$ 6.78	4,140 $\pm$ 13.4	4,300	1.04
5.05 $\pm$ 0.05	2.54	0.5	697.5 $\pm$ 2.83	9.52 $\pm$ 0.25	687.94 $\pm$ 2.87	1,375.88 $\pm$ 5.74	2,720 $\pm$ 11.4	4,200	1.54
3.98 $\pm$ 0.08	0.635	1.0	2,165.3 $\pm$ 7.20	20.04 $\pm$ 0.13	2,145.26 $\pm$ 7.20	2,145.26 $\pm$ 7.20	2,700 $\pm$ 9.06	2,960	1.10
3.98 $\pm$ 0.08	1.27	1.0	1,810.7 $\pm$ 5.10	20.04 $\pm$ 0.13	1,790.66 $\pm$ 5.10	1,790.66 $\pm$ 5.10	2,250 $\pm$ 6.42	2,920	1.30
3.98 $\pm$ 0.08	2.54	4.0	4,772.0 $\pm$ 15.4	80.16 $\pm$ 0.64	4,691.84 $\pm$ 15.41	1,172.96 $\pm$ 3.85	1,480 $\pm$ 4.84	2,850	1.93

TABLE A-2
DATA SUMMARY FOR STAINLESS STEEL ABSORBERS

Source Strength (watts $\pm \sigma$)	Absorber Thickness (cm)	Exposure Duration (hours)	Average Measured Dose (mR $\pm \sigma$)	Average Zero Dose (mR $\pm \sigma$)	Average Net Dose (mR $\pm \sigma$)	Average Measured Dose Rate (mR/hr)	Normalized Measured Dose Rate (mR/hr)	Predicted Dose Rate (mR/hr)	Ratio Predicted to Measured
<u>1. 19 cm.</u>									
10.11 $\pm$ 0.05	0.635	0.25	16,053.0 $\pm$ 182.0	4.22 $\pm$ 0.07	16,048.78 $\pm$ 182.0	64,195.00 $\pm$ 728.0	131,000 $\pm$ 1,492.0	132,000	1.01
10.11 $\pm$ 0.05	1.27	0.25	9,007.0 $\pm$ 51.0	4.22 $\pm$ 0.07	9,002.78 $\pm$ 51.0	36,011.00 $\pm$ 204.0	73,800 $\pm$ 418.0	102,000	1.38
10.11 $\pm$ 0.05	2.54	0.25	3,949.0 $\pm$ 28.0	4.22 $\pm$ 0.07	3,944.78 $\pm$ 28.0	15,779.00 $\pm$ 112.0	32,300 $\pm$ 229.0	59,000	1.83
4.92 $\pm$ 0.05	0.635	0.25	10,754.0 $\pm$ 50.0	4.89 $\pm$ 0.13	10,749.11 $\pm$ 49.9	42,996.44 $\pm$ 200.0	87,400 $\pm$ 406.0	95,400	1.09
4.92 $\pm$ 0.05	1.27	0.25	6,812.0 $\pm$ 44.5	4.89 $\pm$ 0.13	6,807.11 $\pm$ 44.5	27,228.44 $\pm$ 178.0	55,300 $\pm$ 362.0	73,400	1.33
4.92 $\pm$ 0.05	2.54	0.25	3,084.0 $\pm$ 41.2	4.89 $\pm$ 0.13	3,079.11 $\pm$ 41.2	12,316.44 $\pm$ 165.0	25,000 $\pm$ 335.0	41,200	1.65
2.48 $\pm$ 0.05	0.635	0.25	7,147.3 $\pm$ 45.1	11.2 $\pm$ 0.12	7,136.08 $\pm$ 45.1	28,544.32 $\pm$ 180.0	57,400 $\pm$ 363.0	63,600	1.11
2.48 $\pm$ 0.05	1.27	0.25	4,353.3 $\pm$ 38.1	11.2 $\pm$ 0.12	4,342.08 $\pm$ 38.1	17,368.32 $\pm$ 152.0	34,900 $\pm$ 306.0	48,100	1.38
2.48 $\pm$ 0.05	2.54	0.25	7,894.6 $\pm$ 19.0	11.2 $\pm$ 0.12	1,883.38 $\pm$ 19.0	7,533.52 $\pm$ 76.0	15,100 $\pm$ 153.0	26,300	1.74
<u>2. 100 cm.</u>									
15.12 $\pm$ 0.07	0.635	0.5	1,655.3 $\pm$ 6.81	42.3 $\pm$ 0.77	1,613.00 $\pm$ 6.85	3,226.00 $\pm$ 13.7	4,410 $\pm$ 18.7	4,770	1.08
15.12 $\pm$ 0.07	1.27	0.5	995.6 $\pm$ 2.65	42.3 $\pm$ 0.77	953.30 $\pm$ 2.76	1,906.06 $\pm$ 5.52	2,610 $\pm$ 7.55	3,720	1.43
15.12 $\pm$ 0.07	2.54	0.5	459.9 $\pm$ 1.12	42.3 $\pm$ 0.77	417.60 $\pm$ 1.36	835.20 $\pm$ 2.72	1,140 $\pm$ 3.72	2,130	1.87
5.08 $\pm$ 0.05	0.635	0.75	1,262.6 $\pm$ 3.76	6.97 $\pm$ 0.13	1,255.63 $\pm$ 3.76	1,674.17 $\pm$ 5.01	3,290 $\pm$ 9.86	3,430	1.04
5.08 $\pm$ 0.05	1.27	0.75	717.2 $\pm$ 1.57	6.97 $\pm$ 0.13	710.23 $\pm$ 1.57	946.97 $\pm$ 2.09	1,860 $\pm$ 4.11	2,640	1.42
5.08 $\pm$ 0.05	2.54	1.50	491.7 $\pm$ 1.75	13.94 $\pm$ 0.25	477.76 $\pm$ 1.77	318.51 $\pm$ 1.18	627 $\pm$ 2.32	1,480	2.36
3.90 $\pm$ 0.08	0.635	1.5	2,009.1 $\pm$ 11.5	38.67 $\pm$ 0.14	1,970.43 $\pm$ 11.50	1,313.62 $\pm$ 7.67	1,680 $\pm$ 9.82	2,300	1.37
3.90 $\pm$ 0.08	1.27	1.5	1,158.7 $\pm$ 5.79	38.67 $\pm$ 0.14	1,120.03 $\pm$ 5.79	746.69 $\pm$ 3.86	956 $\pm$ 4.94	1,730	1.81
3.90 $\pm$ 0.08	2.54	1.5	481.6 $\pm$ 2.53	38.67 $\pm$ 0.14	442.93 $\pm$ 2.53	295.29 $\pm$ 1.69	378 $\pm$ 2.16	940	2.49

TABLE A-3
DATA SUMMARY FOR LEAD ABSORBERS

Source Strength (watts $\pm \sigma$)	Absorber Thickness (cm)	Exposure Duration (hours)	Average Measured Dose (mR $\pm \sigma$)	Average Zero Dose (mR $\pm \sigma$)	Average Net Dose (mR $\pm \sigma$)	Average Measured Dose Rate (mR/hr)	Normalized Measured Dose Rate (mR/hr)	Predicted Dose Rate (mR/hr)	Ratio: Predicted to Measured
<u>1. 19 cm.</u>									
10.11 $\pm$ 0.05	0.635	0.25	3,085.0 $\pm$ 21.0	1.58 $\pm$ 0.08	3,083.42 $\pm$ 21.00	12,333.68 $\pm$ 84.0	25,300.00 $\pm$ 172.0	27,300.0	1.08
10.11 $\pm$ 0.05	1.27	0.25	850.1 $\pm$ 2.73	1.58 $\pm$ 0.08	848.52 $\pm$ 2.73	3,394.10 $\pm$ 10.9	6,960.00 $\pm$ 22.4	8,720.0	1.25
10.11 $\pm$ 0.05	2.54	1.50	750.5 $\pm$ 3.52	9.48 $\pm$ 0.72	741.02 $\pm$ 3.59	494.00 $\pm$ 2.39	1,010.00 $\pm$ 4.90	1,320.0	1.31
4.92 $\pm$ 0.05	0.635	0.25	1,958.4 $\pm$ 29.7	1.0 $\pm$ 0.03	1,957.40 $\pm$ 29.7	7,829.60 $\pm$ 118.0	15,900.00 $\pm$ 241.0	17,200.0	1.08
4.92 $\pm$ 0.05	1.27	0.50	1,226.3 $\pm$ 7.83	2.0 $\pm$ 0.06	1,224.30 $\pm$ 7.83	2,448.60 $\pm$ 15.7	4,980.00 $\pm$ 31.8	5,270.0	1.06
4.92 $\pm$ 0.05	2.54	3.00	972.9 $\pm$ 3.27	11.9 $\pm$ 0.36	961.10 $\pm$ 3.29	320.30 $\pm$ 1.10	651.00 $\pm$ 2.24	774.0	1.19
2.48 $\pm$ 0.05	0.635	0.25	1,234.7 $\pm$ 13.8	2.18 $\pm$ 0.05	1,232.50 $\pm$ 13.8	4,930.00 $\pm$ 55.2	4,920.00 $\pm$ 111.0	9,970.0	1.01
2.48 $\pm$ 0.05	1.27	0.50	708.3 $\pm$ 3.35	4.36 $\pm$ 0.09	703.90 $\pm$ 3.35	1,407.90 $\pm$ 6.70	2,830.00 $\pm$ 13.5	2,980.0	1.05
2.48 $\pm$ 0.05	2.54	3.50	735.13 $\pm$ 3.46	30.50 $\pm$ 0.63	704.60 $\pm$ 3.52	201.30 $\pm$ 1.01	405.00 $\pm$ 2.03	430.0	1.06
<u>2. 100 cm.</u>									
15.00 $\pm$ 0.05	0.635	1.0	771.80 $\pm$ 2.87	7.19 $\pm$ 0.03	764.67 $\pm$ 2.87	764.67 $\pm$ 2.87	1,050.00 $\pm$ 3.96	970.0	0.92
15.00 $\pm$ 0.05	1.27	2.0	472.80 $\pm$ 0.50	14.38 $\pm$ 0.06	458.48 $\pm$ 0.50	229.24 $\pm$ 0.25	316.00 $\pm$ 0.34	310.0	0.98
15.00 $\pm$ 0.05	2.54	15.0	499.20 $\pm$ 1.85	107.85 $\pm$ 0.45	391.35 $\pm$ 1.90	26.09 $\pm$ 0.13	36.00 $\pm$ 0.18	47.0	1.31
7.52 $\pm$ 0.07	0.635	1.0	485.50 $\pm$ 1.45	4.83 $\pm$ 0.03	480.70 $\pm$ 1.45	480.70 $\pm$ 1.45	639.0 $\pm$ 1.93	614.0	0.96
7.52 $\pm$ 0.07	1.27	3.0	420.30 $\pm$ 1.38	14.49 $\pm$ 0.08	405.84 $\pm$ 1.38	135.28 $\pm$ 0.46	180.0 $\pm$ 0.61	190.0	1.06
7.52 $\pm$ 0.07	2.54	20.0	376.10 $\pm$ 1.05	96.66 $\pm$ 0.54	279.47 $\pm$ 1.16	13.97 $\pm$ 0.06	18.6 $\pm$ 0.08	28.0	1.51
3.86 $\pm$ 0.07	0.635	4.0	1,214.70 $\pm$ 3.59	6.24 $\pm$ 0.03	1,208.43 $\pm$ 3.59	302.11 $\pm$ 0.90	390.8 $\pm$ 1.16	360.0	0.92
3.86 $\pm$ 0.07	1.27	15.0	958.60 $\pm$ 3.91	23.40 $\pm$ 0.11	935.17 $\pm$ 3.91	62.34 $\pm$ 0.26	80.65 $\pm$ 0.34	110.0	1.38
3.86 $\pm$ 0.07	2.54	74.0	660.10 $\pm$ 3.67	115.44 $\pm$ 0.56	544.62 $\pm$ 3.71	7.36 $\pm$ 0.05	9.52 $\pm$ 0.06	15.5	1.63

TABLE A-4
DATA SUMMARY FOR TUNGSTEN ABSORBERS

Source Strength (watts $\pm \sigma$)	Absorber Thickness (cm)	Exposure Duration (hours)	Average Measured Dose (mR $\pm \sigma$)	Average Zero Dose (mR $\pm \sigma$)	Average Net Dose (mR/hr)	Average Measured Dose Rate (mR/hr)	Normalized Measured Dose Rate (mR/hr)	Predicted Dose Rate (mR/hr)	Ratio Predicted to Measured
<u>1. 19 cm.</u>									
10.55 $\pm$ 0.05	0.635	0.25	1925.3 $\pm$ 22.91	0.79 $\pm$ 0.02	1924.54 $\pm$ 22.92	7698.16 $\pm$ 91.7	15,100 $\pm$ 180	16,600	1.10
10.55 $\pm$ 0.05	1.27	0.50	802.5 $\pm$ 4.34	1.58 $\pm$ 0.04	800.89 $\pm$ 4.34	1601.78 $\pm$ 8.68	3,140 $\pm$ 17.0	3,700	1.18
10.55 $\pm$ 0.05	2.54	6.25	842.4 $\pm$ 4.57	19.75 $\pm$ 0.50	822.65 $\pm$ 4.60	131.62 $\pm$ 0.74	258 $\pm$ 1.45	288	1.12
5.13 $\pm$ 0.05	0.635	0.25	1216.7 $\pm$ 13.36	0.357 $\pm$ 0.009	1203.31 $\pm$ 13.36	4813.24 $\pm$ 53.44	9,380 $\pm$ 104	10,200	1.09
5.13 $\pm$ 0.05	1.27	0.75	754.6 $\pm$ 3.32	1.00 $\pm$ 0.027	753.53 $\pm$ 3.32	1004.71 $\pm$ 4.43	1,960 $\pm$ 8.64	2,210	1.13
5.13 $\pm$ 0.05	2.54	22.5	1652.7 $\pm$ 20.3	32.13 $\pm$ 0.765	1620.54 $\pm$ 20.3	72.02 $\pm$ 0.90	140.4 $\pm$ 1.75	167	1.19
2.58 $\pm$ 0.05	0.635	1	2616 $\pm$ 25.85	1.00 $\pm$ 0.03	2615.00 $\pm$ 25.85	2615.00 $\pm$ 25.85	5,060 $\pm$ 50	5,850	1.16
2.58 $\pm$ 0.05	1.27	2	1098.7 $\pm$ 6.20	2.00 $\pm$ 0.06	1096.67 $\pm$ 6.20	548.34 $\pm$ 3.10	1,060 $\pm$ 6	1,230	1.16
2.58 $\pm$ 0.05	2.54	18	765.1 $\pm$ 5.71	18.00 $\pm$ 0.54	747.07 $\pm$ 5.73	41.50 $\pm$ 0.32	80.3 $\pm$ 0.62	92	1.15
<u>2. 100 cm.</u>									
11.97 $\pm$ 0.05	0.635	1.25	379.7 $\pm$ 2.19	1.103 $\pm$ 0.004	378.63 $\pm$ 2.19	302.90 $\pm$ 1.75	524 $\pm$ 3.03	600	1.15
11.97 $\pm$ 0.05	1.27	4.5	273.8 $\pm$ 1.70	3.974 $\pm$ 0.017	269.89 $\pm$ 1.70	59.98 $\pm$ 0.38	104 $\pm$ 0.66	134	1.29
11.97 $\pm$ 0.05	2.54	72.5	298.5 $\pm$ 1.65	64.024 $\pm$ 0.275	234.51 $\pm$ 1.67	3.23 $\pm$ 0.02	5.59 $\pm$ 0.03	10	1.79
5.94 $\pm$ 0.06	0.635	2	361.4 $\pm$ 1.41	0.916 $\pm$ 0.002	360.54 $\pm$ 1.41	180.27 $\pm$ 0.71	303 $\pm$ 1.20	370	1.22
5.94 $\pm$ 0.06	1.27	7	257.1 $\pm$ 0.654	3.207 $\pm$ 0.009	253.85 $\pm$ 0.66	36.26 $\pm$ 0.09	61.0 $\pm$ 0.15	79	1.30
5.94 $\pm$ 0.06	2.54	120	285.9 $\pm$ 0.856	54.984 $\pm$ 0.156	230.95 $\pm$ 0.87	1.93 $\pm$ 0.01	3.25 $\pm$ 0.02	6	1.85
3.10 $\pm$ 0.06	0.635	3	419.4 $\pm$ 1.89	2.397 $\pm$ 0.018	417.01 $\pm$ 1.89	139.00 $\pm$ 0.63	224 $\pm$ 1.02	210	0.94
3.10 $\pm$ 0.06	1.27	18	451.0 $\pm$ 5.16	14.380 $\pm$ 0.108	436.62 $\pm$ 5.16	24.26 $\pm$ 0.29	39.1 $\pm$ 0.47	44	1.13
3.10 $\pm$ 0.06	2.54	168	349.8 $\pm$ 1.08	134.215 $\pm$ 1.008	215.58 $\pm$ 1.48	1.28 $\pm$ 0.009	2.06 $\pm$ 0.02	3.3	1.60

