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THE EFFECTS OF RADIATION ENERGY, DOSE LEVEL, AND THE ATOMIC NUMBER OF THE IMMEDIATE ENVIRONMENT ON THE THERMALLY STIMULATED EXOLECTRON EMISSION FROM BERYLLIUM OXIDE AND LITHIUM FLUORIDE

(Thesis)

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Submitted as a thesis by James F. Wilson to the Graduate Council of The University of Tennessee in partial fulfillment of the requirements for the degree Master of Science

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James F. Wilson

A Thesis Presented to
the Graduate Council of
The University of Tennessee
In Partial Fulfillment of
the Requirements for the Degree
Master of Science

MAY 1971

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ABSTRACT

The effect of thermally stimulated exoelectron emission (TSEE) from very thin layers of ionic crystals provides for the first time an essentially two-dimensional integrating radiation detector. These detectors can be expected to exhibit an energy dependence of their response to x and gamma irradiation which depends as much or more on the effective atomic number of its immediate environment as on that of the detector itself. Because of the unknown contributions of electrons released in the irradiation of the detector's environment and in the detector itself to the total radiation response, the photon energy response of a TSEE material cannot be calculated easily and empirical studies were undertaken.

Two types of TSEE materials, beryllium oxide (BeO) and lithium fluoride (LiF), and two types of detector preparation, mixing of powders with graphite and ceramic disks with metals as conductive and/or sensitizing additives, have been chosen. Their relative sensitivities have been determined for different photon energies, exposure levels, and with covering materials of varying atomic number. The theoretical and practical implications of the results are discussed.

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CHAPTER I

INTRODUCTION

It has been known for several decades that chemical reactions may be accompanied by low energy electron emission in some inorganic ionic crystals and minerals.[1] However, the first systematic study of the basic physical processes involved was not begun until the late 1940's by J. Kramer[2] who, originally assuming exothermic processes to be involved with the emission mechanism, proposed the term "exoelectrons" to describe the emitted electrons. Exoelectron emission (EE) was later found to occur in various systems during or after mechanical deformation, phase changes, changes in crystal structure, and exposure to ionizing radiation. Because of his outstanding contributions to the understanding of this physical phenomena, the general effect of structure dependent, low energy electron emission is also referred to as the "Kramer effect."

Kramer[3,4] observed in several materials (CaF_2, gypsum, and others) that after irradiation a delayed electron emission occurred either spontaneously at room temperature with a more or less rapid decay or when additional energy was transferred to the system by exposure to light or heat (optical or thermal stimulation). The thermally stimulated exoelectron emission (TSEE) was found to exhibit, if the emission rate is determined as a function of temperature, characteristic maxima (Fig. 1). It was later shown by Gobrecht[5] that the exoelectrons found to be emitted from "metals" actually originated from the thin oxide layer on the surface of the material. Clean metal surfaces do not exhibit exoelectron

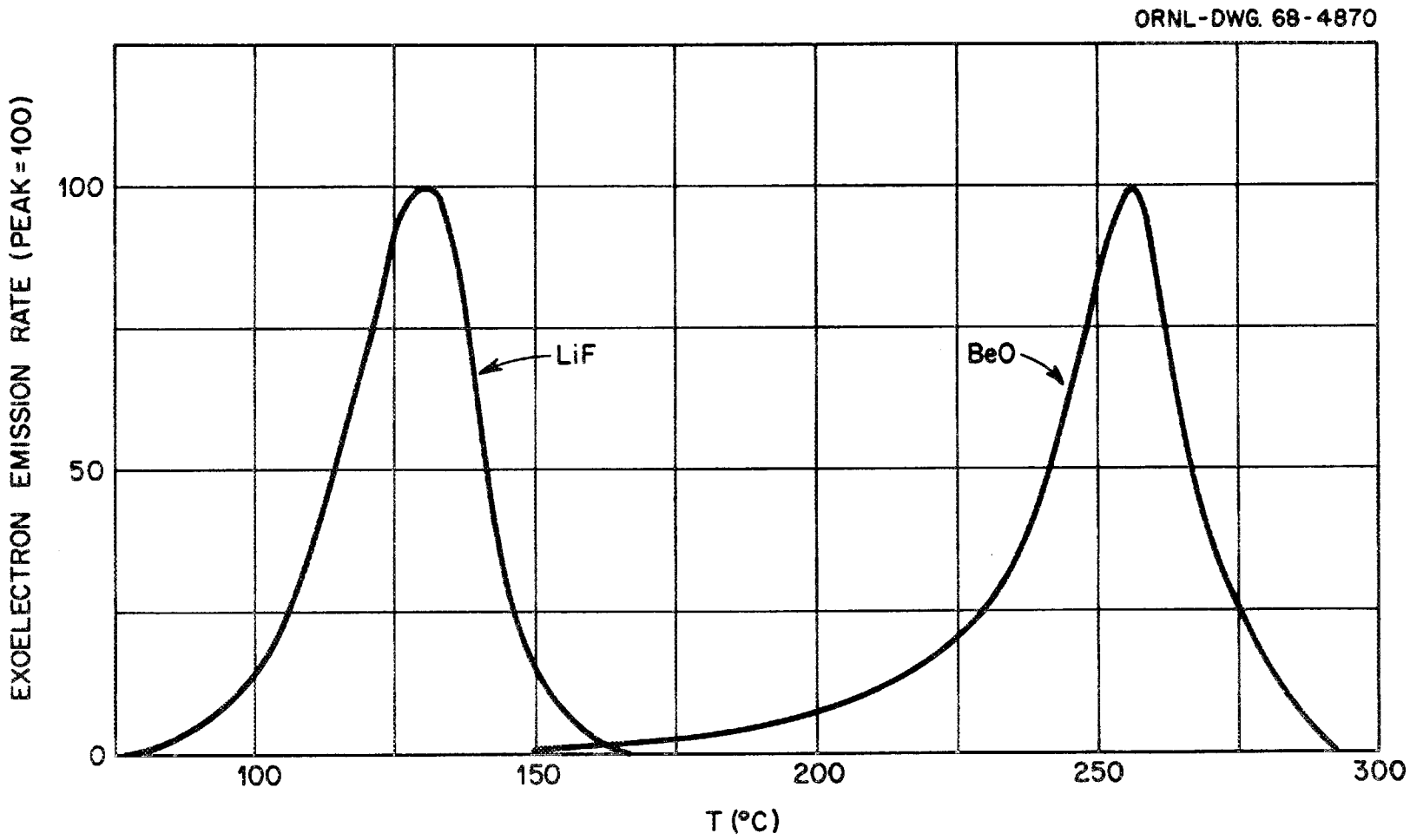

Fig. 1. Typical examples of the exoelectron emission rate as a function of stimulating temperature, showing only the low-temperature maxima, of irradiated LiF and BeO.

emission. This result has been similarly extended to semiconductor materials.

The possibliity of applying stimulated exoelectron emission to the field of radiation dosimetry was first indicated by Kramer[6] and, independently, by Gourgé and Hanle[7] in 1957. Kramer had discovered that there was a proportionality between the observed TSEE and the radiation dose in $CaSO_4$. These and later results were summarized in 1963 in a more detailed paper by Kramer[8] on the dosimetric applications of the stimulated exoelectron emission from $CaSO_4$. In 1966, Hanle et al.[9] found that the radiation sensitivity of $CaSO_4$ could be optimized by preheating the material to 960°C.

The same year, a major breakthrough was made by Kramer[10] when he described a method to obtain a linear relationship between TSEE response and radiation dose. He observed that if the emitting surface was made electrically and thermally conductive by the addition of a material such as graphite, which is not an exoelectron emitter, the radiation response became a linear function over many orders of magnitude of dose. This effect was attributed to the elimination of surface charging effects which interfere with the emission process.

Subsequent investigations of TSEE materials have been limited to a few European laboratories until 1968 when the Health Physics Division of Oak Ridge National Laboratory initiated a research program to explore the dosimetric applications of the TSEE. At ORNL, the TSEE readers initially used (and discussed later in greater detail) consisted of gas-flow G-M counters into which a heating element had been inserted.[11] This system provided for non-linear heating of disk-shaped graphite

mixed detectors to approximately 350°C. Later improvements of this instrumentation permitted reading at higher temperatures, due to the addition of a water-cooling system to the counting volume [12] and the employment of a linear heating device.[13] The present system is being operated in the Geiger region which somewhat limits its ability to measure high TSEE rates and, therefore, high dose levels, due to dead-time losses. High dose levels may be measured by electrometric methods.

Parametric studies on the mechanism and kinetics of fading,[14] the LET dependence,[15] the fast neutron sensitivity[16] of TSEE dosimeters, the relationship between thermoluminescence (TL), TSEE and impurities in several materials such as LiF and BeO,[17] the effect of metal diffusion into the surface of ceramic emitters[18] and the heat sensitization of ceramic BeO[19] have been carried out recently at ORNL. Current efforts include: An analysis of the TSEE of lunar minerals;[20] the search for new EE materials;[21] further investigations and improvements of highly sensitive TSEE dosimeters based on ceramic BeO; and studies on the nature of centers involved in the TSEE process.

