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DOSIMETRY MEASUREMENTS EXTERNAL TO A 10 KILOLITER LIQUID-AIR VESSEL CONTAINING A D-T NEUTRON SOURCE

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ABSTRACT

To determine the radiation attenuation characteristics of liquid air (109 g/cm^2 thick) for 14 MeV neutrons from a central D-T source in a 10 kL vessel, radiation dose measurements were made using a large tissue-equivalent ion chamber external to the vessel. The shielding characteristics of steel spherical shells were also investigated. Experimental results were analyzed and compared with TARTNP Monte Carlo code calculations. Degradation of the ion chamber's response to high-energy neutrons was established and incorporated into the analysis. The dose attenuation factor for the unshielded detector was measured to be 0.073, compared with the calculated value of 0.081. The shielded detector signals were reduced only modestly; with a 10-cm-thick shell, the signal from the filled vessel dropped twofold.

INTRODUCTION

From April 1-8, 1976, experiments were conducted at the Insulated Core Transformer (ICT) accelerator facility at Lawrence Livermore Laboratory to determine the radiation attenuation of liquid air. The apparatus used in these experiments is shown in Fig. 1. It is a large (10-kL) spherical double-walled vessel containing liquid air 4.2 to 129.3 cm from its center. At its center there is a D-T neutron source. Neutrons pass through $\sim 109 \text{ g/cm}^2$ of the liquid air before leaving the vessel. In 1975, this apparatus had been used in experiments with liquid nitrogen.¹ As for these earlier experiments, the primary objective of this study was to measure time-of-flight to infer the emergent neutron and gamma spectra.

Neutron dosimetry measurements were made external to the vessel with a large ($\sim 90\text{-cm}^3$) tissue-equivalent (TE) ion chamber (Fig. 2). For these measurements, integrated signals were used, whereas the pulsed sphere time-of-flight experiments discriminated against any steady-state contribution. Consequently, if processes such as capture-gamma ray production are important, dose values inferred from the dynamic spectral measurements could fall below the neutron doses measured by the ion chamber.

To assess the effect of steel as a shield material, measurements were made with the ion chamber sur-

rounded by spherical steel shells 5.0 and 10.0 cm thick.

Fig. 1. The LLL 10-kL liquid air vessel containing a D-T neutron source used for radiation attenuation studies.

Fig. 2. Schematic representation of the IC-99, the $\sim 90 \text{ cm}^3$ flame-equivalent ion chamber (TEIC) manufactured by EG&G Inc.

DOSIMETRIC MEASUREMENTS AND ANALYSIS

Neutron Source Characteristics

A low-mass D-T source was constructed for use with the large liquid air vessel. The characteristics of this unit are described by Wong.² The neutrons are produced by the pulsing 400 keV deuteron beam that strikes a tritiated disc at the end of the assembly. Neutron production was monitored by counting the associated alpha particles [i.e., $T(d,n)\alpha$] at a well-defined incremental solid angle in the rear direction.

A single tritium target was used during these tests. The geometry of the alpha detector was not considered in determining the conversion factor; rather, an NE213 proton recoil counter of known sensitivity and with an absolute accuracy of $\pm 7\%$ ³ was used as an absolute flux monitor. From April 1-8, 1976, the conversion factor using this method was 2.21×10^6 n/sr· α count. The factor corresponds to neutrons emitted in the forward direction at an angle of 26° with respect to the deuteron beam line. A 6% correction is included to account for air attenuation over a 9.54-m path to the recoil counter. The neutrons are emitted not quite isotropically or isoelectrically with a beam of 400 keV deuterons. The forward direction energy is ~ 15.1 MeV, and the intensity is $\sim 6\%$ above the average.

The source operated at $\sim 10^9$ n/s, which was well-suited to our time-of-flight measurements. At a distance of ~ 2 m, however, this implies a dose ~ 500 mrad/h for the unfilled vessel, and one order of magnitude less for the experiment with the vessel filled with liquid air. Careful attention to electronic details was necessary, so we removed the electrometer from the radiation field and avoided patch panels to maintain drift currents in the 10^{-15} A range.

The experimental pit was a cube 6.1 m on a side and the source was 5.1 m from the forward wall. The pit's concrete walls were 1.9 m thick, and the ceiling's wooden beams were at least 1 m thick.

Tissue-Equivalent Ion Chamber

Our tissue-equivalent ion chamber illustrated in Fig. 2, is a spherical plastic shell mounted on a long aluminum stem with a central electrode. As designed, it is insensitive to orientation with respect to the source. Earlier calibrations with gamma rays and the high-intensity ICT source had indicated a

sensitivity of the chamber to orientation. This has since been found to be due to the electrometer acting alone as an ion chamber. In this study, the electrometer was placed outside the experimental pit and connected by a single double-shielded cable to the ion chamber. For all measurements, the ion chamber was aligned with its stem radially outward from the source.

