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OPERATED BY UNION CARBIDE CORPORATION • FOR THE U.S. ATOMIC ENERGY COMMISSION

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Health Physics Division

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R. B. Gammage, F. F. Haywood, M. H. Lee,** and J. S. Cheka

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ABSTRACT

Situations in which TSEE dosimeters of ceramic BeO can be used profitably for measuring doses of fast neutrons are exemplified by an intercomparison study with other types of nuclear accident monitoring systems and the measurement of dose profiles with high spatial resolution. The sharply changing directional response for neutrons at an angle of incidence of 90°, and the only slight directional dependence for gamma radiation, is made use of in a cubic device which can serve as a personnel accident dosimeter for determining the orientation of the wearer with respect to the neutron beam as well as measuring the neutron and gamma doses. With disks of Thermalox BeO 995, highly sensitized by heat treatment, neutron doses in the range of 2×10^{-2} to 10^{3} rads can be measured. Because the neutron-to-gamma ratio of sensitivities is only about 0.1, the system is of practical use only if the fast neutron dose is at least twice the gamma dose. For repeated use of the BeO detectors, frequent washing in alcohol is advisable to maintain a clean surface. Another difficulty for accurate work is the maintenance of constant detector sensitivity, therefore, recalibration is advisable after using the detectors two or three times.

INTRODUCTION

Dosimeters used as thermally stimulated exoelectron emitters are intrinsically suited for the detection of radiations which are stopped within a short distance of the surface of the detector. The application

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of dosimeters of a commercially available BeO ceramic[†](1,2) has concentrated thus far on the detection of beta radiation from tritium(3) and of recoil protons produced in a hydrogenous cover (polyethylene) by fast neutrons. The response of BeO to fast neutrons is inherently low. The technique introduced by Becker and Crase(4) was used in these studies to measure and distinguish between the gamma and fast neutron components in mixed radiation fields; detectors are employed in pairs, one being covered with polyethylene and the other with Teflon.

There have been several applications of this technique during the last two years, and one is now in a better position to evaluate its strong and weak points and compare the performance of exoelectron emitting BeO dosimeters with other detection systems used to measure fast neutron dose.

OPERATIONAL CHARACTERISTICS OF THE READERS AND DOSIMETERS

The BeO disks, 12.5 mm diameter and 1.5 mm width, were sensitized by a heat treatment at 1400 - 1450°C(1) in excess of 100 hours and stabilized by immersion in water for 100 hours before a final drying at 500°C. The necessity of such a prolonged heat treatment to obtain the highest sensitivity is demonstrated in Fig. 1.

Two readers have been used. The first is based on a gas flow Geiger counter with a linear rate of heating.(5) This reader has been developed for routine use with the operational parameters optimized for maximum reproducibility and sensitivity.(3) A negative potential

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[†]Thermalox 995 produced by Brush Beryllium Co., Elmore, Ohio.

of 90 V has been applied to the seat holding the BeO disk (a platinum-plated nickel cup). This causes the emerging exoelectrons of low energy (less than 1 eV[6]) to be injected into the counting volume with better efficiency and reproducibility.[7] The lower limit of exposure which can be measured is about 0.5 mR when the signal-to-noise ratio is 2. The upper limit is about 200 mR above which the dead time counting losses for this particular counter become excessive.

The second reader is an ionization chamber[8] used for the measurement of higher doses, the signal-to-noise ratio being 2 for an exposure of 2R. The original chamber was slightly modified to permit integral measurement of the TSEE signal with a voltage to frequency converter (VIDAR 2600). Various experimental parameters were investigated for their effect on the TSEE response (such as heating and gas flow rates and the time of flushing with gas before taking a reading) before adopting standard optimized procedures.

Exoelectrons have been accelerated to the collector plates of ionization chambers at voltages between -22[9] and -90 V[10]. The apparent sensitivity of the ionization chamber used in these experiments as a function of collecting potential between -10 and -110 V, with a gap of 10 mm between the BeO and the plate, is shown in Fig. 2. The sensitivity is nearly independent of voltage for high energy β particles, but increases about four-fold for exoelectrons. The ionization chamber used in these studies has been operated at -45 or -90 V.

The performance of the ionization chamber was evaluated for four counting gases: dry air, nitrogen, helium with 0.95% isobutane, and argon with 10% methane. Each time a BeO disk is inserted in the reader, air is also introduced into the ionization chamber. In order to minimize the flushing requirements before taking a reading, it would have been preferable to use dry air as the counting gas, as did Williams[9]. Air, however, was not used for the following reasons. First, and foremost, it often gave rise to a strong, nonradiation induced signal which overlapped the radiation induced signal. These spurious signals were much less pronounced with the other three counting gases. Also, TSEE detectors evaluated in air or N_2 appear to be less sensitive than those read in the other gases; the apparent sensitivities are in the approximate ratio 1:2:3 for nitrogen (dry air), helium-isobutane, and argon-methane, respectively. Consequently, these studies were restricted to either helium-isobutane or argon-methane.

The most consistent results were obtained with the helium-isobutane gas mixture. The lightness of this gas and the geometry of the reader made it easier to effect a thorough flushing after loading a detector (about 8 min. at 3 c.f.h.) than was the case with argon. This same helium-isobutane counting gas was always used in the gas flow Geiger counter.

The response of BeO ceramic detectors heated for a few hours at 1400°C has been reported previously for the exposure range 10^{-4} to 10^{6} R.[1,8] Because the detectors which we now use are more sensitive and the earlier measurements with the ion chamber were less accurate than desired, some of these measurements have been repeated.

Below 10^{-1} R the slope of the response vs. exposure graph was 0.96, a value which is in agreement with an earlier finding.[2] At higher exposures the slope decreases, to 0.82 and 0.79 as measured with the ion chamber and gas-flow Geiger counter, respectively. The results obtained with the ion chamber are plotted in Fig. 3. This property of the slope being less than unity has also been noted by Kriegseis and Scharmann.[11]

We previously expressed the belief that a lack of electrical conductivity in the surface region was causing a depression of the exoelectron emission because of a buildup of positive charge.[2] There is now reason to believe that this is not the entire explanation. For example, a fine conducting grid of evaporated carbon on the BeO surface failed to intensify the TSEE. In fact, the TSEE was reduced in direct proportion to the area of BeO covered by the carbon. Also, it has been reported by Williams[9] that the conductivity of the BeO disk begins to rise rapidly in the temperature region that TSEE takes place, with the current far exceeding the TSEE currents even at zero collection potential. The lack of unit proportionality between the response and the exposure is still lacking in a satisfactory explanation.

It can be seen in Fig. 3 that the effects of saturation first make themselves evident at about 400 R with saturation being complete at about 5000 R. The earlier measurements pointed to a higher saturation level of about 10^4 R.[8] The discrepancy may be related to the several-fold difference in the sensitivity of the two batches of de-

tectors. Using the calibration curve of Fig. 3 for the high-sensitivity detectors, exposures of up to 10^3 R can be measured accurately.