CHAPTER II

THEORY AND BASIC PROPERTIES

Kramer[22] first suggested that there were similarities between the emission phonomena of TSEE and thermoluminescence (TL). After Bohun[23] and Lepper[24] had determined that the exoelectron emission was observed only in those materials whose surface defect structures act as electron traps, Hanle[25] proposed a simple band model for the two processes of TSEE and TL. According to a more recent and somewhat simplified presentation (Fig. 2), electrons which are liberated by the interaction of the radiation with the detector material are transferred from the valance band into the conduction band. These electrons, along with those originating from the immediate environment of the detector which are injected into the crystal, are then captured in the various characteristic electron traps in the sensitive material.

During thermal stimulation, these electrons may be released into the conduction band, producing a transient increase in the conductivity of the material. While in the conduction band the electrons may recombine with defect electron traps, a process which is frequently associated with the emission of a photon (TL). Others of these electrons may be able to overcome the work function and leave the crystal surface by direct ejection (TSEE).

In many cases, however, the two processes are not observed simultaneously (identical TL and TSEE peak shapes and locations) for a number of possible reasons. In spite of this fact there remain enough similarities between the processes of TSEE and TL to ascribe to them the above

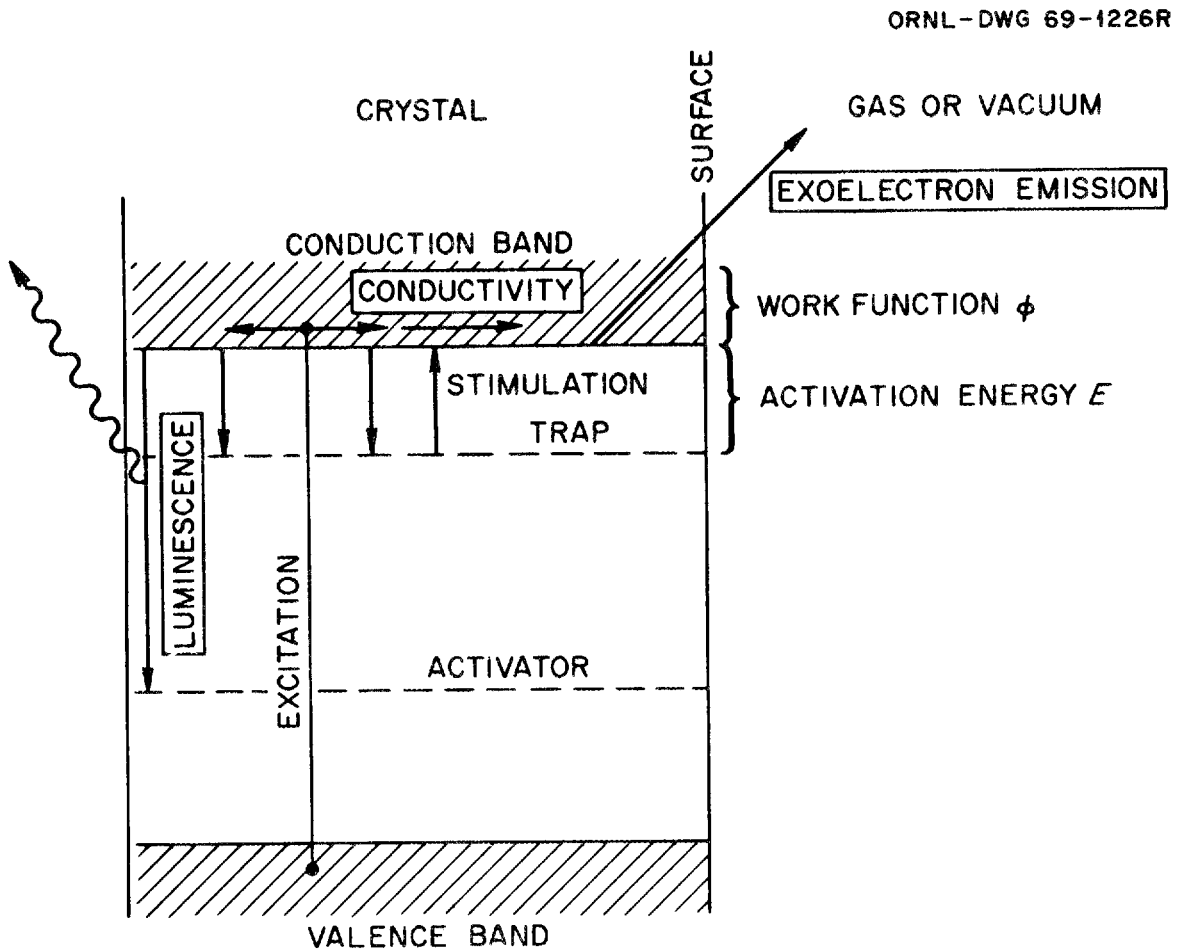

Fig. 2. Simplified band model showing the relation between luminescence, conductivity and exoelectron emission during stimulation of an ionic crystal which has been previously exposed to ionizing radiation.

common basic mechanism. While there is not yet a unified theory that adequately explains these inherent physical processes, the simple band model permits at least an approximate mathematical description of the TSEE kinetics.[26,27]

By its very nature TSEE has been shown to be, in effect, a "surface" phenomenon.* That is, the thickness of the sensitive layer which contributes the electrons that result in the observed TSEE is probably on the order of a mean free path length for the exoelectron in the detector material. Electrons which are released from deeper, "volume" traps would have a negligible probability of reaching the surface. It has been determined by Holzapfel[28] that the surface thickness is approximately 10 to 100 Å. This thickness is small when compared to the range of the electrons which are released by the interaction of photon radiation with the detector and its immediate environment, making the detector essentially two-dimensional. Indeed, it appears that the essential differences between TSEE and other solid state detectors such as those which are based on thermoluminescence (TL) can be attributed to the fact that TSEE is a "surface" effect as compared to the "volume" effects of TL. In thermoluminescence, essentially only the bulk of the material contributes to the observed radiation effect.

The main reasons why TSEE appears to be a promising technique for integrating radiation dosimetry, as compared to the related method of thermoluminescence dosimetry, are:

(1) an easily adjustable, high sensitivity (dose as low as 10^{-5}

*The term "surface" is used in this case to describe the crystal interface and the adjacent thin layer of detector material from which the exoemission results.

rad have been measured with the most sensitive materials);

(2) more stable information storage (TSEE can be recalled from deep traps corresponding to temperatures approaching 600°C, at which point the onset of thermionic emission begins to result in a significant perturbation of the information output. This compares with TL readings which are increasingly obscured by infra-red emission at temperatures as low as approximately 300°C);

(3) little or no spurious effects after proper annealing; and

(4) being a surface effect, TSEE can be used as an essentially two-dimensional detector for microdosimetry to measure short-range types of radiation, etc.

If during a heating cycle of the irradiated material one observes the TSEE rate as a function of temperature, both the integral number of exoelectrons and the peak heights are measures of the effective radiation dose with the former being the more sensitive indicator. The temperature locations of the characteristic emission maxima are heating rate dependent, being shifted to higher temperatures for larger temperature gradients. This effect corresponds to the Randall-Wilkins shift observed in thermoluminescence.[29]

In order to be suitable for TSEE dosimetry, traps in an emitting material should be "deep" enough to retain the TSEE information with negligible fading at ambient temperatures, but "shallow" enough so as not to make the reading process too difficult at temperatures approacing the thermionic emission edge. Also, the different parameters affecting the material's sensitivity should be optimized for the particular dosimetric situation. The sensitivity of the radiation response of a

TSEE detector is mainly affected by:

(1) the concentration of the sensitive material in a mixture with non-emitting additives;

(2) its production history and impurities;

(3) its pre-treatment by heating, irradiation, doping with additional impurities, etc,;

(4) the effective energy and LET of the radiation;

(5) the dose level;

(6) the composition of the immediate detector environment;

(7) the emitting surface area; and

(8) the instrumentation's absolute exoelectron detection efficiency.

