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Prepared by J. C. McDonald, R. V. Griffith, P. Plato, J. Miklos

Pacific Northwest Laboratory
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ABSTRACT

The gamma-ray dose fraction from a moderated ^{252}Cf source was determined by using three types of dosimetry systems. Measurements were carried out in air at a distance of 35 cm from the surface of the moderating sphere (50 cm from the source which is at the center of the sphere) to the geometrical center of each detector. The moderating sphere is 0.8 mm thick stainless steel shell filled with D_2O and covered with 0.5 mm of cadmium. Measurements were also carried out with instruments and dosimeters positioned at the surface of a 40 cm x 40 cm x 15 cm plexiglass irradiation phantom whose front surface was also 35 cm from the surface of the moderating sphere. A-150 tissue-equivalent (TE) plastic ionization chambers and a TE proportional counter (TEPC) were used to measure tissue dose, from which the neutron dose equivalent was computed. The ratio of gamma-ray dose to the neutron dose equivalent was determined by using a relatively neutron insensitive Geiger-Mueller (GM) counter and thermoluminescent dosimeters (TLD). In addition, the event-size spectrum measured by the TEPC was also used to compute the gamma-ray dose fraction. The average value for the ratio of gamma-ray dose to neutron dose equivalent was found to be 0.18 with an uncertainty of about ±18%.

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MEASUREMENTS OF GAMMA-RAY DOSE FROM A MODERATED ^{252}CF SOURCE

INTRODUCTION

The Nuclear Regulatory Commission (NRC) has initiated a testing program for personnel dosimetry processors, and part of this program includes the evaluation of neutron dosimeters. The source of neutrons used for the performance testing program now underway at the University of Michigan (UM) was designed by Schwartz and Eisenhauer (1980) of the National Bureau of Standards (NBS). Earlier work (Griffith 1978) with moderated source of neutrons has also been carried out by Griffith and co-workers at the Lawrence Livermore National Laboratory (LLNL).

The NBS-developed source was designed to provide a neutron spectrum which would simulate the type of spectra encountered in the vicinity of a reactor. In order to do this, a ^{252}Cf source was surrounded by a 30-cm diameter stainless steel shell 0.8 mm thick filled with D_2O. The sphere was also covered with an outer layer of cadmium, 0.5 mm thick. The use of cadmium avoids the calculational difficulties posed by thermal neutrons, which do not contribute significantly to the dose equivalent. The source was fabricated at the Oak Ridge National Laboratory.

Initial results of the first two rounds of testing of the performance of personnel dosimetry processors have been reported (Plato 1980). The neutron source used for the second round of testing was unmoderated ^{252}Cf. Results of the third round of testing, recently completed at the University of Michigan, are reported in NUREG/CR-2891 by Plato and Miklos (1982). For the third round of testing, the neutron source was changed to the moderated ^{252}Cf source described above. Prior to the third round of testing, dosimetry processors were given a copy of the Criteria for Testing Personnel Dosimetry Performance, Draft Standard of the Health Physics Society Standards Committee which, in the Appendix, gave processors the ratio of gamma-ray to neutron dose equivalent of approximately 0.3. During the test, several processors reported to the University of Michigan that this ratio appears to be closer to 0.2.

It was decided that several independent measurements of the gamma-ray dose equivalent rate from the moderated ^{252}Cf source should be obtained using several measurement techniques. Therefore, a joint effort by the Pacific Northwest Laboratory (PNL), LLNL and UM was proposed and executed. This report describes and summarizes these measurements and compares them to previous estimates of the gamma-ray dose equivalent.

DESCRIPTION OF EXPERIMENTAL PROCEOURES

MATERIAL AND METHODS

The D_2O-moderated californium-252 source used at UM is an NBS source (NS-109). This source was used for Test #3 of the NRC pilot study of dosimetry processor performance testing. The neutron emission rate was measured at NBS prior to Test #3. Neutron dose equivalent rate from this source is determined using the NBS-measured neutron emission rate and the conversion factor 9.3×10^{-9} rem cm^2 given in the ANSI N13.11 1982 standard. The neutron dose equivalent rate on June 9, 1982 was calculated using the following equation:

$$\dot{H}_n = \frac{(5.5849 \times 10^9 \text{ n/s})(9.3 \times 10^{-9} \text{ rem}\cdot\text{cm}^2)(60 \text{ s/min})(1000 \text{ mrem/rem})}{4\pi(50 \text{ cm})^2} \qquad \text{(Eq. 1)}$$

$$\dot{H}_n = 99.20 \text{ mrem/min @ 50 cm} \qquad \text{(Eq. 2)}$$

The neutron dose equivalent rate at 50 cm from the source on June 10, 1982 was calcualted to be 99.13 mrem/min.

