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SOME X-RAY AND FAST NEUTRON RESPONSE
CHARACTERISTICS OF SILVER METAPHOSPHATE

GLASS DOSIMETERS

W. T. Thornton
J. A. Auxier

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SOME X-RAY AND FAST NEUTRON RESPONSE CHARACTERISTICS
OF SILVER METAPHOSPHATE GLASS DOSIMETERS

W. T. Thornton and J. A. Auxier

HEALTH PHYSICS DIVISION

Submitted as a thesis to the Faculty of the Graduate
School of Vanderbilt University in partial fulfill-
ment of the requirements for the degree of MASTER OF
SCIENCE in Physics.

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TABLE OF CONTENTS

	Page
ACKNOWLEDGMENTS.....	ii
LIST OF TABLES.....	iv
LIST OF FIGURES.....	v
I. INTRODUCTION AND OBJECTIVES.....	1
II. THEORY.....	3
Radiophotoluminescence of Silver Metaphosphate Glass Calculation of X- and Gamma-Ray Response Calculation of Neutron Energy Absorption by Glass Rods	
III. EXPERIMENTAL APPARATUS AND PROCEDURES.....	24
Fluorimeter Glass Techniques Co ⁶⁰ Calibration Source X-Ray Equipment and Techniques Van de Graaff Experiment	
IV. PRESENTATION OF DATA	32
X-Ray Response Fast Neutron Response Determination of Sensitivity to Gamma Radiation	
V. ANALYSIS AND DISCUSSION	42
Discussion of X-Ray Response Discussion of Experimental and Calculated Deviations in Response of the Shielded Dosimeter as a Function of Photon Energy Analysis of Fast Neutron Response	
VI. CONCLUSIONS AND RECOMMENDATIONS.....	50
BIBLIOGRAPHY.....	53

LIST OF TABLES

Table	Page
1. Sample Calculation of Factors for Evaluating the Integral in Eq. (6)	11
2. Mass Absorption Cross Sections for Glass and Air	13
3. Composition of Glass	14
4. Composition of Air	15
5. Calculated Response of the Glass Rods as a Function of Photon Energy for a Tin-Tantalum Shield	19
6. Effective Neutron Cross Sections for Glass	21
7. Energy Absorbed in Tissue and Glass as a Function of Neutron Energy	23
8. Effective Neutron Cross Sections for Tissue	48

LIST OF FIGURES

Figure	Page
1. Energy Level Diagram for Real and Ideal Crystals	5
2. Diagram of Model for Energy Absorption Calculations	9
3. Comparison of the Theoretical and Experimental Photon Response of the Unshielded Glass	17
4. Microdosimeter Reader - Optical Layout	25
5. Response of Glass with Cadmium and Teflon Shield	33
6. Response of Glass with Tantalum and Teflon Shield	34
7. Response of Glass with Platinum and Teflon Shield	35
8. Response of Glass with 0.015-Inch Tantalum Plus Tin Plus Teflon Shield	37
9. Response of Glass with 0.020-Inch Tantalum Plus Tin Plus Teflon Shield	38
10. Neutron Response of Glass in Various Containers	39
11. Neutron Response of Glass Relative to Co ⁶⁰ Gamma Response	40
12. Sensitivity of Glass to Gamma Radiation	41
13. Filtered X-Ray Spectra Normalized to Unit Dose	43
14. Representation of $R_{E_{eff}}$ as an Area	44
15. Comparison of Theoretical and Experimental Gamma Responses of the Shielded Glass	51

I. INTRODUCTION AND OBJECTIVES

Measurement of the gamma-ray component of radiation fields of co-existent fast neutrons and gamma rays presents some of the most difficult problems in radiation dosimetry. The measurements are particularly difficult for gamma radiation exposure doses intermixed with equal or greater levels of fast neutrons because the neutron response of gamma detectors, such as ion chambers, usually is a linear function of flux or dose. A counting device has been reported for gamma dose rates of up to about 50 r/hr which has a low response to fast neutrons.¹ Anhydrous chemical systems^{2,3} have been used for measuring integrated doses greater than about 25 r, including doses of nuclear weapons radiation. However, the chemical dosimeters have several limitations; they are difficult to manufacture reproducibly, they require elaborate reader systems and techniques, and they are not commercially available.