In general, for volume-type detectors the sensitivity to x and gamma radiation is directly proportional to the ratio of the mass energy-absorption coefficient of the detector to that of air,

$$\frac{\left(\frac{\mu_{en}}{\rho}\right)_d}{\left(\frac{\mu_{en}}{\rho}\right)_{air}}$$

where μ_{en} is the energy-absorption coefficient and ρ is the volume density. If this ratio is then observed as a function of incident photon energy relative to some reference energy (for example the 1.25 MeV (effective) gamma radiation from ^{60}Co), the normalized energy dependence (ED) of the detector over the region of energy can be expressed as

$$ED_{\text{normal.}} \propto \left.\frac{\dfrac{\left(\dfrac{\mu_{en}}{\rho}\right)_d}{\left(\dfrac{\mu_{en}}{\rho}\right)_{air}}}{\dfrac{\left(\dfrac{\mu_{en}}{\rho}\right)_d}{\left(\dfrac{\mu_{en}}{\rho}\right)_{air}}}\right|_{1.25\ \text{MeV}}^{\text{incident photon energy}}$$

The total energy absorption coefficient consists of three components:

$$\mu_{en} = \tau + \sigma_a + K_T$$

where τ = photoelectric cross section in 1/cm;

σ_a = the absorption component of the total Compton cross section (1/cm);

K_T = the cross section for pair production in the field of the nucleus and the atomic electrons (1/cm), which can usually be neglected for photon energies of less than approximately 1.5 MeV.

However, since the process of exoelectron emission has been shown to be a "surface" phenomena, the ED relation, which applies to "volume" processes, may not be valid in this case. Specifically, the observed sensitivity of a TSEE detector is dependent to a large extent on the deposition of energy in the immediate environment and the subsequent electron transfer to the sensitive surface. The actual energy dependence of the sensitivity may be expected to vary substantially from this simple relationship due to this "environmental" contribution.

The photon energy dependence of a given system--either due to the direct interaction of the radiation with the detector, or due to its response to electrons released in its environment--is closely related to the effective atomic number (Z_{eff}) of those materials. The definition of Z_{eff} depends on the photon energy. At low photon energies, where the photoelectric process dominates, the form of the function determining the effective atomic number is

$$Z_{eff} = \sqrt[4]{\sum_i a_i \; Z_i^4}$$

where a_i is the fractional atomic content of the element i, and Z_i is its atomic number. At photon energies up to about 2 MeV, the Compton effect is dominating and the relationship determining Z_{eff} becomes

$$Z_{eff} = \sum_i a_i \; Z_i$$

where a_i and Z_i are as previously defined.

If Z_{eff} in the detector and its environment are very different, the measured energy dependence of the detector can be used as an indicator of the percentage of the total radiation effect caused by electrons from its environment.

Since the detectors are somewhat light sensitive and may be affected by liquid water, they have to be protected by a proper encapsulation if used in the field. The choice of an encapsulation material is simplified by knowing how it affects the energy response. Restricting the scope of the study to the lowest temperature emission maxima from the materials, easily accessible in simple readers, it is the object of this work to consider some of the little investigated parameters affecting TSEE detec-

tor sensitivity. In particular, the effects of photon energy in the 20 keV to 1.25 MeV range, dose level, and the atomic number of the immediate detector environment are examined for two detector materials and compositions.

CHAPTER III

EXPERIMENTAL PROCEDURE

1. INSTRUMENTATION

The exoelectron emission was measured by a gas-flow, straight wire G-M counter modified to permit thermal stimulation of disk-shaped TSEE dosimeters. The samples were heated by a circular element located in the slide tray of the counter.[11,12] The heater was operated by a controlling device which could be adjusted for maximum temperature and linear heating rates from 0.5 to 10 C°/sec.[13] Good thermal and electrical contact for the samples was supplied by a screw-type, brass holding mechanism pressing the detectors onto the heater plate, thermocouple, and grounding strip in the counter. Thermal insulation of the counting volume was enhanced by isolating the heated components with mica or Transite (Mansville Corporation) and by the use of an integrated water cooling system and external fan.

The counter was operated at 1400 volts, well within the plateau region from 1150 to 1600 volts, using a counting gas composed of 99.05% helium and 0.95% isobutane. The dead time of the counter had previously been determined to be 200 μsec., consequently providing negligible counting losses for counting rates less than 10^4 cpm. For quantitative measurements, dose levels and/or heating rates were selected so as to minimize the effect of counting losses.

The counter and associated electronics are shown in a block diagram (Fig. 3). Both the integral number of emitted exoelectrons and the TSEE rate as a function of temperature were recorded during each heating cycle.

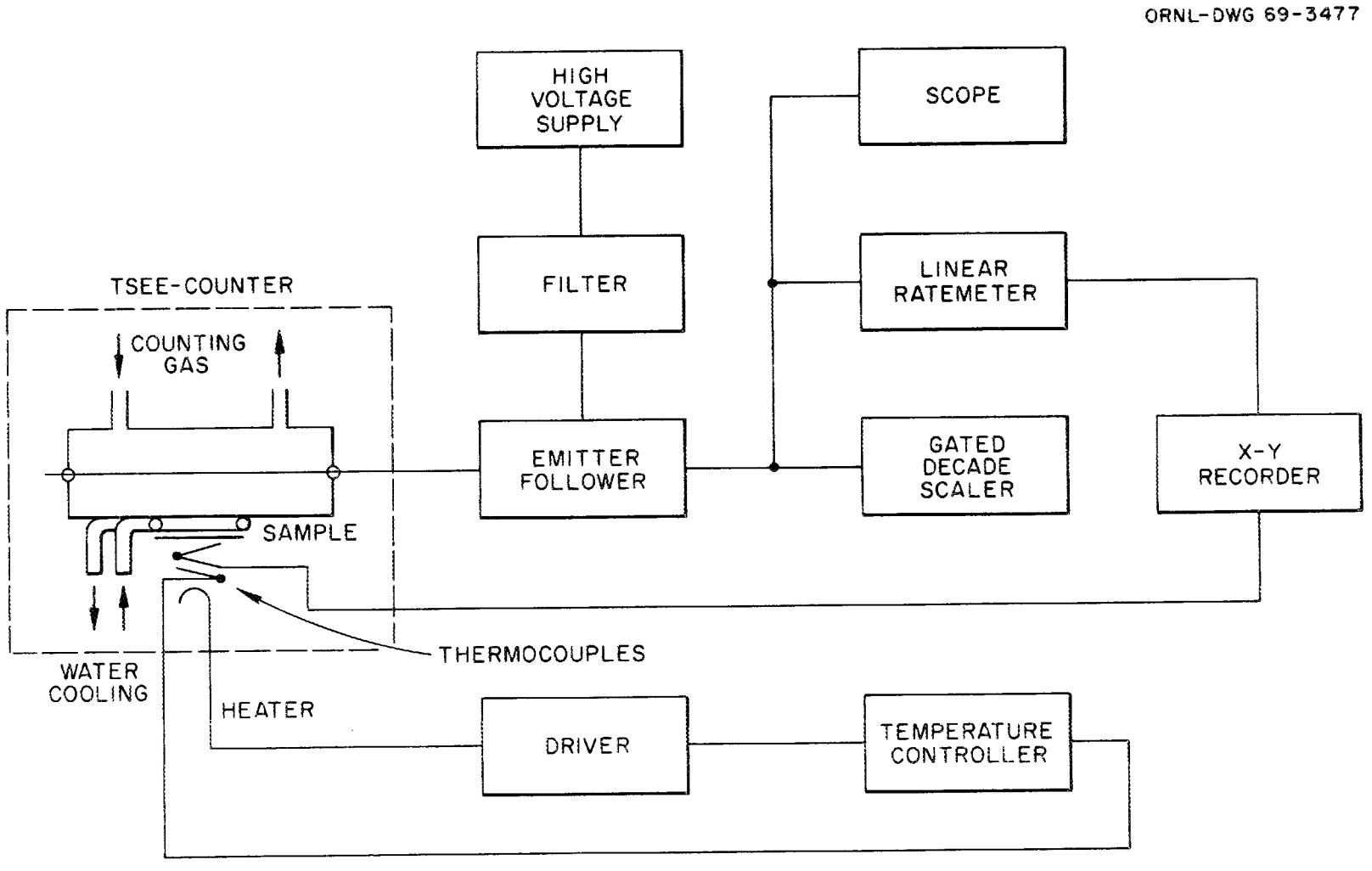

Fig. 3. Schematic block diagram of the complete TSEE reading system

II. DETECTOR PREPARATION

Graphite powder mixtures

Beryllium oxide (UOX powder, Brush Beryllium Co.) and lithium fluoride (J. T. Baker Chemical Co.) were chosen for examination because they are both relatively well studied materials with a low atomic number and they meet satisfactorily the previously stated requirements for a TSEE dosimetry material. An analysis of the impurity constituents of each material is given in Table I, Appendix. In the simplest and most flexible type of preparation, corresponding to a procedure initiated by Kramer, the sensitive material is hand ground to a fine powder and is mixed with powdered reactor grade graphite in a ratio of two parts of BeO or one part of LiF to one part of graphite by weight. Adequate precautions had to be taken in the handling of the loose powders of BeO and LiF, as both materials are toxic, especially the unsintered BeO.