TABLE A-5
DATA SUMMARY FOR DEPLETED URANIUM ABSORBERS

Source Strength (watts $\pm \sigma$)	Absorber Thickness (cm)	Exposure Duration (hours)	Average Measured Dose (mR $\pm \sigma$)	Average Zero Dose (mR $\pm \sigma$)	Average Net Dose (mR $\pm \sigma$)	Average Measured Dose Rate (mR/hr)	Normalized Measured Dose Rate (mR/hr)	Predicted Dose Rate (mR/hr)	Ratio Predicted to Measured
<u>1. 19 cm.</u>									
11.08 $\pm$ 0.05	0.635	1	3115.3 $\pm$ 32.0	1.667 $\pm$ 0.011	3113.63 $\pm$ 32	3113.63 $\pm$ 32.00	5780 $\pm$ 59.8	7,710	1.33
11.08 $\pm$ 0.05	1.27	2	1090.0 $\pm$ 9.24	3.334 $\pm$ 0.022	1086.67 $\pm$ 9.24	543.34 $\pm$ 4.62	979 $\pm$ 8.65	1,170	1.20
11.08 $\pm$ 0.05	2.54	48	3844.0 $\pm$ 20.6	80.016 $\pm$ 0.528	3763.98 $\pm$ 20.61	78.42 $\pm$ 0.43	81.6 $\pm$ 1.43	50	0.61
5.48 $\pm$ 0.05	0.635	1	1961.3 $\pm$ 11.5	0.931 $\pm$ 0.005	1960.37 $\pm$ 11.5	1960.37 $\pm$ 11.5	3540 $\pm$ 21.0	4,650	1.31
5.48 $\pm$ 0.05	1.27	3	1012.7 $\pm$ 8.40	2.793 $\pm$ 0.015	1009.97 $\pm$ 8.40	336.66 $\pm$ 2.80	577 $\pm$ 5.13	680	1.18
5.48 $\pm$ 0.05	2.54	88	5971.3 $\pm$ 34.9	81.928 $\pm$ 0.449	5889.37 $\pm$ 34.90	66.92 $\pm$ 0.40	57.1 $\pm$ 1.39	29	0.51
2.75 $\pm$ 0.05	0.635	1	1230 $\pm$ 18.6	0.743 $\pm$ 0.0065	1229.26 $\pm$ 18.60	1229.26 $\pm$ 18.60	2200 $\pm$ 33.8	2,620	1.19
2.75 $\pm$ 0.05	1.27	4	932.4 $\pm$ 4.17	2.972 $\pm$ 0.0260	929.49 $\pm$ 4.17	232.37 $\pm$ 1.04	386 $\pm$ 1.95	397	1.03
2.75 $\pm$ 0.05	2.54	108	6978 $\pm$ 49.2	80.244 $\pm$ 0.702	6897.76 $\pm$ 49.20	63.87 $\pm$ 0.46	51.1 $\pm$ 1.45	16	0.31
<u>2. 100 cm.</u>									
13.42 $\pm$ 0.06	0.635	2	421.8 $\pm$ 0.863	1.462 $\pm$ 0.0106	420.34 $\pm$ 0.86	210.17 $\pm$ 0.43	324 $\pm$ 0.66	280	0.86
13.42 $\pm$ 0.06	1.27	20	510.9 $\pm$ 1.48	14.62 $\pm$ 0.106	496.31 $\pm$ 1.48	24.82 $\pm$ 0.07	38.3 $\pm$ 0.11	42	1.10
13.42 $\pm$ 0.06	2.54	169	345.4 $\pm$ 0.991	123.53 $\pm$ 0.895	221.87 $\pm$ 1.33	1.31 $\pm$ 0.01	2.02 $\pm$ 0.02	1.8	0.89
7.18 $\pm$ 0.07	0.635	3	498.7 $\pm$ 2.11	2.202 $\pm$ 0.0171	496.53 $\pm$ 2.11	165.51 $\pm$ 0.70	231 $\pm$ 0.97	170	0.74
7.18 $\pm$ 0.07	1.27	24	440.8 $\pm$ 1.71	17.616 $\pm$ 0.1368	423.18 $\pm$ 1.71	17.63 $\pm$ 0.07	24.6 $\pm$ 0.10	25	1.02
7.18 $\pm$ 0.07	2.54	168	405.7 $\pm$ 1.12	123.312 $\pm$ 0.9576	282.42 $\pm$ 1.47	1.68 $\pm$ 0.01	2.34 $\pm$ 0.01	1.0	0.43
3.41 $\pm$ 0.07	0.635	6	436.6 $\pm$ 0.93	3.50 $\pm$ 0.017	433.10 $\pm$ 0.93	72.18 $\pm$ 0.16	106 $\pm$ 0.23	94	0.89
3.41 $\pm$ 0.07	1.27	65	554.2 $\pm$ 1.13	37.92 $\pm$ 0.182	516.28 $\pm$ 1.14	7.94 $\pm$ 0.02	11.6 $\pm$ 0.03	14	1.21
3.41 $\pm$ 0.07	2.54	168	241.2 $\pm$ 0.52	96.26 $\pm$ 0.462	145.00 $\pm$ 0.695	0.863 $\pm$ 0.004	1.26 $\pm$ 0.01	0.58	0.46

APPENDIX B

EXPERIMENTAL APPARATUS AND PROCEDURES

APPENDIX B.1

SOURCE DESCRIPTION

The individual radiation sources used in these measurements consisted of doubly encapsulated stacks of Thulium-170 oxide wafers as depicted in Figure B.1-1. As noted, both the exterior capsule and liner were Hastelloy alloy C.

The thermal and radiation strengths of the sources are given in the following table:

Table B.1-1 Thermal and Radiation Strengths of Thulium-170 Sources

	Thermal Strength (watts)	Radiation Strength (curies)
Source No. 1	5.0 $\pm$ 0.1	2225 $\pm$ 44.5
Source No. 2	10.0 $\pm$ 0.1	4450 $\pm$ 44.5
Source No. 3	20.7 $\pm$ 0.1	9211 $\pm$ 44.5

These thermal strengths were calorimetrically determined on 2 August 1968 by ORNL using instrumentation and techniques described in Appendix D.1. As noted, the accuracy of these measurements is estimated at $\pm$ 2% at the 95% confidence level.

Note: Precise capsule dimensions are presented in Table B.1-5

Figure B.1-1 Representative Thulium Source Capsule

One wafer from the batch irradiated to produce these sources was dissolved by ORNL for analysis under the assumption that it was representative of the batch. A mass analysis made on 10 July 1968 gave the following thulium isotope distribution:

Table B.1-2 Thulium Isotopic Analysis (Thulium Sources)

Tm-169	76.8%
Tm-170	17.7%
Tm-171	5.5%

A chemical analysis of the solution performed on 25 June 1968 determined the ytterbium content to be 17% by weight.

The individual Tm_2O_3 wafers used in the sources are described in the following table:

Table B.1-3 Physical Characteristics of Thulium Sources (Thulia Wafers)

Wafer No.	Weight (gm)	Diameter (in.)	Thickness (mils)	Theoretical Density (%)
1	4.8768	0.736	98.5	80.0
2	4.8500	0.730	100.1	79.6
3	5.045	0.732	99.9	83.7
5	4.5846	0.742	95.5	76.2
6	4.8380	0.735	99.4	78.7
7	4.9690	0.738	100.0	79.9
8	4.9243	0.734	99.5	79.7

The distribution of these wafers in the sources was as follows:

Table B.1-4 Distribution of Thulia Wafers in Thulium Sources
Wafer(s)

Source No. 1	6
Source No. 2	5, 8
Source No. 3	1, 2, 3, 7

The following tabulation gives the precise dimensions (in inches)
of the exterior capsules and their liners.

Table B.1-5 Dimensions of Thulium Sources - Exterior Capsules and Sizes

Capsule No.	Bottom Thickness	Wall Thickness	Capsule OD	Capsule ID	Capsule Height
E X T E R I O R C A P S U L E S					
1	0.0513-0.0518	0.0330-0.0352	0.9210	0.8520	0.960
2	0.0511-0.0514	0.0327-0.0340	0.9200	0.8530	0.960
3	0.0512-0.0515	0.0326-0.0335	0.9190	0.8525	0.959
C A P S U L E L I N E R S					
1	0.0495-0.0503	0.0388-0.0405	0.842	0.760	0.6715
2	0.0495-0.0505	0.0365-0.0380	0.844	0.7674	0.670
3	0.0495-0.0505	0.0385-0.0405	0.842	0.761	0.6684

APPENDIX B.2

DOSE RATE MEASUREMENT APPARATUS

The complete group of equipment used in this experiment to perform Thulium-170 dose rate measurements is shown in Figures B.2-1 through B.2-4. They are also schematically depicted in Figures C.1-6 in the next appendix. Figure B.2-1 is an axial photograph of the measurement fixture showing the source shield cask with the beam tunnel shutter open and the one-quarter inch lead absorber in the absorber holder. Also shown is the radiation scatter shield (described in a later section) and the radiation dosimeters in their holder at the one meter irradiation position. Figure B.2-2 is a disassembled view of the source shield cask and the absorber holder showing the shield cask plug and the aluminum source holder. Also shown is a model fuel capsule. Figure B.2-3 is a top view of the partially assembled source shield cask and absorber holder showing the source holder in position inside the cask. Figure B.2-4 shows the thermoluminescent dosimetry (TLD) reader used in the experiment (Con-Rad Model 5100B) to evaluate the dosimeters. Also shown is a dosimeter holder loaded with five black polyethylene dosimeter capsules and the vibratory instrument (described more fully in Section D. 2 .4) which loads precise quantities of TLD phosphor powder (LiF) into them.

Figure B.2-1 Dose Rate Measurement Fixture

Figure B.2-2 Source Shield Cask (Disassembled)

Figure B.2-3 Top View of a Partially Assembled Source Shield Cask

Figure B.2-4 Thermoluminescent Dosimetry (TLD) Reader

APPENDIX B.3

DOSE RATE MEASUREMENT PROCEDURE

The approved procedure followed in performing these dose rate measurements (SNP - 100017, Rev. 1, Sheets 1 through 22) is attached hereto.

REVISION STATUS OF EACH SHEET

REVISIONS

SHEET			1	2	3	4	5	6	7	8	9	10	11	12	13
REVISION			P			A	B	C	D			E	F	G	H
14	15	16	17	18	19	20	21	22	23	24	25	26	27	28	29
I	J	K	L	M	N	N	P								
30	31	32	33	34	35	36	37	38	39	40	41	42	43	44	45

NOTES

- 1 SHEET ONE REVISION LETTER IS THE IDENTIFYING REVISION FOR THIS MULTISHEET DWG
- 2 SYMBOL ‡ INDICATES VENDOR ITEM - SEE SPEC/SOURCE CONTROL DWG
- 3 INTERPRET DWG PER 815002

LTR	DESCRIPTION	DATE	APPROVED
P	Sheets 1 and 22 redrawn and revised (SEE OBSOLETE MASTER)	10/3/68	J. P. C.

B-9			CONT NO AT-(40-1)-3693			SANDERS NUCLEAR CORPORATION	
			DR			Nashua, New Hampshire	
			CHK		Thulium 170 Oxide Dose Rate Measurements TEST PROCEDURES		
			DEV J. J. Sullivan				
			MECH/ELEC J. L. Cyr				
			PROJ W. J. Fretague		SIZE A	CODE IDENT NO 28428	SNP 100017 Rev. 1
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APPLICATION							

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0 Introduction

The radiation dose rate measurements described in this procedure will be made on thulium-170 to determine the shielded dose rates from thulium oxide and to reliably evaluate calculated effectiveness of candidate shield materials. Wafers of thulium-170 oxide ($Tm_2^{170}O_3$) will be placed behind various shield materials and the transmitted radiations will be allowed to impinge upon thermoluminescent dosimeters for specific time durations. The resulting dose rates will be determined optically and electronically in special "reading" equipment and compared with calculated values.

Specifically, the radiation dose rates from thulium-170 sources through lead, stainless steel, tungsten, aluminum and depleted uranium absorbers will be determined by measurement of the luminescence of lithium fluoride phosphors at high temperatures. The energy imparted to this material as a result of interactions with incident radiation is released as luminescence at elevated temperatures. The particular dosimeter used for these measurements will consist of precise quantities of LiF powder entered into special holders. Three thulium-170 sources in strengths of 0, 20 and 40 thermal watts will be utilized. Three different thicknesses of each shield material (lead, tungsten, stainless steel, aluminum and depleted uranium) will be used varying from 0.250 to 1.000 inches. The dose rate measurements will be made at the shield surface and one meter.

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Representative exposure periods for lead, tungsten and uranium to
 obtain measurement precision of 5% standard deviation at one meter are
 tabulated below:

<u>Field Thickness</u> <u>(inches)</u>	<u>Exposure Time (Minutes)</u>		
	<u>Lead</u>	<u>Tungsten</u>	<u>Uranium</u>
$\frac{1}{4}$	4	6	20
$\frac{1}{2}$	9	17	75
1.0	45	133	834

0 Objective

The dose rate measurements described herein are to be performed to
 reliably evaluate the calculated effectiveness of various absorber materials
 radiation shields for thulium-170.

0 Test Requirements

3.1 Applicable Documents

The measurements described in this procedure will be carried
 out in accordance with the Statement of Work (Article II) of Contract
 No. AT-(40-1)-3693: Section b - Phase I, part 2-C and Phase II, part
 1-C.

3.2 Measurement Conditions

Radiation dose rate measurements will be performed for each of
three thulium sources and each of three different thicknesses of five

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shield materials. Each measurement will utilize five separate dosimeters in accordance with the matrix outlined in Table 3.2.1. The use of five separate TLD dosimeters for each measurement is expected to assure high reliability of test results.