The plastic walls, designated Shonka A-150⁴, have a hydrogen content close to that of living tissue. The chamber was used throughout with tissue-equivalent gas flowing through at a rate of ~ 5 cm³/min. The compositions of Shonka A-150 plastic and TE gas in wt% are⁵: Shonka A-150, H (9.9), C (77.1), N (4.4), O (5.0), other elements (3.6); TE gas, CH₄ (64.51), CO₂ (32.31), N₂ (3.16), impurities (0.02).

The chamber voltage on the sphere was maintained at ± 300 V. When activated by a gamma source, the sensitivity varied by less than 1% for voltages between 200 and 500 V. Drift values were taken for each polarity and corrections were made using the appropriate drift values.

With TE gas flowing through the chamber, the manufacturer's calibration for ⁶⁰Co gamma rays was 3.33×10^7 rad/C (760 mm Hg, 22°C). Our checks agreed within 3% of this value. Placement of a ~ 0.15 -cm-thick plastic sheet in front of the detector made no measurable change in its response to gamma rays.

With no radiation source present, the electrometer used with the ion chamber produced currents of 0 to 4×10^{-15} A, depending on the chamber's polarity. Within the target pit, the reading rose a few femtoamperes, which implied a background level of ~ 0.3 to 0.4 mR/h, consistent with radiation survey meter readings. The radiation source was undoubtedly due to the activation of surrounding walls and structures in the pit. Attainment of these low readings was essential to the experiment: since the signals recorded with the tank filled with liquid air and with the ion chamber shielded were not greatly in excess of background signals.

Temperatures and pressures were recorded regularly to correct the chamber gas density. Reduction of the chamber gas density reduced the ionization for a given radiation field. We took drift measurements frequently, and corrected all integrated currents according to the time elapsed from the initial (zeroing) to the final reading, with regard to chamber polarity.

Dose Measurements, Free-In-Air

Readings were taken with the chamber at ± 300 V. At times, we capped the ion chamber with a plastic cup with a thickness equal to that of the chamber wall. The cap did not attenuate the signal, implying a zero correction for the chamber wall for 15 MeV neutrons. A typical run with cap off is shown in Table 1. From Table 1, we find the instrument's current to be $\sim 8 \times 10^{-13}$ A.

Figure 3 illustrates the results of the free-in-air measurements. Measurements were taken with the ion chamber surrounded by mild steel spherical shells with a 10.0-cm inner radius. The shells were either 5 cm or 10 cm thick. The data is plotted as a function of R^2 rather than R to reveal any trend indicative of a constant background radiation B (i.e., dose $(S/R^2) + B$, where S is the source strength). Additional data shown in Fig. 3 corresponds to measurements with the empty vessel in place.

The free-in-air (unshielded) findings extrapolate to a value of $Q \cdot R^2 / \Delta\alpha = 3.58 \times 10^{-10} \text{ C} \cdot \text{cm}^2 / \Delta\alpha$ for zero spacing. The collected charge Q has been corrected for room background and detector drift,

Table 1. Characteristics of a typical neutron dose measurement

Distance from source to ion chamber center, R	103.4 cm
Number of monitor α counts $\Delta\alpha$	17,886
Voltage on chamber	+300 V
Integrated charge Q	0.641 nC
Time interval of run Δt	13 min
Background current $\cdot \Delta t$	0.047 nC
Temperature	16.2° C
Pressure	748.7 mm Hg
Corrected charge Q_c (22° C, 760 mm Hg)	0.633 nC
Collected charge over distance per alpha $Q_c \cdot R^2 / \Delta\alpha$	0.379 (nC·cm ² /α)

Fig. 3. Time-equivalent ion chamber response vs distance squared (R^2) from neutron source for different detector shieldings and vessel free-in-air. Measurements taken at 760 mm Hg and 22°C.

and is adjusted to 22° C and 760 mm Hg. At a 1-m spacing between source and detector, the room return correction is approximately ~2%.

When the 5-cm thick steel shield was in place, the signal was reduced to ~0.84 of the unshielded value, while the 10-cm thick shield reduced the signal to ~0.64 of the unshielded value.