One of the more important factors for the application of TSEE detectors is their stability.[7] During angular dependence studies the detectors were checked several times for reproducible sensitivity (20 mR ^{60}Co γ) using the GM-counter. These measurements were made over a period of about one month and the results are shown in Fig. 4. The greatest variation in the sensitivity of any one detector was about 30%, while the mean sensitivity of the six detectors drifted within a range of about 15%. This instability made frequent calibration a necessity. Between measurements the BeO disks were annealed at temperatures between 500° and 550°C. A small variation in annealing temperature can significantly alter the sensitivity of a detector. It is likely that this caused some of the fluctuations shown in Fig. 4 and indicates a need to control the annealing temperature (and time) more closely. An example of the effect of annealing temperature on sensitivity is shown in Fig. 5. This phenomenon is being studied in more detail.

Pressing polyethylene and Teflon wafers onto the exoelectron emitting surface of the BeO increases the likelihood of contamination by dirt or fragments of plastic. It is advisable, therefore, to wash the detector ultrasonically in an organic solvent, such as acetone or methanol, a job best done prior to reading a detector.

The fast neutron sensitivity, via recoil protons, was determined for fission spectrum neutrons from the Health Physics Research Reactor

(HPRR) at a distance from the core where the ratio, neutron-to-gamma tissue dose, was known to be 6.5 to 1. The neutron response of five pairs of BeO dosimeters amounted to 11% of the gamma response, a figure which is in fairly good agreement with the 8% reported by Kriegseis and Scharmann,[11] using a ^{241}Am-Be source and BeO 995 detectors sensitized by lithium activator. The fast neutron measurements will be meaningful only where the ratio of neutron-to-gamma doses is appreciably greater than unity; the response of a given detector to a standard dose will normally fluctuate within a σ value of $\pm$ 4% under ideal conditions,[7] and since it is the difference in the responses of two detectors of a pair which represents the response to the fast neutron component, the uncertainty is $\pm$ 8%.

Becker and Crase[4] have reported a more satisfactory figure of merit varying from 18 to 28% in the energy range of neutrons 0.1 to 16 MeV. The reason for this discrepancy is not entirely clear. Their detectors of Thermalox BeO 995, however, were coated with vacuum deposited gold (50 $\mu g/cm^2$), a treatment originally intended to increase the sensitivity and reproducibility, a result now achieved simply by heating the bare detectors at 1400 to 1450°C.[1] Drawbacks of heavy metal coating are the possibility of thermal neutron activation and an enhanced photon energy dependence.

The response of the BeO was also checked for possible dose rate effects. To do this, the reactor was operated in the burst (>10^6 rads/sec) and steady state modes (0.2 rad/sec). The response (for five pairs of detectors) differed by only 1.4% between the burst and steady state operations, indicating, as expected, that dose rate effects are absent.

The variation in the TSEE response as the angle of incidence of neutrons (and gamma rays) onto the readout face of the BeO disk is changed has been examined and made use of to ascertain the orientation of a subject with respect to the neutron beam. The changing response of Teflon and polyethylene covered detectors is shown in Fig. 6 for 14 MeV neutrons and coexistent gamma rays. The response of the detectors surrounded by Teflon, sensitive only to gamma radiation, is not much affected by orientation. It is reduced for backward (180°) rather than frontal (0°) exposure by about 15%. The fast neutron response is reduced little until one exceeds an angle of incidence for neutrons of 45° and then reduces to zero after exceeding 90°. The directional response is more pronounced for the lower energy fission spectrum neutrons as shown by Becker and Razek,[12] whose results for 14 MeV neutrons agreed quite well with those in Fig. 6.

APPLICATION TO FAST NEUTRON MONITORING

TSEE dosimeters were included in a series of intercomparison studies of accident monitoring systems, and some encouraging results were obtained. Because all doses were in excess of 10 rad, only the ion chamber reader was used. The ORNL Health Physics Research Reactor (HPRR), operated in the pulse mode, was used to provide the radiation fields. The reactor was either unshielded, shielded by a 12 cm hydrogeneous barrier, or by a 13 cm steel barrier to provide radiation fields of differing neutron spectrum and neutron-to-gamma ratio. In addition to TSEE, the systems used to measure neutron dose included neutron energy threshold detectors, albedo type badges with TLD and

and activation foils. Dosimeter types used to monitor the gamma fields included RPL, film, TLD, and TSEE.

For TSEE, the counting gases helium-isobutane and argon-methane were used. The results are shown in Table 1. It is evident that the reliability is greater when using helium-isobutane, because the expulsion of air after each sample change is more difficult to achieve when argon-methane is used. The experimental results for the helium-isobutane counting gas are in generally good agreement with those obtained from the mean of the eleven other detection systems employed in this study.[13] The eleven reported values of fast neutron dose were within $\pm$ 30% for all pulses, while all gamma-ray dose measurements were within $\pm$ 26% at one standard deviation. An exception for TSEE is the neutron dose behind the steel shield which is low by 33%. Repetition of this experiment at a later date again gave a low neutron dose which may be explained from the energy dependence of the fast neutron response of polyethylene covered BeO.[4] After passing through the steel shielding, the spectrum is moderated to lower energies and consequently the recoil protons are of lower energy and range.

Five pairs of detectors were employed. They were covered with polyethylene or Teflon of thickness 1 mm which is more than sufficient to establish the maximum recoil proton response.[12] An eleventh detector was used repeatedly (eleven times) to check the response to a standard dose of gamma radiation during the duration of the comparative study. The response was invariant within a standard deviation of 6%.

Table 1. Results of intercomparison studies of nuclear accident monitoring systems: TSEE compared with the average of other detection systems (in parentheses)

Shielding	Unshielded	13 cm steel	12 cm lucite	Unshielded
Fast neutron dose (rad)*	250 (256)	74 (110)	42 (45)	263 (272)
Gamma dose (rad)*	36 (35)	13 (14)	32 (36)	40 (41)
Fast neutron dose (rad)†	324 (340)	137 (135)	54 (66)	236 (392)
Gamma dose (rad)†	30 (57)	16 (24)	37 (60)	51 (66)

* Operation with helium-isobutane counting gas.

† Operation with argon-methane counting gas.

Another application of the BeO fast neutron dosimeters was in a determination of the spatial distribution of fast neutron (>0.1 MeV) and gamma doses at the base of a laminated, polyethylene-borated paraffin shield placed 2 m from the core of the HPRR. The shield was a 40 cm diameter cylinder 30 cm deep with a 10 cm diameter central opening intended for partial body irradiation of mice, whose heads would protrude into the central opening through holes in the inner wall. Pairs of 12.5 mm diameter BeO 995 disks (one polyethylene and one Teflon covered) were used to evaluate the dose profile. The construction and geometry of the shield together with the dose profile are shown in Fig. 7.

