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ORNL/MIT-220

DATE: November 21, 1975 COPY NO.

SUBJECT: Evaluation of BeO Ceramic Disks for Thermally Stimulated Exoelectron Dosimetry

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ABSTRACT

BeO ceramic disks were evaluated for application as dosimeters for both penetrating and non-penetrating radiation. Response to penetrating radiation was measured as thermoluminescence (TL) and to non-penetrating radiation as thermally stimulated exoelectron emission (TSEE). Field experiments demonstrated that both TSEE and TL responses from BeO can monitor diverse radiation fields. BeO disks in a passive dosimeter were found to be sensitive to a lower exposure level of 100 pCi-day/liter of radon. The depth of the more active exoelectron layer in BeO was found to be 4 μm. A second, less active, exoelectron layer extends to a depth of at least 16 μm.

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CONTRACT NO. W-7405-ENG-26

Contents

1. SUMMARY

Beryllium oxide disks were investigated as an environmental source monitor and found to be suitable. The sensitivity of BeO disks as passive radon-222 dosimeter was evaluated as suitable for detecting specified levels of radon-222. The exoelectron layer in BeO was found to consist of a highly active outer surface layer and a less active inner layer. The first layer extends 3 to 4 μm into the surface and the second layer extends beyond 16 μm.

The thermal luminescence (TL) and thermally stimulated exoelectron emission (TSEE) responses of the BeO disks were calibrated with standardized cobalt-60 and cesium-137 sources in the laboratory. Environmental exposures were conducted in a field at varying distances from an environmental pen contaminated with cesium-137 to simulate fallout. Both TL and TSEE produced the expected response of exposure rates varying with distance, with the TL response consistently above the TSEE response, and with the TL-to-TSEE ratio not changing significantly with distance. This may be explained by the changing energy distribution of the photon field and by the geometric placement of the badges relative to the direction of the photon flux.

The sensitivity of the radon-222 dosimeter to an applied chamber voltage was measured with a silicon diode detector. The effect of applied voltage was found to be insignificant. Six beryllium oxide disks were placed in the dosimeter to monitor the effects of various concentrations of radon-222 and were found to have a minimum sensitivity of 100 picocurie/liter for one-day exposure.

The depth of the exoelectron layer in BeO was measured by bombarding the BeO surface with monoenergetic alpha particles. The incident alpha particles from an americium-241 source was moderated with helium at various pressures to provide particles of several energies. The disks were exposed to a constant total energy of incident alpha particles. The TSEE response of exoelectrons/MeV of deposited particle energy decreased sharply at an alpha particle energy of 2 MeV corresponding to a penetration depth of 4 μm and then remained relatively constant below this depth to a distance of 16 μm. The response of an active surface exoelectron layer coupled with a less active inner layer will account for this observation.

Further experiments are needed to determine the effects of the photon energy distribution with distance and of the geometrical arrangement of the badge relative to the photon flux for environmental monitoring. Design changes capable of increasing the sensitivity of the radon dosimeter by a factor of 2 to 5 should be investigated. The depth of the inner exoelectron layer should be measured more precisely. One possible method would be irradiation from the back side of a thin disk with high energy alpha particles.

2. INTRODUCTION

2.1 Background

Ceramic beryllium oxide disks are valuable for radiation dosimetry. Irradiated BeO disks will, upon heating, emit exoelectrons from a thin surface region, or emit photons from within the BeO bulk volume. The first property is known as thermally stimulated exoelectron emission (TSEE), and the second is known as thermoluminescence (TL). Non-penetrating radiation, such as an alpha particle, deposits energy in the surface region of a solid; the TSEE response can monitor the exposure to this radiation. Penetrating radiation, such as a gamma ray, ionizes the BeO disk through the entire volume and can be monitored with TL. These properties form a basis for developing a passive, solid state, integrating dosimeter which can measure and discriminate between penetrating and non-penetrating radiations.

The Health Physics Division at Oak Ridge National Laboratory has an ongoing program of research and evaluation of BeO properties which are suitable for dosimetric applications. Gammage (5) has performed several experiments with calibrated BeO disks near an outdoor cesium-137 field and found a substantial difference in the TL/TSEE ratio in the field tests compared to the laboratory calibration (1.89 to 1.0). This high ratio that was observed in the field required an explanation and was a basis for current experiments. Gammage, Huskey, and Kerr (4) made an exploratory study of BeO disks for monitoring radon-222 and recommended that a passive dosimeter be designed and tested for practical monitoring. As little is known about how exoelectrons occur, the layer in which the exoelectrons originate has been believed to be less than 100 Å deep (7). This depth can be approximately measured using alpha particles of different energies.

2.2 Objectives and Method of Attack

The objectives of this study were to determine the depth of the exoelectron layer and to evaluate BeO disks for dosimetry in low energy photon fields and in a passive radon dosimeter. The method of attack begins with a calibration of the BeO disks to radiation from standardized cobalt-60 and cesium-137 sources. The BeO disks are placed at various distances from a cesium-137 field source and the TL and TSEE responses evaluated. The passive radon-222 dosimeter employing six BeO disks was placed in a radon atmosphere and the sensitivity of the dosimeter determined. The depth of the exoelectron layer for TSEE was measured with an americium-241 alpha source and with different helium pressures to vary the incident alpha particle energy.

The results from the experiments were analyzed to obtain: (1) the calibrated TL and TSEE responses of BeO disks, (2) the apparent TL and TSEE exposure rates at each field position, (3) the TL/TSEE ratio in the field compared to the calibrated source, (4) the optimum operating conditions and sensitivity of the passive radon dosimeter, and (5) the depth of the exoelectron layer in BeO disks based on the TSEE response to different alpha particle energies. The conclusions and recommendations are based on the results obtained.

3. THEORY

Dosimetry is the measurement of exposure rates and total exposure to different types of radiation. Radioactive sources emit non-penetrating radiation such as alpha and beta particles and emit penetrating radiation such as gamma rays. The activity of a source in curies is the number of disintegrations per second in a one-gram mass divided by 3.7 x 10^{10}. Radiation by charged particles is measured by the energy per particle (MeV) and by the number of particles. Gamma photons are measured by the energy per photon quanta (MeV). Charged heavy particles travel only short distances and are easily stopped. Present instruments use ionization effects in gases and solids to determine the energy of radiation and to count the number of particles. Two examples are Geiger counters which measure the ionization from particles traveling through a gas or from photons and a semi-conductor diode which monitors the total amount of ionization produced by charged particles and the number of increments as produced by each incident particle.

Beryllium oxide (BeO) interacts with radiation and traps electrons and holes where ionization has occurred. After irradiation, the beryllium oxide exhibits two effects when heated. Trapped electrons near the surface leave as exoelectrons. This phenomenon is known as thermally stimulated exoelectron emission. Trapped electrons in the bulk volume are released and recombine with positive holes to emit photons. This is known as thermoluminescence. Penetrating radiation affects the entire mass of a BeO ceramic disk and hence, both the TL and TSEE responses. Non-penetrating radiation deposits its energy near the surface of the disk. The TSEE response can monitor this radiation provided the particles stop in a surface active layer. As the depth of this layer of exoelectrons is not well known, applying the TSEE response to monitoring radiation requires further investigation. Energy dependence of both TL and TSEE responses have been reported with appreciable divergence noted at energies below 200 MeV (5). Alpha particles deposit most of their energies at a specified depth, losing energy at a constant rate to this point (8). This property can be used to determine the exoelectron layer depth and an upper limit on the alpha particle energy to be monitored.

Beck (1) has reported that Compton scattering of gamma radiation at large distances lowers the gamma energy 10-15% and increases the spread (dispersion) of the individual energies. Gammage (5) has reported that the TL is enhanced over the TSEE at low energies. Thus, the ratio of TL-to-TSEE can be used to determine the changing energy of the gamma radiation at different distances from a source. In an outdoor environment, natural ground radiation and sky radiation are composed primarily of multiple scattered low energy photons and may also affect dosimeter response.

Another possible application of BeO is the monitoring of activity levels of radon-222 gas. Radon-222 decays by alpha emission (5.49 MeV). The radon daughter products decay by both beta and alpha emission, alpha particles being the primary emission (see Appendix 10.1 for the important decay products). After sampling a radon atmosphere, the activity level of radon-222 can be monitored by measuring the activity of the radon-222 decay products after a time of five half-lives of the daughter products.

4. APPARATUS AND PROCEDURE

4.1 Calibration of BeO Disks with ^{137}Cs and ^{60}Co

The 12.5-mm-diam, 1.5-mm-thick BeO ceramic disks from Brush Beryllium Co., Elmore, Ohio, were mounted in a Lucite rack housed in a light proof ORNL film badge, six disks per badge. Twenty-four disks were exposed and calibrated with cesium-137 and cobalt-60 for use in the subsequent field experiments. Each badge was exposed to 20 mR of gamma radiation at a distance of 20 cm from the standard source - 110 sec exposure time for a 0.10 mCi ^{60}Co source and 8 min:36 sec for a 0.022 mCi ^{137}Cs source. The TL and TSEE responses were calibrated.

The thermoluminescence analysis equipment consisted of a Radiation Detection Co. TLD Reader and an x-y recorder. The photomultiplier voltage was set at 1100 V and the nitrogen gas flow at 2 ft^3/hr. The horizontal axis of the recorder was set at a constant speed of 1 cm/sec. The vertical axis monitors the change in voltage of the photomultiplier tube at a 50 mV/cm sensitivity. Each disk is placed in a pan underneath the photomultiplier tube and heated at a rate of 15 °C/sec. The TL response is plotted by the recorder. As the peak maximum is passed, the heating is stopped and the disk removed from the unit. This process is necessary to reduce fading of the TSEE peak reading which begins immediately after the TL. The disk is allowed to cool to room temperature on a brass plate before placing it back into a badge.

After the analysis for TL, the disks are measured by a TSEE counter. This apparatus has a thermocouple-controlled heating unit, a gas flow counter, and an x-y plotter, as shown in Fig. 1.

The voltage applied to the GM tube HV wire is 1400 V. A helium (99.05%) and isobutane (0.95%) mixture is fed to the counting chamber at a rate of 2 ft^3/hr. Each disk is placed on the heating plate underneath the counter and heated at a rate of 2°C/sec. Counting is started as the temperature reaches 250°C and stopped at 450°C. The heating is continued to 500°C, then stopped, and the apparatus is cooled down. The disk is removed from the pan after the temperature has dropped below 275°C. The integral TSEE count is read from the ratemeter-scaler.

The process of exposing the badges to ^{60}Co and ^{137}Cs sources and analyzing each exposed disk for TL and TSEE was repeated three times to gain statistical confidence in the results and to check reproducibility.