The D_2O-moderated californium-252 source is housed at the UM Willow Run Laboratory. In its irradiation position, the source is approximately 250 cm above the concrete floor, approximately 205 cm from the wooden ceiling, and 400 cm from the nearest concrete block wall. The source is stored in a water filled pit approximately 120 cm in diameter and 120 cm in depth, directly below the moderated sphere.

Table 1 shows some of the radiation properties of the ^{252}Cf source (ICRU 1977). Average neutron and gamma-ray energy will be lowered by the presence of the moderating medium.

TABLE 1. Radiation Properties of ^{252}Cf

Half-life:	2.65 years
Decay Modes:	96.9% α-decay 3.1% Spontaneous fission (average 3.8 neutrons per fission)
Unmoderated	
Mean Neutron Energy:	2.1 - 2.3 MeV
Specific Neutron Emission Rate:	2.31×10^6 n·s^{-1}·μg^{-1}
D_2O + Cd + Stainless Steel Moderated	
Mean Neutron Energy:	0.5 MeV
Specific Neutron Emission Rate*:	2.06×10^6 n·s^{-1}·μg^{-1}

* This number is 0.89 times the unmoderated rate, since about 11% of the neutrons are below the Cd cutoff (Schwartz 1980).

The instruments chosen for these measurements have been used for similar work which has been reported previously (Griffith 1978). LLNL provided spherical ionization chambers constructed of A-150 plastic (ICRU 1977) which were manufactured by Far West Technology Inc. (FWT), Goleta, CA. These chambers were used to measure the total tissue dose (neutrons plus gamma-rays). One chamber used was a Model IC-80 with a volume of approximately 80 mℓ. The inner diameter of this chamber is 5.66 cm and the wall thickness is 0.229. In addition, a FWT Model IC-17 spherical chamber was also used. Its inner diameter is 1.27 cm, its wall thickness is 0.51 cm, and its volume is approximately 1 mℓ. These chambers were operated using a potential of 300 V and readings were taken with both polarities. Methane-based tissue-equivalent (TE) gas (ICRU 1977) flowed through the chambers at a rate of approximately 10 mℓ/min, and, since the chambers were vented to air, appropriate corrections were made for local temprature and atmospheric pressure. Calibrations of these chambers at LLNL agreed to within 2% of the manufacturer's calibration. The LLNL calibration was used for these measurements.

A Geiger-Mueller detector (FWT Model GM-2) from LLNL was also used to measure gamma-ray tissue dose. Since this detector is somewhat sensitive to thermal neutrons (Wagner 1961), data was taken with and without a ^{6}Li shield surrounding the counter tube. It was found that the difference between these conditions was not significant for this source. The response of the G-M detector to fast neutrons from the ^{252}Cf source was estimated to be less than 1% (Klein 1979), therefore this correction factor was assumed to be negligible.

TLDs from LLNL were exposed at the same position as the other detectors. These dosimeters consisted of three ^{7}Li-enriched LiF TLD chips mounted in plastic discs approximately 4 cm in diameter and about 0.4 cm in thickness. These dosimeters are of the standard TLD types worn by workers at LLNL and are normally attached to a security document. Calibrations of each dosimeter were carried out using ^{137}Cs and ^{60}Co sources, and corrections for the fast neutron response of this type of dosimeters were taken from the work of Wingate (1967).

The Pacific Northwest Laboratory (PNL) provided a FWT spherical proportional counter (Model LET 1/2). This counter has an inner diameter of 1.27 cm with a 0.25 cm thick wall, and was filled to a pressure of 68 Torr with propane-based TE gas (ICRU 1977). This pressure corresponds to an equivalent diameter of approximately 2 μm in unit-density tissue, due to the reduction in pressure. This counter provided a simultaneous measurement of both neutron and gamma-ray tissue dose which could be separated in a manner to be described later (Weaver 1977).

