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NEUTRON DOSIMETER

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SITE-SPECIFIC CALIBRATION OF THE HANFORD PERSONNEL NEUTRON DOSIMETER

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INTRODUCTION

A new personnel dosimetry system, employing a standard Hanford thermoluminescent dosimeter (TLD) and a combination dosimeter with both CR-39 nuclear track and TLD-albedo elements, is being implemented at Hanford. Measurements were made in workplace environments in order to verify the accuracy of the system and establish site-specific factors to account for the differences in dosimeter response between the workplace and calibration laboratory. Neutron measurements were performed using sources at Hanford's Plutonium Finishing Plant under high-scatter conditions to calibrate the new neutron dosimeter design to site-specific neutron spectra. The dosimeter was also calibrated using bare and moderated ^{252}Cf sources under low-scatter conditions available in the Hanford Calibration Laboratory. Dose equivalent rates in the workplace were calculated from spectrometer measurements using tissue equivalent proportional counter (TEPC) and multisphere spectrometers. The accuracy of the spectrometers was verified by measurements on neutron sources with calibrations directly traceable to the National Institute of Standards and Technology (NIST).

Three plutonium sources were studied: plutonium tetrafluoride, plutonium oxide, and a plutonium metal alloy. All of the sources were low-exposure plutonium containing about 5% to 6% ^{240}Pu , which is representative of the plutonium processed at Hanford in the past. The plutonium mass varied between 760 grams and 1500 grams in sealed metal containers. The plutonium fluoride source was used for primary calibration because the available source delivers a higher dose rate than the other sources and the highest neutron doses reported for personnel at Hanford historically occurred at the fluorinator hood. The fluoride source was measured at 50 cm and at 1 m, with variations in intervening plastic shielding from 0 in. to 4 in. to simulate shielded glove-box operations. The plutonium oxide and metal sources were measured only at 50 cm with no intervening shielding because of the relatively low neutron dose rates. The dosimeter results were compared with the TEPC and multisphere measurements for correlation.

METHODOLOGY

Measurements at the Plutonium Finishing Plant took place in a glove-box laboratory, which has an open area at one end sufficiently large to reduce interference from materials stored in the glove boxes. The closest glove box is approximately 4 m from the source position. The sources and phantoms were placed on aluminum stands at 1 m above a steel-reinforced concrete floor. The two closest concrete walls were approximately 3 m from the source and the other walls were approximately 8 m and 15 m from the source. The ceiling height was approximately 3 m.

The two phantoms that held the dosimeters were constructed of Lucite acrylic plastic. One phantom measured 40 cm square and the other 30 cm square, both with a thickness of 15 cm. Lucite slabs of 2.54-cm thickness (30 cm square) were taped together for the intervening shielding.

Neutron dose and spectrum measurements were performed with two different types of instruments: multisphere or Bonner sphere spectrometers and TEPCs. A brief description of these instruments is presented below; a more complete description and theory of operation is given in PNL-7881 (Brackenbush et al. 1991).

The multisphere spectrometer provides a coarse measurement of the neutron energy spectrum from thermal energies to 20 MeV, so that neutron dose equivalent can be calculated from the energy spectrum using published fluence-to-dose equivalent conversion coefficients (NCRP 1971). The multisphere spectrometer consists of a ^6LiI scintillator neutron detector that is inserted into polyethylene spheres of various sizes. Using the computer code SPUNIT (Brackenbush and Scherpelz 1983), it is possible to unfold an approximate neutron energy spectrum from the count rates from the different sphere sizes. The accuracy of the technique was verified by performing a spectral measurement with a NIST-traceable ^{252}Cf source and the fluence-to-dose equivalent conversion coefficient given in the National Bureau of Standards (NBS) Special Publication 633 for calibrating neutron dosimeters (Schwartz and Eisenhauer 1982). Several measurements with NIST-traceable neutron sources confirmed that the multisphere spectrometer consistently underestimated the dose equivalent rate by 9%, so the multisphere spectrometer results were increased by a factor of 1.09. The LiI crystal was slightly smaller than the one used for the response function calculations in the SPUNIT unfolding code.