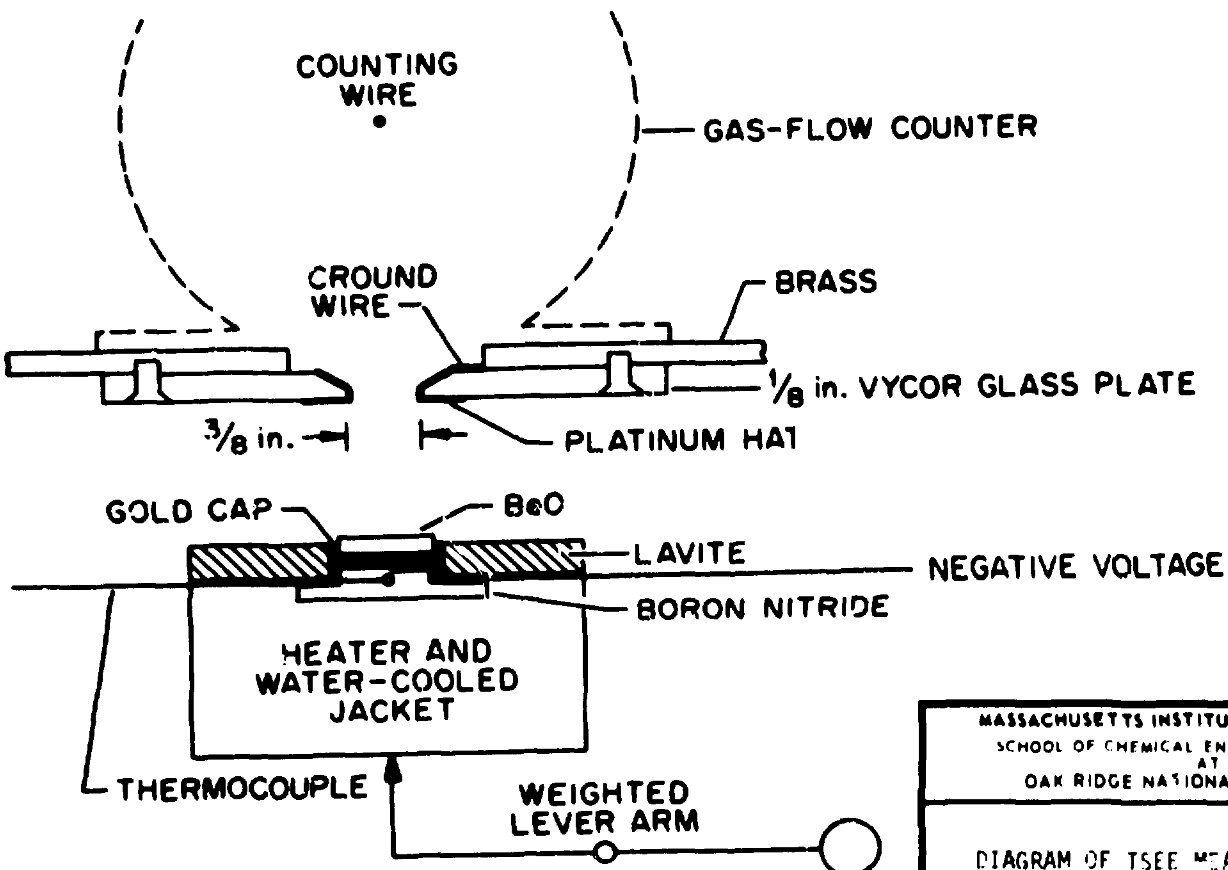

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DIAGRAM OF TSEE MEASURING EQUIPMENT
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4.2 ^{137}Cs Contaminated Field Test

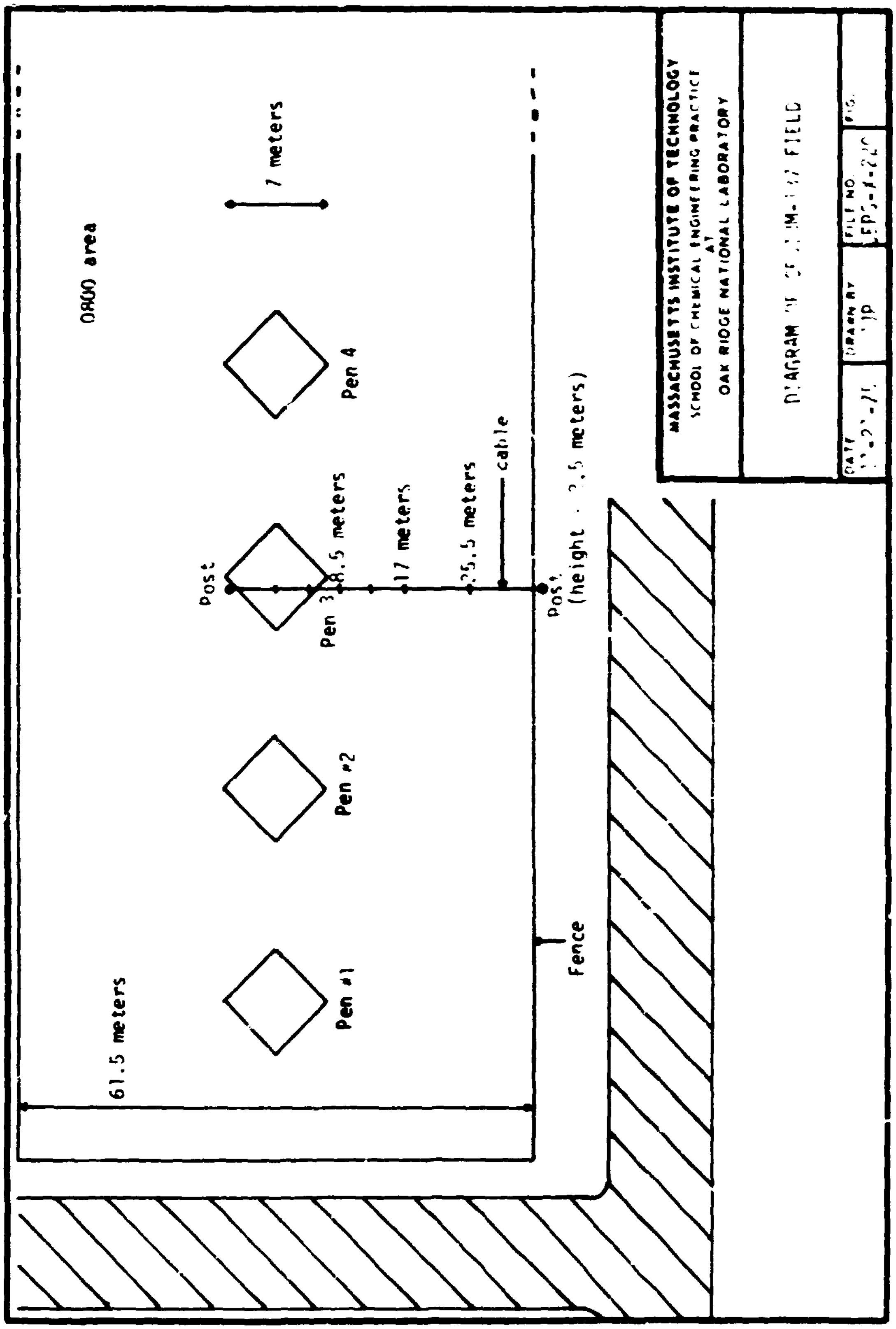
In this experiment, two badges (#1 and #4) were suspended horizontally above the ground in the 0800 area at various distances between the center of the #3 pen and the fence surrounding the 0800 area as shown in Fig. 2. The two badges were taped to the inside surface of a plastic bag; five ounces of silica gel granules were put into each plastic bag to prevent moisture from reaching the surface of the disks. The bag was then heat sealed and taped in an aluminum tray so that the faces of the badges coincided with rectangular holes cut into the bottom of the tray as shown in Fig. 3. A shaped sheet of lead foil was then taped above badge #1.

The basket containing the badges was then hooked on a steel cable stretched between two pulleys mounted on top of two metal poles. One pole was planted on the far side of the pen and the other was planted outside the fence. The bottom of the basket was 1.5 meters above ground level. The basket was suspended at six positions: over the pen, 2.1, 4.3, 8.5, 12.8, 17.1, and 23 meters from the center of the pen to the edge of the field. The basket was suspended for one hour over the pen and in the 2.1 and 4.3 meter positions. The basket was left overnight (15 hr) for the 8.5 and the 12.8 meter positions, and over the weekend (65 hr) for the 17.1 and 23 meter positions.

4.3 Passive Radon-222 Dosimeter

Evaluation of the radon dosimeter is a two-step process. The first is a calibration and evaluation of the design geometry with a silicon diode detector. The second step involves exposing the BeO disks in the dosimeter to the radon-222. The passive dosimeter has a 4-in. air filter with a metal plate on top and the collection area underneath as shown in Fig. 4. The diode is covered with a 0.015-in.-thick aluminized mylar film. The aluminized side faces the radon chamber and is grounded to a high voltage power supply. The dosimeter fits inside a glove box containing 23 lb of uranium ore. Radon-222 gas is released from the ore and accumulates to an appreciable level over a period of 3-4 days. A sample concentration is monitored with a Lucas-Chamber (0.095 liters) and a Low Level Radiation Counting System Model LLR-2. This instrument measures the activity of the Radon-222 and the decay products over a period of time. A sample is taken at the beginning and end of each set of measurements. To measure the activity of the daughter products, the diode is biased between 25 and 100 V. The signal produced by incident alpha particles goes through a pre-amplifier and an amplifier into an ND-110 multi-channel analyzer. This instrument counts the number of alpha particles over a wide energy range and displays the output on an ND-410 monitor or can print the output on paper tape. The counts are usually timed over a 15-min interval by a stopwatch.

The voltage required for optimum operation results from a set of measurements taken with the chamber high voltage ranging from 0 to 3000 V.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FIELD
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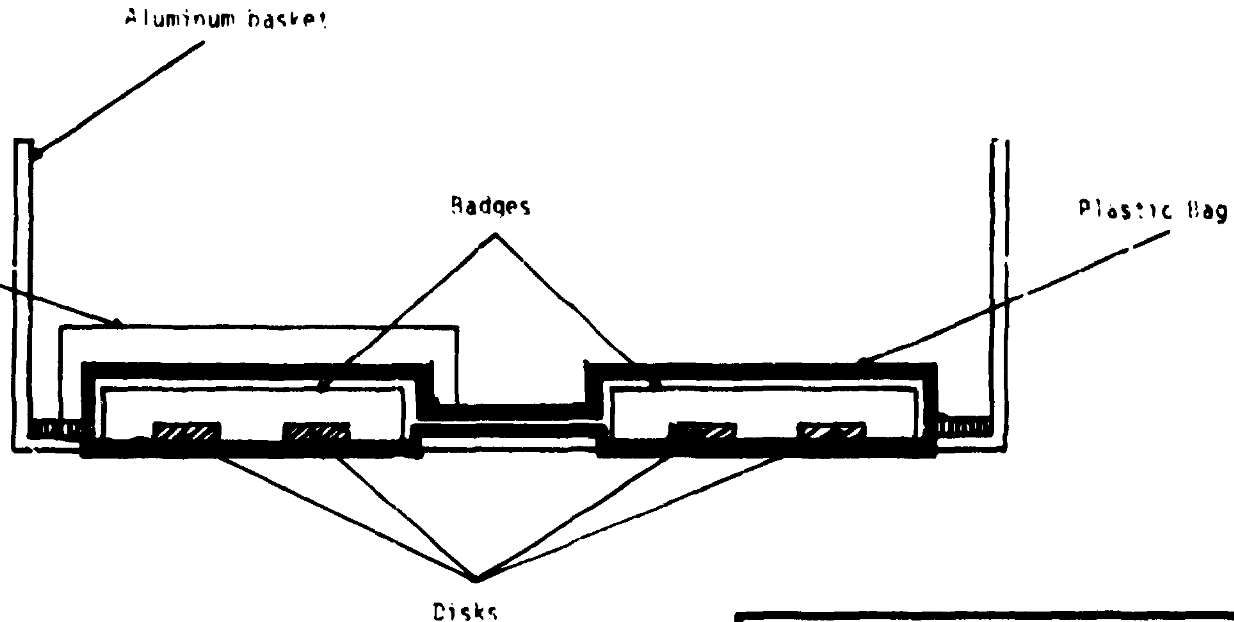
FIG.
ORNL area
7 meters
Pen 4
Post
Pen 3
8.5 meters
17 meters
25.5 meters
cable
Post
Pen #2
Pen #1
Fence
61.5 meters