Pure acetone is added to the mixture of the powders, the resulting suspension ground into a slurry, and final dispersion enhanced by the blending action of a hypodermic syringe. The suspension is then deposited by syringe onto a reactor grade graphite disk 25 mm in diameter and 1.5 mm thick which has a central recess 0.75 mm deep and 19 mm in diameter. The acetone evaporates rapidly and there remains a smooth layer of a few milligrams of material covering the surface of the carrier recess. The sensitivity of the detector is independent of the thickness of the deposited layer in the limit that the surface of the recess is completely covered, but the layer is thin enough to withstand several heating cycles without loss of physical integrity.

The prepared samples are annealed for 10 to 20 minutes at 400°C prior to use. This temperature is well above the low-temperature peaks of LiF (∿150°C) and BeO (∿270°C) considered in this study. The heating eliminates the tribo-effects induced by the sample preparation and drives off the remaining acetone which would otherwise act as a quenching agent in the counting volume.

From a series of freshly prepared detectors, groups containing at least five samples were randomly selected and used for each data point. The reproduciability of these samples within the set was usually within ±10%, and each point in the graphite carrier data represents a minimum of three repeated runs with a set of five dosimeters. When it was found necessary to replace samples from a fresh batch of material, a calibration exposure with a ^{238}Pu alpha radiation source was used to compare the relative response of the new preparation. Appropriate correction factors were then applied to these data.

Metal-impregnated ceramic BeO detectors

Based on the results of previous studies on the preparation of TSEE detectors,[18,19] sintered BeO ceramic disks (Brush Thermalox 995), 1.25 cm in diameter and 1.5 mm thick, were coated with thin layers of gold (0.2 mg/cm^2 and 5 mg/cm^2) and platinium (0.2 mg/cm^2 and 2 mg/cm^2) by vacuum deposition. These metal coatings were then diffused along the grain boundaries of the multicrystalline material by prolonged heating in air.[18,19]

For the gold-coated samples, the heat treatment was for 65 hours at 800°C, followed by 18 hours at 1000°C. It is thought that the small metal concentration on the sensitive surface reduces its work function,

enhancing the ease of electron emission and, therefore, the sensitivity.[18] By diffusing the metals at temperatures which greatly exceed the normal TSEE reading temperature of the ceramics, little change in the concentration of metal atoms in the sensitive layer occurs during consecutive heating cycles.

After the diffusion process has been completed, the detectors were transferred to brass holders, previously heat treated at 650°C in air to establish a surface oxide layer, to simplify the handling of the samples. Since direct handling of the samples may produce "spurious" effects, the brass-framed dosimeters were annealed at 650°C for five minutes to eliminate any induced tribo-TSEE.

III. IRRADIATION PROCEDURES

Samples were exposed to x ray photons using a relatively heavy filtration in order to obtain a narrow spectral distribution in effective energy (Table V, Appendix). The effective energies chosen for this study were 22.4, 35.7, 57, 69, 115.5, and 158.7 keV respectively, using a wide beam geometry. No correction was necessary for the contribution of backscattering on the TSEE response, and adjustments for changes in the geometry of the procedure were considered negligible.

A Victoreen Model 70 Condenser R-Meter was used to measure each exposure; the readings were corrected for ambient temperature, barometric pressure and for the energy dependence of the chamber. Such readings can be considered accurate within approximately ±3%. Exposure rates have been selected which were well within the registration limits of the chambers and electronic equilibrium was established in air for each exposure.

A comparison of these results was made relative to the 1.25 MeV (effective) gamma radiation from a ^{60}Co source. The exposure rate was determined by the application of the radioactive decay law to a source standardization by the National Bureau of Standards. Samples were irradiated in air with electronic equilibrium being established by the several millimeters of plastic comprising the source holder.

For the determination of linearity of response, samples were irradiated with 0.66 MeV gamma radiation from a ^{137}Cs source, the exposure rate of which had been previously determined using glass and TLD dosimetry. In the environmental atomic number response studies, absorbers of varying atomic number (Be, Al, Cu, Sn, and Pb) and thicknesses as necessary for electronic equilibrium have been placed directly over the sample and exposed perpendicular to the radiation.

CHAPTER IV

RESULTS AND DISCUSSION

I. INITIAL STUDIES

Preliminary studies have been carried out on the effect of thermal pretreatment on the sensitivity of BeO and LiF in graphite-mixed detector preparations. It was thought that a heat treatment could affect an increase in the response and sensitivity of these TSEE materials by altering the surface structure or work function, or increasing the number of electron traps which contribute to the exoemission.

An additional advantage can be gained in the heat treatment of "unfired" BeO. The material (UOX, Brush Beryllium Co.) used mostly in this study is highly toxic. Allergic-type reactions to even minute amounts of BeO, if inhaled, result in some persons in a fatal lung disease, berylosis. Both inhalation probability and toxicity are reduced with increased particle size and a more compact surface structure. By heating (sintering) of the powdered BeO, the particles tend to clump together.

The sensitive material was heated for various times at constant temperatures (LiF at 800°C and BeO at 1000°C), and mixed with graphite for the detector preparation. An increase in TSEE sensitivity of a factor of two in LiF and a factor of four in BeO (Fig. 4) were found. There is no easy explanation for the dip observed in the BeO sensitivity versus heating time curve at 48 hours.

Because it was felt at the time that the heat treated BeO exhibited some spurious effects which could complicate experimental procedures, heat-sensitized samples were not used in subsequent experiments.

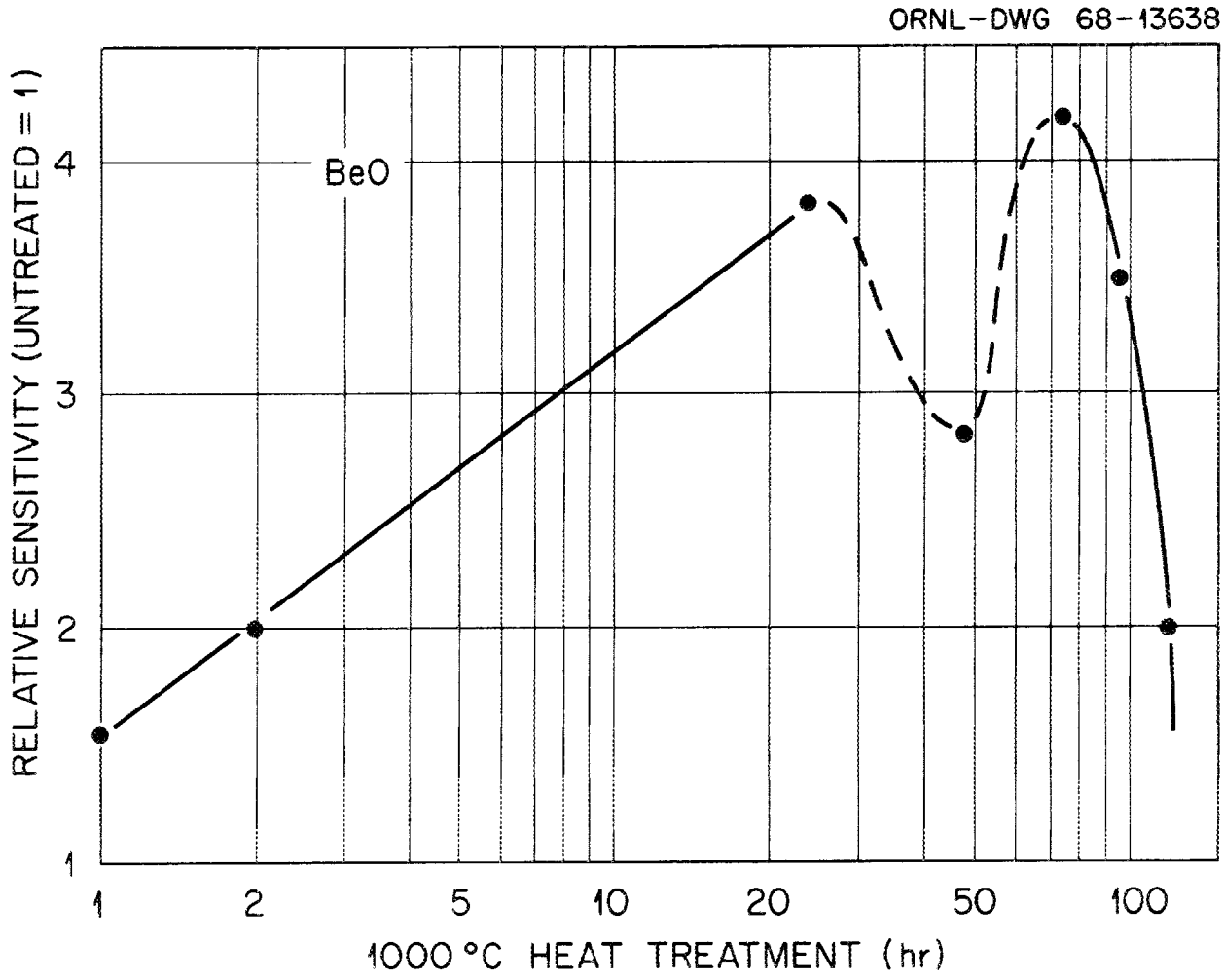

Fig. 4. Effects of thermal pre-treatment on the relative TSEE sensitivity of BeO (Brush UOX) in a graphite-mixed detector preparation.