Since the associated particle recoil counter is calibrated, the value for $Q \cdot R^2 / \Delta\alpha$ may be translated to a more meaningful form:

$$\text{fluence } \Phi = \frac{2.21 \times 10^6 (n/\text{Sr } \Delta\alpha)}{R^2} \times \Delta\alpha,$$

$$Q = \frac{3.68 \times 10^{-10} (C/\text{Sr } \Delta\alpha)}{R^2} \times \Delta\alpha,$$

$$\frac{Q (\text{Coulombs})}{\Phi \cdot n/\text{cm}^2} = 1.67 \times 10^{-16} (C \cdot \text{cm}^2/n)$$

(22°C, 760 mm Hg)

The latter value will be used later in the analysis of the experiment.

One year earlier, we had determined Q/Φ to be $1.825 \times 10^{-16} C \cdot \text{cm}^2/n$. In calculating this value, we incorporated an air-attenuation correction factor of 6%. The present value is 0.915 of the earlier value, suggesting a possible deterioration of detector response to 15 MeV neutrons. On the other hand, the ion chamber response to ^{60}Co gamma radiation appeared unchanged over one year.

Dose Measurements, Vessel Empty

The liquid air vessel we used is described by Sidhu.¹ For the dosimetry experiments, the critical dimensions of the spherical vessel we used are the radial dimensions of the four walls; these are listed in Table 2. With the fluid removed, the vessel walls interpose ~1 cm of metal between source and detector. The removal of neutrons from the high-energy peak of the spectrum estimated from pulsed spheres

is ~12%.⁶ However, the dose attenuation is expected to be more modest since the scattered neutrons and the secondary gamma rays arising from inelastic scattering contribute significantly to the dose.

Figure 3 shows the detector's response with the vessel in place. The vessel here enhances the dose by 4%. On the other hand, experiments one year earlier showed an ~5% attenuation in dose. Generally, fluctuations in the data are in this range, and we conclude that the empty tank has no effect within an uncertainty range of ~±4% in dose.

Fig. 4. Tissue-equivalent ion chamber response vs distance squared (R^2) from neutron source for different detector shieldings and vessel filled with liquid air. Measurements taken at 760 mm Hg and 22°C.

Table 2. Dimensions of liquid air spherical vessel

Wall	Inside radius R_i , cm	Outside radius R_o , cm	Difference ΔR , cm	Density ρ , g/cm ³	Material
I	3.09	3.18	0.081	7.92	Stainless steel #304
II	4.09	4.18	0.081	7.92	Stainless steel #304
III	129.31	129.84	0.533	8.72	Everdur (Cu alloy)
IV	164.86	165.28	0.453	7.85	Mild steel

Dose Measurements, Vessel

From April 1 through 6 1976, the iron vessel was filled with liquid air. Normally, air contains 0.93 molar % Argon and 0.03 molar % CO₂, neither of which was present in the liquid we studied. Our liquid air composition was 21 ± 1 molar % oxygen and 79 ± 1 molar % nitrogen. The liquid density was measured as 0.869 g/cm^3 , compared to the standard value for liquid air (with Ar) of 0.874 g/cm^3 . The $\Delta\rho$ for the liquid is therefore 103.7 g/cm^3 .

Figure 4 illustrates the detector response vs distance squared (R^2) from the neutron source. Here, the spread of data points is greater than those in Fig. 3 because when the liquid air was inserted, the signals were decreased by over one order of magnitude. Instrument drift and background corrections were $\pm 3\%$ for the data shown in Fig. 4. The dispersion of data at common positions is $\sim 7\%$.

Figure 5 summarizes the findings with all data normalized to the free-field findings. If the ion chamber was tissue-equivalent for all neutron energies, this normalization would illustrate the extent of shielding caused by the liquid air. The apparent liquid air attenuation factor is ~ 0.082 for 109 g/cm^2 of liquid air, assuming the metal shells of the vessel to have a net zero effect.

With liquid air in the vessel, the ion chamber was also operated with the shields. Runs were made with the detector successively operated inside the spherical 5- and 10-cm thick iron shields. The results, shown in Figs. 4 and 5 indicate dose transmission factors of 0.71 and 0.54 for the two shields, respectively.

The shielded detector measurements are relevant to the estimation of radiation shielding afforded by military tanks. If the detector readings accurately represent the tissue-equivalent dose, we may assign a dose attenuation mean-free-path of $\sim 15 \text{ cm}$ to a tank exposed to nuclear radiation similar to that emanating from the vessel filled with liquid air. A 2-in. steel wall would therefore transmit 71% of the free-field radiation dose.