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BASKET ARRANGEMENT FOR ^{137}Cs FIELD EXPERIMENT			
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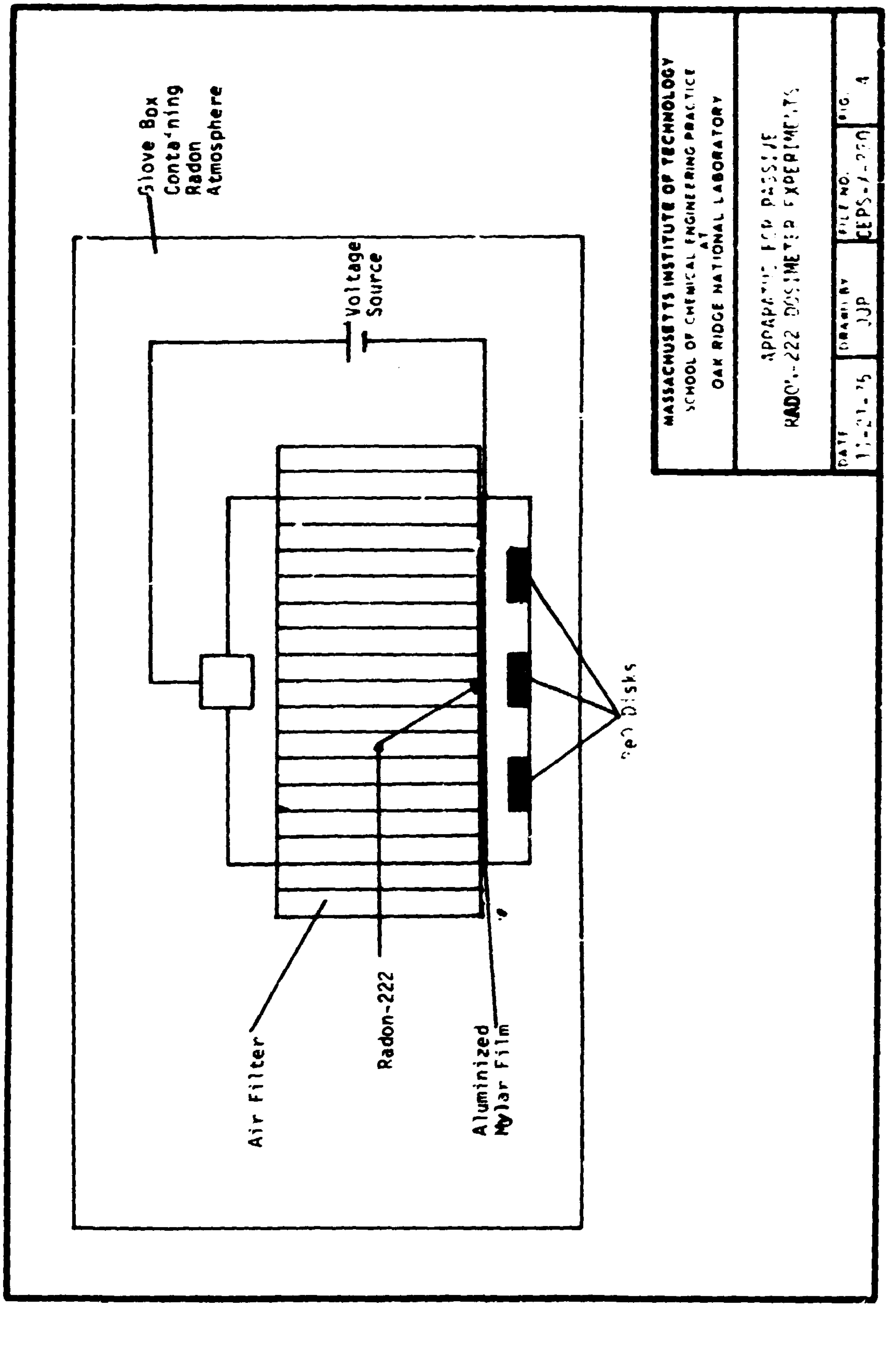
Glove Box Containing Radon Atmosphere
Voltage Source
Air Filter
Radon-222
Aluminized Mylar Film
Disks
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DOSIMETER EXPERIMENTS
DATE
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FIG.
4

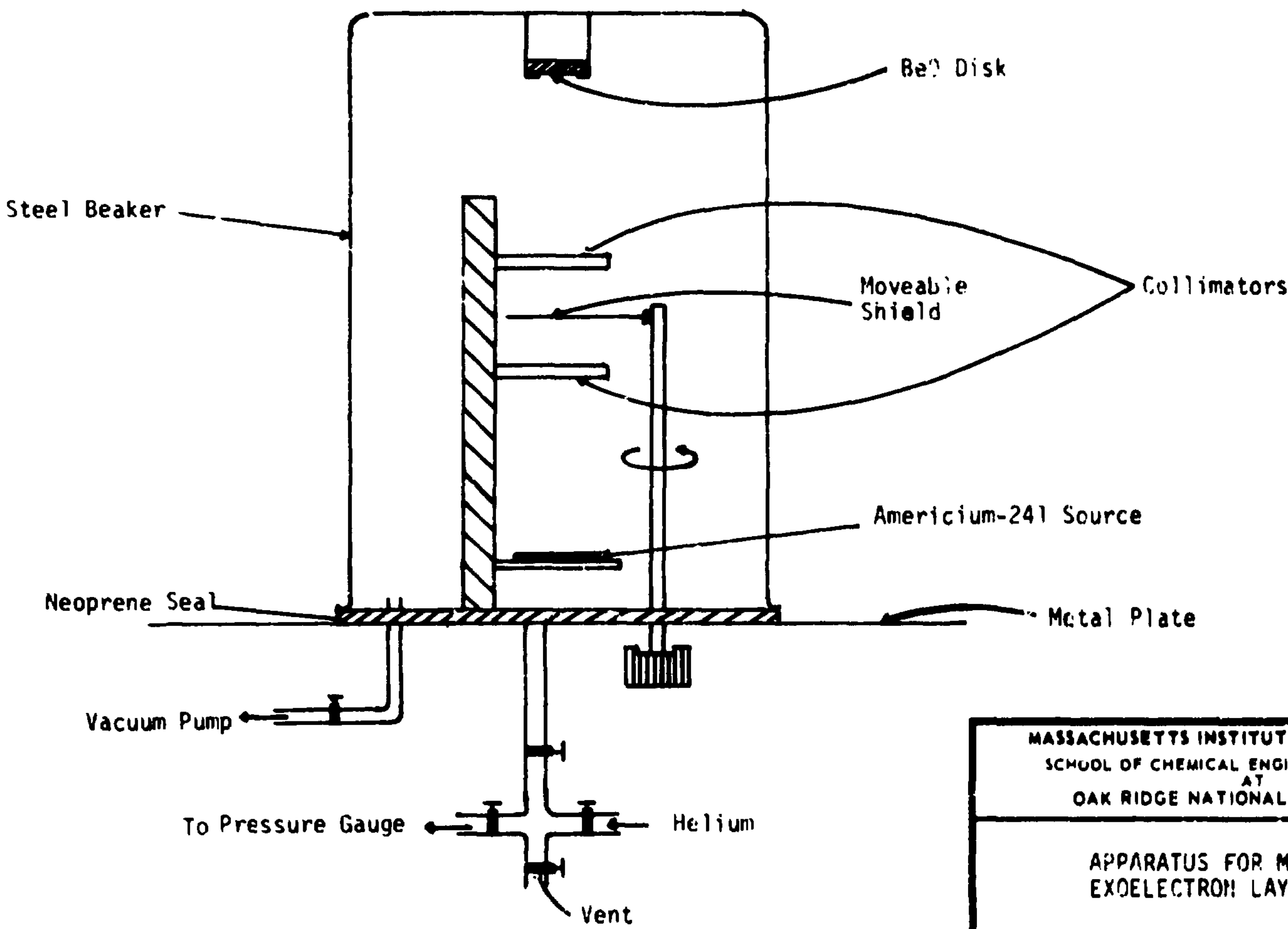
After setting each applied voltage, 15 min was allowed before counting the diode output to allow the first daughter product, ^{218}Po, to deposit on the Mylar film. The recorded activity is a measure of the sensitivity and efficiency of the design.

After the applied voltage was determined to have negligible effect using the silicon diode, the BeO disks were placed in the dosimeter and covered with aluminized Mylar film. A 300-V battery was connected across the chamber - negative to the aluminized film and positive to the top plate. The uranium ore was removed from the glove box. The dosimeter and battery were placed in the box containing radon-222 and a sample of the atmosphere was taken and counted using the Lucas chamber in the LLR-2. The dosimeter was left overnight for a timed period, removed, and the TL and TSEE response measured. The procedure was repeated for multiple measurements. Radon-222 concentrations were measured at the end and beginning of each timed exposure.

4.4 Measurement of Exoelectron Layer Depth

BeO disks were exposed to americium-241 alpha particles of varying energies. The energy of the 5.4 MeV alpha particle was attenuated by varying the helium pressure between the americium source and the BeO disks. The equipment for this experiment, shown in Fig. 5, consisted of a post with a platform for the americium source, a moveable shield to control the exposure time, and a series of collimators to restrict the width of the alpha particle beam. The BeO disk is held in a plastic tube which is attached to the inside center of a metal beaker that covers the whole apparatus.

To calibrate the equipment, a silicon diode was placed in the top of the metal chamber. The total energy of 1000 incident alpha particles was counted using the diode at different pressures of helium. The time necessary to achieve a count of 1000 particles was noted. Subsequent exposures of BeO disks were also for this same time and, hence, the same number of events. After the disk had been placed inside the metal beaker, the beaker was bolted to the metal plate and over the post with the ^{241}Am source. The beaker was evacuated, refilled with the helium, and re-evacuated. The beaker was then filled with helium to the pressure appropriate to the particular test. The test was carried out by removing the shield from between the ^{241}Am source and the target disk for a pre-calculated time. The beaker was brought back to atmospheric pressure by venting. The BeO disk was analyzed on the TSEE counting equipment in the same manner as that described in Sect. 4.1.