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DOSE RATE MEASUREMENT MATRIX

Source 1
(75)

Source 3
(75)

Thickness

- 1 - 10 watts
- 2 - 20 watts
- 3 - 40 watts

1 - $\frac{1}{4}$ "
2 - $\frac{1}{2}$ "
3 - 1.0"

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3.3 Dosimeter Readout

After exposure the thermoluminescent dosimeters will be read by placing them in a specially designed heating device which raises them to a specified temperature at which light is emitted proportional to the amount of radiation absorbed by the dosimeter. This light emission is detected by a photomultiplier tube, the integrated output current of which is electronically converted into a dose reading. TLD dosimeters utilizing LiF (TLD-700) are available either in preformed geometrical configurations or as loose powder; the ranges of measurement precision vs. exposure dose for each is tabulated below:¹

TABLE 3.3.1
PRECISION AS A FUNCTION OF EXPOSURE FOR
LiF POWDER (TLD-700) AND LiF-TEFLON DOSIMETERS

Exposure Dose (roentgens)	LiF-Teflon Discs	Standard Deviation (%) TLD-700 Powder
0.01	--	± 20
0.05	± 15	--
0.30	± 10	--
0.10	--	± 5
1.0	± 5	± 2
3.0	± 3	± 2
10.0	± 3	± 2
50 to 10 ⁵	± 3	± 2

1. Bjarngard, B.E., et al: Lithium fluoride - teflon thermoluminescence dosimeters. Proceedings: International Conference of Luminescence Dosimetry. Stanford University, Stanford, California. June 1965.

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* note : capsule mat'l to be HASTELLOY C
capsules to be scribed -
to source strength as

SNC
TM-170
No. 1

and numbered according
indicated in FIGURE 3.21

FIGURE 4.1.1

REPRESENTATIVE
SOURCE CAPSULE

THULIUM

B-18

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3.4 Measurement Failure Criteria

A given dose rate measurement result will be deemed void if the value thereof deviates from the group mean value by a factor greater than the standard deviation listed in Table 3.3.1.

3.5 Test Results

Dose rates will be calculated for each test measurement using computed values of source specific activity and published¹ values of bremsstrahlung emission spectra appearing in the literature. Computed dose rates will be compared with experimental test results and recommendations drafted respecting how anomalies, if any, might be resolved.

3.6 Reports

Monthly status letter reports and a final report will be prepared as stated in the contract.

4.0 Procedure

4.1 Receipt of Thulium-170 Sources

It is anticipated that the sources upon which these measurements are to be performed will arrive at Sanders Nuclear Corporation in

Arnold, E.D. Handbook of Shielding Requirements and Radiation Characteristics of Isotopic Power Sources for Terrestrial, Marine and Space Applications. ORNL-3576. 1964.

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appropriately shielded transportation casks. They will be deposited inside the hot cell where any necessary preliminary operations for source removal will be performed. The hot cell will then be closed and secured. Using remote manipulators in conjunction with a travelling hoist, the casks will be uncapped and the sources removed. Once removed, the sources will be visually inspected and wipe tests will be made on their exterior surfaces. If necessary, weight and dimensional measurements will be made on each source to assure their compatibility with the test fixtures. The radiation level in the hot cell and at the operators station will be continuously monitored and recorded during these operations.

The individual radiation sources will consist of doubly encapsulated stacks of thulium-170 oxide wafers as depicted in Figure 4.1.1. As noted, both the exterior capsule and the lever will be of Hastelloy C to the dimensions indicated. Since each wafer will comprise approximately 10 watts of thermal energy, it is anticipated that the three sources strengths of 10, 20 and 40 watts will consist of 1, 2 and 4 wafers, respectively.

4.2 Dosimeter Exposure Fixture

Having completed preliminary operations with the sources, they will then be individually placed into position on a special radiation

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exposure measurement fixture. During all operations with any one source, all other radioactive sources will be stored inside their shipment casks or storage pigs to reduce extraneous radiation.

The measurement fixture will consist of a rectangular base platform upon which will be mounted: (1) a shielded enclosure for the radiation sources, (2) a fixed holder for the various absorber specimens and (3) another fixed holder for the thermoluminescent dosimeters. This fixture is schematically depicted in Figure 4.2.1. The shield enclosure will incorporate lead walls of adequate thickness that with the strongest anticipated thulium-170 source contained within it, the dose rate at one meter from the enclosure will be no greater than 10 mr/hr. In addition, the enclosure will have a removeable (or hinged) lead cover over the source cavity of likewise appropriate thickness. The cavity itself will be so constructed and aligned with a radiation beam "tunnel" that with the thulium capsule(s) in place, only the axial radiation emission will be allowed to exit through the tunnel and be incident upon the absorber specimen. The source shield enclosure and the absorber holder are depicted in Figure 4.2.2. Both the absorber and dosimeter holders will be axially aligned with the source capsule/beam tunnel centerline. In addition, the dosimeter holder (shown in Figure 4.2.3) will be located so that the dosimeters may be positioned at various distances from the vertical source centerline. Furthermore,

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Figure 4.2.1 - Dose Rate Measurements Schematic Arrangement

Figure 4.2.2 - Source Shield Enclosure Arrangement

Figure 4.2.3 - Dosimeter Mounting Arrangement

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the shield enclosure will incorporate a removeable (or hinged) lead shutter in front of the collimator slit of appropriate thickness that the preceding dose rate specification is maintained.

4.3 Preliminary Test Operations

Prior to commencement of any exposure procedures, the following operations will be performed.

4.3.1. Radiation Absorber Acceptability Criteria

Each of the various absorbers to be used in the measurements will be visually inspected for cleanliness and freedom from surface defects and/or blemishes. These absorbers will then be arranged in special trays (equipped with hinged covers capable of being opened and closed with the hot cell remote manipulators) which will then be placed next to the measurement fixture inside the hot cell within easy reach of the manipulators.

4.3.2. Dosimeter Acceptability Criteria

Each dosimeter to be used in the measurement experiment will be visually inspected for cleanliness and freedom from surface defects and/or blemishes. These dosimeters will then be arranged in special trays (equipped with opaque covers) in groups of eight dosimeters per tray. Three of these trays (24 dosimeters, total) will then be placed within easy reach of the remote manipulators for each dosimeter exposure sequence with a particular absorber

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material. (See Table 3.2.1). In addition, the dosimeters will be located as far as practicable from the stored radiation sources and will be protected by sufficient auxiliary radiation shielding that they will receive minimal spurious radiation exposure during source transfer operations and during purposeful source exposure procedures.

4.3.3 Dosimeter Reader Calibration

The TLD dosimeter reader will be calibrated against a traceable, vendor-supplied standard. This calibration will be repeated at appropriate intervals throughout the performance of measurements.

4.3.4 Dosimeter Geometry Calibration

To assure reproducibility of the geometrical configuration exposure of calibrated TLD dosimeters will be repeated after each change in configuration and/or experiment interruption.

4.4 Test Operations

Having completed the preliminary operations described in Section 4.3, the dosimeter exposure operations will be initiated.

4.4.1 The first radiation source to be measured will be removed from its storage cask and transferred into the uncovered shield enclosure, the beam-hole shutter of which shall be closed. The shield enclosure cover will then be replaced.

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4.4.2 The particular thickness of absorber to be utilized shall be removed from its storage tray and positioned in its special holder in front of the beam-hole shutter.

4.4.3 The first tray of dosimeters to be exposed shall be opened and each dosimeter shall be individually subjected to the following procedures:

a) each dosimeter to be placed in position in the dosimeter holder which will then be positioned in the configuration being measured (as required).

b) the shutter in front of the radiation beam-hole to be opened for a predetermined length of time.

c) the shutter to be closed and the dosimeter to be removed from its holder, the dosimeter being replaced into its original position in its tray. (At all times the tray will be positioned and shielded such as to minimize non-intentional exposure of dosimeters).

4.4.4 Succeeding trays of dosimeters will be opened and the sequence of Section 4.4.3 will be repeated, for the successive absorber thicknesses until the transmitted radiation has been monitored with five dosimeters at each thickness for a total of fifteen exposure readings.

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4.4.5 The radiation source will be replaced into storage and the exposed dosimeters will be removed from the hot cell. Additional dosimeters required for the next exposure will be brought in.

4.4.6 The source will be returned to the shield enclosure and the first thickness of the next absorber material placed into the special holder. The complete exposure sequence of Section 4.4.3 and 4.4.4 will be repeated for a total of fifteen exposures, after which the procedure of 4.4.5 will be repeated.

4.4.7 The sequence described in paragraph 4.4.6 will be repeated for the remaining absorber materials, resulting in a total of 75 exposures for the five absorber (candidate shield) materials.

4.4.8 Sections 4.4.3 through 4.4.6 will be repeated for the remaining two sources for a total of 225 exposures.

4.4.9 Having completed the procedures of Sections 4.4.3 through 4.4.8 for each of the three thulium sources and for all absorber specimens, these procedures will then be repeated for each of the remaining geometrical configurations for a total of 450 exposures.

4.4.10 The exposed dosimeters will be inserted into appropriate reader equipment and their absorbed doses measured as detailed above (Section 3.3). The resulting data will be recorded on data sheets, a sample of which is presented in Figure 4.4.10.

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4.4.11 These dosimeters will be annealed after reading to eliminate any thermoluminescence remaining after the irradiation and readout process. For loose LiF powder, this consists of heating it to 400°C for one hour.

4.5 Confirmatory Measurements

Selected configurations of source/absorber/geometry will be duplicated and dose rate measurements repeated for comparison with original data to assure reproducibility of results.

4.6 Data Analysis Operations

The data obtained above will be statistically corrected to yield a single representative value of absorbed dose from each group of five measurements. Comparison of this value of integrated dose with the exposure time in each case will yield the dose rate at a given separation distance due to a known strength of thulium-170 behind a known thickness of absorber material. This result may then be compared with calculated values for identical conditions.

4.7 Measurement Termination

Having completed all the exposures detailed in Sections 4.4 and 4.5 the measurement operations will be terminated by removal of all sources to their storage casks. These will be securely closed and identified. Procedures for ultimate disposal of these sources will

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be determined at the conclusion of all experimentation.

0 Sample Data Sheet

(Attached)

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A			
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SANDERS NUCLEAR CORPORATION
Radioactive Operations Data
Figure 4.4.10

Project _____

Date _____

Project Engineer _____

Page _____ of _____

Radioactive Source _____

Source/Dose Point Separation _____

Control Dosimeters
Exposure _____
Absorbed Dose
(m rad)

Absorber _____
Exposure _____
Absorbed Dose
(m rad)

Absorber _____
Exposure _____
Absorbed Dose
(m rad)

Absorber _____
Exposure _____
Absorbed Dose
(m rad)

1.
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Project Engineer _____
TLD Surveyor _____

SCALE	A	SIZE
	94117	CODE IDENT NO
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LTR	DESCRIPTION	DATE	APPROVED

APPENDIX B.4

DOSE RATE MEASUREMENT DATA REDUCTION PROCEDURE

The statistical analyses procedure (Sheets 1 through 10) followed in treating the dose rate measurement data is attached hereto.

Examples of application of the various equations are presented in Appendix E. The summary results of such applications to all the measurement data obtained are tabulated in Appendix A.

Furthermore, these measurement data were also corrected as well for Thulium-170 radioactive decay since 2 August 1968, and thereby normalized to that date. The factor used in each case was derived from the basic expression describing radioactive decay as follows:

$$A(t) = A_0 e^{-\lambda t} \quad (B.4.1)$$

where: $A(t)$ = activity after time (t)
 A_0 = initial activity
 λ = decay constant

Therefore, if the decay time (t) is known as well as the activity (measured dose) after this time, and it is desired to obtain the initial activity as of a particular date, Equation B.4.1 becomes:

$$A_o = A(t) e^{\lambda t} \quad (B.4.2)$$

or,

$$A_o = A(t) \exp \left(\frac{0.693}{T_{1/2}} t \right) \quad (B.4.3)$$

In each of the measurements described the particular dose rate obtained was corrected by an appropriate exponential factor (Equation B.4.3). These results are presented in Appendix A also.

SANDERS NUCLEAR CORPORATION

NASHUA, NEW HAMPSHIRE

STATISTICAL ANALYSIS OF RADIOACTIVE DECAY MEASUREMENT DATA

SNTR-1007-BVP
ROTR-5001

11 October 1968

Prepared by: Joseph L. Cyr
Joseph L. Cyr

Modern theories of radioactive decay involve the assumption that in an assembly of nuclei of a given type, e.g., thulium-170, all the nuclei are identical, independent and each has a definite and constant probability of decaying in unit time. Since these conditions are exactly the same as the necessary and sufficient conditions for Poisson's distribution, it is customary to employ various relationships derived from the theory of this distribution in the statistical analysis of observed radioactive decay data. In addition, the fact that individual nuclear processes are random in character and hence obey Poisson's distribution means that it is possible to apply certain statistical tests to determine the "goodness of fit" between observed data and this distribution and thus to validate particular data acquisition, procedures and/or instrumentation.

Consider an experiment in which the radioactive decay rate of an isotope is being observed. A finite number of observations are made during each of which a given number of decay events (counts) are recorded over a given time interval. From this finite number it is proposed to estimate the results of an essentially infinite number of observations. In particular, it is proposed to estimate the average count rate (counts per unit time) and the distribution of observed count rates about that average from the acquired data..

If the "true" mean (average value) from an infinite number of observations is m , then from a finite number of observations the best estimate of this average value is simply the arithmetical mean given by

$$m \approx \bar{x} = \frac{1}{N} \sum_{i=1}^N x_i \quad (1)$$

where N is the number of observations.

The "expectation value" of any statistical variable is the mean value which would be obtained for the variable in a very large number of observations. Thus, the expectation value of x is m .

The distribution of observed results about the true mean value is a measure of the precision of the acquired data and is expressed quantitatively by the standard deviation σ . This parameter (often abbreviated as S.D.) is defined as the square root of the average value of the square of the individual deviations from the true mean for an infinite number of observations, i.e.,

$$\sigma^2 = \overline{(x_i - m)^2} = \frac{1}{N} \sum (x_i - m)^2 \quad (2)$$

The square of the S.D. (usually called the variance) is thus seen to be simply the second moment of the distribution of observed values about the true mean.

For the Poisson distribution, moreover, the S.D. has a definite value in terms of the true mean which is the only parameter of this distribution, the usual form of which is

$$P(x) = \frac{m^x}{x!} e^{-m} \quad (3)$$

in which $P(x)$ is the probability of observing x events when the average for a very large number of trials is m events. From this it can be shown that

$$\sigma^2 = m \quad (4)$$

Hence, for the Poisson distribution, the S.D. of individual observations is simply

$$\sigma = \sqrt{m} \approx \sqrt{\bar{x}} \quad (5)$$

In practice, however, the usual situation is the performance of a finite number of observations from which the value of the true mean m can never be determined exactly. Therefore, neither can the S.D. be exactly determined since equation (2) is only valid for an infinite or very large number of observations. Under ordinary circumstances, therefore, the best possible estimate of the variance is

$$\sigma^2 = \frac{1}{N-1} \sum_{i=1}^N (x_i - \bar{x})^2 \quad (6)$$

This expression for the variance differs from equation (2) only in its denominator $(N-1)$ and in the use of $\bar{x}$ in place of $\underline{m}$. The reason for this denominator is that the estimation of $\underline{m}$ by $\bar{x}$ from a finite number of observed values of $\underline{x}$ reduces by one the number of independent data available for further computation which use this $\bar{x}$. In the extreme case of only one observation, $\bar{x} = x$ and the S.D. (σ) is then indeterminate.