Inside the shield about a ten-fold reduction in the fast neutron dose was achieved. The dose dropped rapidly at the edge of the wall, and the small detector size enabled the doses close to the wall of the shield to be evaluated with good spatial resolution. The ten-fold reduction in fast neutron dose was confirmed by irradiating 38 mm diameter sulphur foils placed in shielded and unshielded regions. The 1 cm thick lead section of the shield reduced the gamma dose by 32%.

The findings on angular dependence for fast neutrons led to our use of a cubic device, shown in Fig. 8, for locating the direction of an incident beam of neutrons. A recess was machined into each face of a 2.5 cm Teflon cube and a BeO disk was lowered into each hole with the reading face pointed outwards. Plugs were pushed gently into the recesses and against each detector, four being made of polyethylene and the other two of Teflon (using two opposing holes). Those two

BeO disks surrounded entirely by Teflon responsed only to the gamma component of the mixed field.

With this arrangement, it was easy to locate the quadrant within which the neutron beam was incident upon the cube. In addition to direction finding, the effects of the absorption and scattering of 14 MeV neutrons by a thorax phantom could be observed after placing the cube at different positions on the phantom (Fig. 9).

Analogous experiments were performed for fission spectrum neutrons from the HPRR at a standard dose of 0.3 rad with the cubic detector holder attached to the phantom, and with a 13 cm steel or a 12 cm lucite shield interposed between the detectors and the reactor core (see Fig. 10). These same shields were used in the earlier mentioned intercomparison studies and the fractional reduction in neutron dose by each shield was about the same as that indicated in Table 1.

Not too much reliability can be placed on the value of dose caused by backscattered neutrons. The excess TSEE counts due to neutrons in the case of the steel shield amount to only 10% of the total counts which is not enough for a meaningful evaluation of the neutron dose.

The Teflon cube, while permitting easy alignment to the neutron beam for making the angular dependence measurements, is obviously too large and heavy as a personnel dosimeter. Much better would be a Teflon cylinder with about six recesses for BeO disks, about the size of a regular ionization chamber. When clipped to a breast pocket, it could serve as a personnel accident dosimeter for measuring neutron and gamma doses as well as the orientation of the wearer to the neutron beam. This information could be obtained fairly quickly after the exposure, the detector readings taking about 30 minutes.

Acknowledgments:

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FIGURE CAPTIONS

Fig. 1. Exoelectron emission from Thermalox BeO 995 dosimeters (20 mR ^{60}Co γ, 12.5 mm diameter BeO disks) as a function of the time of heat treatment.

Fig. 2. Effect of collection voltage of the ionization chamber on the exoelectron counting efficiency.

Fig. 3. Mean TSEE response of ten Thermalox BeO 995 detectors to gamma radiation.

Fig. 4. Changes in the sensitivity to gamma radiation of BeO ceramic dosimeters used to measure fast neutron doses over a period of one month. The disks were washed in alcohol and annealed at 500° to 550°C before each calibration.

Fig. 5. TSEE from a BeO disk which had been soaked in water and dehydrated at progressively higher temperatures. The heating time was standardized at 30 minutes, the exposure at 20 mR ^{60}Co γ.

Fig. 6. Directional response of ceramic BeO disk TSEE dosimeters to 14 MeV neutrons and gamma radiation, using thick polyethylene and Teflon radiator covers.

Fig. 7. Fast neutron and gamma dose profile below a shield designed for the partial body irradiation of mice.

Fig. 8. Teflon cube, 2.5 cm in diameter, with six recesses and plugs of polyethylene or Teflon to hold six BeO ceramic disks 1.25 cm in diameter. The device is used for locating the direction of a neutron beam as well as for measuring the fast neutron and gamma doses.

Fig. 9. Fast neutron induced TSEE response of polyethylene covered BeO detectors held in four sides of a Teflon cube mounted on the front, back, and side of a thorax phantom. The gamma response was measured by Teflon covered BeO detectors in the remaining two faces of the cube.

Fig. 10. Fast neutron response of BeO detectors mounted in the Teflon cube placed on the front of a thorax phantom. Fission spectrum neutrons were moderated either by air, a 12 cm lucite shield, or a 13 cm steel shield.

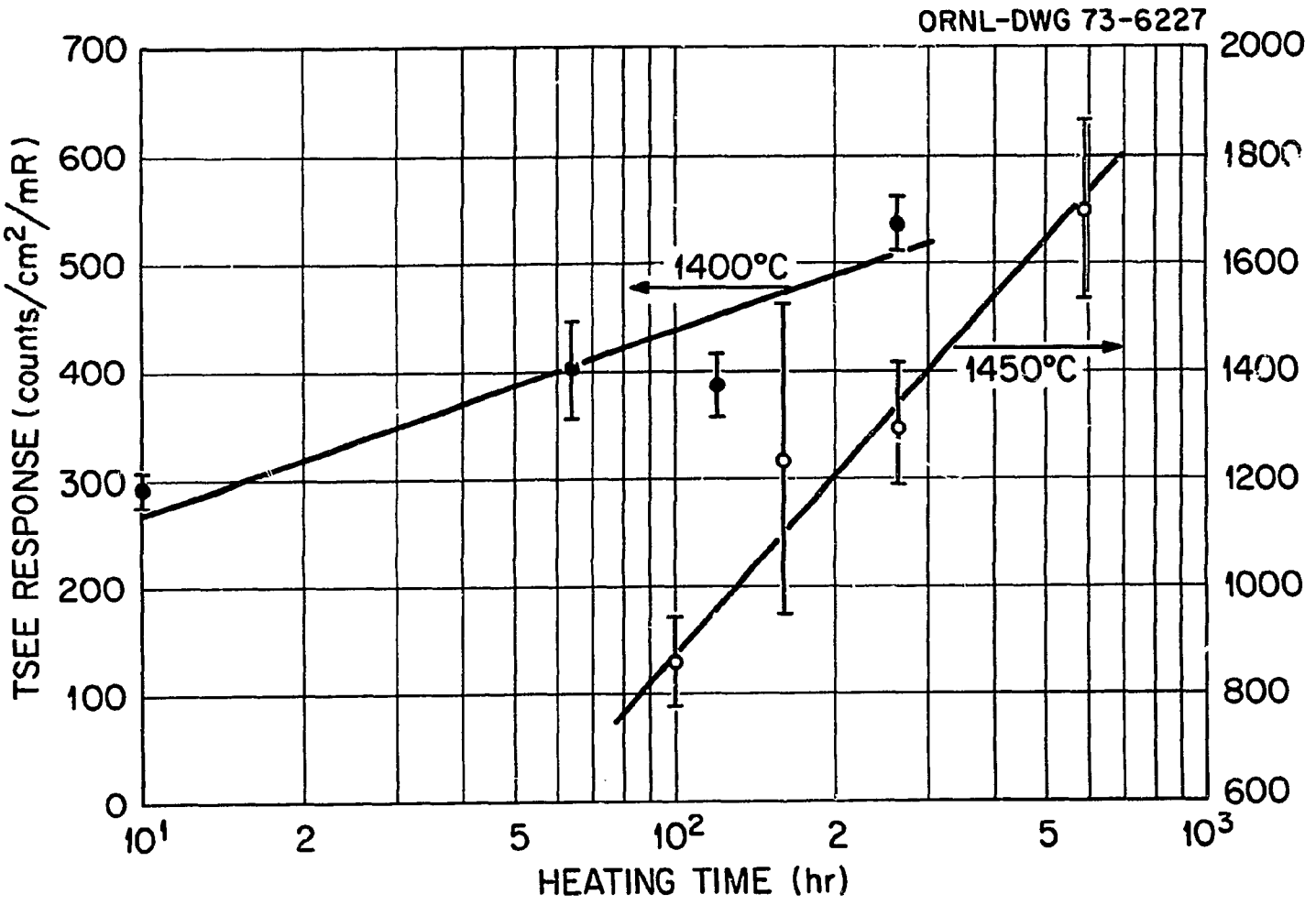
ORNL-DWG 73-6227
TSEE RESPONSE (counts/cm²/mR)
HEATING TIME (hr)
1400°C
1450°C
700
600
500
400
300
200
100
0
2000
1800
1600
1400
1200
1000
800
600
10¹
2
5
10²
2
5
10³