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APPARATUS FOR MEASURING
EXOELECTRON LAYER DEPTH

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5. RESULTS

5.1 Calibration and Field Tests

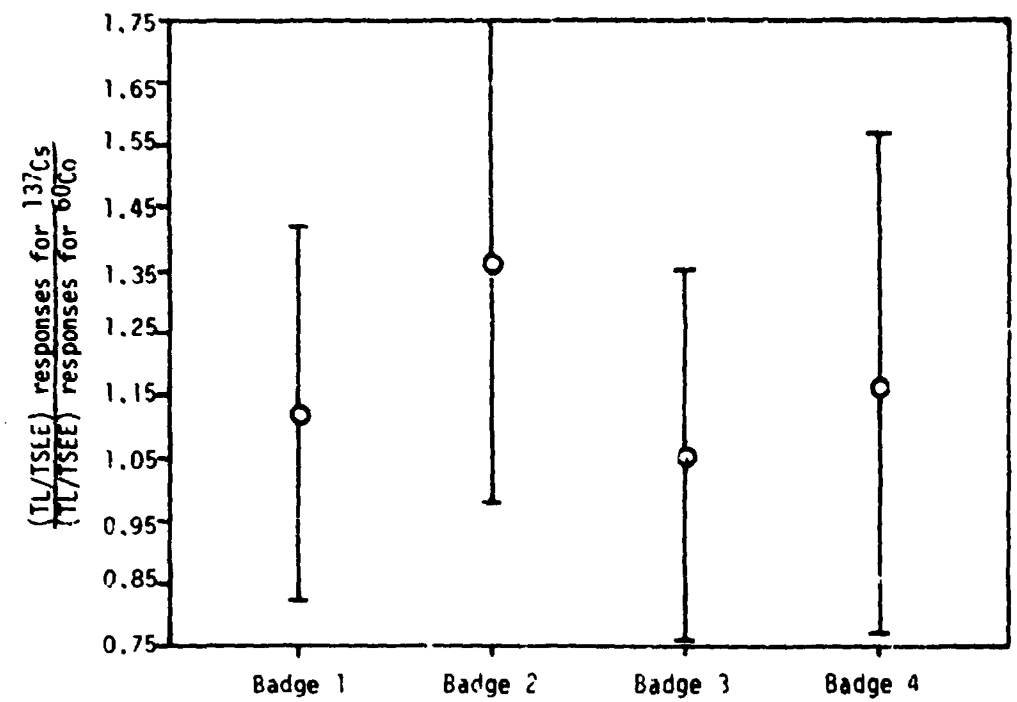
The purpose of the calibration was to determine the TL and TSEE response of each set of BeO disks at 20 mR from a standardized source. Exposures were then measured in the environmental ^{137}Cs field based on the TL and TSEE sensitivities/mR in the standard ^{60}Co and ^{137}Cs photon fluxes. Two sources of gamma radiation, ^{60}Co (0.10 mCi) and ^{137}Cs (0.022 mCi), were used as standards. Each badge was exposed at least three times to each exposure, and the TL and TSEE responses measured. The mean TL and TSEE responses and their standard deviations were then calculated. These results are presented in Table 1. The TL response to ^{137}Cs is enhanced above the TL response to the same exposure from ^{60}Co. The TSEE responses from each source are equal within experimental error. To quantify this effect, the TL-to-TSEE ratio for ^{137}Cs was compared to the TL-to-TSEE ratio for ^{60}Co with associated error limits and is shown in Fig. 6.

Table 1. Average Response of Badges* Containing Six BeO Disks to 20 mR from Calibrated ^{60}Co and ^{137}Cs

Badge	TL (^{60}Co) (mV)	TL (^{137}Cs) (mV)	TSEE (^{60}Co) (electrons)	TSEE (^{137}Cs) (electrons)
1	260.9 ± 31.0	295.7 ± 23.5	12,455 ± 1312	12,574 ± 2510
2	252.9 ± 15.9	298.3 ± 19.4	14,318 ± 1912	12,353 ± 2922
3	279.6 ± 43.9	308.2 ± 29.6	15,220 ± 1955	15,894 ± 2843
4	368.3 ± 56.5	423.7 ± 81.8	16,350 ± 2521	16,094 ± 3782

*Each response is the mean value ± one standard deviation.

After calibration the BeO disks were exposed at various distances from a cesium-137 field source under natural climatic conditions as described in Sect. 4.2. This experiment was designed to measure the effect of a natural radiation field on the TL and TSEE responses of the BeO disks. One of the two badges was covered with a lead shield to determine the effect of sky radiation on the responses. The apparent exposure rate determined at various distances from the cesium-137 pen is presented in Fig. 7. The ratio of TL to TSEE in the field was compared to the calibrated laboratory ratio and is plotted as a function of distance in Fig. 8.



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TL TO TSEE RESPONSE RATIO FOR ^{137}CS NORMALIZED TO ^{60}CO

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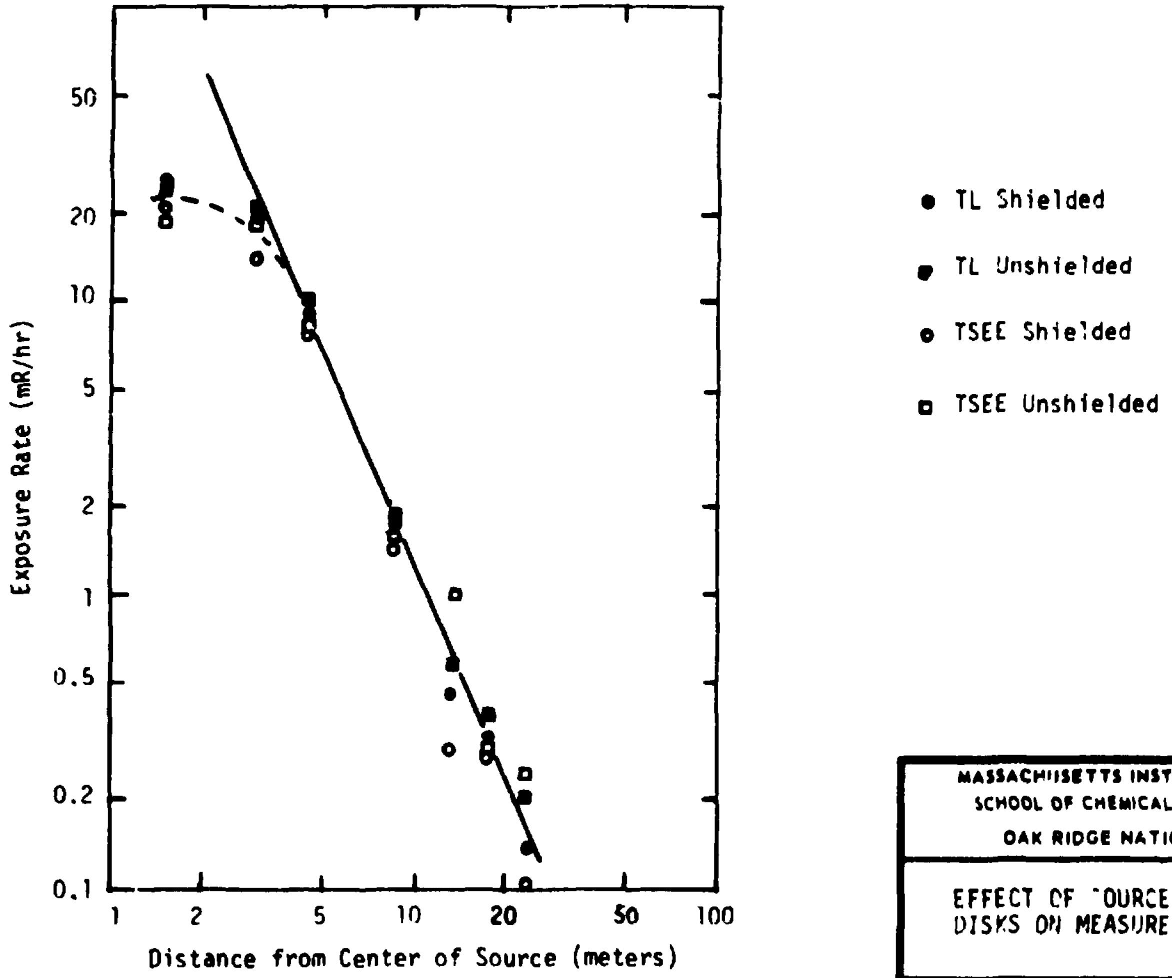

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EFFECT OF SOURCE DISTANCE FROM BeO DISKS ON MEASURED EXPOSURE RATES

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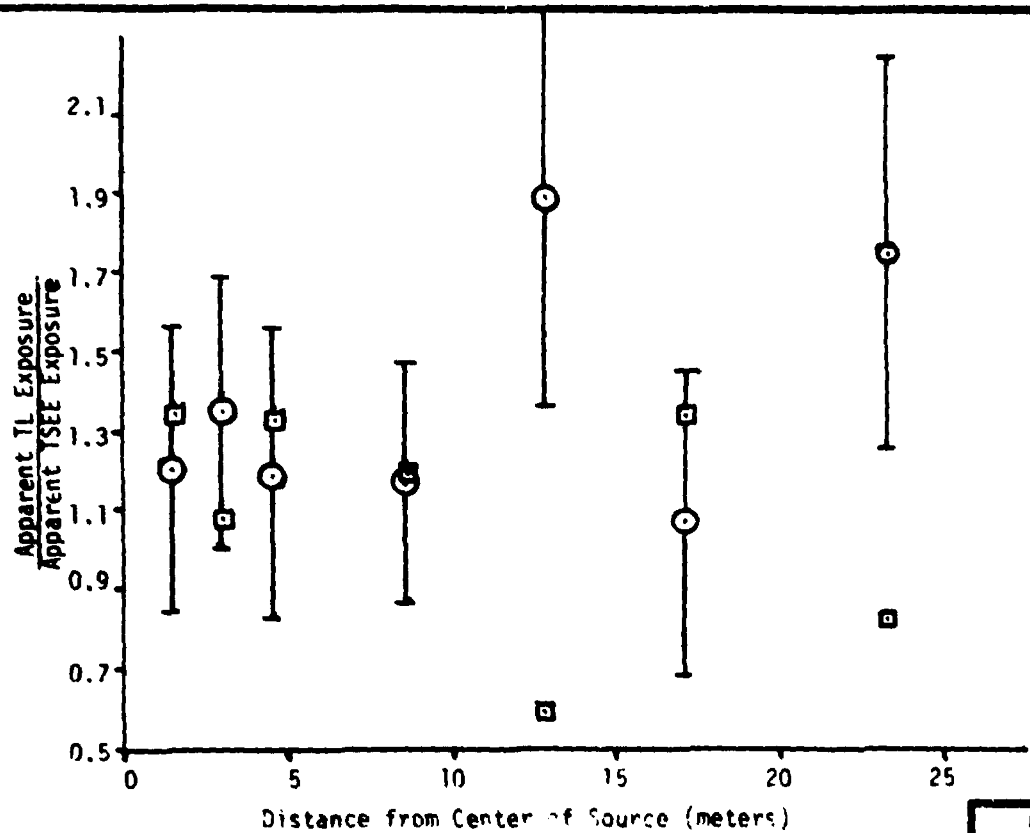