Data Analysis

The ion chamber we used is designed to exhibit tissue-equivalent behavior to all forms of radiation. The 1-cm³ tissue-equivalent ion chamber regularly used at the ICT facility has been extensively studied and found to exhibit a tissue-equivalent response at 15 MeV neutron energy.⁷ A uniform response to neutron energy is especially desirable so that one

may take ratios of measurements with the liquid air filled vessel to those measurements free-in-air (and thereby sidestep considerations of absolute detector efficiency, which would cancel out).

We carefully examined the large ion chamber's absolute response to 15 MeV neutrons and determined the kerma per unit fluence, as shown in Appendix A. We find that at a neutron energy E_n of 15 MeV, the ion chamber's response is 0.79 of the desired tissue-equivalent value.

One year earlier, the value of Q/Φ was $\sim 9\%$ higher, which suggests that the detector deteriorated in response to 15 MeV neutrons.

We explored independent means to judge whether the chamber response was below design expectations. Measurements were made using ⁶⁰Co sources and a ²⁵²Cf spontaneous fission source, with the large ion chamber used here and the 1-cc ion chamber, the subject of the study in Ref. 7. Recently, the two chambers were compared at the ICT facility using 15 MeV neutrons. In Table 3 are listed the ratios of charge Q collected for a common integrated radiation flux. Corrections are made for the greater wall thickness of the smaller ion chamber (i.e., 0.51 vs 0.23 cm).

The relative response of the large ion chamber to 15 MeV neutrons is approximately .82 of that for ⁶⁰Co or ²⁵²Cf radiation, which is in close agreement with the value derived in Appendix A from the Bragg-Gray relation.

In this study, the major correction stems from the fact that our ion chamber was not tissue equivalent when used at 15 MeV neutron energy. Since we have made a comparison between measurements with the vessel filled to that free-in-air, the latter data is subject to the full correction for the chamber's lack of tissue-equivalency. To unfold the correct attenuation factors would require prior knowledge of the

Table 3. Detector response ratio
 $Q(\text{IC-80})/Q(\text{IC-17})$

Radiation source	Filling gas	Ratio	Corrected ratio
⁶⁰ Co (γ)	Air	89.6 ^a	88.7
⁶⁰ Co (γ)	TE	87.9 ^a	87.0
⁶⁰ Co (γ)	TE	91.1	90.0
²⁵² Cf (n, γ)	TE	93.6	90.0
15 MeV (n)	TE	72.3	71.0
15 MeV (n)	Air	75.7	74.0

^aManufacturer's ⁶⁰Co calibration values.

Fig. 5. Response of 80 cm³ TEIC vs distance squared (R^2) from neutron source. Q is collected charge, $\Delta\alpha$ is alpha particle counts, and R is distance from source to detector.

spectral nature of the radiation. We have therefore taken advantage of our Monte Carlo calculations to fold the detector response function into the appropriate calculations to permit a direct experimental comparison and to avoid tedious unfolding process. This procedure treated below then allows us to generate a dose-attenuation factor that we would find experimentally with a fully tissue-equivalent ion chamber.

Since the maximum correction upon measurement is encountered at 15 MeV neutron energy, we may anticipate that the largest adjustment to the dose-attenuation factor will be ~20%. In all cases,

the correction will decrease the apparent dose attenuation factor. Where the spectrum falling on the detector has a weak high neutron energy component (i.e., $E_n > 10$ MeV) for both of the configurations being compared, the net correction should be << 20%.

To summarize, the free-in-air readings are simply interpreted as suppressed by a factor of .79 to .82 with respect to full tissue-equivalency. In free-field conditions, the uncorrected experimental dose-attenuation factors for the 5- and 10-cm thick steel shells were 0.84 and 0.64, respectively. The corresponding factors for the filled vessel were 0.71 and 0.54, respectively.

MONTE CARLO CALCULATIONS AND ANALYSIS

As of June 22 1976, the TARTNP Monte Carlo neutron/photon transport code⁸ with the current Evaluated Neutron Data Library⁹ was used for calculations for both the unshielded and shielded ion chamber measurements previously discussed. Since the code geometry input for these two cases varied considerably and the data from them is not directly comparable, each will be presented separately.

Unshielded Detectors

Neutron Source and Geometry

Here, data were derived using the neutron source described earlier. This source, which closely matches the characteristics of the ICT source, is distinguished by a ~6% above average neutron energy in the forward direction, i.e., ~15.1 MeV.

Where included in the Monte Carlo calculations, the liquid air container, the ICT target pit, and the concrete pit walls were described as a series of concentric spherical shells with the dimensions taken from Table 1.

Free-In-Air

To permit direct comparison of calculations to experimental data, it was necessary to first determine the calculated kerma factor K per unit fluence Φ at the origin (see Appendix A). This removed effects such as air attenuation. This factor may be derived directly from the nuclear cross-sections and reaction energies incorporated in the code. We found excellent agreement between the two procedures.