The TEPC consists of a hollow sphere of tissue equivalent plastic filled with tissue equivalent (TE) gas. It measures the energy deposition in a known mass of TE gas and thus provides a direct measure of absorbed neutron dose. Using appropriate algorithms (Brackenbush et al. 1985), it is possible to determine the distribution of absorbed dose as a function of linear energy transfer (LET). Because quality factors are defined as a function of LET, it is possible to determine average neutron quality factors, and hence dose equivalent, directly from a single TEPC measurement. The accuracy of the TEPC was verified by measurements on a ^{252}Cf source with calibrations directly traceable to NIST in the Pacific Northwest Laboratory (PNL) Calibration Laboratory. At the beginning of the measurements, the dose equivalent determined by the TEPC was within 4% of the delivered dose equivalent from the NIST-traceable ^{252}Cf source. However, the TEPC experiences gain shifts with changes in temperature and impurities diffusing into the TE gas from the TE plastic wall, so accuracy deteriorates with time. In most instances, the TEPC determines dose equivalent within $\pm 15\%$ after several months. An internal alpha source was used to check for gain shifts before and after each TEPC measurement was completed.

The TEPCs were placed on opposite sides of the source at 1 m from the floor, and the phantoms were placed on opposite sides 90° from the TEPCs. The distance from the source varied from 50 cm to 1 m, coinciding with the distance at which the dosimeters were placed from the source. The TEPC measurements were taken with all dosimeter irradiations for the entire duration of each exposure. Irradiation times varied from overnight to more

than two weeks, depending on the source strength and the distance from the source. Background measurements with the sources removed from the room were also taken for correlation. Gamma measurements with a microrem meter were taken with each source at the same distances as above in order to quantify the contributing gamma dose. This was performed for each source and was repeated without the source for background measurement determination.

DOSIMETER DESIGN

The new Hanford neutron dosimeter consists of a package of two dosimeter designs which are physically attached to each other: 1) a beta/photon dosimeter with four TLD elements and 2) a neutron dosimeter with four TLD elements and two CR-39 foils.

The beta/photon dosimeter is based on the Harshaw 8825 design with four TLD-700 elements. The filtration is described in Figure 1 and in Table 1. The 8825 was modified from the standard design by substituting a TLD-700 for the TLD-600 neutron-sensitive element, since this dosimeter will be used only in combination with the 8816 neutron dosimeter. This allows for better discrimination of low- and mid-energy photons.

The neutron dosimeter is the Harshaw 8816 design with one TLD-700 and three TLD-600 elements. The filtration is described in Figure 2 and Table 2. Two CR-39 foils have been included in the design in order to provide capability for a technology with better neutron-energy response. The 8816 dosimeter will be used for the personnel neutron dose of record when the 8816 reports a neutron dose equivalent less than 50 mrem or greater than 1000 mrem. Otherwise, the dose equivalent calculated from the processed CR-39 elements will be used for the neutron dose of record.

DOSE CALCULATION

The basic methodology for neutron dose calculation for the 8816 dosimeter is based on the albedo effect. The NIST traceability is maintained for all sources used for beta, gamma, x-ray, and neutron calibrations.

The 8816 dose calculation algorithm can distinguish between bare and moderated ^{252}Cf spectra and will apply the appropriate calibration factors. Element 1 (TLD-700) is used for gamma background subtraction from the other three elements. After subtraction of the gamma component, the ratios of elements 2 to 4 and 3 to 4 are used for energy determination and dose calculation.

The 8825 beta/gamma dose calculation is a proprietary Harshaw algorithm that uses chip ratios to determine what calibration factors to apply.