Figure 1

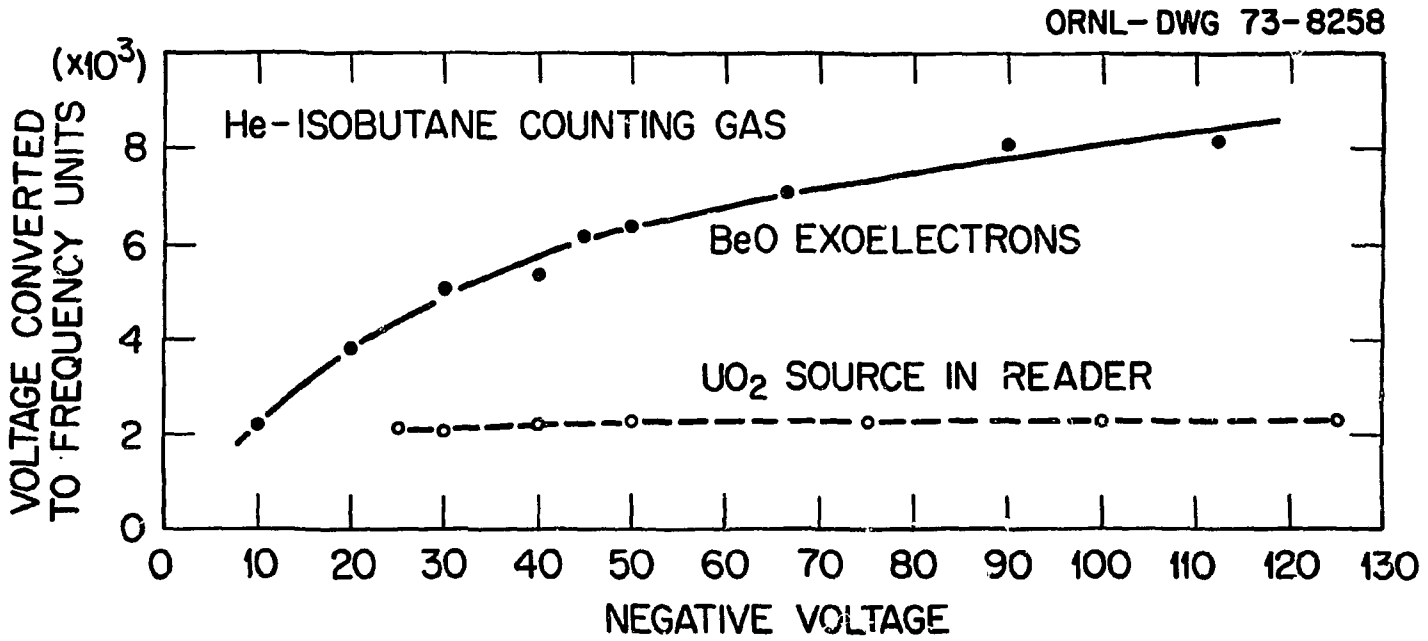
ORNL-DWG 73-8258
(x10³)
VOLTAGE CONVERTED TO FREQUENCY UNITS
He-ISOBUTANE COUNTING GAS
BeO EXOELECTRONS
UO₂ SOURCE IN READER
0
2
4
6
8
0
10
20
30
40
50
60
70
80
90
100
110
120
130
NEGATIVE VOLTAGE

Figure 2

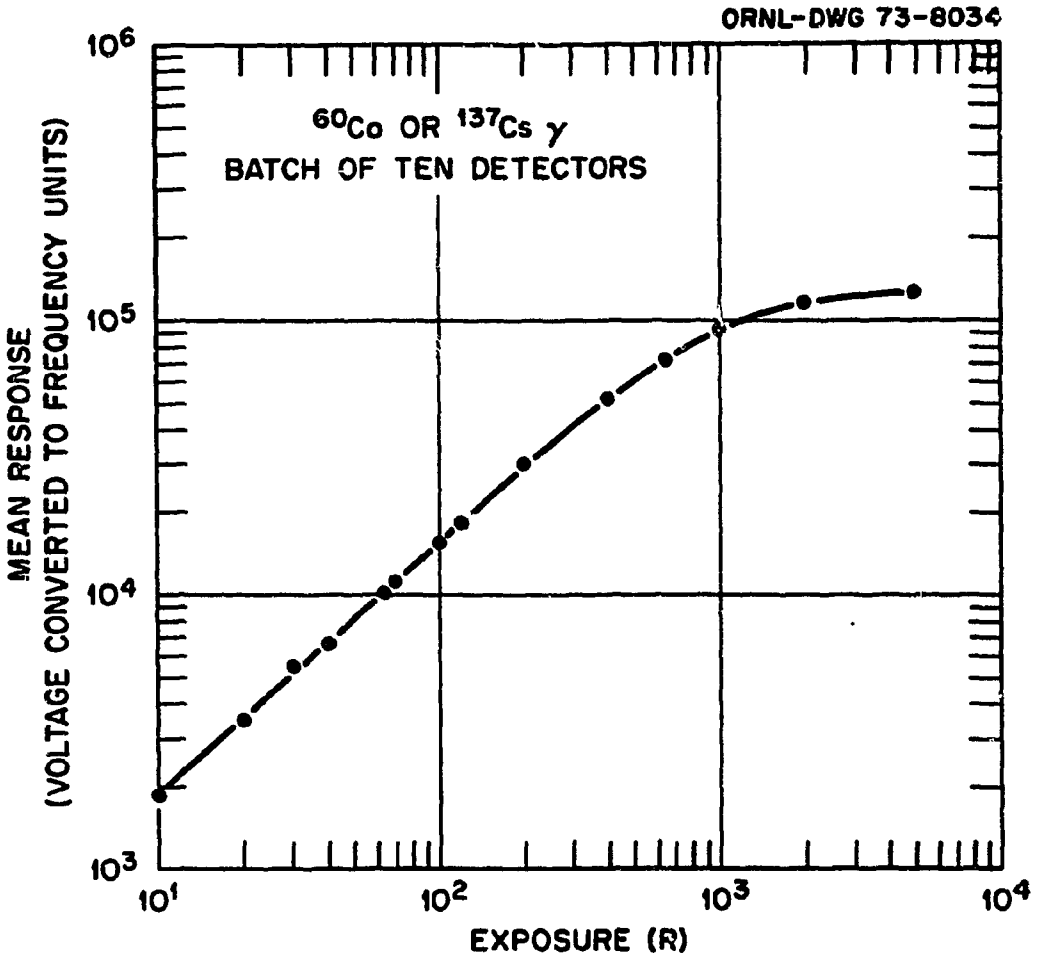
ORNL-DWG 73-8034
^{60}Co OR ^{137}Cs γ
BATCH OF TEN DETECTORS
MEAN RESPONSE
(VOLTAGE CONVERTED TO FREQUENCY UNITS)
10^6
10^5
10^4
10^3
10^1
10^2
10^3
10^4
EXPOSURE (R)