The effects of particle size on the TSEE response was briefly considered. Fractions of LiF particles having diameter ranges greater than 149μ, between 149μ to 44μ, and smaller than 44μ were examined. No changes in radiation sensitivity were discernable in these preliminary measurements, but further analysis is warranted before definite conclusions can be drawn.

II. DOSE RESPONSE CHARACTERISTICS

The dose response of the graphite-plated detectors has been measured previously[10] and was found to be a linear function of dose. This was confirmed in preliminary experiments.

The sensitivity of ceramic TSEE detectors (Brush BeO Thermalox 995), expressed in total counts/R, are presented as a function of dose in a semi-log plot in Figures 5 and 6. The standard deviation of the mean ($\bar{\sigma}_{\bar{x}}$) of individual readings of the same detector was determined by

$$\bar{\sigma}_{\bar{x}} = \frac{1}{M} \sum_{j=1}^{M} \sigma_{\bar{x}^j}$$

where M = number of data points in each series of measurements. The results are given in Table II, Appendix.

It should be noted, however, that the actual surface mass densities of the deposited metal layers are not well known and cannot be reproduced with great precision. Each of these early metal-plated ceramic detectors was, therefore, unique.

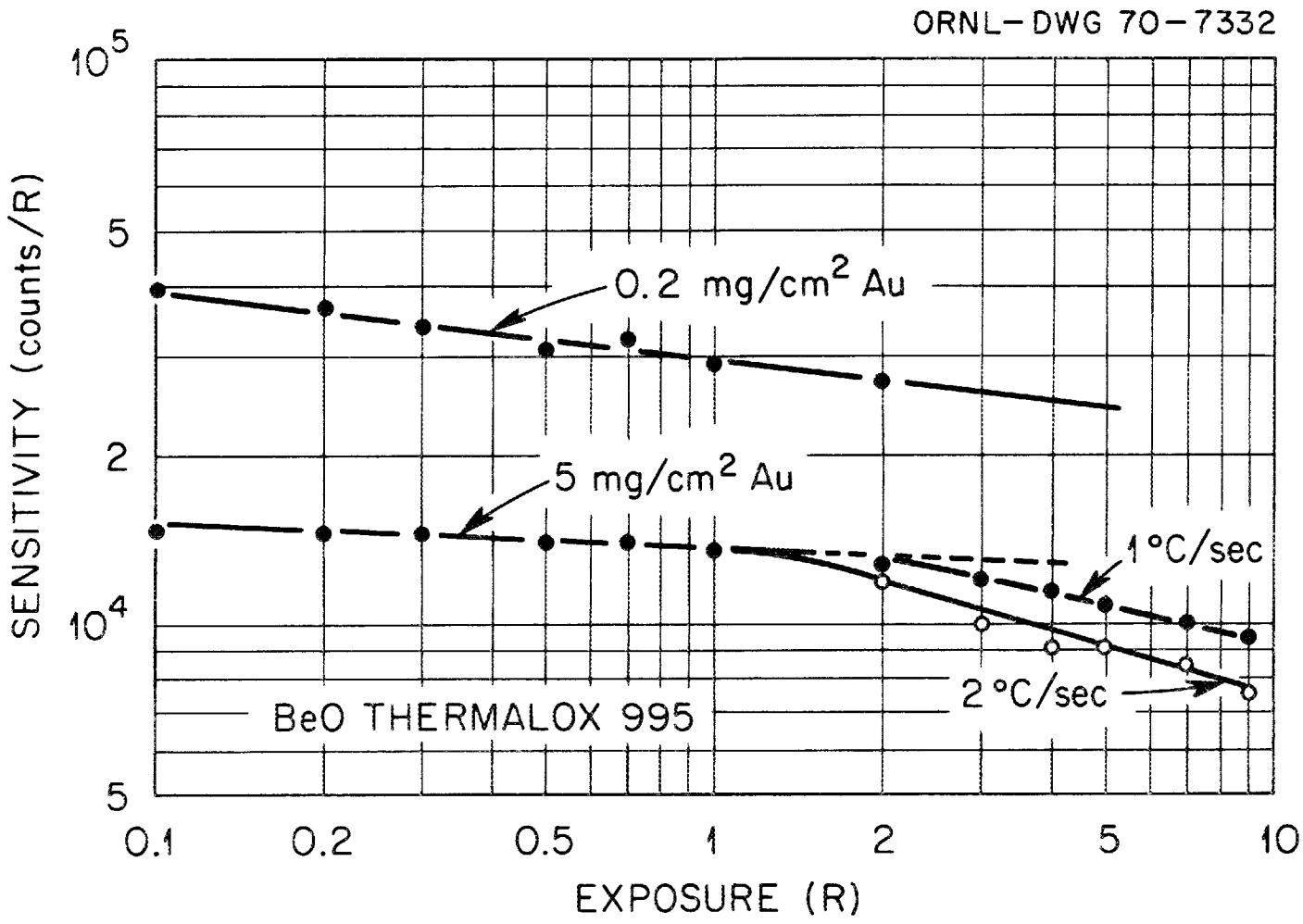

Fig. 5. The relative TSEE sensitivity of gold-treated Brush Thermalox 995 Ceramic BeO detectors as a function of gamma dose

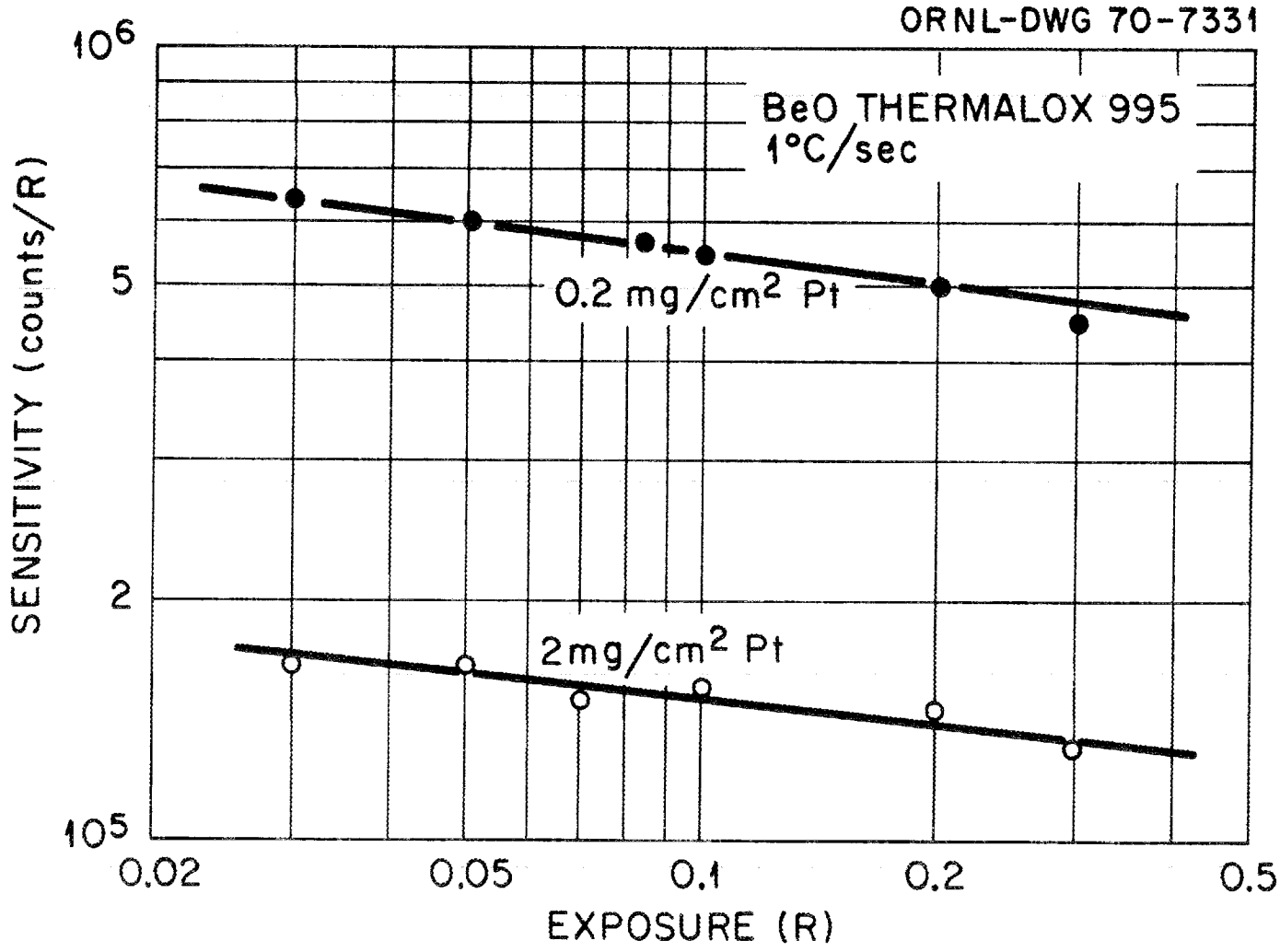

Fig. 6. The TSEE sensitivity of platinum-treated Brush Thermalox 995 Ceramic BeO detectors as a function of gamma dose

III. ENERGY RESPONSE CHARACTERISTICS

At present, there has been only one published work on the energy dependence of a TSEE material. This was done by J. Kramer[31] in 1968 when he examined the photon energy response of a BeO-graphite mixture.