The spherical proportional counter was operated at two potentials, +475 V for neutron portion and +645 V for the gamma-ray portion. Pulses were amplified using an Ortec Model 142-PC preamplifier, followed by approximately 25 m of low-noise cable which was connected to a Canberra Model 1412 spectroscopy amplifier. Shaped pulses were then accumulated using a Canberra Series 40 multi-channel analyzer.

Calibration of the counter was carried out in two ways. The built-in ^{241}Am alpha particle source produces a distribution of pulses with an energy deposition estimated to be 88 keV/μm ±3% (McDonald 1979). In addition, the position of the "proton-edge," which occurs because of the maximum in the proton stopping power versus energy relationship (Barkas 1964), was estimated to occur at approximately 105 keV/μm (Glass 1972). These two reference points were used to establish the energy deposition scale.

The output of an electronic pulser was set to coincide with the alpha source peak. The main amplifier gain was then increased, in order to acquire the gamma-ray portion of the spectrum, and the pulser peak was then noted to obtain the calibration of this portion of the spectrum.

The energy deposition scale is expressed in terms of lineal energy, y. This quantity is the amount of energy deposited, by charged particles, divided by the mean chord length in the volume of interest. In the case of a sphere this is 2/3 the diameter. A useful method for presenting the data is to form the product of the number of counts, or frequency, f, with the lineal energy, y, squared. When this product is plotted versus the lineal energy on a semi-log scale, the area under the curve is proportional to dose. This property follows from the fact that the derivative of ℓn y is 1/y, so that:

$$\int_a^b y^2 \cdot f(y)\, d(\ell n\ y) = \int_a^b y \cdot f(y)\, dy \qquad \text{(Eq. 3)}$$

Since $y \cdot f(y)\, dy$ is equal to the energy imparted in the lineal energy range between y and y + dy, the second integral is proportional to the dose deposited by particles with lineal energies between the limits a and b.

The data obtained in this experiment is shown in Figure 1, and this presentation clearly shows the separation of events due to gamma-rays and neutrons. This separation occurs because the secondary charged particles created by gamma-ray interactions (which are electrons) cannot deposit more than about 20 keV/μm. The separation is quite clear in this case, since there is a "valley" in the curve at about 10 keV/μm. The events above this point are predominately due to recoil protons created by neutrons interactions in the counter wall.

The hatched lines indicate the spread in the raw data, and the decrease in this spread, shown at about 4 keV/μm, is due to a gain change in the pulse amplifier. The dashed lines indicate the smoothed curves that were passed through the data in order to delineate the areas of gamma-ray and neutron events.