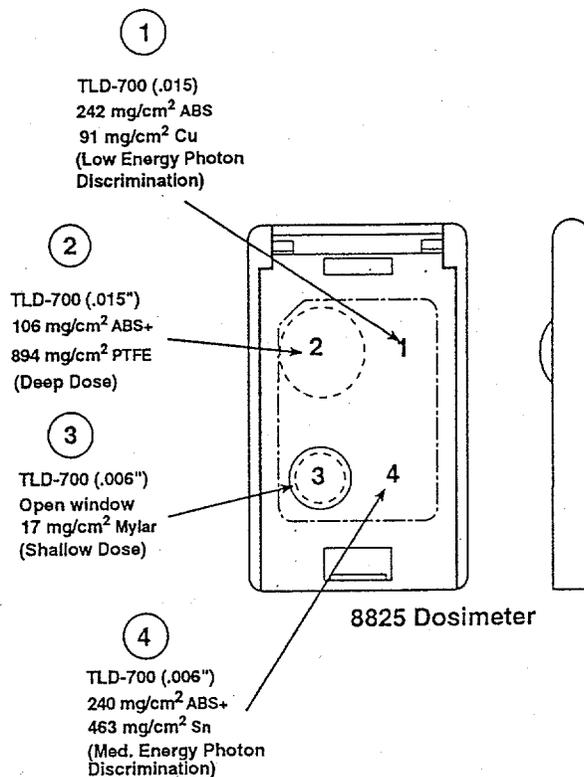


FIGURE 1. Hanford Combination Beta/Gamma Dosimeter

TABLE 1. Hanford Combination Neutron Dosimeter Holder Design

Dosimeter Position	Phosphor Type	Thickness, mil (mass density)	Total Holder Filtration ^(a)	
			Front	Back
1	TLD-700	15 (100 mg/cm ²)	464 mg/cm ² Sn plus 80 mg/cm ² ABS plastic	464 mg/cm ² Sn plus 80 mg/cm ² ABS plastic
2	TLD-600	15 (100 mg/cm ²)	461 mg/cm ² Cd plus 80 mg/cm ² ABS plastic	464 mg/cm ² Sn plus 80 mg/cm ² ABS plastic
3	TLD-600	15 (100 mg/cm ²)	464 mg/cm ² Sn plus 80 mg/cm ² ABS plastic	461 mg/cm ² Cd plus 80 mg/cm ² ABS plastic
4	TLD-600	15 (100 mg/cm ²)	464 mg/cm ² Sn plus 80 mg/cm ² ABS plastic	464 mg/cm ² Sn plus 80 mg/cm ² ABS plastic

(a) Values include Teflon (2 mil) used to enclose chips.