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EFFECT OF DISTANCE ON
TL/TSEE RESPONSE RATIO

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11-21-75	JRB	CEPS-X-220	8

5.2 Passive Radon-222 Dosimeter

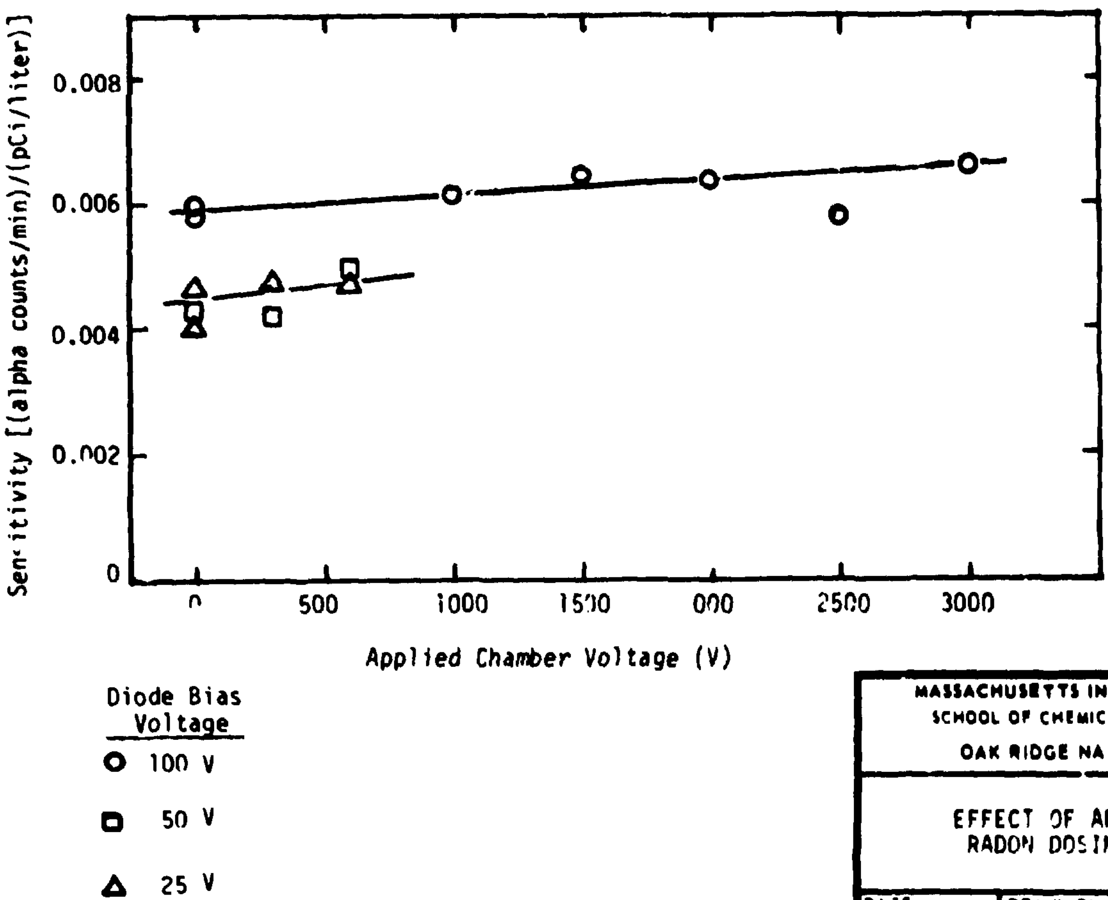
The passive radon-222 dosimeter was assembled with a silicon diode detector (surface area of 1.23 cm^2) to determine an optimum chamber voltage for the deposition of the radon-222 decay products. The silicon diode was connected to a multi-channel analyzer which sorts the diode output into 127 channels covering a range of alpha energies from 0 to 10 MeV. Each channel counts the number of alpha particles in a particular energy range. The final channel output after 15 min is printed out on paper tape and the total counts added. Increasing the bias voltage on the diode increases the sensitivity to lower energy particles. The measurements with the diode at different bias voltages and at different voltages applied to the chamber are shown in Fig. 9. These diode measurements result in a negligible effect of the applied chamber voltage to the alpha activity. Based on this result and past work, a 300-V battery was applied to the dosimeter for testing the BeO disks and the results are shown in Fig. 10.

5.3 Exoelectron Layer Experiment

The amounts of energy associated with 1000 incident alpha particles deposited in a diode mounted in place of the BeO disk and the times required for this deposition of alpha particles on the silicon detector are given in Table 2.

Table 2. Calibration of Alpha Particle Energies to Helium Pressures

Helium Pressure (mm Hg)	Total Incident Alpha Energy (MeV)	Time (sec)
~0	5.48×10^3	377.1
150	4.99×10^3	388.2
300	4.52×10^3	394.3
450	4.03×10^3	381.6
600	3.51×10^3	374.3
750	2.90×10^3	386.1
900	2.20×10^3	397.3
1050	1.18×10^3	393.8



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EFFECT OF APPLIED VOLTAGE ON RADON DOSIMETER SENSITIVITY

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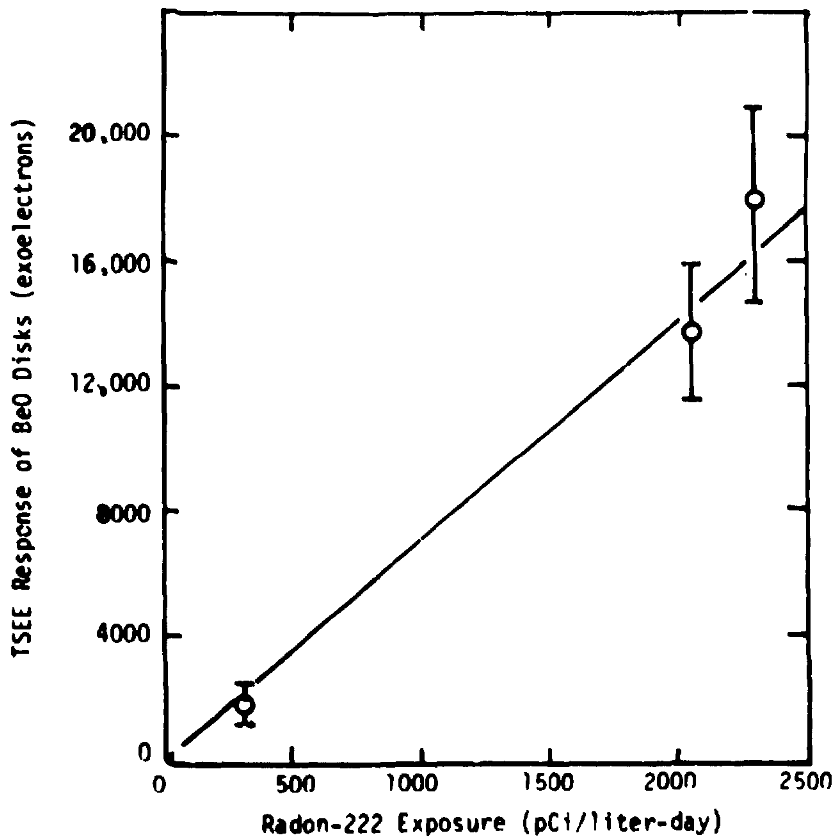

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TSEE RESPONSE OF BeO
TO RADON-222 EXPOSURE

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The total amount of energy absorbed by the diode at a particular helium pressure is given by the total energy deposited in the diode at full vacuum, multiplied by the ratio of the diode outputs at each helium pressure. The exposure time of the disks is calculated from the data in Table 2 to expose each disk to the same amount of absorbed energy (see Appendix 10.2.4 for sample calculation).

The TSEE responses of the exposed disks are presented in Fig. 11. Disks 318 and 273 gave consistent results; however, disk 317, having very erratic behavior, was deemed unrepresentative of BeO disks and was not included in the analysis.

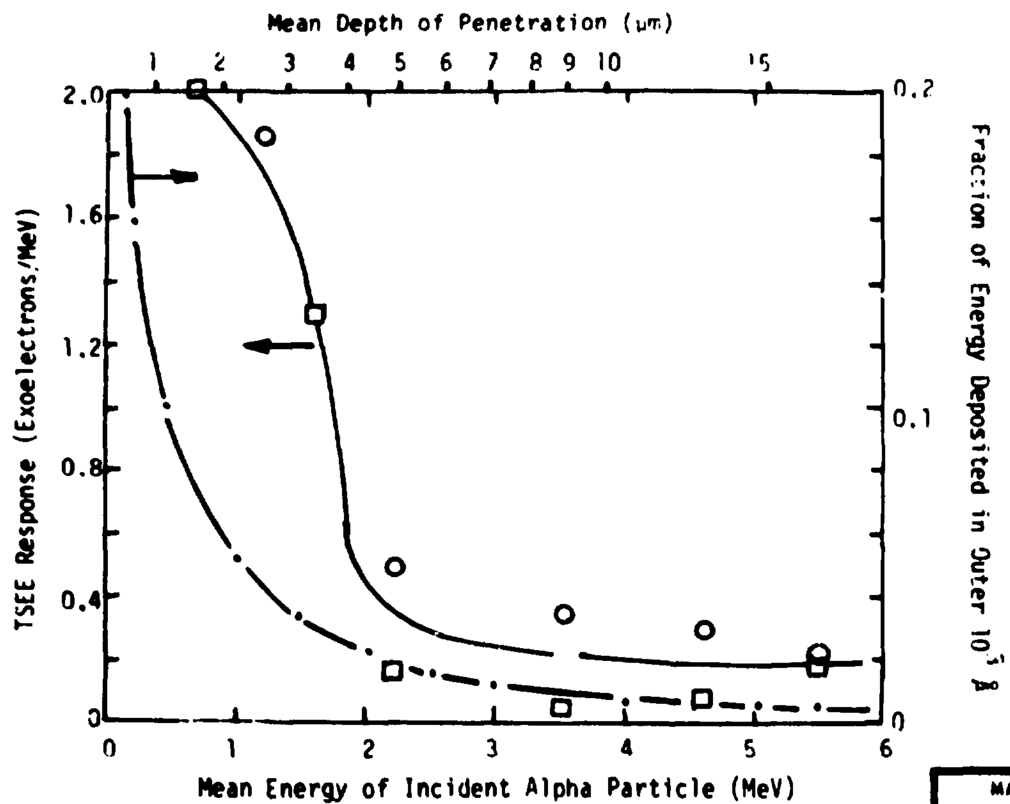
A final experiment was made in which a BeO disk was exposed to the americium-241 alpha source with the back side of the disk facing the source. The TSEE response from this experiment gave a negligible reading when measured from the front side.