The sensitivities of the graphite-mixed and ceramic detectors are presented as a function of effective energy in a semi-log plot in Figures 7, 8, 9, and 10. These data were normalized to the 1.25 MeV (effective) gamma radiation from ^{60}Co. The calculated curves of the constituents of the detectors (graphite, BeO, and LiF) are from a previous work by Becker.[32] An anomalous and as yet unexplainable result was found for the BeO-graphite detectors: Between 50 and 120 keV, the experimental response of the BeO/C mixture was consistently lower than the calculated response for each of the constituents.

The error bars for the graphite carrier data were determined by averaging the response deviations of each detector set from the mean. They are shown on the curves in Figures 7 and 8. The standard deviation of the ceramics was determined as previously described. The results are shown in Table III, Appendix. One should, however, realize that the actual experimental errors exceed the statistical fluctuations indicated by the error bars due to additional inaccuracies in the dose determination, etc.

The data for ceramic detectors followed the generally predictable result of an over-sensitivity in the low energy portion of the curve due to the high atomic number of the diffused metal. In this region the mass-energy absorption coefficients are predominated by the photoelectric cross-section which varies as the fourth power of the atomic number of the material.

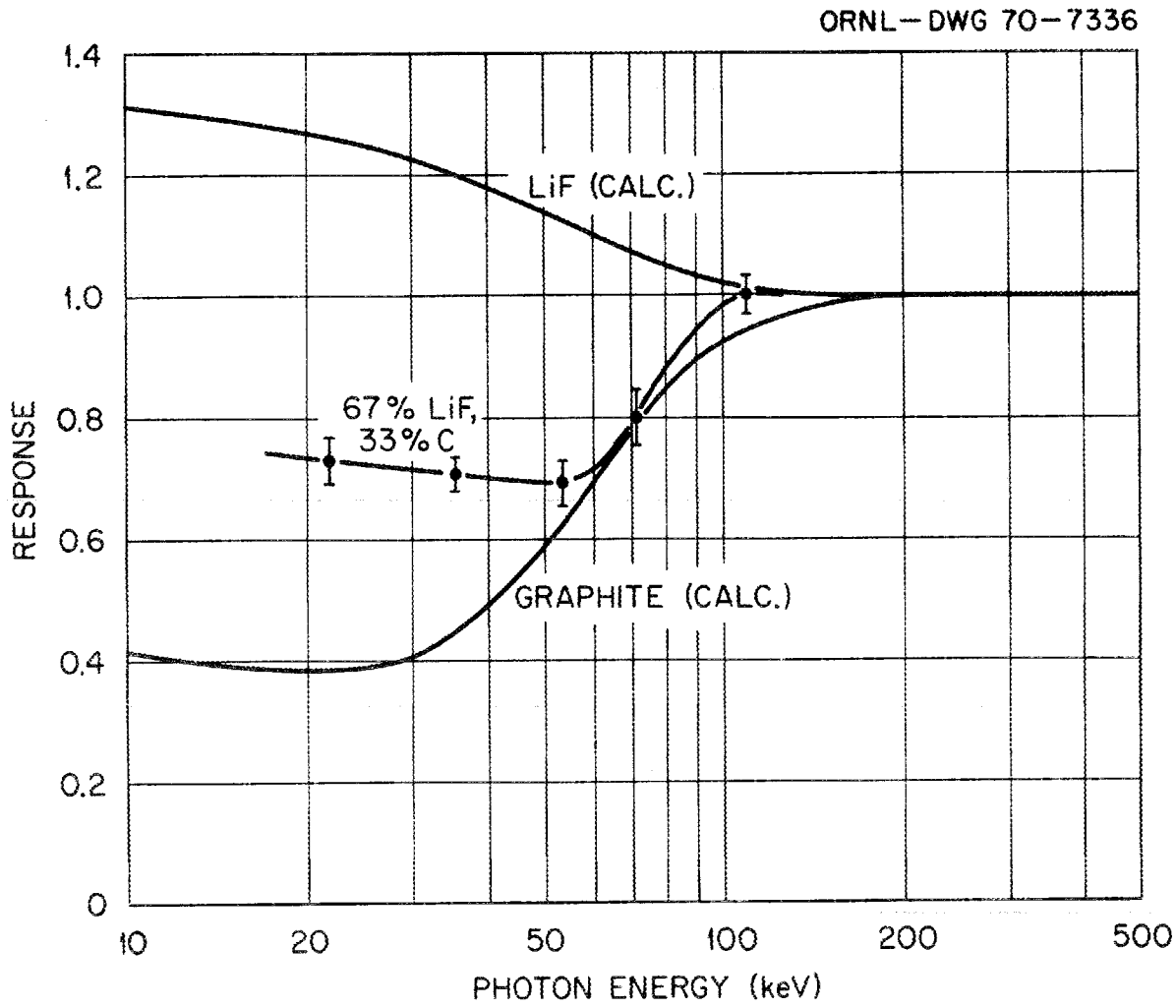

Fig. 7. Energy dependence of a LiF graphite mixture, normalized for ^{60}Co gamma radiation, and calculated energy response of LiF on C (after 32)

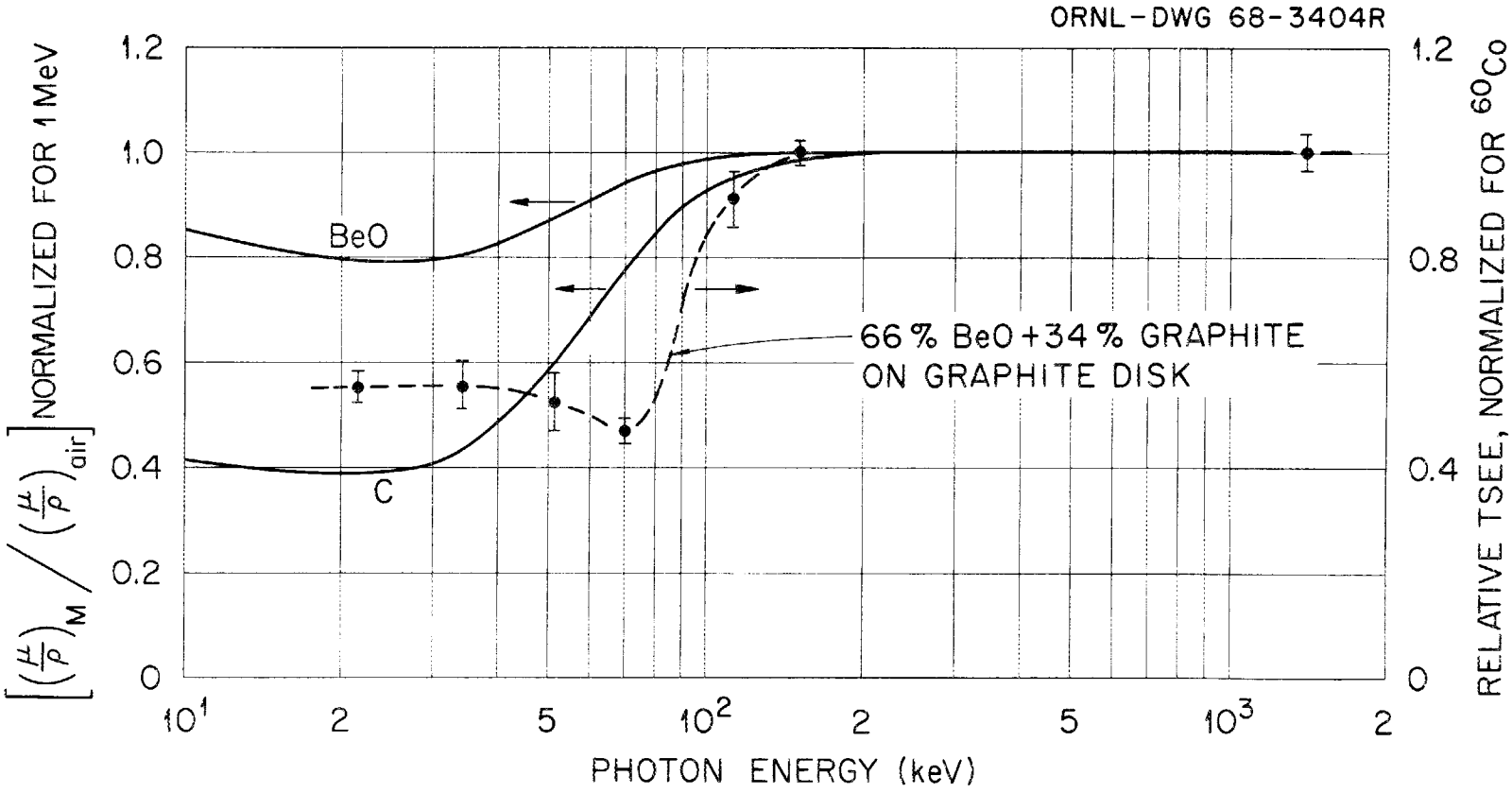

Fig. 8. Photon energy dependence of BeO graphite mixture, normalized for ^{60}Co, and calculated energy response of BeO and C (after 32)