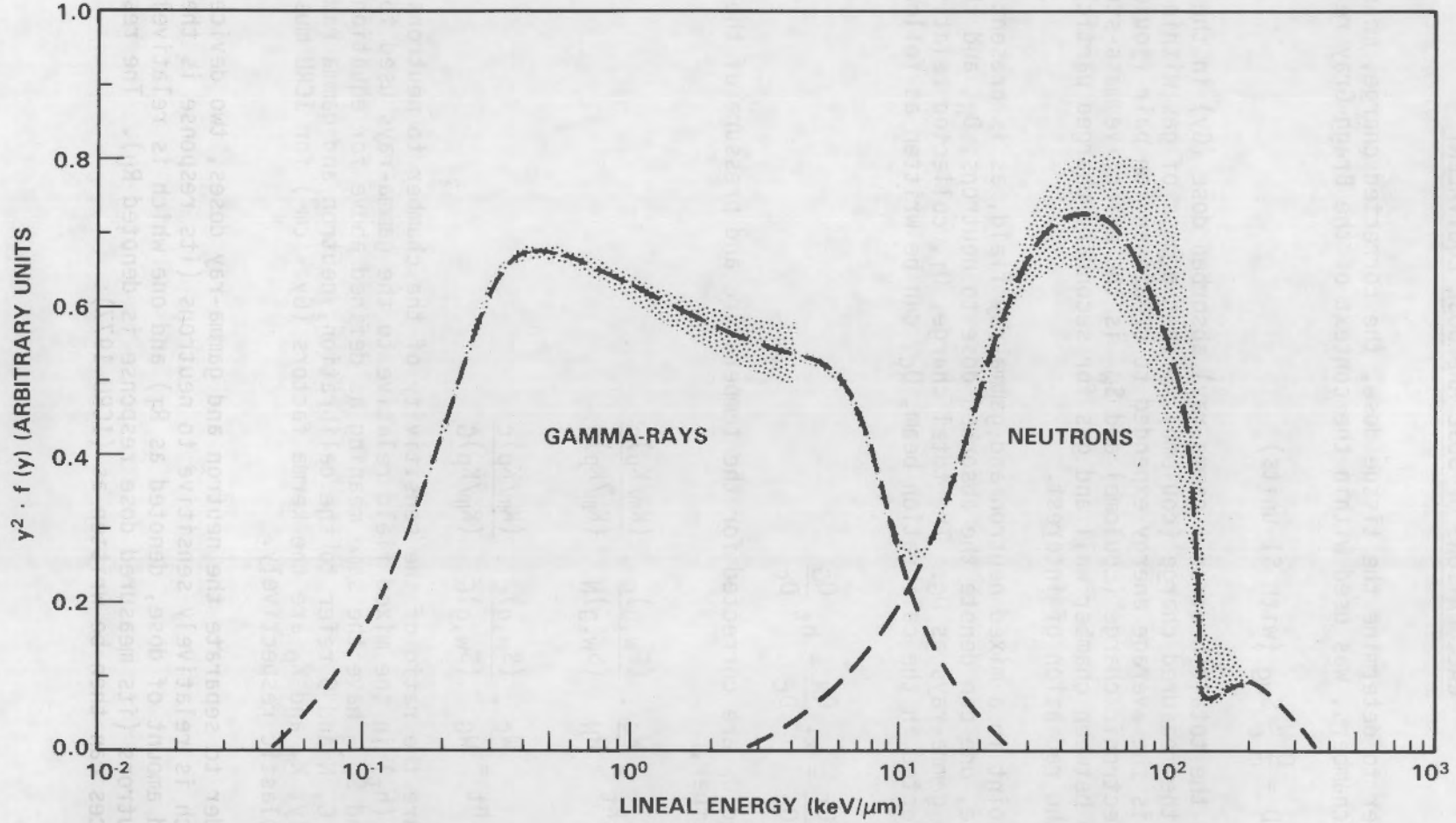

FIGURE 1. Event-Size Distribution from Moderated ^{252}Cf

CALCULATIONS OF DOSE AND DOSE EQUIVALENT

In order to determine the tissue dose, the corrected charge, measured with the TE ion chambers, was used within the context of the Bragg-Gray relationship:

$$D = \frac{Q}{m}\frac{W}{e} S_{wg} \text{ (with SI units)} \qquad \text{(Eq. 4)}$$

where, D is the total (neutron + gamma-ray) absorbed dose (Gy) in the chamber wall, Q is the measured charge (coulomb), m is the mass of gas within the chamber (g), W is the average energy expended to create an ion pair (joules/coulomb), e is the electronic charge (coulomb) and S_{wg} is the effective mass-stopping power ratio between chamber wall and gas for secondary charged particles generated by the radiation of interest.

At a point in a mixed neutron and gamma-ray field, as is present near a ^{252}Cf source, one can denote the absorbed dose to neutrons, D_N, and the absorbed dose due to gamma-rays as D_G. The total charge, Q_T, collected relative to the charge collected in the calibration beam, Q_C, can be written as follows (ICRU 1977):

$$\frac{Q_T}{Q_C} = k_t \frac{Q_N}{D_C} + h_t \frac{Q_G}{D_C} \qquad \text{(Eq. 5)}$$

where Q_T and Q_C are corrected for the temperature and pressure of the gas in the ion chamber,

and,
$$k_t = \frac{W_C}{W_N} \cdot \frac{(S_{w,g})_C}{(S_{w,g})_N} \cdot \frac{(K_m/K_p)_C}{(K_m/K_p)_N} \qquad \text{(Eq. 6)}$$

$$h_t = \frac{W_C}{W_G} \cdot \frac{(S_{w,g})_C}{(S_{w,g})_G} \cdot \frac{(K_m/K_p)_C}{(K_m/K_p)_G} \qquad \text{(Eq. 7)}$$

k_t and h_t are the ratio of the sensitivity of the chamber to neutrons (k_t) and gamma-rays (h_t) in the mixed field relative to the gamma-rays used for calibration. W and $S_{w,g}$ have the same meaning as defined above for equation 4 and the subscripts C, N and G refer to the calibration, neutron and gamma radiations, respectively, K_m and K_p are the kerma factors ($Gy \cdot cm^2$) for ICRU muscle-tissue and A-150 plastic respectively.