In the preceeding discussions the problems under consideration were the estimation from $\underline{N}$ observations of the results which would be realized from a very large number of observations. It is now necessary to discuss the precision of these estimates which should not be confused with the precision of the observed data, although the two are related. The main interest here is the distribution of values of $\bar{x}$ obtained from many sets of experiments, each with a finite $\underline{N}$. It is well known in the theory of errors that a series of $\underline{k}$ mean values, $\bar{x}_1, \bar{x}_2, \bar{x}_3 \dots \bar{x}_k$, each based on $\underline{N}$ observations, will tend to exhibit a normal distribution about their grand average $\bar{\bar{x}}$. This is true if $\underline{N}$ is sufficiently large, even if the parent population $\underline{x}_i$ is not normally distributed but is, for example, an asymmetric Poisson distribution. In general, the distribution of mean values tends to be much more nearly normal than the parent population. Based upon this fact, it can be shown that in a large series of $\underline{k}$ measurements of the mean value of $\bar{x}$, each

based on N measurements, of $\underline{x}$, the grand average $\bar{\bar{x}}$ approaches the true mean $\underline{m}$ and that the S.D. of $\bar{x}$ depends upon N in the following way

$$\sigma_{\bar{x}}^2 = \frac{1}{k} \sum_{j=1}^k (x_j - m)^2 = \frac{\sigma^2}{N}$$

Thus, the results of a single series of N measurements of $\underline{x}$ may be reported as $(\bar{x} \pm \sigma_{\bar{x}})$ where

$$\begin{aligned} \bar{x} &= \frac{1}{N} \sum_{i=1}^N x_i \\ \sigma_{\bar{x}} &= \frac{\sigma}{\sqrt{N}} \approx \sqrt{\frac{1}{N(N-1)} \sum_{i=1}^N (x_i - \bar{x})^2} \end{aligned} \quad (7)$$

In general, a repetition of a series of N measurements would yield a different mean value $\bar{x}$ but the probability that this new value would occur within plus or minus one S.D. of the previous mean is 0.683. The parameter $\sigma_{\bar{x}}$, the S.D. of the mean $\bar{x}$, is often called the standard error, the use of which likewise signifies a probability of 0.317 that the new mean value differs from the true mean $\underline{m}$ by more than one S.D. ($\sigma_{\bar{x}}$), if the distribution of $\bar{x}$ is normal.

Instead of quoting a standard error about a mean, it is more usual to report a quantity called the probable error, which is likewise derived from the S.D. The probable error is exactly as likely to be exceeded as not, i.e., the probability that the new value of $\bar{x}$ is inside or outside one S.D. about the mean is equally 0.500.

Again, this is under the provision that the distribution of $\bar{x}$ is normal. From tables of areas under the normal distribution curve, the probability that an error r has exactly a 0.500 chance of being exceeded in this distribution is

$$r = 0.6745\sigma \quad \text{and,} \quad r_{\bar{x}} = 0.6745\sigma_{\bar{x}} \quad (8)$$

It should be noted that the specification of a physical result as $(\bar{x} \pm r_{\bar{x}})$ is exact only for a symmetric normal distribution.

Any asymmetry in the actual distribution (as in the Poisson distribution) results in a different probable positive error than a probable negative error. However, the fact that the asymmetry of the Poisson distribution becomes small and the distribution approaches the normal distribution in the vicinity of the mean value when $m \gg 1$ validates the application of the probable error concept to this distribution under these conditions. Thus the use of equation (8) in connection with Poisson distributed values results in a conservative estimate of probable error and is a safe procedure to follow.

There are two principal statistical tests which may be applied to determine the "goodness of fit" between observed data and the distributions assumed in their analysis. The first of these is the Lexis Divergence Coefficient (Q^2) which is defined as the ratio of the average of the squares of the deviations to the arithmetical mean, or,

$$Q^2 = \frac{1}{\bar{x}} \left[\frac{1}{N} \sum_{i=1}^N (x_i - \bar{x})^2 \right] \quad (9)$$

Insertion of equation (6) into the preceeding yields

$$Q^2 = \frac{N-1}{N} \frac{\sigma^2}{\bar{x}} \quad (10)$$

which for the normal distribution can have any value since σ is a parameter of the normal distribution and can itself assume any value.

For the Poisson distribution, however, Q^2 has a definite value because σ is expressible in terms of the mean value m . Thus, if the data were in perfect agreement with Poisson's distribution, the divergence coefficient becomes

$$Q^2 = \frac{\sigma^2}{m} = 1 \quad (11)$$

It is now possible to compute Q^2 from experimental data using equation (9) and if the result is close to unity to conclude that the data seem to follow the Poisson distribution.

Beyond this, it is necessary to determine how far from unity the divergence coefficient may be before the randomness of the data becomes suspect. This determination may be made by application of the second statistical test which is called the Chi-square Test (χ^2).

This test determines the probability P that a repetition of a series of observations would show greater deviations from the expected values of the frequency distribution which has been assumed to govern the data. If it is assumed that for any distribution, in a series of N observations of a process over equal time intervals, the expected value in each interval is constant and equal to the mean value $\bar{x}$ for all intervals, then

$$\chi^2 = \sum_{i=1}^N \frac{1}{\bar{x}} (x_i - \bar{x})^2 = N Q^2 \quad (12)$$

The next step in this test is to determine the number of degrees of freedom F which is the number of independent expected values which the data may assume in N observations.

Then, using these values of χ^2 and F, reference to tabulations of Chi-square (5) will determine P, which is the probability that χ^2 should exceed its observed value. The usual interpretation for this value is that if P lies between 0.1 and 0.9, then it may be concluded that the observed data seem to follow the assumed distribution. If,

however, P is less than 0.02 or more than 0.98, the data do not follow the distribution, and the data and/or the assumed distribution should be re-examined. Stipulating the randomness of radioactive decay, it is usually not the assumed (Poisson) distribution which is inadequate but rather the data gathering procedures or the instrumentation being used which require re-evaluation.

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APPENDIX C

DOSE RATE PREDICTION

METHODS AND STUDIES

APPENDIX C.1
CALCULATION PROCEDURE

The objectives of the calculation format described below were to provide theoretical values of radiation dose rates from Thulium-170 sources under various combinations of exposure conditions which were subsequently to be experimentally measured as well. These exposure conditions, based upon axial source irradiation, are:

- Each of three source strengths; 5.1, 10.0 and 20.7 thermal watts
- Each of four representative photon energy groups; 0.1, 0.2, 0.475 and 0.775 MeV
- Self-absorption of emitted radiations within the sources
- Radiation attenuation within the source encapsulation material (Hastelloy alloy C)
- Radiation attenuation by each of three thicknesses (0.250, 0.500 and 1.0 inch) of each of five absorber materials; aluminum, stainless steel, lead, tungsten and depleted uranium

- Radiation attenuation by inverse square law for each of two source-to-dose point separation distances; 19 and 100 cm
- Radiation attenuation by the intervening air

In order to evaluate the residual flux expressions (C.1.2a, 2b) for the various combinations of the preceeding irradiation conditions, and to calculate the associated absorbed radiation dosages with equation C.1.3, a computer program was generated and perfected. The results of the computer calculations modified by the appropriate infinite build-up factors are presented graphically here in Figures C.1-1 through C.1-5. In these figures, the radiation dose rates are presented as a function of absorber thickness for each of the five absorber materials.

The three Thulium-170 source strengths and two source-to-dose point separation distances were used as the parameters of these analyses. The particular values of source strengths given above were chosen because they closely approximate the actual values determined at ORNL as of 2 August 1968. The source-to-dose point separation distances used above were chosen because they were thought to represent meaningful and usual irradiation configurations.

69-H65987-006

Figure C.1-1 Radiation Dose Rate Computed Data for Thulium-170 Using Aluminum Absorbers

69-H65987-007

Figure C.1-2 Radiation Dose Rate Computed Data for Thulium-170 Using Stainless Steel Absorbers

Figure C.1-3 Radiation Dose Rate Computed Data for Thulium-170 Using Lead Absorbers

69-H65987-009

Figure C.1-4 Radiation Dose Rate Computed Data for Thulium-170 Using Tungsten Absorbers

69-H65987-010

Figure C.1-5 Radiation Dose Rate Computed Data for Thulium-170 Using Uranium Absorbers

These computer analyses did not take into account radiation build-up within absorber samples. Instead, build-up factors were obtained from the report of Goldstein and Wilkins⁽¹⁾ for various photon energies in infinite absorber media, and the computed dose rate results modified accordingly. Despite the fact that the build-up factors thus obtained are rigorously applicable only to point isotropic sources in infinite absorber mediums, corresponding values for distributed volume sources in finite media do not generally exist, and the usual practice is to employ the infinite media values. Consequently, it was anticipated that the results of the dose rate measurements to be performed might provide useful information regarding the actual build-up factors for the irradiation conditions.

Two different approaches were investigated to obtain analytic expressions for the radiation exposure conditions described. The first was to develop the exact differential equation that describes the uncollided radiation flux incident upon a dose point from a volume radiation source through selected absorber thicknesses.

The schematic geometry and corresponding expression for the radiation flux resulting from this approach are given below:

(1) Goldstein, H. and Wilkins, J.E. Jr., Calculations of the Penetration of Gamma Rays, NDA 15C-41, 1954.

$$\phi(E_o) = \frac{Sv}{2} \int_0^h dx \int_0^R \frac{rdr}{r^2 + (x+a)^2} \exp\left(-\mu_1 t \sqrt{1 + \frac{r^2}{(x+a)^2}}\right) \dots$$

$$\exp\left[-\mu_2 \left(\sqrt{r^2 + (x+a)^2} - a \sqrt{1 + \frac{r^2}{(x+a)^2}}\right)\right] \dots$$

$$\exp\left[-\mu_3 (c+e) \sqrt{1 + \frac{r^2}{(x+a)^2}}\right] \quad (C.1.1)$$

where:

$\phi(E_o)$ = radiation flux of energy E_o , photons/cm² · sec

μ_1, μ_2, μ_3 = linear attenuation coefficient of the shield material, source and air, respectively, for gamma photons of energy E_o , cm⁻¹

Sv = volumetric source strength, photons/cm³ · sec

This expression was not developed further owing to the fact that its evaluation would have required intensive numerical analysis at each point.

The second approach was to make use of approximate expressions for the uncollided radiation flux transmitted axially by a right circular cylindrical source through a given thickness of absorber which are presented in Rockwell (2). The schematic geometry of this approach and corresponding flux equations are presented below:

a. upper limit radiation flux:

$$\phi = \frac{Sv}{2\mu_2} \left\{ E_2(b_1) - E_2(b_3) + \frac{E_2(b_3 \sec \theta_1)}{\sec \theta_1} - \frac{E_2(b_1 \sec \theta_1)}{\sec \theta_1} \right\} \quad (C.1.2a)$$

b. lower limit radiation flux:

$$\phi = \frac{Sv}{2\mu_2} \left\{ E_2(b_1) - E_2(b_3) + \frac{E_2(b_3 \sec \theta_2)}{\sec \theta_2} - \frac{E_2(b_1 \sec \theta_2)}{\sec \theta_2} \right\} \quad (C.1.2b)$$

(2) Rockwell, T., Reactor Shielding Design Manual, D. Van Nostrand Co., New York, N. Y., 1956.

where:

$E_2(b)$ = exponential integral

$$= b \int_b^{\infty} \frac{e^{-t}}{t^2} dt$$

$$b_1 = \sum_{i=1}^3 \mu_i t_i$$

$$b_2 = b_1 + \mu_2 Z$$

$$b_3 = b_1 + \mu_2 h$$

Z = self-attenuation distance (cm.)

Subsequent analysis (see Section C.2) regarding the solid angle subtended by the source at the dose point utilizing the actual experimentally measured dimensions revealed:

$$\left. \begin{array}{l} \sec \theta_1 = 1.00108 \\ \sec \theta_2 = 1.00098 \end{array} \right\} \therefore \sec \theta_1 \approx \sec \theta_2$$

Therefore, equations C.1.2a and 2b become essentially indistinguishable, and either can be used with very little loss of accuracy to yield calculated values of the uncollided radiation flux at the dose point.

Then, the absorbed dose rate in human tissue (mrad/hr) can be determined from these fluxes through the following expression:

$$D(E_o) = 5.767 (10^{-2}) \left[\frac{\mu_a}{\rho} \right] E_o B(E_o, \mu t) \phi(E_o) \quad (C.1.3)$$

where:

$D(E_o)$ = dose rate from exposure to uncollided gamma photons of energy E_o , including buildup contribution, millirads/hr.

μ_a / ρ = energy absorption mass attenuation coefficient for human tissue, cm^2/gm .

E_o = photon energy, Mev

$B(E_o, \mu t)$ = buildup factor for gamma radiation of initial energy E_o and a shield with a thickness of μt relaxation lengths.

In addition, investigations were conducted into the alternate modes and possible effects of radiation scatter in the experimental geometry. Consideration of Figure C.1-6 reveals that in operation, the Thulium-170 sources were well shielded in their experimental cask, and the only exit path for the emitted radiation was the beam tunnel which was designed to the same diameter as the source wafers themselves. Thus, the solid angle beam of radiation emerging from this tunnel was well defined, and the absorber specimens were designed to completely intercept this beam in their experimental positions. Therefore, since the scatter of these radiations in air is insignificant, the possible sources of such scattering became:

- Single scatter by the absorber specimen to dose point (build-up)
- Double scatter by the absorber and a part of the experimental fixture
- Double scatter by the absorber and a hot cell structural member
- Single backscatter off the rear wall to the dose point.

Figure C.1-6 Tm-170 Dosimetry Fixture

Single scatter by the absorber specimen to the dose point constituted the build-up factor discussed previously . Continued examination of Figure C.1-6 and the diagrammatic sketch below reveals that only the radiations scattered by the absorbers approximately straight ahead and in the downward direction had any probability of encountering

a part of the experimental fixture (notably the aluminum base channel) and thus had any real probability of second scatter to the dose point. Therefore, to prevent this from occurring, a lead scatter shield was designed and installed to intercept and attenuate all such scattered radiations (identified in Figure C.1-6 as the "radiation scatter shield").

The size of the solid angle of radiation beyond the absorber was such that no radiation could be directly incident upon any of the side walls, the ceiling or the hot cell floor. Thus, only double scatter by the absorber and one of these structural portions could possibly result in scattered radiation incident at the dose point as illustrated below:

To calculate the intensity of radiation doubly scattered out of the original flux to the dose point (I/I_0), reference is made to the scattering equation (discussed more fully below).

$$\frac{I}{I_0} = \left\{ 7.94 (10^{-26}) \eta_1 F_1(E_0, \theta_1) \frac{1}{4\pi r_0^2} \frac{1}{r_1^2} \Delta V_1 \right\} \\ \times \left\{ 7.94 (10^{-26}) \eta_2 F_2(E_1, \theta_2) \frac{1}{r_2^2} \Delta V_2 \right\}$$

where:

I = intensity of scattered radiation, $\text{Mev}/\text{cm}^2\text{-sec}$

η = number of electrons per cm^3 of scattering material

$F(E, \theta)$ = scatter probability for particle energy E (Mev) and scattering angle θ

r = length of scattering path of radiation, cm

ΔV = volume of scattering volume, cm^3

It is assumed that all radiation scatter occurs at the surfaces of the structural members. The following are the pertinent dimensions of the assumed scattering system:

$$\eta_1 = \eta_2 = 2.2 (10^{24}) \text{ electrons/cm}^3$$

$$r_o \approx 15 \text{ cm}$$

$$r_1 \approx 90 \text{ cm}$$

$$r_2 \approx 120 \text{ cm}$$

$$\Delta V_1 \approx 24.6 \text{ cm}^3$$

$$\Delta V_2 \approx 1.7 (10^4) \text{ cm}^3$$

$$E_o \approx 0.5 \text{ Mev}$$

Upon insertion of these into the appropriate supplementary equations likewise discussed below, the intensity of radiation doubly scattered to the dose point becomes:

$$\frac{I}{I_o} \approx 1.65 (10^{-10})$$

which is negligible except for very high fluxes incident over lengthy irradiation intervals. It should be noted that the highest strength sources with the thinnest absorbers also had the briefest

irradiation intervals. Therefore, this scattering mechanism was not thought to constitute a significant source of measurement error. The final case was that of backscatter (see illustration below) off the rear wall to the dose point.