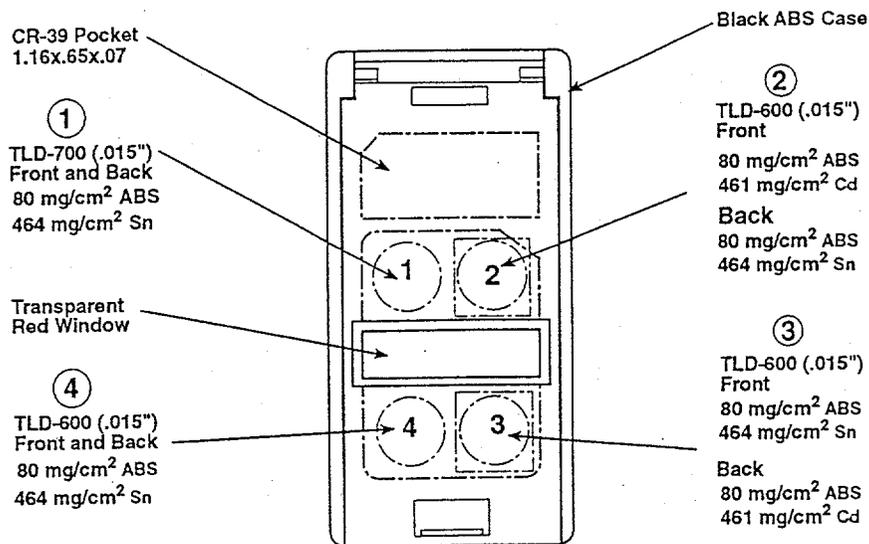


FIGURE 2. Hanford Combination Neutron Dosimeter

TABLE 2. Hanford Combination Beta/Gamma Dosimeter Holder Design

Dosimeter Position	Phosphor Type	Thickness, mil (mass density)	Total Holder Filtration ^(a)	
			Front	Back
1	TLD-700	15 (100 mg/cm ²)	242 cm ² ABS ^(b) plastic plus 91 mg/cm ² copper	173 mg/cm ² ABS
2	TLD-700	15 (100 mg/cm ²)	1000 mg/cm ² ABS and PTFE ^(c) plastic	173 mg/cm ² ABS
3	TLD-700	6 (40 mg/cm ²)	17 mg/cm ² Teflon and Mylar	173 mg/cm ² ABS
4	TLD-700	15 (100 mg/cm ²)	240 mg/cm ² ABS plastic plus 463 mg/cm ² tin	173 mg/cm ² ABS

(a) Values include Teflon (2 mil) used to enclose chips.

(b) ABS = acrylonitrile-butadiene-styrene.

(c) PTFE = polytetrafluoroethylene.

RESULTS

Table 3 shows the dose information for each irradiation that was made for each source, along with the pertinent geometry information.

TABLE 3. Dose Equivalent Rates as a Function of Distance for Shielded Plutonium Sources

Source/ Distance	Shielding	Neutron, mrem/h				Gamma, mrem/h	
		8816	TEPC	Multisphere	CR-39	8825	Rem meter
PuF ₄ 1 m	No shield	9.45	5.79	5.62	6.46	0.90	1.10
	1-in. shield	5.83	3.99	4.22	4.16	0.89	0.88
	2-in. shield	N/A	3.11	2.93	N/A	N/A	0.70
	3-in. shield	N/A	2.14	1.66	N/A	N/A	0.60
	4-in. shield	N/A	1.61	1.25	N/A	N/A	0.50
PuF ₄ 50 cm	No shield	35.4	20.6	19.6	21.5	3.10	4.00
	1-in. shield	15.5	15.2	14.9	12.8	2.90	3.20
	2-in. shield	18.0	9.93	9.46	9.94	2.80	2.80
	3-in. shield	10.3	6.21	6.50	4.97	2.22	2.10
	4-in. shield	10.9	3.94	3.83	4.08	2.12	1.90
PuO ₂ 50 cm	No shield	0.45	0.42	0.41	0.47	1.83	2.00
Metal 50 cm	No shield	0.70	0.53	0.50	0.59	1.76	2.20
Background (no source)		0.12	0.12	0.13	0.02	0.15	0.20
Glove Box 9		0.52	0.12	0.12	0.09	7.61	3.65

ANALYSIS

Table 4 shows the field calibration ratios of TLD results divided by measured dose for the plutonium tetrafluoride source and for the Glove Box 9 work area. Figure 3 overlays the source spectra for each of the sources used, with plutonium tetrafluoride shown as the bare source and as having 4-in. shielding.

The dosimeters are calibrated under low-scatter conditions with a bare ²⁵²Cf source. This radiation geometry and low-scatter condition are required for conformance to U.S. Department of Energy Laboratory Accreditation Program (DOELAP) testing criteria. But in the workplace, dosimeters are exposed to a variety of different sources under high-scatter conditions. This is evidenced by Figure 4, which compares the measured spectra of the californium calibration source and the spectra of a glove box in the Plutonium Finishing Plant, which is representative of typical plutonium operations at Hanford. The lower energy neutrons in the workplace spectra produce a greater neutron dosimeter response per unit dose equivalent. Thus, it is necessary to introduce field-specific or site-specific correction factors to account for the differences in response to the neutron energy spectra. Note that much of the response of the TLD-albedo dosimeter originates from low-energy scattered neutrons with energies below about 10 keV. The scattering conditions in work areas are very

TABLE 4. Field Calibrations for Plutonium Tetrafluoride Source at 1 m and 50 cm and for Glove Box 9 Work Area