TARTNP code calculations were run with a geometry that included neutron source, air, and target pit walls. The walls were represented as spherical, and the detectors as thin spherical caps of 30° half-angles in the forward direction. The doses are plotted in Fig. 6. The units given for $D \cdot R^2$ are radiation dose absorbed (rad)·cm²/source neutron.

Extrapolation of these curves to the origin gave a value of $\sim 6.07 \times 10^{-10}$ rad·cm²/n, which we used to normalize all TARTNP data for the unshielded detector calculations. Corrected for source anisotropy, this value corresponds to 7.19×10^{-7} erg·cm/g·n, or about 1.3% higher than the value

Fig. 6. TARTNP code calculations of radiation dose absorbed with and without ICT pit wall vs distance R from neutron source.

calculated for A-150 plastic. This discrepancy is likely due to statistical error.

Extrapolation of the curves to the ICT pit wall ($R = 514.68$ cm) revealed a dose enhancement by the wall ~ 1.7 times a simple $1/R^2$ dependence at that radius. The decrease of $D \cdot R^2$ from a horizontal line for the free-in-air measurement without the pit wall was due to detector and air attenuation.

Empty Vessel

In Fig. 7, we illustrate normalized values of TARTNP and mean experimental doses external to the empty vessel as a function of distance from source to ion chamber center. For comparison, we also plotted normalized TARTNP calculated doses for the free-in-air case. A comparison of the values obtained from our experiments to our calculations showed excellent agreement in normalized dose determinations relatively close to the vessel surface (170 to 300 cm from the source): the TARTNP values were within 1% of our mean experimental values. Although we suspected a degradation in the detector's response to high energy neutrons, the comparison of normalized values is not significantly affected by this since the presence of the empty vessel does not appreciably alter the neutron spectrum. Careful examination of Fig. 8 reveals a slight dose enhancement very close to the vessel (< 200 cm). This is due to scattering of the emergent radiation by the vessel walls. Generally, the calculated effect of the vessel's presence was $< 1\%$ increase in dose over free-field calculations.

A considerable spread of $\sim 7.7\%$ between measured and calculated values at approximately 430 cm distance is probably attributable to an increased dose from room return as a result of describing the room walls as a spherical shell in the Monte Carlo calculations.

Fig. 7. Comparison of TARTNP code calculations with experimental data of absorbed dose vs distance R from neutron source.

Vessel with Liquid Air

The effect of the liquid air (density $\rho = 0.869$ g/cm³) is illustrated for normalized TARTNP-calculated doses for the empty and filled vessel in Fig. 8. At 200 to 300 cm from the source, where vessel and room wall effects are minimized, the calculated average dose attenuation factor was 0.0814, which is remarkably close to our experimental value of 0.082.

A valid comparison between calculated and experimental values requires a correction of the latter, since from Table 2, the ion chamber's response is degraded for neutrons with energies above a fission spectrum. In Appendix A, an $\sim 20\%$ reduction in response was inferred for neutrons with energies of ~ 15 MeV. We did not know the exact nature of the degradation or the energy region over which it applies, so we assumed a 20% reduction in response for the following two extreme cases:

1. Degraded response of 80% for $E_n > 12$ MeV. About 25% of the total dose external to the filled vessel results from such neutrons.
2. Degraded response of 80% for $E_n > 2$ MeV. About 50% of the total dose external to the filled vessel results from such neutrons.

Rather than attempting an unfolding process, we used data taken from Monte Carlo calculations in conjunction with the two detector response functions given above to generate attenuation factors that could be compared directly to experimental values.

In applying this folding-in procedure, we generated the following corrected values of measured dose attenuation with the vessel filled:

For case 1, 0.0714 ($E_n > 12$ MeV).

Fig. 8. TARTNP code calculations comparing absorbed dose for empty and filled vessel vs distance R from neutron source.

For case 2, 0.0750 ($E_n > 2$ MeV), with a degraded response of 0.8 employed. A reasonable estimate of the corrected experimental dose attenuation should be between these two extremes; the mean value of 0.073 with an experimental uncertainty of $\pm 15\%$ seems appropriate to us. The TARTNP-calculated dose attenuation of 0.0814 falls within this $\pm 15\%$ uncertainty, the experimental mean value being $\sim 10\%$ lower than the Monte Carlo value. Even this disagreement is not unreasonable since it evolves over some 2.5 dose-attenuation, mean-free paths. A 4% increase in the attenuation cross sections in the calculations would produce agreement. The discrepancy falls within the experimental uncertainty.