Figure 3

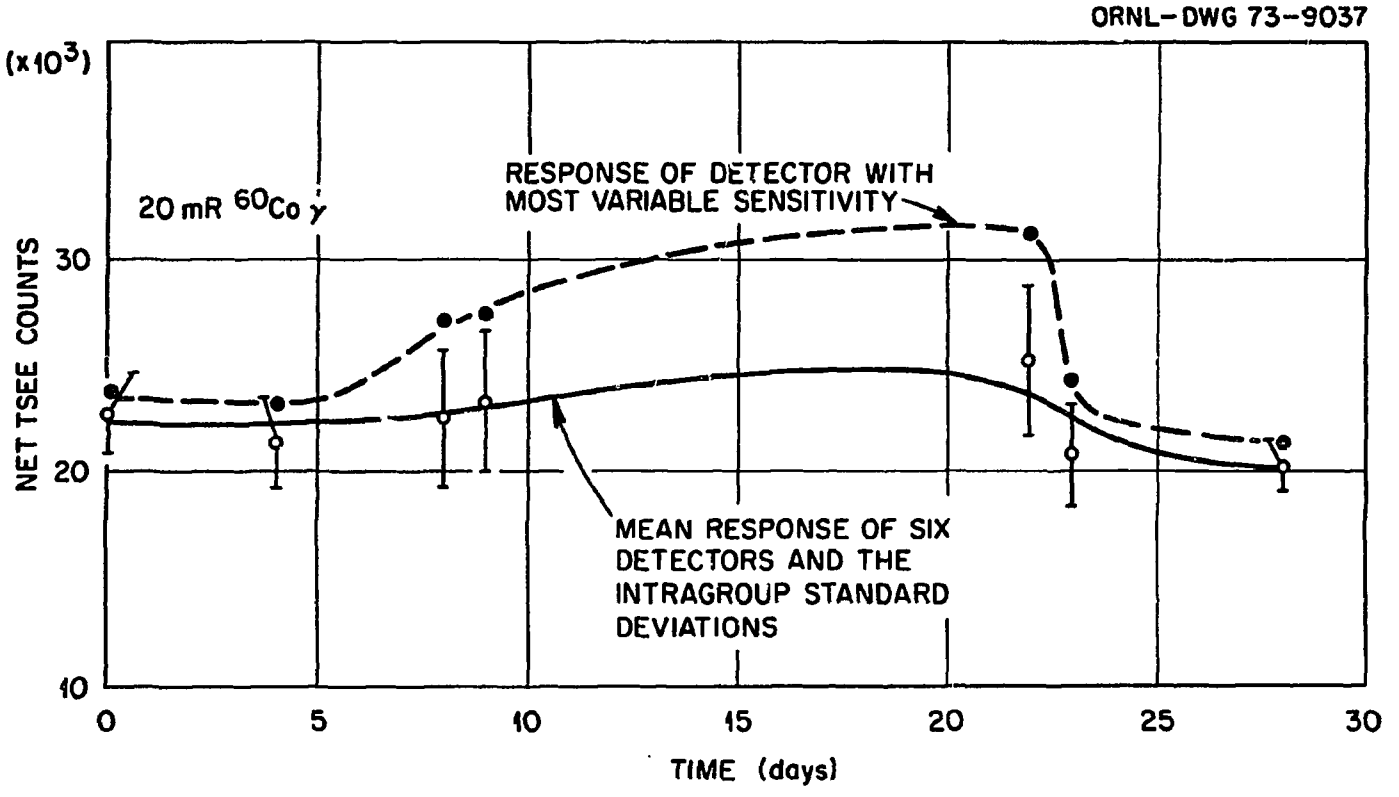
ORNL-DWG 73-9037
(x10³)
NET TSEE COUNTS
30
20
10
0
5
10
15
20
25
30
TIME (days)
20 mR ⁶⁰Co γ
RESPONSE OF DETECTOR WITH MOST VARIABLE SENSITIVITY
MEAN RESPONSE OF SIX DETECTORS AND THE INTRAGROUP STANDARD DEVIATIONS