6. DISCUSSION OF RESULTS

6.1 Calibrations and Field Experiments

Calibration with a known source is necessary to evaluate the field results. Two aspects of the calibration are discussed: the standard deviation of the measurements and the divergence of the TL/TSEE response. The standard deviation of the calibration measurements is about 10%. This result is comparable to literature reports for standard deviations of 3-20% for groups of BeO ceramic disks (2). Large variations in absolute sensitivity of the disks need to be controlled to a smaller range for reduction in the standard deviations. This can be accomplished through quality control in manufacturing the disks. The energy dependence of TL and TSEE responses as shown in Fig. 12 explains the changing TL to TSEE ratio (3). The TL response increases above the relative TSEE responses as the photon energy decreases below 1.0 MeV. The cesium-137 gamma photon is 0.66 MeV which is far below the photon energy from cobalt-60 (1.17 and 1.32 MeV); the TL and TSEE responses from Fig. 12 give a TL-to-TSEE ratio of 1.02. This ratio is also within the error limits of the TL-to-TSEE ratio obtained in the field experiments as shown in Fig. 6. The expected value (1.02) disagrees with the average of the field ratio (1.17) possibly due to a lowering of the 0.66 MeV photon energy of ^{137}Cs by multiple scattering in air.

The apparent field exposure rate is presented as a function of the distance of the badges from the ^{137}Cs source in Fig. 7. The apparent exposure rate is calculated from both TL and TSEE field responses and the ^{137}Cs calibration results for both shielded and unshielded badges. One badge was shielded to determine the effect of radiation from above the badges on both TL and TSEE responses. Comparison shows no significant difference between the exposures for the shielded and unshielded badges. The values for the above sky radiation are less than one mR/hr with an



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TSEE RESPONSE OF BeO DISKS TO INCIDENT ALPHA PARTICLE ENERGY			
DATE 11-25-75	DRAWN BY JRB	FILE NO. CEPS-X-220	FIG. 11

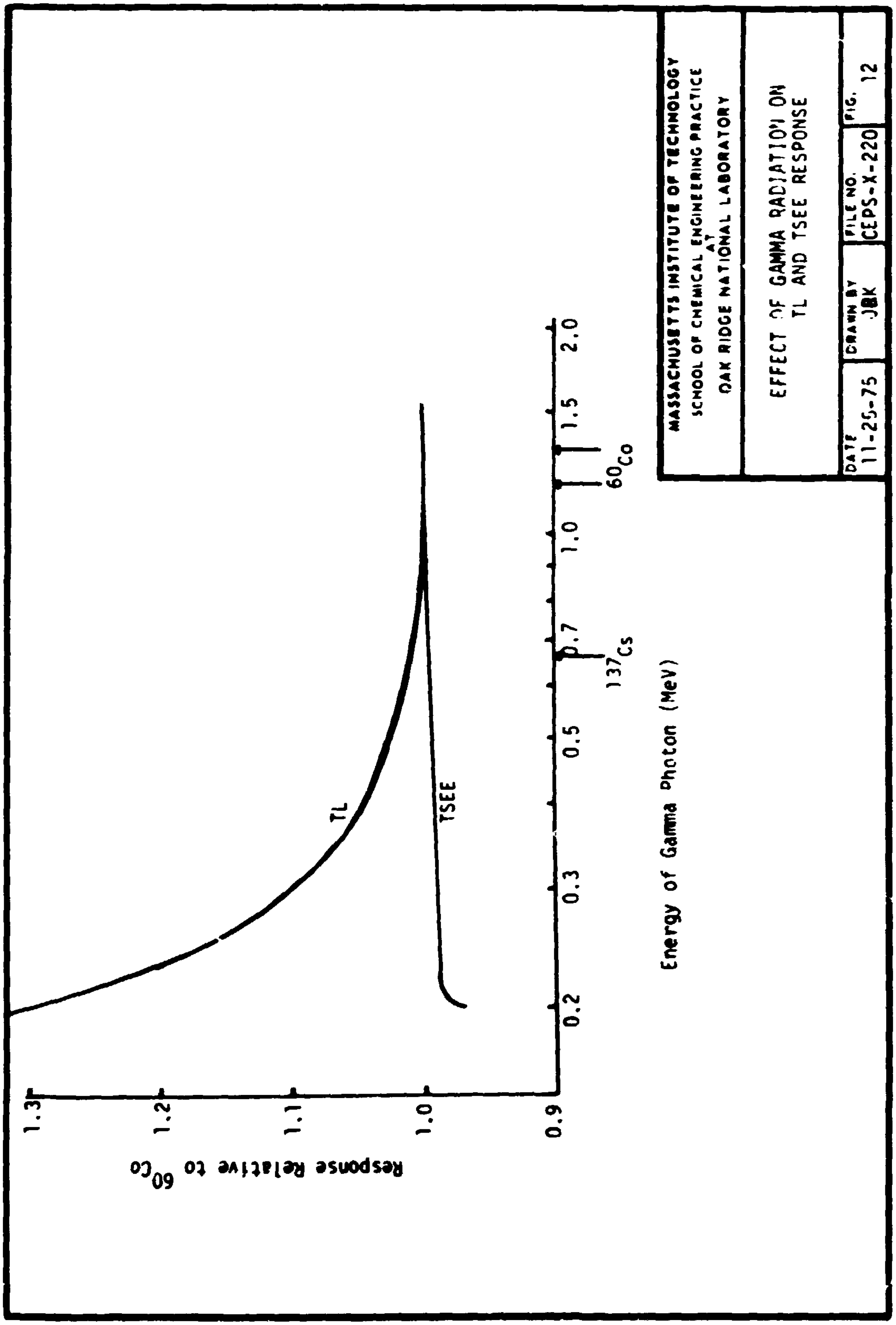
Response Relative to ^{60}Co
1.3
1.2
1.1
1.0
0.9
0.2
0.3
0.5
0.7
1.0
1.5
2.0
TL
TSEE
^{137}Cs
^{60}Co
Energy of Gamma Photon (MeV)
MASSACHUSETTS INSTITUTE OF TECHNOLOGY
SCHOOL OF CHEMICAL ENGINEERING PRACTICE
AT
OAK RIDGE NATIONAL LABORATORY
EFFECT OF GAMMA RADIATION ON TL AND TSEE RESPONSE
DATE 11-25-75
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FIG. 12

error of ±1000%. The low confidence inherent in determining the effects of sky radiation results from the small differences between relatively large TL and TSEE values, both having large significant errors. Beck (1) has calculated that the sky radiation should contribute about 10 to 15% of the exposure rate at large distances (~30 meters) from a field source. As only the upper portion of the badge was shielded, the lead shield may not have been effective in eliminating all the sky radiation. Radiation scattered and reflected from the ground has not been controlled and may have considerable effect. Thus no conclusion can be reached concerning the effects of sky radiation.

The apparent exposure rate compares well with the qualitatively expected results. The first two positions are over the contaminated ^{137}Cs pen, whereas the remainder lie progressively further away. A flat response over the contaminated area was expected, followed by a decrease in exposure proportional to the inverse square of the distance from the source. The TL and TSEE responses of BeO placed over the pen is constant as shown in Fig. 7. The linear portion beyond this has a slope of -2.5 that should compare to an expected slope of -2 for the inverse square relationship. In explanation, the badges face the ground at all locations as they were drawn away from the pen. The area is thickly covered with tall weeds that may moderate the photon flux. The rate measured by the TL response is consistently higher than that measured by TSEE as shown by the TL/TSEE exposure rates plotted in Fig. 8.

The high value of the ratio would imply a lowering photon energy field. The errors associated with these ratios are too large for any definite conclusions to be drawn. The plot of TL to TSEE indicates that the field ratio is consistently above the laboratory result and that it does not vary significantly with distance, showing a fairly uniform photon energy spectrum.

The TL-to-TSEE ratio should show an enhancement due to a lower incident photon energy different from the calibration responses such as shown in Fig. 12. In a field environment, a high percentage of the photons reaching distances a few meters from the source have lower energies due to Compton scattering (1). Thus an increase in the TL response relative to the TSEE response would be expected. In a field of gamma photons from cesium-137, the photons are expected to have 5-10% lower energies at distances to 30 meters from the source (1). However, the TL-to-TSEE ratio can also be affected by the geometry of the badge position relative to the source. In the experiments conducted in the field, the badges are parallel to the ground. Thus, the photons strike normal to the surface only when directly over the Cs-137 pen. At larger distances, the photons strike at an angle between 90° and 4° and change the TL and TSEE responses due to path length in the surface and a smaller flux area.

Gammage (5) has monitored the same field at 30 meters with BeO disks normal to the incident radiation rather than parallel to the ground in the presently described experiments. The TL to TSEE ratio from his results was 1.89. His result and the ratios obtained from the current experiments (1.17) indicate a strong directional dependence. Further experiments to measure the distribution of the photon energy at different distances, as well as angular dependence measurements, should be performed to provide precise analysis of the differences in the TL to TSEE ratios.

Any effect of geometry could be measured by conducting several experiments in which the incident angle, θ, of the photon flux is controlled and the total exposure kept constant. The TL and TSEE responses would respond differently to changes in θ if geometry has an effect.

6.2 Passive Radon-222 Dosimeter

The silicon diode measurements with the radon-222 dosimeter were designed to select the optimum chamber voltage for collection of radon decay products on an aluminized Mylar film. The number of alpha particles counted at a given radon concentration varies negligibly with applied chamber voltage as shown in Fig. 9. Previous work with different applied chamber voltages on one and five-liter systems shows a greater initial effect below 500 V and negligible change at higher voltages. The initial voltage effect not being present in the current design may be due to either the small size of the chamber or the high velocity of the charged particles. The heavy charged particles have a recoil energy of about 100 keV. This energy corresponds to an initial energy of 1.6×10^{-13} joules. As the plate spacing in the dosimeter is 5 cm, the particle cannot travel much farther than this distance before striking either the detector or a wall. The change in particle energy that may result from the electrical field is equal to the work required to move a doubly charged particle a distance, d, in an electrical field, V/d, and is given by

$$\Delta E = W = 2e\left(\frac{V}{d}\right)d = 2\,eV = 2(1.6 \times 10^{-19}\ \text{coulombs})(300\ \text{V}) \qquad (1)$$

$$= 9.6 \times 10^{-17}\ \text{joules}$$

Hence, the electrical field will have a negligible effect on the motion of the charged particle. Thus, the high initial energy of the particle is the major reason for a negligible effect of voltage.

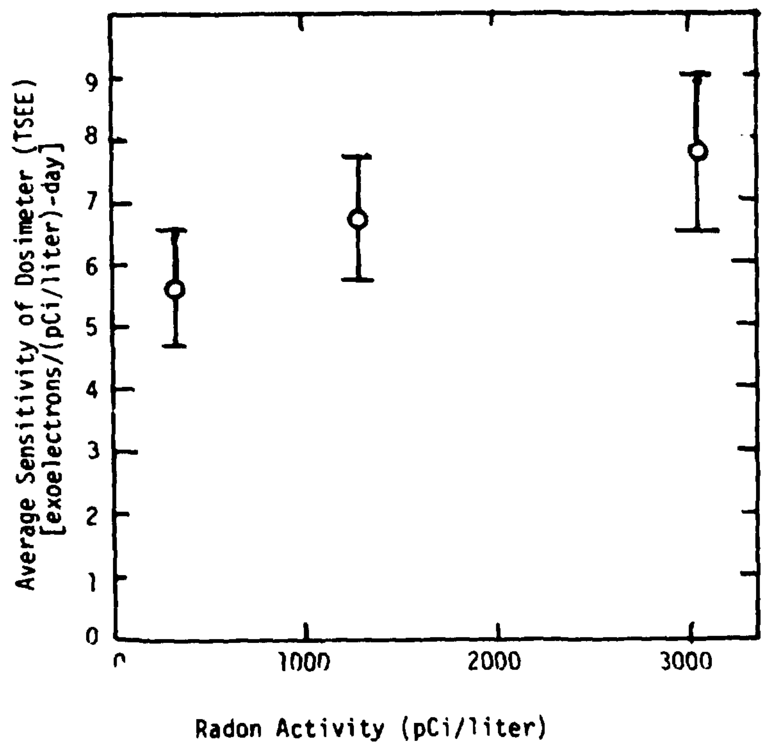
The concentration or activity of the radon-222 gas in the glove box is measured with a Lucas chamber. This chamber counts the number of alpha particles from the radon daughters, ^{218}Po and ^{214}Po. Five half lives (4.13 hr) are required to reach 99% of equilibrium with the radon gas in the chamber. The chamber counting technique is calibrated to give the radon activity within 5%. The sample from the glove box is removed from the opposite end of the box from which the passive dosimeter is placed and at the same height as the dosimeter. The error in measuring the concentration and activity of the radon gas may vary as much as 10-15% at high concentrations and up to 50% at very low concentrations ($\sim$1-100 pCi/liter).