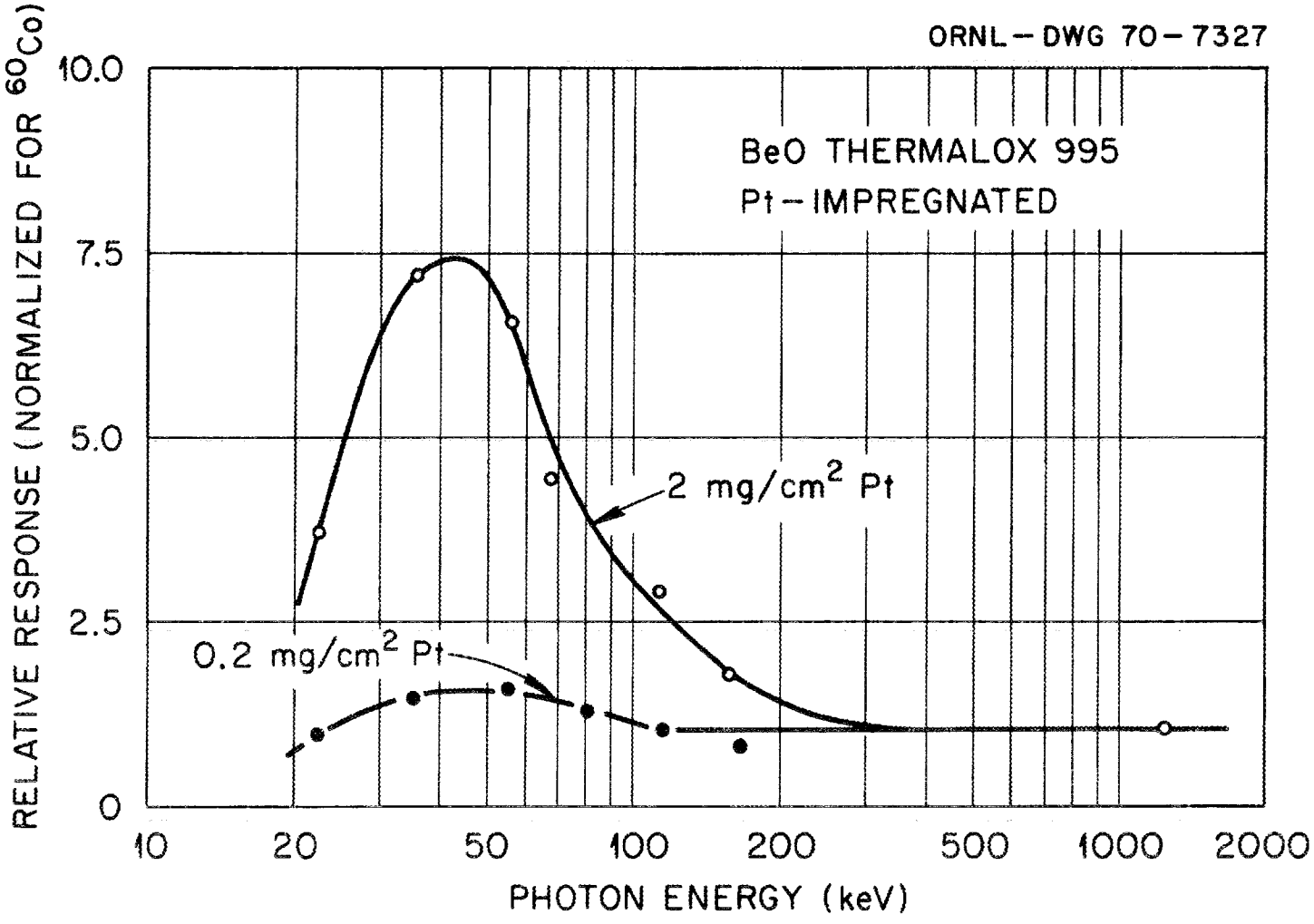

Fig. 9. Photon energy dependence of Pt treated Brush Thermalox 995 Ceramic BeO, normalized for ^{60}Co

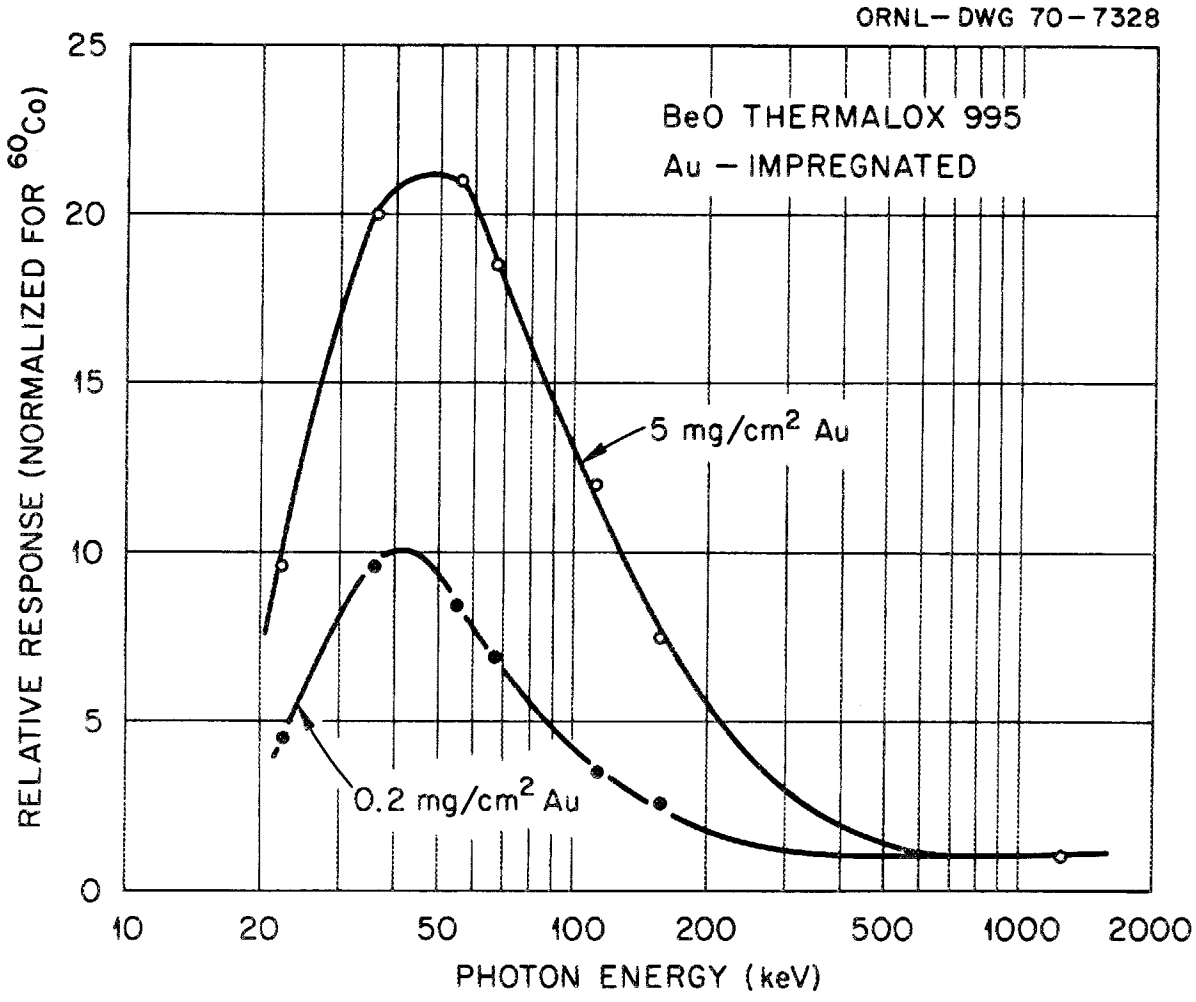

Fig. 10. Photon energy dependence of response of Au-treated Brush Thermalox 995 Ceramic BeO, normalized for ^{60}Cs

After performing the energy dependence studies with the ceramics, it was felt that the perturbation of the radiation effect due to the brass holder could have altered the radiation response of the detectors. To determine the magnitude of this effect, a new holder was made from graphite (Z=6), the detector inserted, and the experiment repeated. A correction of 1.6 in the energy region of maximum oversensitivity was found and these corrections were applied to the response curves for the ceramics. Additional corrections to the curves were applied to account for the non-linearity of the dose response.