The calculation to determine the intensity of radiation scattered in this way was performed with the aid of the following diagram:

The intensity of the radiation scattered within the unit volume of scattering medium through an angle (θ) is:

$$dI = I_o \times (7.94 \times 10^{-26}) \eta F_1(E_o, \theta) \left(\frac{e^{-\mu r_2}}{4\pi r_1^2} \right) \left(\frac{e^{-\mu r_3}}{r_4^2} \right) dV$$

$$\therefore \frac{I}{I_o} = \frac{7.94 \times 10^{-26} \eta}{4\pi} \int_{vol} F_1(E_o, \theta) \frac{e^{-\mu(r_2+r_3)}}{(r_1^2)(r_4^2)} dV \frac{\text{Mev}}{\text{cm}^2\text{-sec}}$$

where the quantities are as previously defined and where:.

$$F_1(E_o, \theta) = \frac{p^3}{2} \left(\frac{1}{p} + p - \sin^2 \theta \right)$$

$$p = \frac{E}{E_o} = \frac{1}{1 + \frac{E_o}{m_0 c^2} (1 - \cos \theta)}$$

From the geometry of the experiment, as illustrated below:

*Note

$$\sin \theta = \sin(180^\circ - \phi) = \sin \phi$$

$$\cos \theta = \cos(180^\circ - \phi) = -\cos \phi$$

The approximate scattering intensity was calculated as follows

(E_0 assumed as 0.50 MeV):

$$P = \frac{1}{1 + (1 + \cos 4.5)} \approx \frac{1}{1 + 2} \approx \frac{1}{3} = 0.33$$

$$F_1 = \frac{1}{2} (p^2 + p^4 - p^3 \sin^2 \theta) = 6 \times 10^{-2}$$

Since the back wall has a steel surface, all scattering was assumed to take place at this surface:

$$\therefore e^{-\mu(r_2 + r_3)} = 1$$

$$\eta = 2.2 \times 10^{24} \text{ e}^-/\text{cm}^3 \text{ (for steel)}$$

$$r_1 = 3.4 \times 10^2 \text{ cm}$$

$$r_4 = 2.5 \times 10^2 \text{ cm}$$

For the maximum scattering case, a circular disc volume with radius = 75.1 cm and 1 cm thick was assumed.

$$\therefore \Delta V = 1.7 \times 10^4 \text{ cm}^3$$

and,

$$\frac{I}{I_0} = \frac{7.94 \times 10^{-26}}{4\pi} \eta F_1(E_0, \theta) \frac{1}{(r_1^2)(r_4^2)} \Delta V$$

$$\frac{I}{I_0} \approx \boxed{1 \times 10^{-9}}$$

Note: This is a conservative case; therefore, the scattering from the wall of the hot cell was neglected.

In summary, therefore, it was concluded that with the exception of build-up, no significant contributions to the radiation dose rates would arise due to scattering.

APPENDIX C.2

AXIAL RADIATION FLUX CONSIDERATIONS

From the geometry, the most significant solid angle is subtended at the smallest separation distance of source and dose point:

$$\tan \theta_1 = \frac{0.375}{7.5} = 0.0500$$

$$\tan \theta_2 = \frac{0.375}{7.5 + 0.4} = 0.0476$$

$$\therefore \theta_1 = 2^\circ 52'$$

$$\theta_2 = 2^\circ 43'$$

$$\sec \theta_1 = \frac{1}{0.99875} = 1.00108$$

$$\sec \theta_2 = \frac{1}{0.99888} = 1.00098$$

$$\therefore \sec \theta_1 \approx \sec \theta_2$$

therefore:

Upper limit $\approx$ lower limit

and, either expression may be used to calculate the gamma radiation flux at the dose point.

APPENDIX D

CALIBRATION, TRACEABILITY AND
STANDARDS

APPENDIX D.1

CALIBRATION OF RADIATION SOURCES

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June 4, 1969

Radiation sources were calibrated by measurement of their thermal output using a calorimeter. The calorimeter and the procedures are described in references 1 and 2. The calorimeter was calibrated in the cell just prior to use. The calibration was made using a precision heater and a calibrated wattmeter and recording the thermocouple potentiometer reading as a function of power input to the heater. The most recent calibration report for the wattmeter is attached. It was unchanged from the calibration in effect at the time of use.

The same potentiometer and standard cell were then used to measure the heat output of the radiation sources. Since the calorimeter used was a less refined model than the type described in the references, the accuracy is estimated at 2% at the 95% confidence level.

-
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Instrument Department

Standards Laboratory

CALIBRATION REPORT

Traceable to the National Bureau
of Standards, Test # 211.02/184345
1.1/G-28370

Instrument AD-DC Wattmeter

Manufacturer Weston

Type Electrodymeter

Model 310

Serial No. 13949

X No. 45337

The indications are within $\frac{1}{4}$ of 1 percent of full scale when used with corrections and within $\frac{1}{2}$ percent without the corrections.

Date calibrated May, 27, 1969

Calibrated by M. H. Cooper, Jr.
M. H. Cooper, Jr.

Calorimetry for Measuring Radioactivity

By J. C. Posey

The use of calorimetry at the ORNL Isotopes Development Center was started as a part of the program to produce large radioactive heat sources. A calorimetric method was desired since it would give direct measurements of the rates of heat production and thus would eliminate errors associated with uncertain knowledge of the isotope decay schemes. It is the only feasible method of measuring directly the radioactivity of large samples that produce only alpha or beta radiation. The dependability of calorimetric methods and the fact that they have generally greater accuracy than competing counting procedures have led to their increasing use.

Calorimetry as applied to radioactive sources differs in one basic respect from conventional calorimetry: the quantity measured is power, or rate of heat production, and is measured in such units as watts or calories per second; in conventional calorimetry a quantity of heat is measured in units such as watt-hours or calories. Because of this, most commercial calorimeters cannot be used for the assay of radioactive sources.

A number of calorimeters for assaying the power of radioactive sources are described in Refs. 14 and 15; the first was described by Rutherford and Barnes¹⁶ in 1904. These calorimeters vary greatly in detail. However, the operation is in all cases based on one of the following measurements:

1. Rate of temperature increase of a mass of material of known heat capacity
2. Temperature gradient produced by the conduction of heat through a path of known properties
3. Temperature increase of a constant stream of cooling fluid
4. Rate of vaporization of a liquid

Most of the calorimetric work in the ORNL Isotopes Development Center has been done with simple calorimeters developed in its own laboratories; these are of type No. 2 mentioned above. They occupy a minimum of hot-cell space and are easy to operate, resistant to damage, and low in cost.

The basic parts of these calorimeters are shown in Fig. VI-4. The upper part is a metal cup mounted on a metal shaft, at the base of which is a heat sink cooled by a stream of water at constant temperature. The heat produced by radioactive decay of the source is absorbed by the cup and conducted down the shaft to the heat sink. At steady state the temperature difference between the top and the base of the shaft is nearly proportional to the rate at which heat is produced by the radioactive source. The temperature difference is measured by a thermopile with junctions attached to the surface of the shaft. Copper wires are attached to the thermopile at the hot end of the shaft. Extraneous heat loss is minimized by enclosing the cup and the shaft in an inverted Dewar flask.

The construction materials were selected to maintain nearly exact proportionality between the thermopile output and the thermal power of the source. Iron-constantan couples were chosen for the thermopile because their sensitivity (millivolts per degrees centigrade) varies only slightly within the range of temperatures measured. Copper was selected for the shaft because its thermal conductivity is relatively constant. The departures from constancy of thermopile sensitivity and thermal conductivity which do occur are opposite in effect to the small loss of heat through the Dewar flask.

The calorimeters are calibrated before use by means of an electric heater placed in the cup in substitution for the radioactive source. The voltage drop across the heater, the heater current, and the thermopile output are measured with precision electrical equipment of commercial manufacture. These electrical measurements during calibration and those during use are the only instances during which a high degree of precision must be attained. Comparatively loose tolerances are permissible in fabrication.

The design of a calorimeter of this type involves a suitable compromise between accuracy and speed of operation. The approach to steady state is described approximately by

$$\theta = \frac{C}{U} \ln \frac{\Delta T_{ss}}{\Delta T_{ss} - \Delta T}$$

where θ = elapsed time

C = heat capacity of the sample cup and upper part of the Dewar flask (the primary assumption is that these are at the same temperature)

Fig. VI-4 Basic parts of calorimeter used for measuring rates of heat production by radioisotope sources.

ΔT = difference in temperature between ends of shaft

ΔT_{ss} = difference in temperature at steady state

U = thermal conductivity of shaft

The time required for ΔT to reach a given fraction of ΔT_{ss} is thus inversely proportional to the thermal conductivity of the shaft. However, ΔT_{ss} is inversely proportional to U . Consequently, since the error caused by fluctuations in cooling-water temperature and by limits of precision of the measurement of the thermopile output are approximately inversely proportional to ΔT_{ss} , the increase in speed of operation is accompanied by a loss of precision. Furthermore, if U is increased until the re-

sistances to heat flow from the sample to the cup and from the cup to the Dewar flask are significant compared to the shaft resistance, the time required will not decline as rapidly as predicted by the equation.

Four of the calorimeters that have been constructed are shown in Fig. VI-5, minus the Dewar flasks and thermopiles. The first (upper left) was designed for measuring the thermal powers of Sr^{90} samples of 5 to 50 watts. The shaft ΔT is $\sim 1.5^\circ\text{C}$ per watt.

The reproducibility of results obtained with this calorimeter is illustrated by the calibration data given in Table VI-1. No individual value of the calibration constant differs from the average by more than 0.13%. If deviations from the average are treated as random errors,

the 95% confidence limit of a single determination is 0.2%.

The performance of the calorimeter is only moderately sensitive to the rate of flow or to the temperature of the cooling water provided both are constant during any given determination. Variation of the flow rate from 1.1 to 8 liters/min produced a change of only 0.4% in the calibration constant. A change in the cooling-water temperature from 10 to 27°C lowered the constant by 1.4%. A lowering of 1.56% was calculated from the known change of the iron-constantan thermopile and the copper shaft with temperature. The agreement is within experimental error.

The second calorimeter (upper right) was designed for use with Cs^{137} samples of 5 to 50 watts. The cup is constructed of metallic uranium and has walls $1\frac{1}{16}$ in. thick. The gamma-ray absorption is nearly complete; more than 99.8% of the total energy is absorbed.

Because of the heat capacity of the thick-walled cup, a much longer time (7 to 8 hr) is required for this calorimeter to reach steady state than is required for calorimeters designed for use with only alpha- or beta-emitting materials. The results are no less accurate, however, being reproducible within 0.2%.

The third calorimeter (lower left) is designed to allow more rapid measurements of gamma-

emitting sources. The thinner walls absorb a known fraction of the radiation (94.85% of the total energy from Cs^{137}). Thus the thermal powers of gamma-emitting samples can be determined in approximately the same time (1.5 to 2 hr) as that required with calorimeters de-

Table VI-1 CALIBRATION DATA OBTAINED WITH CALORIMETER SHOWN IN UPPER LEFT-HAND CORNER OF FIG. VI-5

Thermal power, watts	Thermopile output, mv	Calibration constant, watts/mv
24.18	6.594	3.667
24.32	6.626	3.671
18.38	5.005	3.673
18.08	4.929	3.669
12.27	3.339	3.674
12.32	3.354	3.675
6.17	1.682	3.676
		3.672 (av.)

signed for use with alpha- and beta-emitting materials. Some uncertainty is added because of differing degrees of self-absorption of gamma radiation by the samples themselves.

The last calorimeter was designed for use with small samples of alpha- or beta-emitting materials held in a standard capsule used for irradiation of samples in the hydraulic tubes of the Oak Ridge Research Reactor. Measurements are accurate to 0.005 watt in the range 1 to 10 watts. A similar calorimeter with thick uranium walls has been constructed for gamma-ray absorption. It has been used primarily for the assay of small Ir^{192} samples.

A calorimeter for assaying alpha-emitting samples of 30 to 300 watts is being made. It will extend the range of sample power that can be measured. Since the shaft has a much larger thermal conductivity than that of any of the calorimeters previously constructed, it will be a test of the degree to which the time required to reach steady state can be reduced by this means.

Flow Calorimeters

Assay of sources in the form of long, thin rods is sometimes necessary. The calorimeters described above are not suitable for such assays, and two calorimeters have been designed, one for alpha- or beta-emitting sources and one for gamma-emitting sources, which

Fig. VI-5 Thermal-conductivity calorimeters.

Fig. VI-6 Calorimetric decay study of irradiated Am²⁴¹ (Cm²⁴²).

may be used with long sources. The increase in temperature of a constant and accurately measured stream of water is used as a measure of the rate of heat production by the source in these new devices.

Applications of Calorimetry

Calorimetric methods are used for the final check of the magnitude of the heat output of the radioactive heat sources produced by the Isotopes Development Center and are for control analysis during manufacture of the materials of these sources.¹⁷ Calorimetry also provides a convenient measurement of the degree of conversion of samples exposed in atomic reactors. The samples are removed from the reactor, and the thermal power is measured without removing the sample from its container. This method has been used in the curium development program.

One notable project carried out in support of the above work was a decay study of an irradiated Am²⁴¹ sample. Although the thermal power of this sample was largely produced by the Cm²⁴² product, significant fractions were produced by fission products and by long-lived transuranic impurities. Ordinarily the quantities of these other radioactive materials present in

samples are calculated, so it was desired to check the accuracy of the methods of calculation.

A decay study was carried out over a period of 65 days (Fig. VI-6). The total or measured powers were corrected by the normal method for the contribution of fission-product decay. Since the decline of the corrected power with time could be attributed within experimental error entirely to the decay of Cm²⁴² to Pu²³⁸, these values could be correlated with time by a least-mean-squares equation, using literature values for the half-lives and energies of decay of Cm²⁴² and Pu²³⁸. Agreement of this derived equation with the data was excellent; the standard deviation of the points from the equation was only 0.0023 watt, or 0.06% of the average power of the sample.

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HOT-CELL CALORIMETRY FOR ROUTINE DETERMINATION OF THERMAL POWER GENERATED BY KILOCURIE SOURCES

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Calorimeters have been developed to measure the thermal power of multi-kilocurie radioactive sources of Sr^{90} and Cs^{137} . The calorimeters are resistant to accidental damage, occupy little hot-cell space, and are simple to operate. Measurements are accurate to $\pm 0.20\%$ when measuring ~ 20 watts of thermal power.