Source Distance/ Work Area	Shielding	8816	Average TEPC + Multisphere	Ratio
PuF ₄ 1 m	No shielding	9.45	5.71	1.63
	1-in. shielding	5.83	4.11	1.42
PuF ₄ 50 cm	No shielding	35.4	20.3	1.74
	1-in. shielding	15.5	15.1	1.03
	2-in. shielding	18.0	9.70	1.86
	3-in. shielding	10.3	6.36	1.62
	4-in. shielding	10.9	3.89	2.80
			Average	1.73
PuO ₂	No shielding	0.45	0.41	1.09
Pu-Ga alloy	No shielding	0.70	0.52	1.35
Background	No source/shield	0.12	0.12	0.99
Glove Box 9		0.52	0.12	4.33

Multisphere Flux Versus Energy

Overlaid spectra - 50 cm

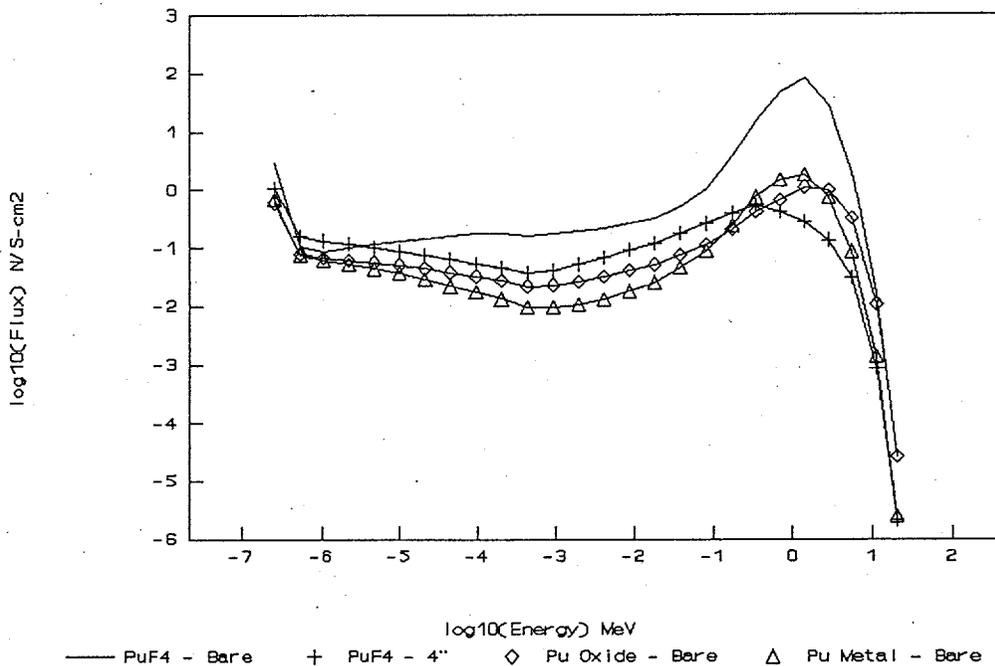


FIGURE 3. Overlaid Plutonium Source Spectra at 50 cm

Multisphere Flux Versus Energy

Overlaid spectra - Glove Box and Cf-252

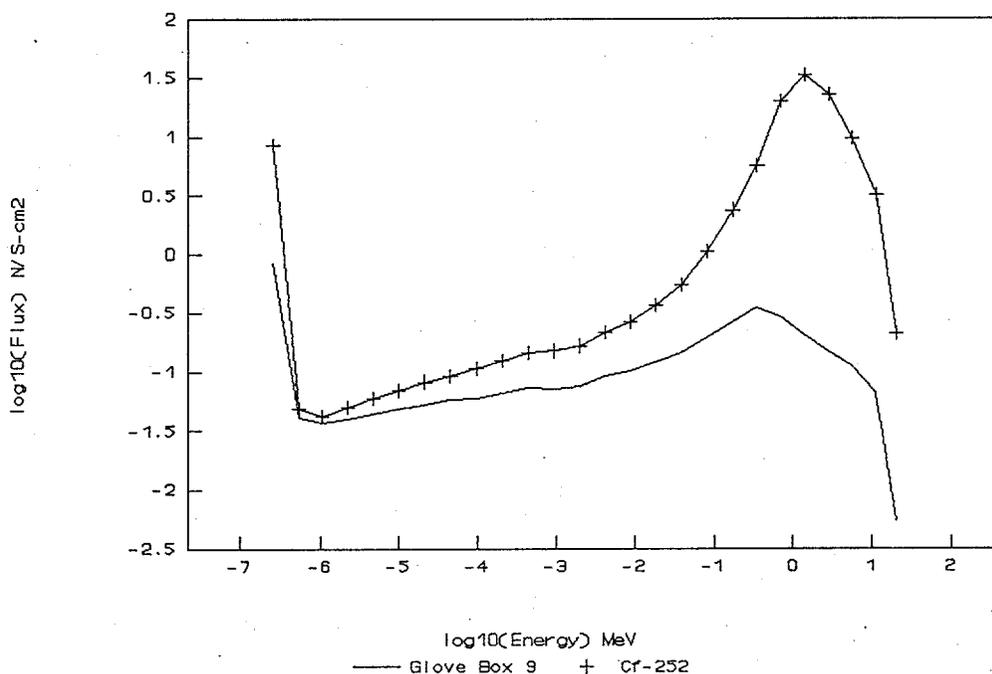


FIGURE 4. Neutron Energy Spectra from Glove Box 9 and Bare ^{252}Cf

important in determining dosimeter response. Any additional moderator, such as acrylic plastic walls of glove boxes or thick concrete floors and walls, can increase the TLD-albedo dosimeter response per unit dose equivalent.

CONCLUSION

Traditionally, the highest neutron exposures at Hanford have been the result of fluorination of plutonium. To minimize workers' doses, additional shielding has been placed around fluorination operations, which has produced changes in the neutron energy spectra and TLD-albedo dosimeter response per unit dose equivalent, as evidenced by Figure 3 and Table 4.

But the Hanford mission has changed from plutonium production, and plutonium fluoride is no longer being produced. Workers are now exposed to neutrons as the result of handling plutonium oxide or metal, and the neutron energy spectrum shown in Figure 4 is typical of glove-box operations at Hanford. Dosimeters were also exposed at 50 cm from cans of plutonium metal and plutonium oxide under the high-scatter conditions typical of workers packaging and moving plutonium. These spectra include low-energy neutrons scattered from the concrete floor and walls and room background.