As the diode measurements show no change in sensitivity to the daughter products at high chamber voltages, a 300-V battery was applied across the chamber for testing the BeO detectors in the dosimeter. The disks were exposed for a measured time with the activity of the radon gas measured initially after the dosimeter was placed in the box and immediately before removing the dosimeter. Thus the radon level can be determined from each individual measurement and the decay constant. The responses of the BeO disks in the dosimeter are shown in Fig. 13 for different radon activity levels. The average response of the disks shows an increase in sensitivity with increasing activity of radon; however, the large error associated with each measurement could also indicate a constant sensitivity. Further experiments are necessary to establish the sensitivity over wider ranges. In Fig. 10 the time exposures of BeO disks to radon-222 give exoelectron counts for the sensitivity of the TSEE reader. For long-term monitoring at low concentrations (0.1 - 4.0 pCi/liter) in homes (6), it would be necessary to monitor for 100 to 300 days. This is near the lower sensitivity of the BeO disk. The sensitivity could possibly be improved by changing the chamber size and shape, and by investigating an optimum design for concentration of the daughter products onto the detector area. An increase in sensitivity by a factor of 2 or 3 would be sufficient for practical long-term monitoring. The sensitivity of BeO to radon-222 is sufficient to justify further investigation into design improvements.

6.3 Measurement of Exoelectron Layer Depth

The purpose of this experiment was to determine the depth of the region near the surface of BeO which emits exoelectrons after irradiation. The reasoning behind the experiment is based on the manner in which the energy of a particle is deposited as it progresses into the solid. The rate of energy deposition by the particle is constant until the particle is almost completely stopped at which point the rate increases to a maximum, then decreases to zero. This maximum energy deposition is known as the Bragg peak. If the energy of a particle is sufficient to place the Bragg peak in the region past the TSEE active zone, the TSEE response per incident alpha particle will be constant with increasing energies. If the particle energy is low enough such that the Bragg peak occurs within the active region, the TSEE response increases with increasing energies. By exposing a disk to alpha radiation of varying energies and constant total energy, one can determine the alpha particle energy with which the TSEE response increases dramatically (which will reflect the fact that the Bragg peak has entered the active region). Once the critical alpha particle energy is known, the depth of penetration of the alpha particle can be calculated from data on the stopping powers of beryllium and oxygen as shown in Fig. 14 (10).

The results of the experiment, as shown in Fig. 11, have two conclusions. First, the sharp break occurring at the 1.8 MeV particle energy demonstrates that the depth of the highly active TSEE layer in BeO is 3-4 μm. This is significantly deeper (x 400) than has been predicted



Values are mean values from six disks with error bars corresponding to plus and minus one standard deviation.

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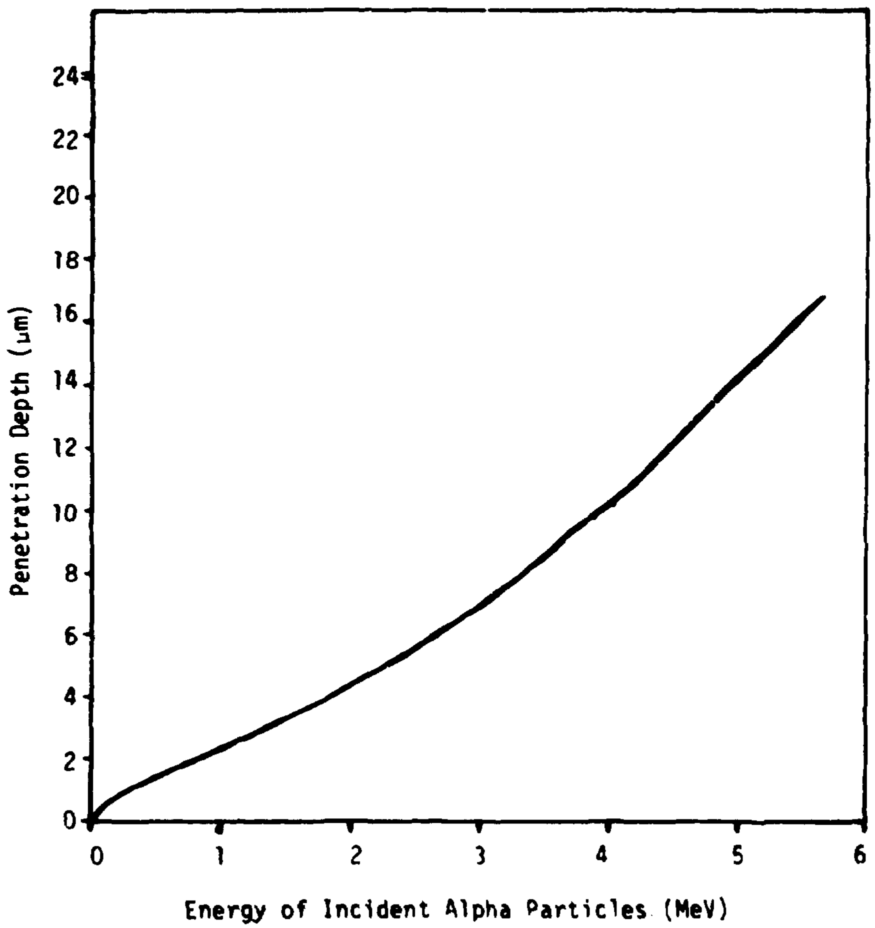
EFFECT OF RADON-222 ACTIVITY ON SENSITIVITY OF BeO DOSIMETER

DATE	DRAWN BY	FILE NO.	FIG.
11-25-75	JRB	CEPS-X-220	13



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PENETRATION DEPTH OF ALPHA PARTICLES IN BeO DISKS (26)

DATE	DRAWN BY	FILE NO.	FIG.
11-25-75	JJP	CEPS-X-220	14

(~100 Å) (9). Second, the occurrence of a relatively constant response of TSEE/MeV to increasing particle energies above 1.8 MeV (up to 5.49 MeV, the maximum energy obtainable) is produced by a second, less active, TSEE layer extending from 4 μm to at least 16 μm into BeO.

The second conclusion stems from an analysis of the rate of energy deposition as a function of distance into the BeO (10) (for the first 4 μm of BeO). The fractional amount of energy deposited in the highly active, first exoelectron layer decreases as the alpha particle energy increases above 1.8 MeV (10). If only one exoelectron layer existed, the TSEE/MeV response would continue to decrease after the Bragg peak had passed the first layer. As the TSEE response does not decrease, the exoelectrons must originate from a second deeper active layer. Since the amount of energy deposited in the solid is constant, and the amount of energy deposited in the first layer decreases as the alpha particle energy increases, then, by difference the fractional amount of energy deposited in this second layer must increase. This would cause the TSEE response of the second layer to increase as the alpha particle energy increases (assuming that the TSEE response of the second layer does not depend on the depth of the Bragg peak within the layer). As shown in Fig. 15, calculations based on stopping power of BeO show that this is true. Thus the TSEE response per MeV should be constant as the alpha energy increases.

The second exoelectron layer extends at least 16 μm into the solid. From the experiment in which the BeO disk was irradiated from the back side and gave no response when analyzed from the front side, the second layer does not extend through the solid, 1500 μm. Further experiments with higher energy alpha particles are necessary to find the actual depth of this second TSEE active layer.

7. CONCLUSIONS

1. The beryllium oxide disks can be employed to monitor environmental sources of cesium-137 by either TL or TSEE. A knowledge of the photon energy flux is necessary to evaluate the TL-to-TSEE ratio.

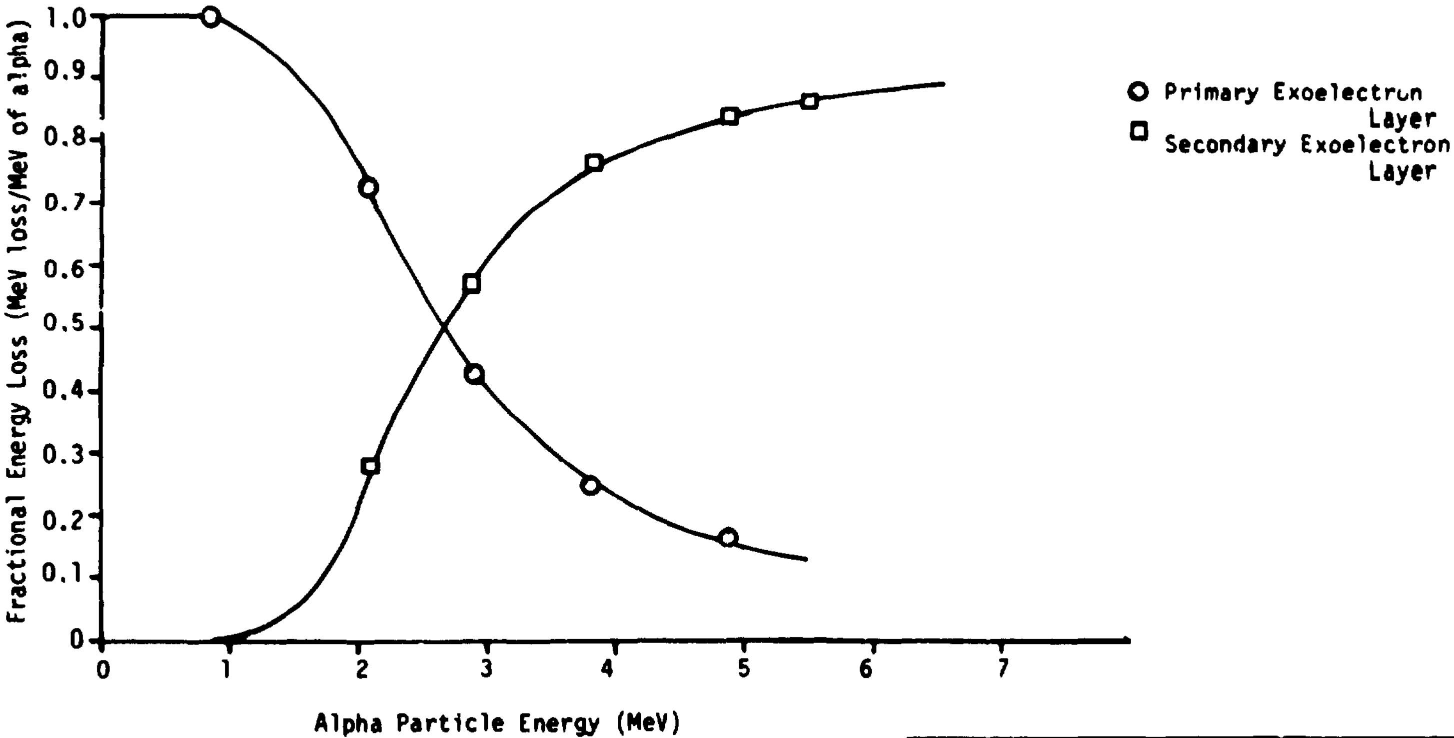
2. Effects of sky radiation are negligible.