INTRODUCTION

The heat produced by the decay of large radioactive sources is being used increasingly in special applications. One of the most important uses to date is in thermoelectric generators in the SNAP (Systems of Nuclear Auxiliary Power) program to furnish power for electronic equipment in devices such as automatic weather stations in remote areas. In order to match the source to the thermoelectric conversion unit, it is necessary to make an accurate determination of the thermal power generated by the source. Direct calorimetric measurement is preferred for this purpose, since (1) it involves no assumption or calculation of the average beta energy; (2) it does not depend upon the certainty with which the radioactive decay scheme is known; (3) it is independent of radiochemical assay methods and ordinarily more accurate.

Requirements of the Calorimeter. The calorimeters were designed to meet the following criteria:

1. A minimum of hot cell space.
2. A small number of simple manipulations in the cell.
3. Equipment which is resistant to damage.
4. Equipment which requires a minimum amount of instruction prior to operation.
5. Low initial investment and operating costs.

Types of Calorimeters. The thermal power of a radioactive sample was successfully measured by calorimetric means as early as 1904.⁽¹⁾ Since then, many successful calorimeters have been designed, based on measurements such as:

1. The rate of temperature increase of a mass of material of known heat capacity.
2. The temperature gradient produced by the steady-state conduction of the heat from the sample through a path having known properties.

Operated for the U. S. Atomic Energy Commission by the Union Carbide Corporation.

Fig. 1. Basic design of calorimeter

3. The temperature increase of a constant stream of cooling fluid.
4. The rate of vaporization of a liquid.

An evaluation of the characteristics of the types of calorimeters suitable for the intended application suggested that the thermal conductivity type, operated at steady-state conditions, is preferable. Only the measurements of the temperatures at the terminals of the path through which the heat is conducted are required; and, if the thermal conductivity of the heat-path material is nearly constant over the range of temperatures involved, only the measurement of the temperature difference between the terminals of the heat conductor (ΔT) is needed. A single measurement, of course, reduces the chance of human or instrument error.

DESCRIPTION OF EQUIPMENT

Basic Design. The basic design of the calorimeters is shown in Fig. 1. The upper portion consists of a metal cup mounted on a metal shaft. At the base of the shaft is a heat sink cooled by a stream of water at constant temperature. The heat produced by the radioactive decay of the source is absorbed by the cup and conducted down the shaft to the heat sink. At steady state the temperature difference between the top and the base of the shaft, ΔT , is essentially proportional to the rate at which heat is produced by the radioactive source. Extraneous heat loss is minimized by enclosing the cup and shaft in an inverted Dewar flask.

The temperature difference between the ends of the shaft is measured by a thermopile with the junctions attached to the upper and lower extremities of the shaft. The junctions of the thermopile are evenly distributed about the circumference of the shaft at either end. A single layer of plastic-impregnated glass tape provides electrical insulation from the shaft; a second layer holds the junctions in place. The copper lead wires are connected to the thermopile at the hot-junction end of the shaft. One of the iron-constantan junctions is replaced by an iron-copper and a constantan-copper junction. The lead wires pass under the edge of the Dewar flask through a channel machined in the base.

The materials of construction were selected to maintain as nearly as possible exact proportionality between the thermopile output and the thermal power of the source. Iron-constantan couples were chosen for the thermopile because the sensitivity (mv/°C) varies only slightly within the range of temperatures measured. Copper was selected for the shaft because the thermal conductivity is relatively constant.

Design Parameters. The time required for this type of calorimeter to reach steady-state conditions is approximately directly proportional to the combined heat capacities of the cup for holding the sample, plus the sample. The mass of the cup, therefore, should be as small as feasible. A calorimeter designed for use with gamma-ray emitters (e.g., Cs^{137}) requires thick walls to absorb the gamma-ray energy. Since only beta rays are emitted by Sr^{90} and its short-lived daughter, Y^{90} , the cup walls need to be only heavy enough to absorb the maximum beta energy and to insure adequate mechanical strength.

Errors in measurements of ΔT can arise as a result of variations in the heat-sink temperature and from limitations of the instrumentation. The percent error introduced in ΔT from these two sources decreases as ΔT increases. However, the time required to reach steady state increases as the value of ΔT per unit of thermal power increases. Since an increase in this value can be achieved only by decreasing the thermal conductivity of the principal heat path, extraneous heat losses from the unit become important.

The calorimeters were designed for ΔT values of $\sim 1.5^\circ\text{C}/\text{watt}$ and $\sim 0.5^\circ\text{C}/\text{watt}$ for Sr^{90} and Cs^{137} , respectively, in order to allow accurate measurement of ΔT with readily available potentiometers. The shaft lengths and cross-sectional areas were chosen to give the desired values of ΔT and to allow the calorimeters to be insulated by Dewar flasks of standard sizes.

Strontium-90 Calorimeters. Two different calorimeters were fabricated for the measurement of the thermal power produced by Sr^{90} decay. The first differed from that shown in Fig. 1 in that the base had a less efficient heat transfer coefficient. The sample cup

was copper, with walls $1/4$ " thick, and was large enough to hold cylindrical samples measuring ~ 2 " in diameter by 1" long; it weighed 1.6 lbs. The shaft was 1" in diameter and $5-3/8$ " long. The temperature gradient of the shaft was measured by a four-junction thermopile. The base was cooled by flowing tap water without attempting to control the temperature to a precise value.

The design of the second Sr^{90} calorimeter was changed to incorporate the improved heat sink design shown in the Cs^{137} calorimeter in Fig. 1.

Cesium-137 Calorimeter. Approximately 60% of the energy derived from Cs^{137} decay is emitted in the form of gamma rays, in contrast to the pure beta radiation from the Sr^{90} sources. It is therefore necessary that the sample cup have sufficient mass to absorb the gamma rays and convert them to thermal energy. Uranium was chosen for the cup material since it has the lowest heat capacity per unit mass and the highest mass-absorption coefficient for the Cs^{137} gamma rays of any readily available metal. The cup had a wall $1-1/16$ " thick and a cavity of $2-5/8$ " in diameter by $1-3/4$ " high. Calculations using gamma-absorption data (2,3) showed that 99.8% of the total energy (including beta) would be absorbed and converted to heat in this cup. The total weight of the uranium cup was about 30 lbs. The ΔT was measured by a 12-junction thermopile to increase the millivolt response. The cooling-water temperature was controlled by a commercial hot water heater and a Leeds and Northrup proportional temperature controller.

Calibration. The calorimeters were calibrated with a 20-ohm resistance heater substituted for the radioactive sample. The thermopile output and the heater voltage and amperage were measured with a Rubicon double-range potentiometer. The measured thermal power of the heater was accurate within $\pm 0.13\%$ at the 20-watt level.

Production Instrumentation. The thermopile output of the calorimeters was measured by a 2-7 millivolt recording potentiometer. The visible record aided the operator in determining that a steady state existed. Also, the water temperature was recorded to enable the operator to recognize spurious results caused by a drift in the cooling-water temperature.

PERFORMANCE DATA AND DISCUSSION

First Sr^{90} Calorimeter. As a result of less than ideal heat transfer from the base of the shaft to the cooling water, the thermopile output from the first calorimeter was not exactly proportional to the thermal-power input. This effect also made the calibration sensitive to variations in cooling-water flow rate. Thus the use of a single calibration constant for the calorimeter was not possible and it was necessary to determine the constant as a function of the thermopile output, and to control the flow of cooling water. At a fixed cooling-water flow rate, and with the use of a calibration chart, the measurements of several samples varying from 7.74 to 28.50 watts were reproducible and had a standard error of only 0.3%, which included errors in weighing as well as those in the calorimetric measurement.

Second Sr^{90} Calorimeter. Calibration data for the second Sr^{90} calorimeter are shown in Table I. The cooling-water flow rate was maintained at 3.4 liters/min during the determination of these data. Within the range of conditions covered, the improved heat transfer between the shaft and the base almost completely eliminated the tendency of the calibration constant to change with the thermal power. No individual value differs from the average value by more than 0.13%. If the deviations from the average of the individual determinations are treated as random errors, the 95%-confidence limit of a single determination is 0.2%. Further experiments showed that the calibration constant changed only 0.4% when the water flow was varied between 1.1 and 8 liters/min. Consequently, only approximate control of the water flow was necessary.

The use of cooling water at 27°C instead of 10°C lowered the calibration constant by 1.4%. This compares favorably with 1.56% as calculated from the known change in properties of the iron-constantan thermopile with temperature, and the change in the thermal conductivity of the copper shaft.

TABLE I

Thermal Power (watts)	Thermopile Output (mv)	Calibration Constant (watts/mv)
24.18	6.594	3.667
24.32	6.626	3.671
18.38	5.005	3.673
18.08	4.929	3.669
12.27	3.339	3.674
12.32	3.354	3.675
6.17	1.682	3.676
Average		3.672

Cesium-137 Calorimeter. Because of the much larger external dimensions of the sample cup, a larger Dewar flask was needed to cover the Cs¹³⁷ calorimeter. This increased the area through which heat could escape from the system. Inefficient heat transfer between components of the cup in the Cs¹³⁷ calorimeter caused the temperature of the top of the cup to rise considerably above the ambient temperature of the cell. In this case the escape of heat from the system was great enough that it could no longer be neglected. Consequently it was necessary to calibrate the calorimeter at conditions which were near those expected during the actual sample measurements.

Table II lists data obtained by the calorimetric measurement of the thermal power of Cs¹³⁷ sources prepared from homogeneous batches of powder. The overall standard deviation of the specific thermal power is 0.0002 watts/g. This deviation includes errors in the sample preparation as well as those in the calorimetric measurements. Duplicate measurements of the same sample were carried out in the case of two Cs¹³⁷ sources; the values differed by 0.18% in one case and 0.08% in the other for ~ 10-watt samples.

The time required to complete the thermal assay of a source is a function of the total heat capacity of the calorimeter and sample. In the case of the Sr⁹⁰ calorimeters, steady state was reached in approximately 1-1/2 hrs. In practice at least three hours was allowed to insure equilibrium conditions. However a total of only about 30 minutes of actual work is needed to load the sample, start the apparatus, and record the data in any one determination of the thermal power of a sample. The Cs¹³⁷ calorimeter required ~ 7 hrs to reach steady state due to the greater heat capacity of its heavy cup, but the time to obtain the data was essentially the same. However, if a series of similar samples are assayed consecutively, both the total time and the man-hours of labor can be appreciably reduced -- especially in the case of the Cs¹³⁷ calorimeter -- since samples can be changed quickly enough to prevent much drop in temperature of the heavy cup.

TABLE II

Source of Cs ¹³⁷ Powder	Thermal Power of Sample (watts)	Specific Thermal Power (watts/g)
Batch 1	21.16	0.0670
Batch 1	21.19	0.0670
Batch 2	25.13	0.0815
Batch 2	23.00	0.0813
Batch 2	23.31	0.0818

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APPENDIX D.2
THERMOLUMINESCENT DOSIMETRY (TLD) SYSTEM

D.2.1 Thermoluminescent Dosimetry Readout System Description

The thermoluminescence dosimetry system used in obtaining the radiation dose rates in this experiment consisted of the following:

- Con-Rad Model 5100B Readout Instrument
- Con-Rad Model PDAV Vibratory Phosphor Dispenser
- Thermoluminescent Dosimetry Powder Type TLD-700 (LiF-7)

In the operation of the readout instrument, the phosphor sample or dosimeter is heated and the amount of light emitted within a fixed time period is integrated and this integral is presented digitally. The instrument can be adjusted in such a manner that the dose can be read directly in dose units over a broad dose range.

The concept of the instrument is indicated in block diagram form by Figure D.2.1-1.

The system employs reusable and exchangeable heating elements. When measuring loose powder dosimeters, high-resistivity heating elements are used onto which the powder is poured. These heating elements are inserted by a slide into the instrument where the edges become gripped by copper-beryllium alloy contacts during heating. The Constant Current Heater Control module provides a constant current through the heating element and ensures a reproducible heating process.

Figure D.2.1-1 Block Diagram of Readout Instrument

A photomultiplier tube selected for a low dark current, a large range of measurable light fluxes, and a high stability is the first stage in the measuring system. A Teflon tube socket effectively prevents leakage currents. The High Voltage Power Supply provides the photomultiplier with voltage. It is continuously adjustable, and the setting can be controlled to ± 0.3 volts, which simplifies exact adjustment of the overall light sensitivity of the instrument.

The anode current of the photomultiplier is integrated on a polystyrene capacitor in the integrator subunit. The voltage on this capacitor can be divided by successive factors of ten by pressing a scale factor switch-button. An electrometer circuit, also placed in the integrator, is used as an intermediate stage when the voltage on the capacitor is measured, to provide the instrument with a long time constant in the retention of the signal. A Low Voltage Power Supply provides this part of the circuitry with the necessary voltages.

The final measurement of the collected charge from the photomultiplier and the display of the result is made with a Digital Voltmeter.

The readout cycle is controlled by a synchronous motor driven Cam Timer, which is programmed to operate a series of relays once the

"Start Bar" is actuated. This feature makes routine operation simple and reliable.

A Regulating Transformer compensates effects of primary voltage fluctuations on all components of the apparatus. Among other things, it assists in keeping the heating current constant.

For measurements of small doses, the instrument is equipped with a gas inlet so that the readout chamber can be filled with an appropriate gas, normally prepurified nitrogen. The use of this feature makes it possible to measure with LiF down to 10 mR with a standard error of less than $\pm 20\%$. Among the accessory equipment is a recorder, for glow curve registration, which can be attached without disturbance on the normal integration procedure.

The controls and indicators on the Front Panel of the readout instrument are designated in Figure D.2.1-2, and identified in Table D.2.1-1 which follows.

Figure D.2.1-2 Controls and Indicators on Front
of Readout Instrument

TABLE D.2.1-1

CONTROLS AND INDICATORS
ON FRONT OF READOUT INSTRUMENT

	<u>Designation</u>	<u>Type</u>	<u>Function</u>
1	Digital Readout	Digital Voltmeter	Presents readout value, proportional to charge on integrating capacitor.
2	Reset	Press Button Switch	Discharges integrating capacitor.
3	Zero Adjust	Potentiometer	Fine zero adjustment of digital voltmeter.
4	Power On	Switch	Connects main power to instrument.*
		Light	Light indicates main power to instrument.
5	Read Cycle	Light	Light indicates readout cycle in process.
6	Scale Factor	Switch	Divides Reading by Factors of 10.
	X10 Light		When pressed once, upper light indicates division by 10.
	X100 Light		When pressed twice, lower light indicates division by 100.
7	Slide Inserter		Inserts heating element and dosimeter for measurement. Inserts light source for checkout.
8	Start Bar		Initiates readout cycle, the first stage of which is resetting instruments.

* The low voltage power supply (L.V.P.S.) is independent of the "power on" switch. When instrument is plugged into mains, L.V.P.S. is provided with power.

The operation of the phosphor dispenser and its accuracy are discussed in Section D.2.4.

The thermoluminescent phosphor itself, TLD-700, is LiF powder which has been refined so that it is essentially free of the Lithium-6 isotope (i.e., it consists almost entirely of Lithium-7). The range of radiation dosage measurable with this phosphor extends from 10 mR to 100,000 R for gamma, x-ray, beta and electron radiation. The radiation dose measurement precision attainable with this phosphor as a function of dose is tabulated below:

<u>Dose</u>	<u>Measurement Precision</u>
10 mR	$\pm 20\%$ S.D.
100 mR	$\pm 5\%$ S.D.
1R - 10^5 R	$\pm 2\%$ S.D.