Photon Dose

In addition to total (neutron + photon) dose, TARTNP calculations provided information on photon dose alone. The photons contributing to the total dose were created in the vessel walls, room walls, room air, liquid air, and detectors. The results of the Monte Carlo calculations are summarized here.

Free field calculations. Photons created in the room air and "detectors" contributed from 0.1 to 0.5% of the total dose. The photon dose contribution due to the presence of the ICT pit walls varied from $\sim 0.1\%$ near the source to $\sim 10\%$ near the walls.

Empty vessel calculations. An empty vessel increased the photon total dose an additional 2 to 3%.

Vessel with liquid air calculations. Photons contributed about 24% to the total dose, of which only $\sim 1\%$ was due to room return.

Shielded Detector

Compared with the dimensions of the ICT target pit and the liquid air vessel, the relatively small size of the steel shields and ion chamber made it impractical to merely add these features to the geometry used for the Monte Carlo calculations for the unshielded detector. Such an approach would have required an inordinate amount of computer time to obtain reasonable statistical data. The technique described below provided rapid and reliable results and is particularly justified because of the spherical geometry of the detector and shields.

Neutron and Gamma Sources

Since the dose of radiation through the liquid air vessel had already been calculated for the unshielded detector, we saved considerable computer

time by using available data. Radiation incident on the shields included neutrons emerging from the vessel or those scattered by system components such as the walls, and secondary gamma rays created throughout the system.

The neutron and gamma sources used for most of these calculations were generated from the forward-directed spectra, external and close to the outer surface of the vessel filled with liquid air. The sources were described as collimated beams fully irradiating the shields.

Geometry

The tissue-equivalent ion chamber and steel shields were described as concentric spherical shells. Baseline unshielded calculations were run for this geometry using only the A-150 plastic/TE gas ion chamber. For shielded calculations, we first added the inner 5-cm thick steel shell ($R_i = 10$ cm) and then the outer 5-cm thick steel shell.

Free-in-Air

The TARTNP calculations for the free-in-air condition were made using a 15-MeV neutron source. The experimental and calculated shield transmission factors are summarized in Table 4. The corrected experimental values were derived using the same general procedures as described earlier when corrections were made for unshielded ion chamber measurements.

The corrected experimental measurements of dose transmission for the 80-cm³ TEIC are approximately 6 and 15% lower than the Monte Carlo calculated values for the 5 and 10 cm shields, respectively. We checked for potential causes of these differences. Iron removal (nonelastic) cross sections used in the TARTNP calculations were checked against experimental results.⁶ For this Monte Carlo calculation, a 14 MeV neutron isotropic point source was placed inside a 10-cm thick iron shell ($\rho = 7.87$ g/cm³). Results compared in Table 5 show no significant differences; here, the discrepancy is within experimental uncertainty.

Table 4. Transmission factors:
free-in-air geometry

Experimental measurements	5 cm steel	10 cm steel
90-cm ³ TEIC (corrected)	0.78 $\pm$ 0.09	0.57 $\pm$ 0.11
1-cm ³ TEIC ¹⁰	0.76	0.53
TARTNP calculations	0.83	0.67

Table 5. Comparison of experimental data and TARTNP calculations for iron removal cross sections per unit mass

	Σ/ρ (cm ² /g)	
	Experiment	TARTNP
For removal from high-energy peak	0.0153	0.0158
For neutron kinetic energy above 2 MeV	0.0099	0.0106

Table 6. Transmission factors external to liquid air-filled vessel

Experimental measurements	5 cm steel	10 cm steel
90-cm ³ TEIC (corrected)	0.70 ± 0.02	0.52 ± 0.03
TARTNP calculations		
Neutrons and secondary gammas created in shield	0.846	0.681
Secondary gammas incident on shield	0.379	0.134
Total	0.72	0.53

Vessel with Liquid Air

Summarized in Table 6 are the experimental and calculated transmission factors for the steel shields

external to the vessel filled with liquid air. The corrected experimental values were derived as previously described.

Here, the agreement between corrected experimental values and code calculations is close, the corrected measurements falling 2 to 3% below the results calculated by TARTNP. This is a very significant comparison since the dose results from radiation transported through both the liquid air and the steel shields. The detector's degraded response to high-energy neutrons was not a significant factor in this case, since the high-energy neutron peak was strongly attenuated by the liquid air.