Figure 4

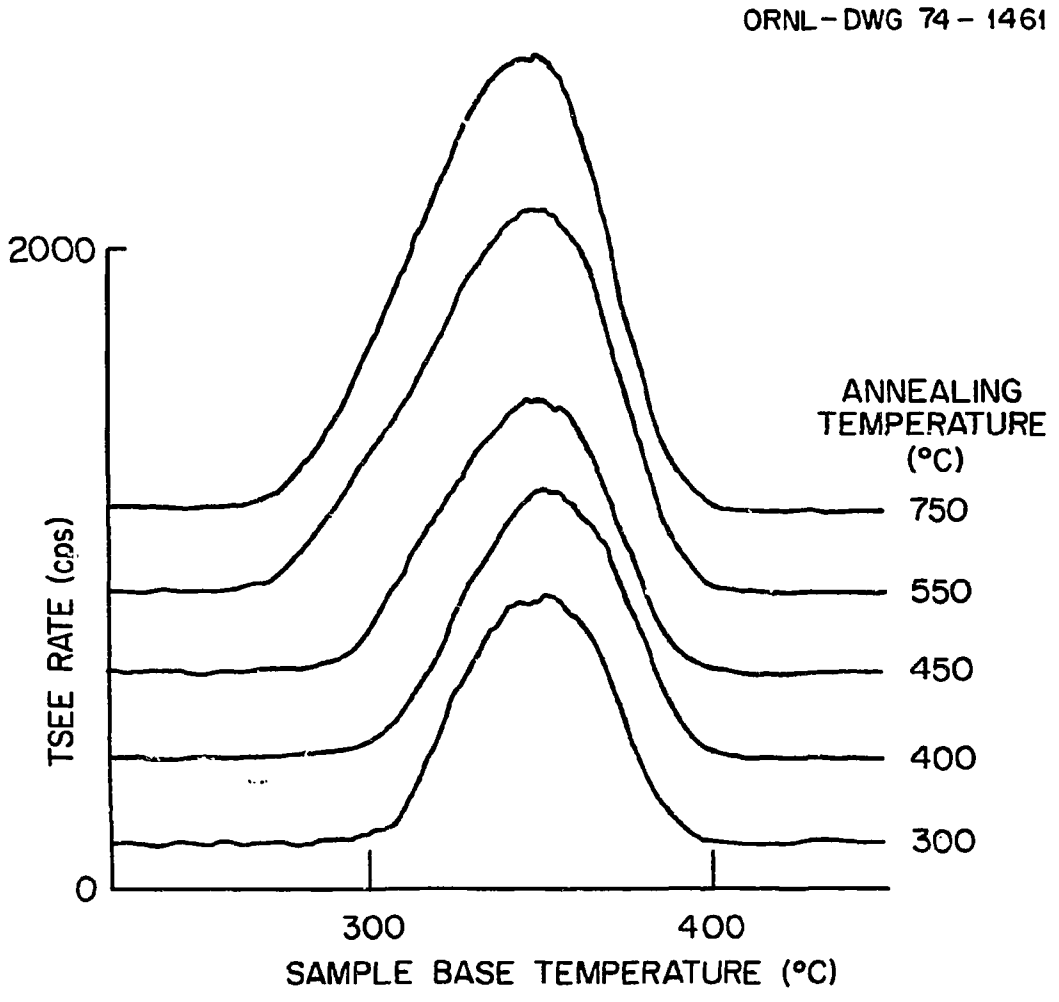
ORNL-DWG 74-1461
2000
0
TSEE RATE (cps)
ANNEALING TEMPERATURE (°C)
750
550
450
400
300
300
400
SAMPLE BASE TEMPERATURE (°C)

Figure 5

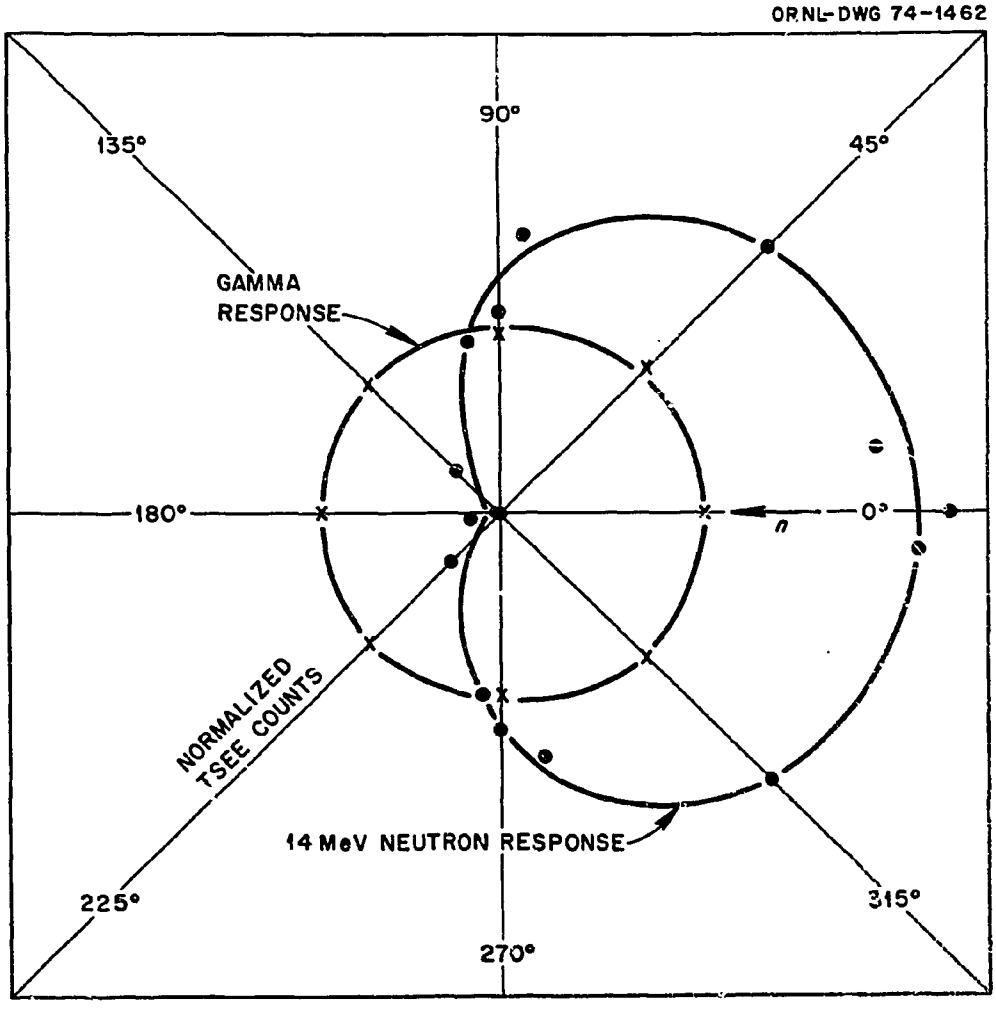
ORNL-DWG 74-1462
90°
135°
45°
GAMMA
RESPONSE
180°
n
0°
NORMALIZED
TSEE COUNTS
14 MeV NEUTRON RESPONSE
225°
315°
270°