In addition, the response of LiF phosphor is shown in Figure D.2.1-3 to be practically independent of incident photon energy down to about 100 keV.

However, overall accuracy of dose measurement depends not only upon dosimeter precision, but also upon the accuracy of calibration of the readout instrument and dosimeter system. Under very favorable conditions, Con-Rad claims the overall accuracy of its TLD system to be within $\pm 5\%$.

Figure D.2.1-3 Response of Con-Rad Lithium Fluoride TLD Dosimeters to Radiation Energy

D.2.2 TLD Readout Instrument Calibration

Field calibration of Con-Rad TLD Readout Instruments was performed by means of two light sources provided with the instrument, which were used to set the instrument sensitivity for dosimetry phosphor obtained from Con-Rad. These light sources have been fabricated so that they emit the same light intensity within $\pm 2\%$ of their corresponding standards at the Con-Rad Laboratory. The TLD phosphor obtained for the dose rate measurements along with the instrument was accompanied by an Instrument Calibration Form (shown in Figure D.2.2-1) which provided the recommended light source readings to permit calibration of the instrument so that readout sensitivities of 0.001, 0.10 and 10 R/digit could be obtained with the phosphor. The initial calibration procedure employed upon receipt of the instrument is detailed below.

The instrument was energized and permitted to warm up for two hours to achieve stability. After this, light source A was inserted into the readout chamber and output readings taken for photomultiplier high voltages of 400, 600, 800 and 1000 volts. These readings were found to be in excellent agreement with corresponding curves provided by the manufacturer for the instrument. Similarly, light source B was next inserted and readout

ISOTOPES, INC.

A TELEDYNE COMPANY

The sensitivity of LiF-7 Phosphor lot 760
has been determined and the following light source readings should be
set with Isotopes/Con-Rad standard light sources to obtain the required
instrument calibration.

	0.001 R/digit	0.1 R/digit	10 R/digit
Light Source	B	A	A
Reading	<u>185</u>	<u>306</u>	<u>3</u>

NOTE: The method utilizing the light source to calibrate the
instrument is described in information sheet #8.

TLD/CALIB/1

IMPORTANT—Name & Address Change
CON-RAD - - - - ISOTOPES
Correspondence, etc. formerly directed to Controls for Radiation, Inc. or Con-Rad Laboratories, Iso- topes, Cambridge, Mass., should be directed to
Isotopes, A Teledyne Company Laboratory Products Division 50 Van Buren Avenue Westwood, N J 07675

Figure D.2.2-1 Instrument Calibration Sheet Supplied
With TLD Phosphor Used in this Experiment

values obtained at 1000, 1200, 1400, 1600 and 1800 volts. Again, the agreement with the specification curves was very good. Also, as part of these readings, the zero point of the digital voltmeter, as well as the light source readout values, were confirmed to be stable, thereby indicating instrument stability. Finally, the instrument was calibrated to obtain readout sensitivity of 0.001 R/digit by reinsertion of light source B and adjustment of the photomultiplier high voltage to obtain digital readout of 185, as specified on the Instrument Calibration Form. Subsequently, ten additional readings were performed and the average value confirmed to be the required value. In addition, the standard deviation of these readouts was calculated and confirmed as not exceeding $\pm 1\%$, thereby verifying reproducibility of the measurements.

In summary, although this light source technique of readout instrument calibration is simple and effective, the measurement accuracy obtained thereby is limited by the following factors:

- The light source readings for the desired sensitivity are valid only for the particular batch of phosphor with which they are supplied
- The readings are valid only for dosimeter phosphor which has not been annealed

- The calibration is based upon exposures to Cobalt-60 gamma radiation. For photon energies less than about 100 keV, corrections should be applied for phosphor energy dependence
- The fundamental accuracy of this calibration technique is determined by:
 1. The accuracy of calibration by Con-Rad of batches of dosimetry phosphor which is about $\pm 4\%$
 2. The accuracy of the light source used compared with that of the light source standard maintained at Con-Rad laboratories to establish dosimeter sensitivities which is $\pm 2\%$
 3. Inaccuracies created by the influence of ambient temperature on photomultiplier spectral sensitivity, variations in positioning of light source and in setting of heater current in both Con-Rad Laboratory and field instruments which should contribute less than $\pm 3\%$.

As a result of these sources of error, an overall accuracy of about $\pm 9\%$ or better was expected for the Thulium-170 dose rate measurements subsequently performed.

D.2.3 TLD Phosphor Calibration

As mentioned previously, the accuracy of the TLD system calibration is partially limited by the fact that after the phosphor has been used and subsequently annealed, its sensitivity to radiation changes. Consequently, repeated exposures of a given quantity of phosphor to identical irradiation configurations would yield varying dose indications. Accordingly, since it was anticipated that it would be necessary to conduct several annealing operations during the course of these dose rate measurements, a small quantity of unirradiated phosphor was initially set aside and not subjected to any exposure at all. Subsequently, after each annealing operation, the zero dose reading of a small sample from "clean" phosphor was compared with that of the freshly annealed stock. The ratio of the clean to annealed zero dose readings was calculated and used as a factor to modify subsequent dose rate measurement results so that all values thus obtained uniformly reflected the initial phosphor sensitivity.

Furthermore, it was anticipated that the measured dose values thus obtained could only be as precise as the phosphor response to radiation would permit. Accordingly, a calibrated quantity of TLD phosphor was obtained from Con-Rad which consisted of

500 milligrams of LiF-7 powder irradiated to 1 roentgen within $\pm 4\%$.

This irradiation was performed by Con-Rad with a Cobalt-60 source, the exposure rate of which is known to be within $\pm 3\%$. This source has been calibrated using a Landsverk Roentgen Chamber, which itself has been calibrated with a precision of $\pm 2\%$ against standards periodically checked by the National Bureau of Standards, and frequently cross-checked within Con-Rad Laboratory. The calibration certificate for the Landsverk R-meter is illustrated in Figure D.2.3-1.

The general procedure followed in making use of this calibrated phosphor to determine the response precision of its experimental counterpart is as follows:

1. Prepare separate dosimeter capsules containing identical quantities of calibration and experimental phosphor;
2. Irradiate these dosimeters simultaneously at the same separation distance from an experimental source; and,
3. Evaluate these dosimeters.

Ultimately, a calibration factor for the experimental phosphor was derived from these results according to the following equation:

ISOTOPES, INC.
123 WOODLAND AVENUE
WESTWOOD, NEW JERSEY 07675

Job No. 11297
November 22, 1967

Landsverk
CALIBRATION CERTIFICATE

Exposure Symbol *		Co 60	M	L	K	J	H
Applied KV (DC)			250	200	150	100	60
Inherent Filter			3mm al	3mm al	3mm al	3mm al	3mm al
Added Filter	mm al	**	1.0	1.0	1.0	1.0	0
	mm cu		1.0	0.50	0.25	0.	0
HVL mm cu			2.1	1.2	0.60	0.19	0.08
Approx. KV Eff.		1000	120.	80.	65.	46.	30.
Range	Serial #						
10 MR	624	1.02					
*100 MR	754	.99	.96	.95	.93	.87	.70
1 R	644	.96					
10 R	572	.98					
100 R	554	.91					
1000 R	531	.92					
10,000 R	250	.94					
* Replaced #667 (100 MR) with #754 of same range.							

Measurements were made on Roentgen Meter Model L-64, Serial No. 499

The values that are given in the table are sensitivities. The chamber reading divided by the sensitivity gives the dose.

Landsverk Roentgen Chambers are calibrated with a precision of $\pm 2\%$. Standards are checked periodically by the National Bureau of Standards and are frequently cross-checked in our laboratory.

All measurements above 250 kilovolts effective are made with a 3/16" thick bakelite sleeve over the ionization chamber. No air temperature or pressure corrections are necessary.

We certify that we have calibrated your Roentgen Chambers and Roentgen Meter which are identified above by their serial numbers and that the sensitivities are as indicated in the table.

* As per techniques laid down by the Bureau of Standards.

** No filter. Equilibrium sleeve around chamber.

D-29

$$C.F. = \frac{R_{cal} - R_{exp}}{R_{cal}}$$

where:

$$R_{cal} = \bar{X}_{cal} \pm \frac{\sigma}{\bar{X}} = \text{average net calibration phosphor dose readout and probable error}$$

$$R_{exp} = \bar{X}_{exp} \pm \frac{\sigma}{\bar{X}} = \text{average net experimental phosphor dose readout and probable error}$$

The data accumulated as a result of the foregoing procedure are tabulated in Appendix E.2 along with their associated statistical analyses. The calibration factor derived as a result is:

$$C.F. = 0.10 \text{ or } 10\%$$

Thus, the measurement precision attained with the experimental phosphor is about 10%.

D.2.4 TLD POWDER DISPENSER CALIBRATION

An investigation was conducted regarding the reproducibility of measurement of thermoluminescent dosimetry (TLD) phosphor powder into the dosimeter capsules being used in the dose rate measurement exposures. The instrument being used to measure calibrated quantities of TLD phosphor (LiF-7) into the capsules is the Con-Rad Phosphor Dispenser - Vibrator (Model PDAV-1). (See Figure D.2.4-1). This instrument is claimed to measure 43.7 milligrams of phosphor with a reproducibility of ± 0.1 percent standard deviation. The dispenser consists of a horizontal cylindrical plunger with a small vertical hole, i.e., a metering volume, normal to its axis. This plunger is closely fitted into a hole in a block of dissimilar metal which forms the dispenser body. The top of the body is machined to provide a reservoir for the powder incorporating a conical bottom with a small outlet hole which is aligned with the plunger metering volume. When the plunger is pushed, the metered volume is moved under the reservoir. A simultaneously actuated vibrator assures thorough packing of a calibrated quantity of phosphor into the cavity. After a certain vibratory packing interval, the vibrator ceases and the plunger is then pulled out against its stop whereupon the metering volume is aligned with the dispenser tube under which is placed the receptacle for the powder. Again, the vibrator is actuated during the dispensing cycle ensuring thorough removal of the calibrated phosphor quantity.

Figure D.2.4-1 Phosphor Dispenser Vibrator

To check the reproducibility of this instrument under actual conditions, a series of thirty measurements were taken of the weights of clean, dry phosphor powder dispensed each time. These measurements were made with an Ainsworth analytical balance (Model 21 N). The tabulated data thus acquired are presented in the Table D.2.4-1 along with associated statistical evaluation calculations. The net result of these measurements is that the average weight of phosphor dispensed by the instrument is:

average weight = $43.89 \pm 0.22\%$ S.D. mg.

This value appeared in good agreement with the manufacturer's claimed reproducibility of phosphor measurement, and was therefore assumed to be satisfactory. Subsequent examination of the literature^(1,2) also revealed that owing to this comparatively large nominal phosphor weight per dosimeter (i.e., 43.7 mg) the slightly higher experimental value obtained herein represented only a few additional grams of phosphor per dosimeter and any radiation dose measurement error introduced thereby is insignificant.

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 - (2) Attix, F. H., ed., "Luminescence Dosimetry", United States Atomic Energy Commission Conference Report No. CONF-650637, Proceedings International Conference of Luminescence Dosimetry, Stanford, Calif., 1965.

TABLE D.2.4-1

SANDERS NUCLEAR CORPORATION
RADIOACTIVE OPERATIONS WEIGHT MEASUREMENT DATA

PROJECT

Experimental Verification of Reproducibility of TLD Phosphor Measurement by the Con-Rad Vibrator Tool.

PROJECT ENGINEER

J. L. Cyr

EXPERIMENTAL DATA

Test No.	Sample Weight, x (mg)	Test No.	Sample Weight, x (mg)
1	43.9	16	43.9
2	44.0	17	44.1
3	44.0	18	43.8
4	43.9	19	43.9
5	43.9	20	43.9
6	44.0	21	43.8
7	43.9	22	43.7
8	43.9	23	43.8
9	43.8	24	43.7
10	43.8	25	43.9
11	43.8	26	43.9
12	44.0	27	43.9
13	44.0	28	43.8
14	44.1	29	43.9
15	43.9	30	43.9

STATISTICAL ANALYSIS OF DATA

1. Arithmetical Average

$$\bar{X} = \frac{1}{N} \sum X = 43.893$$

2. Sum of Squares of Deviations

$$\Sigma (x - \bar{x})^2 = 0.278$$

3. Standard Deviation

$$\sigma = \pm \sqrt{\frac{1}{N-1} \Sigma (x - \bar{x})^2} = \pm 0.098$$

or,

$$\sigma = \pm \frac{0.098}{43.89} (100) = \pm 0.22\%$$

APPENDIX E
SAMPLE DATA ANALYSES
AND SUPPLEMENTARY
MEASUREMENTS

APPENDIX E.1 DOSE MEASUREMENT DATA AND ANALYSES FOR
THULIUM-170 WITH LEAD ABSORBERS

SANDERS NUCLEAR CORPORATION

Radioactive Operations Data

Project USAEC Contract No. AT-(40-1)-3693

Date 18-23 SEPT 68

Project Engineer J. Cyr

Page 1 of 1

Source/Dose Point Separation #1. 100 CM

Source #1 Control Dosimeters Exposure <u>93 hrs</u> Absorbed Dose (m rad)		Source #1 Absorber PB-1($1/4''$) Exposure <u>4 hrs</u> Absorbed Dose (m rad)		Source #1 Absorber PB-2($1/2''$) Exposure <u>15 hrs</u> Absorbed Dose (m rad)		Source #1 Absorber PB-3(1.0'') Exposure <u>74 hrs</u> Absorbed Dose (m rad)	
1.	146	1.	1240	1.	977	1.	632
2.	142	2.	1220	2.	993	2.	628
3.	148	3.	1200	3.	968	3.	645
4.	140	4.	1200	4.	888	4.	634
5.	148	5.	1230	5.	940	5.	640
6.	142	6.	1220	6.	929	6.	638
7.	146	7.	1180	7.	944	7.	686
8.	145	8.	1210	8.	940	8.	676
9.	149	9.	1220	9.	946	9.	667
10.		10.	1250	10.	937	10.	688
11.		11.	1220	11.	981	11.	666
12.		12.	1210	12.	992	12.	665
13.		13.	1200	13.	952	13.	690
14.		14.	1240	14.	962	14.	672
15.		15.	1180	15.	959	15.	674

Project Engineer _____

TLD Surveyor _____

SANDERS NUCLEAR CORPORATION
RADIOACTIVE OPERATIONS DATA ANALYSIS

Radioactive Source - Thulium-170, 3.86 ± 0.07 watts, 1700 ± 31 curies

Radiation Absorber - Lead, 1/4 in., 1/2 in., and 1 in.