In order to separate the neutron and gamma-ray doses, two devices are used, one of which is relatively sensitive to neutrons (its response is the observed or measured amount of dose, denoted as R_T) and one which is relatively insensitive to neutrons (its measured dose response is denoted R_U). The response of these devices can then be written as (ICRU 1977):

$$R_T = k_T D_N + h_T D_G \quad \text{(Eq. 8)}$$

$$R_u = k_u D_N + h_u D_G \quad \text{(Eq. 9)}$$

where h_u and k_u refer to the neutron insensitive device and correspond to the quantities h_t and k_t which were defined previously. Solving for D_N and D_G, and making the assumption that $h_T = h_u = k_T = 1$ yields the following solutions:

$$D_N = \frac{R_T - R_u}{1 - k_u} \quad \text{(Eq. 10)}$$

$$D_G = \frac{R_u - k_u R_T}{1 - k_u} \quad \text{(Eq. 11)}$$

The assumption of setting $h_T = h_u = k_T = 1$ is justified since the response of both the neutron sensitive and insensitive devices is nearly the same to gamma-rays and k_T is close to unity for these radiations. In equations 10 and 11, R_T and R_u are measured and k_u is set equal to zero for these instruments (Klein 1979).

In order to obtain tissue dose a correction was applied to account for the absorption of neutrons in the wall of each chamber. The numerical values for all factors used in this work are contained below in the Corrections and Approximations section.

The gamma-ray dose in tissue was computed from the G-M detector measurements with the following equation:

$$D_G = \text{Counts} \cdot R_{GM} \cdot C_D \cdot C_T \quad \text{(Eq. 12)}$$

where D_G is the gamma-ray dose, R_{GM} (Roentgens/Count) is the resonse of the G-M detector to the calibration gamma-ray source, C_D is the dead-time correction, and C_T is the factor to convert from roentgen to tissue rads.

The neutron and gamma-ray dose ratios were determined using the TEPC data by computing the area under the respective portions of the distributions of $y^2 \cdot f(y)$ versus log y shown in Figure 1. The same type of corrections applied to the ionization chambers were applied to the TEPC. There dose(D) was determined from the TEPC data using the following formula:

$$D = \text{Constant} \sum_{h_1}^{h_2} \frac{c \cdot h \cdot N(h)}{\text{MeV}} \cdot \rho \cdot V \quad \text{(Eq. 13)}$$

where h is the MCA channel, N(h) is the number of pulses in h, h_1 and h_2 are the limits of either the neutron or gamma-ray region, c is the instrument calibration, ρ is the density of the gas and V is the chamber volume (ICRU 1977).

DOSE EQUIVALENT CONVERSION FACTORS

The spectrum of gamma-rays emitted from the source was not measured, however an estimate of their mean effective energy was determined to be approximately 400 keV by Plato and Miklos using the response of TLDs. Due to the rather large thickness of absorber (15 cm of D_2O + 0.08 cm stainless steel + 0.05 cm Cd) it is unlikely that there were large numbers of low energy gamma-rays present at the measurement position. The detectors have a flat response above about 40-50 keV and it is assumed in this work that gamma-ray dose equivalent is identical to the gamma-ray dose.

Since the measurement was carried out using a unidirectional, indirectly ionization radiation having secondary charged particles of short range in tissue, the product of the tissue dose times an effective quality factor was used to compute dose equivalent (ICRU 1976). The neutron dose equivalent was computed from the product of tissue dose and the conversion factor from tissue dose to dose equivalent. For measurements on phantom in the moderated ^{252}Cf field a value of 10.2 for the quality factor was taken from the work of Ing and Cross (1982).