3. An applied chamber voltage has negligible effect on sensitivity of a passive radon dosimeter.

4. Dosimeter is sensitive at a minimum level of 100 (pCi/liter)-day.

5. The primary exoelectron layer in BeO was measured experimentally to be 3 to 4 μm deep.

6. A second exoelectron layer was found that extends to a depth of at least 16 μ.



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FRACTIONAL ENERGY LOSS IN
EXOELFCTRON LAYERS OF BeO

DATE	DRAWN BY	FILE NO.	FIG.
11-25-75	JJP	CEPS-X-220	15

8. RECOMMENDATIONS

1. Further experiments are required to measure the actual photon energy field and its effect on the TL-to-TSEE ratio and to show any effect of angle of incidence of radiation on response.

2. Further investigation into an optimum design with larger chamber volume is recommended to increase the sensitivity of the passive radon-222 dosimeter for practical applications.

3. The secondary exoelectron layer should be measured to establish total depth and to determine how electrons travel such large distances. Irradiation from the back side using high energy alpha particles and reading from the front side may accomplish this.

4. A quality control in manufacturing BeO disks is needed to establish uniform sensitivity for practical application.

9. ACKNOWLEDGMENTS

We thank R.B. Gammage, G.D. Kerr, F.F. Haywood, and J.H. Thorngate for their help and effort in setting up and understanding the experiments.

10. APPENDIX

10.1 Radiation Sources

The decay particle of each of the experimental sources and their respective energies are shown in Table 2.

Table 3. Radiation Sources

Source		Radiation	Energy (MeV)
Cobalt-60		β^-	0.314
		γ	117, 133
Cesium-137		β^-	0.514
		γ	0.662
Americium-241		α	5.49
Radon-222		α	5.49
Important Decay Products:	^{218}Po	α	6.00
	^{214}Pb	β^-/γ	0.69, 0.79/0.29, 0.35
	^{214}Bi	β^-/γ	2, 3.26/0.61, 1.12, 1.76
	^{214}Po	α	7.69

10.2 Sample Calculations

10.2.1 Calibration of BeO Disks

To calibrate the BeO disks, they were divided into four groups of six disks each, one badge consisting of six disks. These badges were then exposed at least three times to 20 mR from ^{60}Co and ^{137}Cs (6 exposures). The resulting TL and TSEE responses of each disk were measured. The average, shown in Table 4, and standard deviation were computed as follows:

Table 4. Calibration of BeO Disks to ^{60}Co (20 mR)

Disk	TL (mV)	TL (mV)	TL (mV)
E317	237	263	273
E295	246	235	241
E272	284	231	234
E273	245	258	254
E281	250	244	249
E318	256	276	276

The average is 253.

The standard deviation is found from

$$S = \sqrt{\frac{(x_i - \bar{x})^2}{n - 1}} = 16.0$$

The TSEE/TL ratios were calculated from the average TL and TSEE responses. For badge 2 exposed to ^{60}Co this ratio was

$$\frac{TSEE}{TL} = \frac{1432}{253} = 56.6$$

This ratio is compared to the ratio for ^{137}Cs.

$$\frac{\left(\frac{TSEE}{TL}\right)_{^{60}Co}}{\left(\frac{TSEE}{TL}\right)_{^{137}Cs}} = \frac{56.6}{41.4} = 1.37$$

For an error analysis in calibration, consider a function (y) of several experimental variables:

$$y = f(x_1, x_2, \ldots.) \qquad (2)$$

The variance in computing y is given by

$$(\Delta y)^2 = \left(\frac{\partial f}{\partial x_1}\right)^2 \Delta x_1^2 + \left(\frac{\partial f}{\partial x_2}\right)^2 \Delta x_2^2 + \ldots\ldots \quad (3)$$

where Δx_i and Δ_y are the standard deviations associated with x and y. After the standard deviation is found from Eq. (3), all subsequent errors in ratios are found with the expression. If

$$w = y_1/y_2$$

then

$$(\Delta w)^2 = \left(\frac{\partial w}{\partial y_1}\right)^2 \Delta y_1^2 + \left(\frac{\partial w}{\partial y_2}\right)^2 \Delta y_2^2 = \left(\frac{1}{y_2}\right)^2 \Delta y_1^2 + \left(-\frac{y_1}{y_2^2}\right)^2 \Delta y_2^2$$

$$= w^2\left(\frac{\Delta y_1^2}{y_1^2} + \frac{\Delta y_2^2}{y_2^2}\right)$$

For the TSEE/TL ratio (badge 2, ^{60}Co),

$y_1 = 1432$ $\quad\Delta y_1 = 1913$

$y_2 = 253$ $\quad\Delta y_2 = 16.0$

$w = 56.6$

$$(\Delta w)^2 = \frac{(1913)^2}{(253)^2} + \frac{(56.6)^2(16.0)^2}{(253)^2} = 57.2 + 12.8 = 70.0$$

$$\Delta w = 8.4$$

This general procedure was used to compute the error of all quotients.

10.2.2 Cesium-137 Field Experiment

The apparent exposure rate in the field was determined as follows:

$$\frac{\left(\frac{\text{field response}}{\text{lab response for 20 mR}}\right)20\ \text{mR}}{\text{time exposed}}$$

For example, for badge 1, exposed 4 hr, the average TL response was 540.6 $\pm$ 5.4 mV. From the laboratory calibration with ^{137}Cs the response to 20 mR was 295.7 $\pm$ 3.49 mV. Then the apparent exposure rate is

$$\frac{\frac{540.6\ \text{mV}}{295.7\ \text{mV}}(20\ \text{mR})}{4\ \text{hr}} = 9.1\ \text{mR/hr}$$

The same method applies to TSEE response.

The error can be calculated as in Eq. (3). The TL/TSEE response ratio is calculated as follows:

$$\text{apparent TL exposure} = \frac{540.6}{297.7}(20) = 36.6\ \text{mR}$$

$$\text{apparent TSEE exposure} = \frac{19{,}061.7}{12{,}374.9}(20) = 30.8\ \text{mR}$$

19,061.7 and 12,374.9 are average responses from data minus 200 for background counts.

$$\frac{\text{apparent TL response}}{\text{apparent TSEE response}} = \frac{36.6}{30.8} = 1.19$$

10.2.3 Lucas Chamber Counting for Radon-222

A Lucas chamber measures the activity level of radon-222. The chamber gives an output of alpha counts per minute from a volume of 0.095 liters. This can be changed to an activity level, A_c, in pCi/liter by:

$$A_c = \left[\frac{\text{alpha counts/min}}{(\text{efficiency})(\text{chamber volume})}\right]\left[\frac{1\ \text{radon disintegration}}{2\ \text{alpha particles}}\right]\left[\frac{100\ \text{pCi}/\ell}{3.7\ \text{dis/sec}}\right]\left[\frac{1\ \text{min}}{60\ \text{sec}}\right]$$

For a count of 1500 alphas/min and an efficiency of 0.885 counts/alpha, the activity of the radon gas is:

$$\left[\frac{1500}{(0.095)(0.885)}\right]\left(\frac{1}{2}\right)\left(\frac{100}{3.7}\right)\left(\frac{1}{60}\right) = 4018\ \text{pCi/liter}$$

10.2.4 Exposure Levels to Radon

The exposure level, E_ℓ, of radon is the exposure over a period of time to given concentrations of radon. If an initial concentration, C_o, is present, the activity will decrease with time as determined by the decay constant for radon:

$$E_\ell = \int_0^t C_0 e^{-\lambda t} dt = C_0 \frac{1}{\lambda}(1 - e^{-\lambda t}) \quad (4)$$

The decay constant, λ, is 2.094×10^{-6} sec^{-1}. For an initial radon activity concentration of 3150 pCi/liter and an exposure time of 19.67 hr, the exposure level is:

$$E_\ell = 3150[\frac{1}{2.094 \times 10^{-6}}][1 - e^{-(2.094 \times 10^{-6})(70,812)}]$$

$$= 2.073 \times 10^{8}(pCi/liter)\text{-}sec = 2399\ (pCi/liter)\text{-}day$$

10.2.5 Exoelectron Layer Depth

Alpha particle energy at various helium pressures was measured experimentally and reported in Table 2. A correlation of depth of penetration in BeO to alpha particle energy was provided by Thorngate (10) and is shown in Fig. 14. The only calculation necessary to determine the depth of the exoelectron layer was determination of exposure time for constant deposition of energy.

The alpha particle energy is determined from the analyzed diode response in mV, D. This response as a function of energy is described by the line

$$D = 36.12\ (energy) + 2.304$$

At the 750 mm helium pressure, the response of the diode was 107 which gives an energy of

$$E = \frac{107 - 2.304}{36.12} = 2.898 \text{ MeV}$$

The experiment on alpha particle energy also gave the time for 1000 alpha particles to reach the detector. Hence, the time for a constant total energy deposition is found as

$$t = \left(\frac{\text{total energy deposited}}{\text{avg energy/alpha particle}}\right)\left(\frac{\text{particles}}{\text{(sec)(area of diode)}}\right)\left(\frac{\text{area of diode}}{\text{area of disk}}\right)$$

At 750 mm Hg of helium pressure, the average energy per alpha particle is 2.898 MeV/particle, and the time is 386 sec for 1000 alpha particles to deposit. The area ratio of diode to disk is 2.11. For 15,000 MeV to be deposited, the time required is

$$\left(\frac{15{,}000\text{ MeV}}{2.898\text{ MeV/alpha particle}}\right)\left(\frac{0.386\text{ sec}}{\text{alpha particle}}\right)(2.11) = 4216\text{ sec}$$

10.3 Location of Original Data

The original data can be found in ORNL Databook A-7549-G, pp. 1-56 on file at the MIT School of Chemical Engineering Practice, Bldg. 3001, ORNL. All original TL and TSEE recordings are bound and labeled in a notebook in the possession of R.B. Gammage.

10.4 Nomenclature

A area of plates in radon dosimeter, cm^2

A_c activity level of radon-222, pCi/liter

D diode output for exoelectron calibration, mV

d distance between plates, cm

e charge on an electron

E_ℓ exposure level to radon-222, pCi/liter-day

C_0 initial concentration of radon gas

t time, sec

V applied chamber voltage, V

λ decay constant = 2.094×10^{-6} sec^{-1} for radon-222

θ angle of incident gamma photons, degrees

10.5 References

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