IV. ENVIRONMENTAL ATOMIC NUMBER RESPONSE CHARACTERISTICS

The TSEE response to ^{137}Cs gamma radiation of LiF and BeO in graphite-mixed detector preparations is shown as a function of the atomic number of the cover layer in a semi-log plot in Fig. 11. The response is shown in relative units (the BeO detectors exceeded the sensitivity of the LiF by approximately a factor of 10). The standard deviations of detector response are given in Table IV, Appendix.

At the incident photon energy studied, 0.66 MeV, the Compton cross-section predominates the mass energy-absorption coefficients. In the range of energies from 0.1 to 1 MeV, the mass energy-absorption coefficients will, consequently, vary as the ratio of the atomic number divided by the atomic weight, Z/A. The total absorbed dose, D (and to a first approximation the TSEE response) would be expected to vary as,

$$D_T \propto \frac{(Z/A)_T}{(Z/A)_{air}}$$

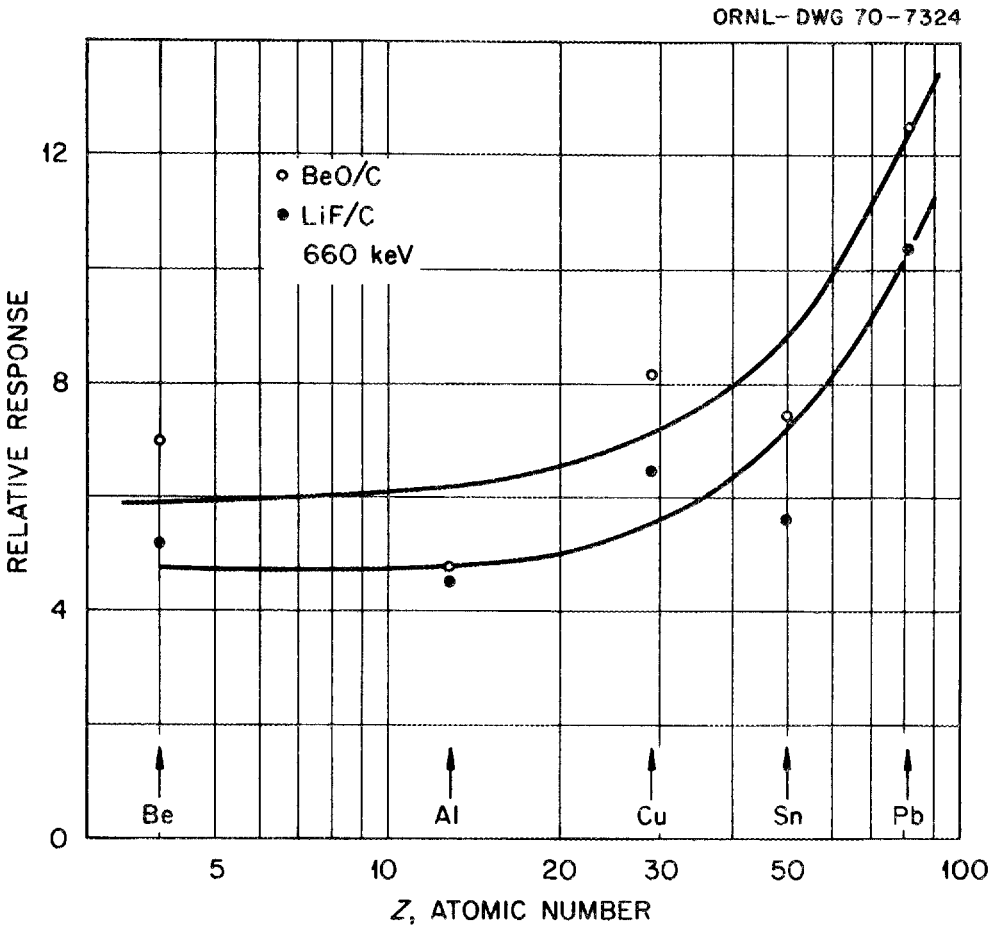

Fig. 11. Effect of the atomic number of the covering material on the relative TSEE response of BeO/C and LiF/C to ^{137}Cs radiation

Little variation of the absorbed dose with energy or atomic number is, therefore, expected in this energy region. The ratio of the atomic number to the atomic weight of the covering material comprising the detector's immediate environment varies by approximately 20 per cent from Be to Pb. Since the composition of the detector did not change, the total absorbed dose was not expected to show a marked variation and, consequently, no great change in the TSEE response should result.

This is in contrast to the experimental results (Fig. 11). Extrapolation of the curve to an atomic number of "zero" should give a measure of the contribution of the electrons liberated in the detector to the total TSEE response at 660 keV. The environmental contribution can be estimated as follows: (1) Be, 0%; (2) Al, 5%; (3) Cu, 13%; (4) Sn, 31%, and (5) Pb, 52%. The results may be complicated by the degraded secondary photons which may be in an energy region where photoelectric processes dominate. In this energy range the absorbed dose varies as the fourth power of the ratios of the effective atomic number of the detector and environment to that of air.

$$D_T \propto \frac{(Z_{eff})_m^4}{(Z_{air})^4}$$

V. CONCLUSIONS

Initial studies on the thermal pre-treatment of LiF and BeO resulted in some improvements in their sensitivity. Only slow changes, if any, can be expected to occur in heat-treated material's sensitivity over many usages, if the treating temperature was significantly higher than the normal reading temperature of the detectors.

Increasing the thickness of a thin Pt or Au layer on the BeO surface results in a more linear dose response accompanied with a reduction in detector sensitivity. One possible explanation is that the probability of an exoelectron penetrating the thicker metal layer is reduced, resulting in a decrease in detector sensitivity. The improvements in linearity can be attributed to the reduction of local charge effects at the dosimeter surface during reading.

The energy dependence studies showed some variation from the expected behavior: In both LiF and BeO, the resulting energy dependence curve is lower that would be expected from the energy dependence calculated for the constituents. By coating with very small amounts of high-Z metals, the energy response of BeO can be adjusted to simulate that of air over a large range of photon energy.

Efforts to separate from the total TSEE response the components related to the detector and its immediate environment have been partially successful. As expected, the contributions of the environmental components are largely independent of the type of sensitive material.

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APPENDIX

TABLES

Table I. Impurities in BeO and LiF[17]

Element (in ppm by wt.)	Brush BeO UOX	Element (ppm wt.)	LiF J. T. Baker "analyzed reagent"
Al	4	Al	7
Ca	4	Ba	___
Cr	4	Cr	5
Cu	___	Ca	7
Fe	___	Cu	20
Mg	4	Fe	___
Mn	___	Mg	10
Si	___	Mn	___
Sr	1	Si	100
Ti	___	Sr	2
Zn	___	Ti	___
Zr	___	V	10

Table II. Dose Response Standard Deviation of Brush Thermalox 995

Metal	Pt	Pt	Au	Au
Surface Mass Density (mg/cm^2)	0.2	2.0	0.2	5.0
$\overline{\sigma}_{\bar{x}}$ (%)	1.8	1.9	2.5	1.3

Table III. Energy Response Standard Deviation of Brush Thermalox 995

Metal	Pt	Pt	Au	Au
Surface Mass Density (mg/cm^2)	0.2	2.0	0.2	5.0
$\overline{\sigma}_{\bar{x}}$ (%)	2.1	2.4	2.5	1.4

Table IV. Environmental Atomic Number Response Standard Deviation of BeO and LiF

Detector	$\overline{\sigma}_{\bar{x}}$
LiF-graphite	3.3
BeO-graphite	2.8

Table V. Kilovolt Settings, Standard Filtration, and Effective Energy[30]

X-Ray Machine Dial Setting (keV)	Filtration (mg/cm^2)*				Effective Energy (keV)
	Lead	Tin	Copper	Aluminum	
238	2,363	861	959	279	200.9
188	716	1,863	929	279	158.7
138	---	913	2,790	279	115.5
92	548	---	---	279	68.6
44	---	---	233	133	37.7
18	---	---	---	133	22.4

*Inherent tube filtration equivalent to 661 mg/cm^2 of Aluminum as determined from absorption curves with no added filtration, using data given by Taylor in "Physical Foundations of Radiology" by Glasser, O., Quimby, E. H., Taylor, L. S., and Weatherwax, J. L., (Paul B. Hoeber, Inc., New York, Second Edition, 1952) pp. 245-249

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