Figure 6

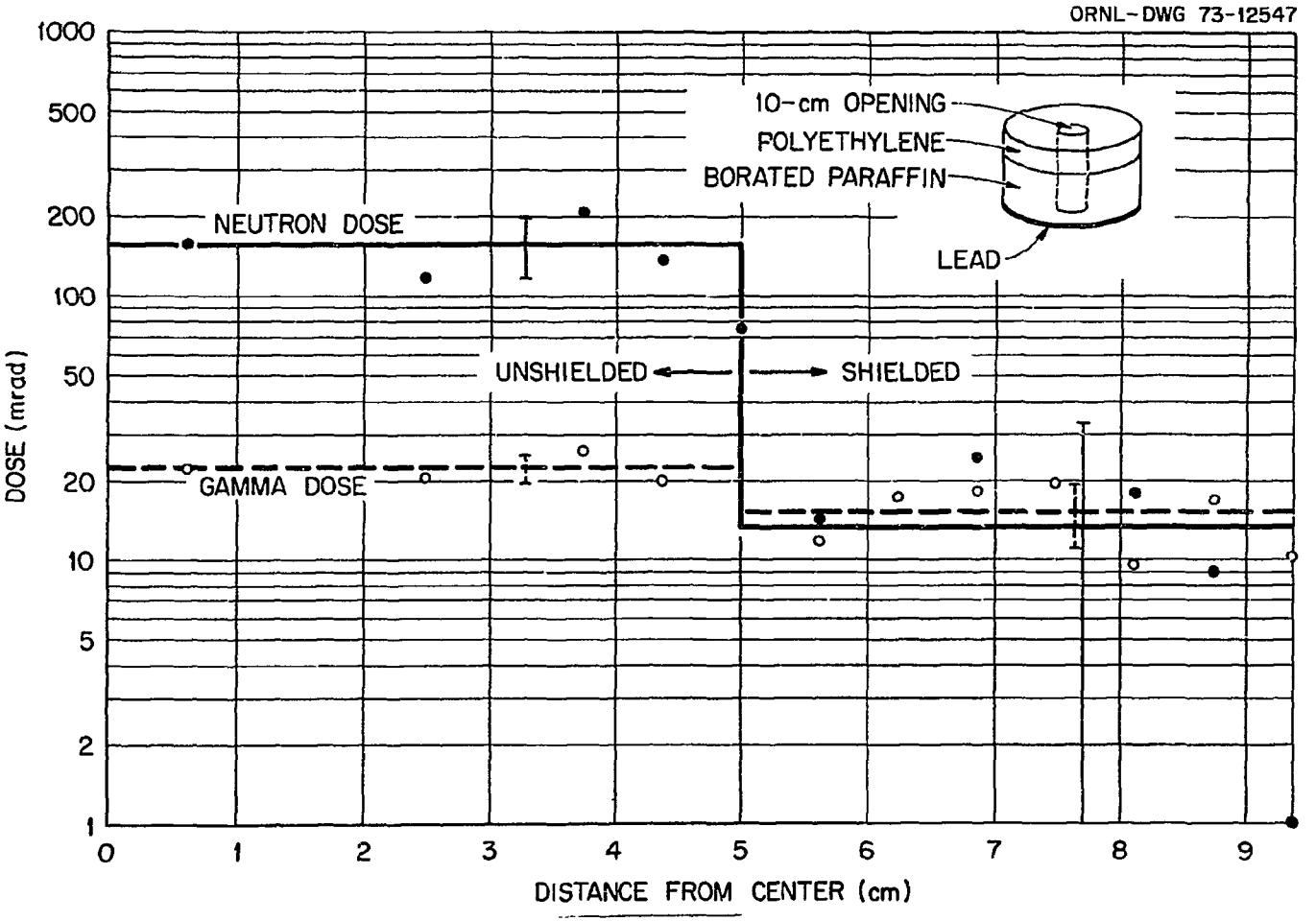
ORNL-DWG 73-12547
1000
500
200
100
50
20
10
5
2
1
DOSE (mrad)
0
1
2
3
4
5
6
7
8
9
DISTANCE FROM CENTER (cm)
10-cm OPENING
POLYETHYLENE
BORATED PARAFFIN
LEAD
NEUTRON DOSE
GAMMA DOSE
UNSHIELDED
SHIELDED