Comparison to Time-of-Flight Experiments

At the time of our study, Farley¹¹ measured the neutron and gamma spectra emerging from the liquid air vessel using time-of-flight techniques. Their findings were also compared to TARTNP code results. The dose from neutrons was inferred by folding appropriate kerma factors into both calculated and measured leakage spectra; the experimental and calculated gamma-ray, pulse-height spectra were used to directly derive the gamma dose. The experimentally derived total dose was found to be 5% greater than the calculations predicted. By contrast, in this study the experimental dose was 10% less than that calculated by TARTNP. Similarly, a reliability of 15% was attributed to the dose calculations by Farley's group. The difference in total dose determinations falls within the uncertainties we cite.

SUMMARY

- A large (~90 cm³) tissue-equivalent ion chamber was used at the ICT facility to make radiation dose measurements external to a 10-kl spherical vessel filled with liquid air. From a low-mass D-T source, neutrons were generated at the vessel's center.

- This ion chamber responded at ~80% of its expected level when exposed to 15 MeV neutrons. The reduced performance was determined by the Bragg-Gray relation and by comparison to the response of a 1.0 cm³ ion chamber, which had been independently shown to behave as tissue-equivalent.

- The ~90 cm³ ion chamber appeared to be tissue-equivalent in the field of a ²⁵²Cf source.

- When filled, the spherical vessel contained $\rho \Delta r = 109$ g/cm² of liquid air (~21 molar % O₂, 79

molar % N₂). The observed dose when corrected for deviation from tissue equivalence was reduced to 0.073 ± 0.011 of the free-field dose by the filled tank.

- In free-field geometry, spherical iron shells of 5.0 and 10.0 cm thickness and 10.0 cm inner radius, reduced the dose due to 15 MeV neutrons to 0.78 ± 0.09 and 0.57 ± 0.11 of the unshielded dose, respectively. These results were corrected for detector deviation from tissue equivalence.

- For the radiation field external to the liquid-air-filled vessel, the 5- and 10-cm thick shields reduced the dose to 0.70 ± 0.02 and 0.52 ± 0.03 of the unshielded dose, respectively. Here, the incorporated detector response corrections were modest.

● The Monte Carlo calculations confirmed experimental findings of the shielding properties for liquid air and steel shells, agreement being within the experimental uncertainties of the measurements.

● The code calculations of dose attenuation factors were higher than the experimental results by as much as ~10%. The results of our measurements and calculations are summarized in Table 7.

Table 7. Summary of results

Measurement	Corrected experimental values	Monte Carlo calculations	Experiment/ calculations
Vessel free-in-air			
Kerma per unit fluence ($\text{erg}\cdot\text{cm}^2/\text{g}\cdot\text{n}$)	5.58×10^{-7}	7.19×10^{-7}	0.78
5-cm steel shield attenuation factors	0.78 ± 0.09	0.83	0.94
10-cm steel shield attenuation factors	0.57 ± 0.11	0.67	0.85
Vessel filled with liquid air			
Liquid air attenuation factors	0.073 ± 0.011	0.081	0.90
5-cm steel shield attenuation factors	0.70 ± 0.02	0.72	0.97
10-cm steel shield attenuation factors	0.52 ± 0.03	0.53	0.98

APPENDIX A

Derivation of Kerma Per Unit Fluence for Large Ion Chamber

In an ion chamber, the energy absorbed in the gas (per gram of gas), when exposed to gamma rays, according to the Bragg-Gray relation is as follows:

$$D_{\gamma}^{\text{gas}} = \frac{Q_{\gamma} W_e}{M_{\text{gas}}},$$

where Q_{γ} is the collected charge and W_e is the energy absorbed per ion pair. The energy deposited in the gas is due mainly to primary ions formed in the chamber walls, and the energy per gram deposited in the walls reflects the stopping power of the primary ions in the two regions:

$$\begin{aligned} D_{\gamma}^{\text{TE plastic}} &= \frac{(dE/dX)_{\text{pl}}^0}{(dE/dX)_{\text{gas}}} \cdot D_{\gamma}^{\text{gas}} \equiv (S_e^p)_e D_{\gamma}^{\text{gas}} \\ &= (S_e^p)_e \frac{Q_{\gamma} W_e}{M_{\text{gas}}}. \end{aligned} \quad (\text{A-1})$$

When the detector is calibrated in a known field, the results are given in units such as coulombs/roentgen (C/R), where the R field refers to the free-field reading with air as reference:

$$D_{\gamma}^{\text{air}} (\text{rads}) = 0.869 R.$$

In such a field, the energy deposited per gram of plastic would vary as its energy absorption coefficient:

$$D_{\gamma}^{\text{TE}} = \frac{(\mu/\rho)_{\text{TE}}}{(\mu/\rho)_{\text{air}}} \cdot D_{\gamma}^{\text{air}} = \frac{(\mu/\rho)_{\text{TE}}}{(\mu/\rho)_{\text{air}}} \cdot 0.869 R.$$