Separation Distance - 100 cm Exposure date - 18-23 Sept 1968

A. CONTROL ANALYSIS

93 hours

<u>Test</u>	<u>X</u>	<u> X-$\bar{X}$ </u>	<u>(X-$\bar{X}$)²</u>
1	146	0.89	0.792
2	142	3.11	9.672
3	148	2.89	8.352
4	140	5.11	26.112
5	148	2.89	8.352
6	142	3.11	9.672
7	146	0.89	0.792
8	145	0.11	0.012
9	<u>149</u>	3.89	<u>15.130</u>
Total	1306		78.886

Arithmetical Average

$$\bar{X}_c = \frac{1}{N} \sum X_1 = \frac{1306}{9} = 145.10$$

Sum of Square of Deviations

$$\sum (X - \bar{X})^2 = 78.88$$

Standard Deviation

$$\sigma = \sqrt{\frac{1}{N-1} \sum (X - \bar{X})^2} = \sqrt{\frac{78.89}{8}} = 3.14$$

Standard Error

$$\sigma_{\bar{X}} = \frac{\sigma}{\sqrt{N}} = \frac{3.14}{\sqrt{9}} = 1.05$$

Probable Error

$$r_c = 0.6745 (1.05) = 0.7082$$

Average zero dose = 145.0 ± 0.70 MR/93 hours

B. LEAD ABSORBER NUMBER ONE (1/4 in.)

4 hours

<u>Test (N)</u>	<u>X</u>	<u> X - $\bar{X}$ </u>	<u>(X - $\bar{X}$)²</u>
1	1240	25.33	641.60
2	1220	5.33	28.40
3	1200	14.67	215.20
4	1200	14.67	215.20
5	1230	15.33	235.00
6	1220	5.33	28.40
7	1180	34.67	1202.00
8	1210	4.67	21.80
9	1220	5.33	28.40
10	1250	35.33	1248.20
11	1220	5.33	28.40
12	1210	4.67	21.80
13	1200	14.67	215.20
14	1240	25.33	641.60
15	<u>1180</u>	34.67	<u>1202.00</u>
Total 18,220			5973.20

Arithmetical Average

$$\bar{X} = \frac{1}{N} \sum X_1 = \frac{18,220}{15} = 1214.67$$

Sum of Square of Deviations

$$\Sigma(X-\bar{X})^2 = 5973.20$$

Standard Deviation

$$\sigma = \sqrt{\frac{1}{N-1} \Sigma(X-\bar{X})^2} = \sqrt{\frac{5973.20}{14}} = 20.66$$

Standard Error

$$\sigma_{\bar{X}} = \frac{\sigma}{\sqrt{N}} = \frac{20.66}{\sqrt{15}} = 5.33$$

Probable Error

$$r_{\bar{X}} = 0.6745 (5.33) = 3.59$$

Divergence Coefficient of Data

$$Q^2 = \frac{1}{\bar{X}} \frac{\Sigma(X-\bar{X})^2}{N} = \frac{5973.20}{1214.67 \times 15} = 0.327$$

Chi-Square Test of Data

$$\chi^2 = \frac{1}{\bar{X}} \Sigma(X-\bar{X})^2 = \frac{5973.20}{1214.67} = 4.91$$

$$F = 15-1 = 14$$

$$\therefore P \cong .99$$

Analysis Results

1. Thulium-170 measured dose at one meter with 1/4 inch lead absorber =
1214.67 $\pm$ 3.59 MR/4 hours

$$\begin{aligned} 2. \text{ Average net dose} &= \bar{X} - \bar{X}_c \pm \left[r_{\bar{X}}^2 + r_c^2 \right]^{1/2} \\ &= 1214.67 - 6.24 \pm [(3.59)^2 + (0.03)^2]^{1/2} \\ \text{net dose} &= 1208.43 \pm 3.59 \text{ MR/4 hours} \end{aligned}$$

3. Average dose rate = 302.11 ± 0.90 MR/hr

$$4. \text{ Average dose rate normalized to 2 Aug 1968} = (302.11 \pm 0.90) \exp - \left(\frac{0.693}{128} 47 \right)$$

$$= 391 \pm 1.16 \text{ MR/hr}$$

C. LEAD ABSORBER NUMBER TWO (1/2 in.) 15-HOUR IRRADIATION

<u>Test</u>	<u>X</u>	<u> X - $\bar{X}$ </u>	<u>(X - $\bar{X}$)²</u>
1	977	18.43	339.66
2	993	34.43	1185.42
3	968	9.43	88.92
4	940	18.57	344.84
5	929	29.57	874.38
6	944	14.57	212.28
7	940	18.57	344.84
8	946	12.57	158.00
9	937	21.57	465.26
10	981	22.43	503.10
11	992	33.43	1117.56
12	952	6.57	43.16
13	962	3.43	11.76
14	<u>959</u>	0.43	<u>0.18</u>
Total	13,420		5689.36

Arithmetical Average

$$\bar{X} = \frac{13420}{14} = 958.57$$

Sum of Square of Deviations

$$\Sigma(X - \bar{X})^2 = 5689.36$$

Standard Deviation

$$\sigma = \sqrt{\frac{1}{N-1} \Sigma(X-\bar{X})^2} = 20.92$$

Standard Error

$$\sigma_{\bar{X}} = \frac{\sigma}{\sqrt{N}} = \frac{20.92}{\sqrt{13}} = 5.81$$

Probable Error

$$r_{\bar{X}} = 0.6745 (5.81) = 3.91$$

Divergence Coefficient of Data

$$Q^2 = \frac{1}{\bar{X}} \frac{\Sigma(X-\bar{X})^2}{N} = \frac{5689.36}{958.57 \times 14} = 0.423$$

Chi Square Test of Data

$$\chi^2 = \frac{1}{\bar{X}} \Sigma(X-\bar{X})^2 = \frac{5689.36}{958.57} = 5.93$$

$$F = 14 - 1 = 13$$

$$\therefore P \cong .95$$

Analysis Results

1. Thulium-170 measured dose at one meter with 1/2 inch lead absorber =
958.57 $\pm$ 3.91 MR/15 hr

$$\begin{aligned} 2. \quad \text{Average net dose} &= \bar{X} - \bar{X}_c \pm \left[r_{\bar{X}}^2 + r_c^2 \right]^{1/2} \\ &= 958.57 - 23.40 \pm [(3.91)^2 + (0.11)^2]^{1/2} \\ \text{net dose} &= 935.17 \pm 3.91 \text{ MR/15 hr} \end{aligned}$$

3. Average dose rate = 62.34 $\pm$ 0.26 MR/hr

4. Average normalized dose rate = 80.61 $\pm$ 0.34 MR/hr

D. LEAD ABSORBER NUMBER THREE (1.0") 74 HOUR IRRADIATION

<u>Test</u>	<u>X</u>	<u> X - $\bar{X}$ </u>	<u>(X - $\bar{X}$)²</u>
1	632	28.06	787.36
2	628	32.06	1027.84
3	645	15.06	226.80
4	634	16.06	257.92
5	640	20.06	402.40
6	638	22.06	486.64
7	686	25.94	672.88
8	676	15.94	254.08
9	667	6.94	48.16
10	688	27.94	780.64
11	666	5.94	35.28
12	665	4.94	24.40
13	690	29.94	896.40
14	672	11.94	142.56
15	<u>674</u>	13.94	<u>194.32</u>
Total	9901		6237.68

Arithmetical Average

$$\bar{X} = \frac{9901}{15} = 660.06$$

Sum of Square of Deviations

$$\Sigma(X - \bar{X})^2 = 6237.68$$

Standard Deviation

$$\sigma = \sqrt{\frac{1}{N-1} \Sigma(X - \bar{X})^2} = \sqrt{\frac{6237.68}{14}} = 21.11$$

Standard Error

$$\sigma_{\bar{X}} = \frac{21.11}{\sqrt{15}} = 5.45$$

Probable Error

$$r_{\bar{X}} = 0.6745 (5.45) = 3.67$$

Divergence Coefficient of Data

$$Q^2 = \frac{1}{\bar{X}} \frac{\Sigma(X - \bar{X})^2}{N} = \frac{6237.68}{660.06 \times 15} = 0.630$$

Chi Square Test of Data

$$\chi^2 = \frac{6237.68}{660.06} = 9.45$$

$$F = 15 - 1 = 14$$

$$\therefore P \cong .80$$

Analysis Results

1. Thulium-170 measured dose at one meter with 1.0" lead absorber =
 660.06 ± 3.67 MR/74 hr

$$\begin{aligned} 2. \quad \text{Average net dose} &= \bar{X} - \bar{X}_c \pm \left[r_{\bar{X}}^2 + r_c^2 \right]^{1/2} \\ &= 660.06 - 115.44 \pm \left[(3.67)^2 + (0.56)^2 \right]^{1/2} \\ &= 544.62 \pm 3.71 \text{ MR/74 hr} \end{aligned}$$

3. Average dose rate = 7.36 ± 0.05 MR/hr

4. Average normalized dose rate = 9.52 ± 0.06 MR/hr

E.2 SUPPLEMENTARY MEASUREMENTS AND ANALYSES FOR DEPLETED URANIUM ABSORBERS

Examination of the preliminary dose rate measurement results with the depleted uranium absorbers revealed several discrepancies which were not traceable to the build-up factor as previously described. Furthermore, it was apparent that these anomalies were even more pronounced at the 19 cm source-to-dose point separation distance. It was subsequently concluded that the reason for these discrepancies was the fact that depleted uranium absorbers were themselves radioactive and hence they were making spurious contributions to the measured dose rates. Accordingly, it was decided to measure the magnitude of these contributions thereby enabling the correction of the Thulium-170 source dose rate results. In conducting these experiments, all conditions of the previous measurements were duplicated even to the insertion of ORNL source number three in position inside the shield cask. The shutter, of course, was left closed. In this way, it was anticipated that despite the exact nature of the spurious radiation (e.g., beta or bremsstrahlung), subsequent subtraction of these results from the original data would completely negate their contributions. Accordingly, the data accumulated as a result of these measurements together with the net dose rates generated thereby are presented below in Table E.2-1. The summary data tabulations for depleted uranium (Table A-5) have been corrected using these results.

TABLE E.2-1

DOSE MEASUREMENT DATA AND ANALYSES FOR DEPLETED URANIUM

SANDERS NUCLEAR CORPORATION
RADIOACTIVE OPERATIONS DATA

4 Jan 1969

Page 1 of 3

Project: USAEC Contract No. AT(40-1)-3693
 Project Engineer: J. Cyr
 Source: Dose Point Separation ~13 cm

<u>Source</u> <u>Control Dosimeters</u> <u>Exposure 168 hr</u>	<u>Source: U-1</u> <u>Absorber: none</u> <u>Exposure: 168 hr</u>	<u>Source: U-2</u> <u>Absorber: none</u> <u>Exposure: 168 hr</u>	<u>Source: U-3</u> <u>Absorber: none</u> <u>Exposure: 168 hr</u>
471	6084	6114	10191
255	6083	5764	10194
405	6201	6513	10894
281	6484	6691	12493
285	6824	6654	10591
249	7013	7024	10894
303	6511	7053	12994
233	6564	6441	11593
336	6774	5884	11791
	7013	6704	9994
	6541	6883	11194
	6954	6171	12793
	6724	6874	11691
	6653	6124	11294
	-	6053	9894

TABLE E.2-1

DOSE MEASUREMENT DATA AND ANALYSES FOR
DEPLETED URANIUM (CONT)

Page 2 of 3

1. Control Dosimeter Analysis

$$\bar{X} = 313$$

$$\sigma = \pm 79$$

$$\sigma_{\bar{X}} = \pm 26$$

$$r_{\bar{X}} = \pm 18$$

Average control reading = 313 ± 18 MR/168 hr2. Uranium Source No. 1 (1/4 in.)

$$\bar{X} = 6602$$

$$\sigma = \pm 314$$

$$\sigma_{\bar{X}} = \pm 84$$

$$r_{\bar{X}} = \pm 57$$

Average 1/4 in. uranium reading = 6602 ± 57 MR/168 hr3. Uranium Source No. 2 (1/2 in.)

$$\bar{X} = 6463$$

$$\sigma = \pm 420$$

$$\sigma_{\bar{X}} = \pm 109$$

$$r_{\bar{X}} = \pm 74$$

Average 1/2 in. uranium reading = 6463 ± 74 MR/168 hr

TABLE E.2-1

DOSE MEASUREMENT DATA AND ANALYSES FOR
DEPLETED URANIUM (CONT)

Page 3 of 3

4. Uranium Source No. 3 (1.0 in.)

$$\bar{X} = 11,233$$

$$\sigma = \pm 997$$

$$\sigma_{\bar{X}} = \pm 258$$

$$r_{\bar{X}} = \pm 174$$

Average 1.0 in. uranium reading = $11,233 \pm 174$ MR/168 hr5. Net Uranium Source Readings

$$1/4 \text{ in. uranium - net dose} = 6289 \pm 60 \text{ MR/168 hr}$$

$$1/2 \text{ in. uranium - net dose} = 6150 \pm 76 \text{ MR/168 hr}$$

$$1.0 \text{ in. uranium - net dose} = 10920 \pm 175 \text{ MR/168 hr}$$

6. Net Uranium Source Dose Rates

$$1/4 \text{ in. uranium - net dose rate} = 37.4 \pm 0.4 \text{ MR/hr}$$

$$1/2 \text{ in. uranium - net dose rate} = 36.6 \pm 0.4 \text{ MR/hr}$$

$$1.0 \text{ in. uranium - net dose rate} = 65.0 \pm 1.0 \text{ MR/hr}$$

E.3 TLD PHOSPHOR CALIBRATION MEASUREMENTS DATA AND ANALYSES

SANDERS NUCLEAR CORPORATION RADIOACTIVE OPERATIONS DATA

Project: USAEC Contract No. AT-(40-1)-3693
 Project Engineer: J. Cyr
 Source: Dose Point Separation: 19 cm

A. CALIBRATION PHOSPHOR

Source
 Control Dosimeters
 Exposure: 4 min
Absorbed Dose (m Rad)

1475

1484

1495

$$\bar{X} = 1485$$

$$\sigma = \pm 10$$

$$\sigma_{\bar{X}} = \pm 6$$

$$r_{\bar{X}} = \pm 4$$

∴ average control calibration phosphor dose =
 1485 ± 4 MR/4 min.

Source: ORNL No. 3 (Tm-170)
 Absorber: none
 Exposure: 4 min
Absorbed Dose (m Rad)

4154

4095

4235

3905

3874

3895

$$\bar{X} = 4026$$

$$\sigma = \pm 155$$

$$\sigma_{\bar{X}} = \pm 63$$

$$r_{\bar{X}} = \pm 47$$

∴ average irradiated calibration phosphor dose =
 4026 ± 47 MR/4 min.

B. EXPERIMENTAL PHOSPHOR

Source
Control Dosimeters
Exposure: 4 min.
Absorbed Dose (m Rad)

259

383

329

$$\bar{X} = 324$$

$$\sigma = \pm 62$$

$$\sigma_{\bar{X}} = \pm 36$$

$$r_{\bar{X}} = \pm 24$$

average control experimental
phosphor dose =
 324 ± 24 MR/4 min

Source: ORNL No. 3
Absorber: none
Exposure: 4 min.
Absorbed Dose (m Rad)

2145

2105

2224

3065

3075

3045

$$\bar{X} = 2610$$

$$\sigma = \pm 497$$

$$\sigma_{\bar{X}} = \pm 203$$

$$r_{\bar{X}} = \pm 137$$

average irradiated experimental
phosphor dose =
 2610 ± 137 MR/4 min

C. COMPARATIVE ANALYSIS

$$\left. \begin{array}{l} \text{net calibration} \\ \text{phosphor dose rate} \end{array} \right\} = 4026 - 1485 \pm \sqrt{4^2 + 47^2}$$

$$= 2541 \pm 48 \text{ MR/4 min}$$

$$\left. \begin{array}{l} \text{net experimental} \\ \text{phosphor dose rate} \end{array} \right\} = 2610 - 324 \pm \sqrt{24^2 + 137^2}$$

$$= 2286 \pm 139 \text{ MR/4 min}$$

$$\text{Calibration Factor (C.F.)} = \frac{2541 - 2286}{2541}$$

$$= 0.10$$

$$\therefore \text{C.F.} = 10\%$$