Figure 7

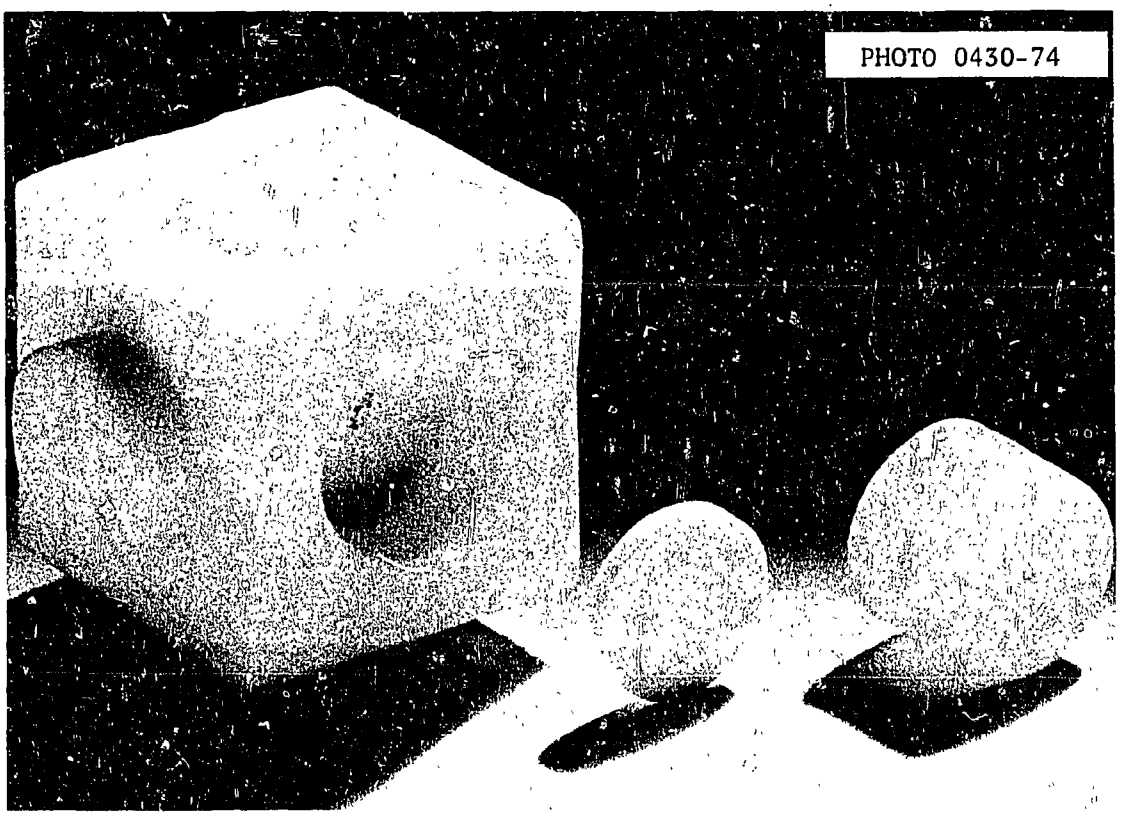

Figure 8

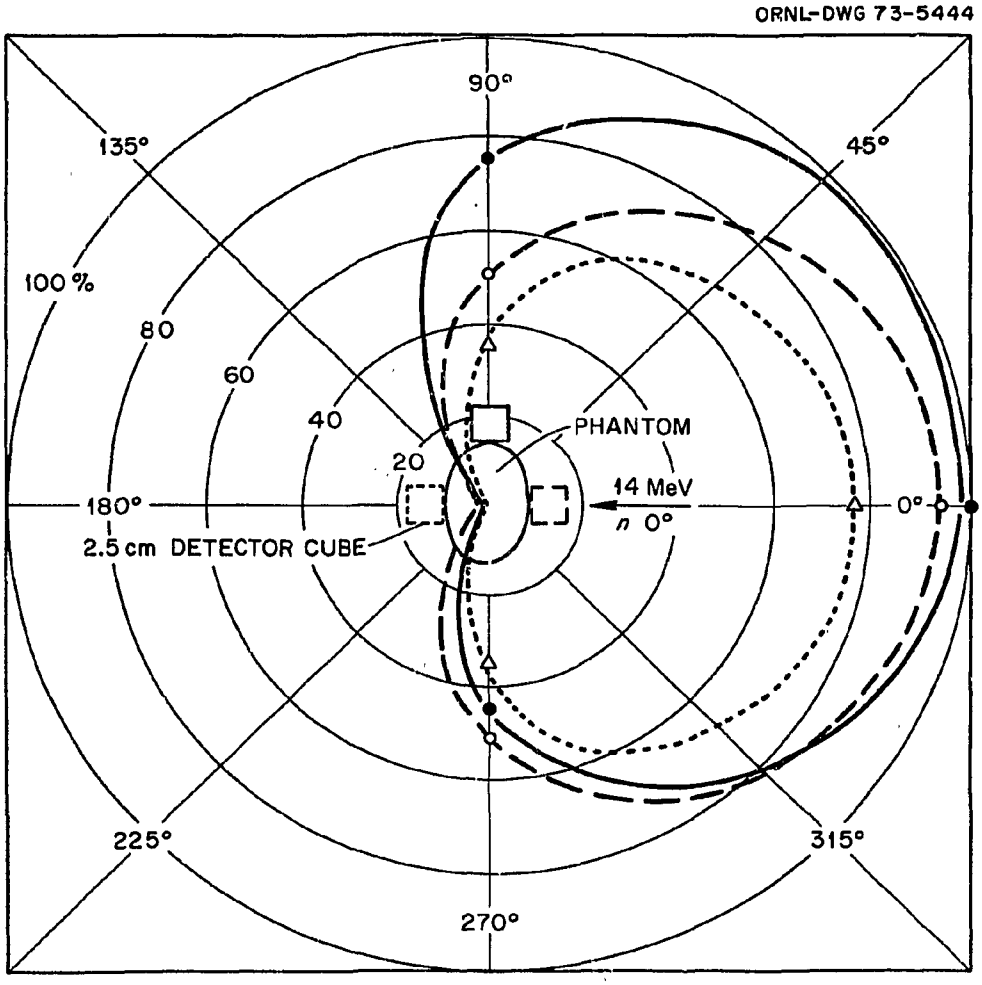
ORNL-DWG 73-5444
90°
135°
45°
100 %
80
60
40
20
PHANTOM
14 MeV
n 0°
180°
0°
2.5 cm DETECTOR CUBE
225°
315°
270°

Figure 9

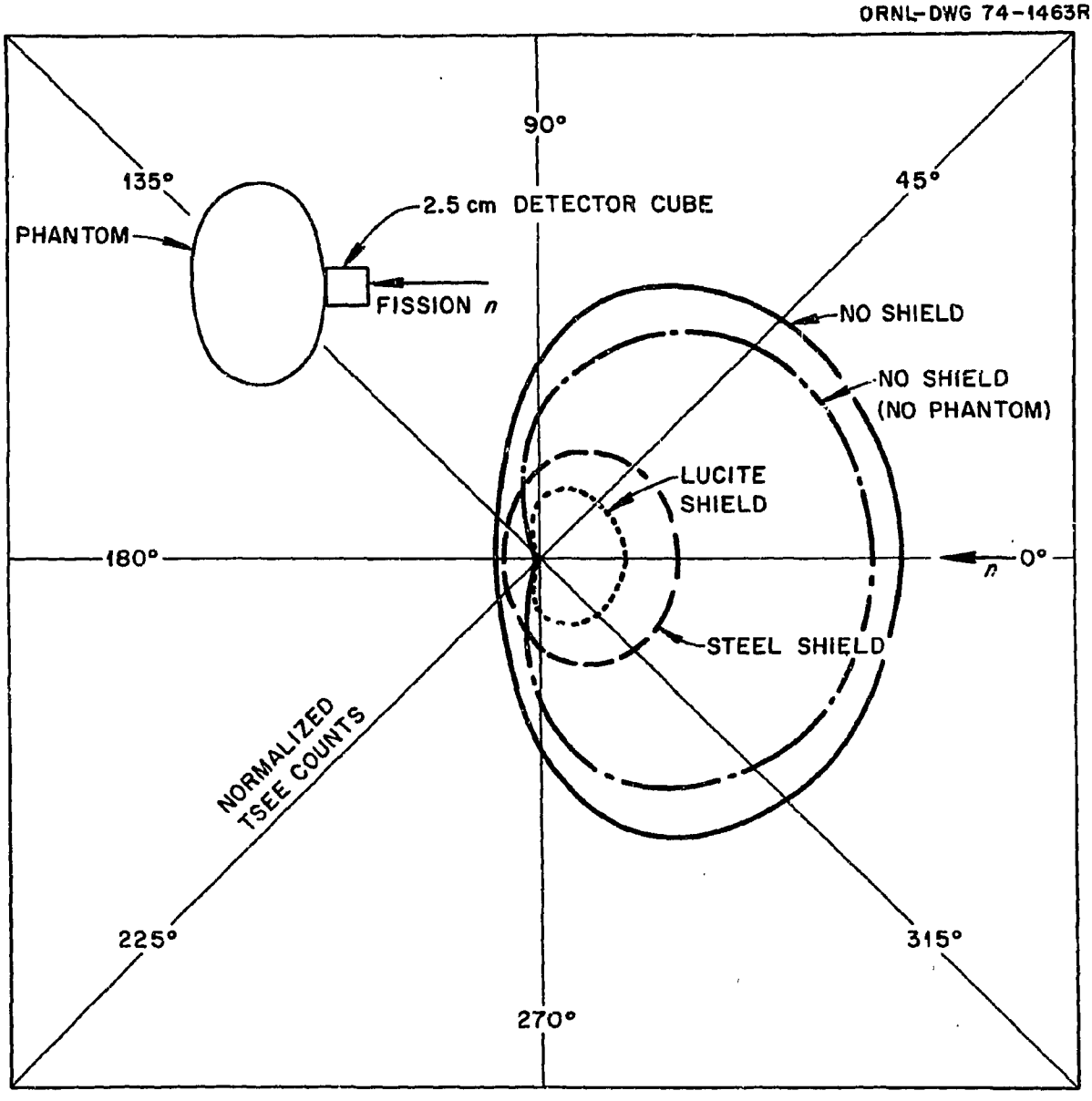
ORNL-DWG 74-1463R
90°
135°
45°
2.5 cm DETECTOR CUBE
PHANTOM
FISSION n
NO SHIELD
NO SHIELD
(NO PHANTOM)
LUCITE
SHIELD
180°
0°
n
STEEL SHIELD
NORMALIZED
TSEE COUNTS
225°
315°
270°

Figure 10