The average correction factor for all of the measured spectra is 1.7 ± 0.5 . When the TLD-albedo dosimeter is calibrated to a bare ^{252}Cf source at 50 cm in a low-scatter calibration facility, the indicated dose equivalent

must be reduced by a factor of 1.7 to account for the shift in neutron energy spectra.

The CR-39 dosimeters received sufficient dose to provide good counting statistics. Under these conditions, the ratio of the dose indicated by the CR-39 dosimeter divided by the delivered dose as measured by the average of the TEPC and multisphere spectrometer measurements was 0.99 ± 0.16 for all the measurements. If the delivered dose is above about 100 mrem and the sources are lightly shielded, the CR-39 dosimeter provides accurate results. But highly scattered spectra, such as those from the background measurements, have a large percentage of neutrons with energies below 100 keV. In one instance, the CR-39 dosimeter indicated a dose of 16% of that indicated by the TEPC and multisphere measurements. The CR-39 dosimeter is insensitive to neutrons below about 100 keV, so it may give questionable results. But this is an energy range in which the TLD-albedo dosimeter responds well, so that the ratio of TLD-albedo and CR-39 results may be used as a spectral indicator.

In general, the combination TLD-albedo and CR-39 nuclear track detector provided a reasonably accurate indication of dose equivalent when exposed to a wide variety of plutonium sources (metal, oxide, and fluoride) with varying amounts of moderator (0 in. to 4 in. of acrylic plastic).

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