CORRECTIONS AND APPROXIMATIONS

The following table contains the various correction factors applied to the dosimeters. The symbols have been defined previously (ICRU 1977).

$$\frac{W_N}{W_G} = 1.075^{(a)}$$

$$\frac{(S_{wg})_N}{(S_{wg})_G} \sim 1$$

$$\frac{K_m}{K_p} = 1.021^{(a)}$$

The correction (F) to standard temperature and pressure was computed using the measured temperature (T) and pressure (P) as follows:

$$F = \frac{273.2 + T}{295.2} \cdot \frac{P}{760}$$

The wall attenuation for neutrons for the chambers was:

1 mℓ chamber	80 mℓ chamber
17%	7.5%

The conversion from exposure to tissue dose was taken as (ICRU 1977):

0.0096 Gy/R (D.96 rad/R)

UNCERTAINTIES

The uncertainties for the ionization chamber measurements are mainly due to uncertainties in: W-values, of approximately 3%, the stopping power ratios of approximately 1.5%, the kerma factors, of approximately ±2% and in the chamber calibration ±2.5%. An additional uncertainty of about ±5% is assigned to the separation of neutron and gamma-ray dose along with ±5% experimental uncertainties. When compounded in quadrature, the overall uncertainty for the ionization chambers are estimated to be about ±9%.

The TEPC has errors contributed from counting statistics, approximately ±5%, lack of exact knowledge of the counter volume, approximately ±3%, uncertainty in the pressure and density of the gas, about ±2% and about ±3% in the calibration, and about ±5% experimental uncertainties leading to an overall

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(a) Private communication, L. J. Goodman, National Bureau of Standards (1982).

value of about ±9%. The G-M detector has about a ±3% uncertainty due to its calibration approximately ±2.5% due to its neutron sensitivity, ±5% due to counting statistics and ±5% experimental uncertainty leading to an overall uncertainty of about ±8%. The TLDs have an uncertainty in their neutron sensitivity which contributes about ±5%, and the uncertainty in calibration contributes about ±5-7% leading to an overall uncertainty of ±9%.

The conversion factors used to calculate dose equivalent from tissue dose represented a systematic error for both the chambers and the TEPC, and probably had an uncertainty of ±15%.

The overall uncertainties in the specifications of dose and dose equivalent are conservatively estimated to be as follows:

	GM	TEPC	TLD
Gamma-Ray Tissue Dose	±8%	±9%	±9%

	Ion Chamber	TEPC
Neutron Tissue Dose	±9%	±9%

	Ion Chamber and GM	TEPC	Ion Chamber and TLD
Ratio of Gamma-Ray to Neutron Dose Equivalents	±18%	±18%	±18%

RESULTS

Table 2 lists the results of the measurements. All values are in terms of tissue dose rate (mrad/min) given at a distance of 50 cm from the center of the D_2O filled moderator sphere to the geometrical center of each detector. The values indicated were determined from the LLNL ion chamber, TLD's and G-M along with the PNL TEPC system. Each value is the average of from two to four individual measurements.

TABLE 2. Results

Gamma-Ray Tissue Dose Rates (mrad/min)

	GM	TEPC	TLD
Free Air	13.6	15.2	12.4
Phantom Surface	15.8	18.1	15.4

Neutron Tissue Dose Rates (mrad/min)

	Ion Chamber and GM	TEPC	Ion Chamber and TLD
Free Air	7.2	7.98	8.87
Phantom Surface	8.11	8.82	8.56

Ratio of Gamma-Ray to Neutron Dose Equivalents

	Ion Chamber and GM	TEPC	Ion Chamber and TLD
Free Air	0.185	0.187	0.137
Phantom Surface	0.191	0.201	0.176

Plato and Miklos have measured the gamma-ray percentage for the University of Michigan ^{252}Cf source at the phantom surface using TLDs. The value they obtained was approximately 0.16, which is in good agreement with the values obtained using the G-M and TEPC.

The values of the neutron tissue dose rate measurements were 10-15% lower than the rate computed from an NBS-based neutron emission determination used by the University of Michigan (82.7, 90 and 87.3 versus 99.2 mrem/min). Although this is just within the expected uncertainties, it may indicate that the value of the quality factor used (10.2) should have had a larger uncertainty, or should have been somewhat larger itself.

The techniques used in this study yielded slightly different results, but the differences were within the expected uncertainties. The overall average value for the gamma-ray dose equivalent ratio at the surface of the irradiation phantom was computed by taking an arithmetic mean of the results indicated in Table 2, and this value was found to be 0.18 with an uncertainty of ±18%.

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17. KEY WORDS AND DOCUMENT ANALYSIS

17a. DESCRIPTORS

Gamma-ray, dose fraction,
moderated, ^{252}Cf source,
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