Substituting, we find that

$$(S_e^p)_e \frac{Q_{\gamma} W_e}{M_{\text{gas}}} = \frac{(\mu/\rho)_{\text{TE}}}{(\mu/\rho)_{\text{air}}} \cdot 0.869 \cdot R.$$

The chamber volume is then

$$V = \frac{M}{\rho} = \frac{(S_e^p)_e Q_{\gamma} W_e}{\frac{(\mu/\rho)_{\text{TE}}}{(\mu/\rho)_{\text{air}}} \cdot 0.869 \cdot \rho_{\text{air}} \cdot R}.$$

The values for S_e and W_e , taken from Ref. 7, for air in the chamber are

$$(S_e^p)_e = 1.140,$$

$$W_e = 33.7 \text{ eV/ion pair},$$

and

$$\frac{(\mu/\rho)_{TE}}{(\mu/\rho)_{air}} = 1.102.$$

The manufacturer's calibration gives

$$Q/R = 2.88 \times 10^{-8} \text{ C/R},$$

at standard temperature and pressure. Therefore, we find a chamber volume of 89.4 cm^3 .

The walls are 0.23 cm thick so self attenuation of the gamma rays leads to an $\sim 1\%$ correction, raising the volume to 90.25 cm^3 .

When the instrument with tissue-equivalent gas filling is ^{60}Co -calibrated and parameters for that filling are taken from Ref. 7, a chamber volume of 87.0 cm^3 is found. An average volume of 88.6 cm^3 compares reasonably well to 89.2 cm^3 , determined by comparison of the response of a calibrated 1 cm^3 ion chamber. The average volume of 88.9 cm^3 will be used. With the chamber filled with tissue equivalent (TE) gas of density 1.046 g/l at 25°C , at our calibration temperature and pressure of 22°C and 760 mm Hg , we calculate a gas mass of 0.0939 g .

In Ref. 7, the correction dealing with the difference between gas and solid in heavy ion stopping-power is discussed. For protons, the stopping power of TE gas is taken to be 1.06 times that for TE solid; for electrons, the ratio is 1.03. The net effect is to reduce the K/Φ value by 3%.

Finally, the flux depression due to self-absorption by the detector must be estimated. The chamber wall is 0.229 cm thick, with a density of 1.12 g/cm^3 .¹² The equilibrium cap had a thickness of 0.15 cm teflon and 0.10 cm plastic. The areal densities were 0.37 g/cm^2 plastic and $0.31 \text{ g/cm}^2 \text{CF}_2$. Reasonably, we assume that neutrons do not multiply scatter in the chamber, so that the removal cross sections cited by Logan and Komoto⁶ are appropriate. They calculated

$$0.37 \times 0.0778 + 0.31 \times 0.0292 = 0.038.$$

Therefore, an $\sim 4\%$ attenuation is appropriate. On the other hand, in a bulky target such as that studied by Maruyama and Bouts,¹³ a relaxation length of 24 cm in polyethylene would suggest a $\sim 2\%$ attenuation. We will assume the mean of these two, and apply a correction of 1.03.

Equation (A-1) is now used to evaluate the kerma per unit fluence. The dose in the plastic may be equated to the kerma—i.e., energy deposited by heavy charged particles, with no contribution from the primary or secondary gamma rays within the field. This expression also holds in a neutron field. Dividing both sides by the fluence Φ and including the corrections we have discussed, we derive:

$$(K/\Phi)_{\text{wall}} = \frac{(Q/\Phi) (C \cdot \text{cm}^2/\text{n}) \cdot 31.4 (\text{eV/ion pair}) \cdot 1.601 \times 10^{-12} \text{ erg/eV} \cdot 1.03}{1.601 \times 10^{-19} \text{ C/esu} \cdot M_{\text{gas}} \cdot 1.03}.$$

The value for Q/Φ was found earlier to be $1.67 \times 10^{-16} \text{ C} \cdot \text{cm}^2/\text{n}$ (22°C , 760 mm Hg), and the TE gas mass was 0.0939 g .

We therefore calculate:

$$(K/\Phi)_{\text{wall}} = 5.58 \times 10^{-9} \text{ rad cm}^2/\text{n},$$

in A-150 plastic at 22°C and 760 mm Hg .

The value derived from basic cross-sections is $7.07 \times 10^{-9} \text{ rad} \cdot \text{cm}^2/\text{n}$. The detector response at $E_n = 15 \text{ MeV}$ appears then to be 0.79 times the tissue-equivalent value.

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