A small metaphosphate glass detector has been studied by Schulman^{4,5} and others.⁶⁻⁸ Recent developments include a commercially

¹J. A. Auxier, G. S. Hurst, and R. E. Zedler, Health Phys. 1, 21 (1958).

²S. C. Sigoloff, Nucleonics 14, No. 10, 54 (1956).

³D. B. Ott, Los Alamos Scientific Laboratory Report LA-2249 (1959).

⁴J. H. Schulman, R. J. Ginther, and C. C. Klick, J. Appl. Phys. 22, 1479 (1951).

⁵J. H. Schulman and H. W. Etzel, Science 118, 184 (1953).

⁶N. J. Kreidl and G. E. Blair, Nucleonics 14, No. 3, 82 (1956).

⁷H. W. Etzel, R. D. Kirk, and J. H. Schulman, Ra-Det 8, No. 2, 49 (1955).

⁸A. L. Riegart, H. E. Johns, and J. W. T. Spinks, Nucleonics 14, No. 11 134 (1956).

available reader suitable for 1 mm diameter by 6 mm long glass rods.⁹ The glass detectors offered a means of overcoming a large part of the mixed radiation field problem if the neutron response were sufficiently low; the absence of elements of low atomic number indicated that this might be the case because the maximum energy transferable per neutron atom collision is an inverse function of atomic weight. A method for decreasing the adverse photon energy dependence in the photoelectric region⁸ was also needed.

In general, the objective of this study was to determine the adequacy of the metaphosphate glass as a dosimeter for gamma radiation coexistent with fast neutrons. Specific objectives were to determine the fast neutron response in the energy region encompassing a major portion of the fission spectrum and to develop a method of decreasing the photon energy dependence. The improvement in the response of this glass as a function of photon energy, in order to be very useful, should be such that errors in measuring gamma radiation in the energy region of 100 kev to 3 Mev would not exceed 10%.

⁹Manufactured by Bausch and Lomb Optical Company, Rochester, N. Y.

II. THEORY

Radiophotoluminescence of Silver Metaphosphate Glass

Although this paper deals specifically with the radiophotoluminescence of silver-activated metaphosphate glass, many substances exhibit radioluminescent properties. Metals such as sodium and mercury represent the simplest type of fluorescence centers. In the gaseous state and at sufficiently low pressure, these atoms exhibit resonance fluorescence, i.e. wave lengths characteristic of the atomic energy levels, emitting a spectrum of discrete lines. These lines represent the permitted transitions from various energy levels of electronic excitation to the ground state. The spectrum, however, loses its discrete characteristic when the fluorescence center is too much disturbed by energy interaction with neighboring atoms. Studies of the fluorescence phenomenon have revealed that fluorescence occurs only when the atom absorbing the exciting radiation is in an "energetically isolated" state¹⁰ because isolation prevents energy dissipation in the form of additional thermal motion.

Energy isolation of this sort is easily obtained for metals in the gaseous state at low pressure, but it is of interest here to understand the interaction processes when fluorescence centers are formed in the lattice network of a crystalline substance. Dauvillier¹¹ in 1920

¹⁰W. A. Weyl, *Ind. and Eng. Chem.* 34, No. 9, 1035 (1942).

¹¹M. A. Dauvillier, *Compt. Rendus Acad. Sci.* 171, 627 (1920).

and Przibram¹² in 1923 suggested that luminescence could be produced by the reduction of metal ions to neutron metal atoms by electrons ejected from anions in the lattice. Then Przibram noticed the importance of lattice defects in luminescence production. In ideal crystals there are forbidden energy zones between the permitted energy levels, and due to this periodic field of potential, irradiation in the ultraviolet absorption band has no permanent effect on the crystal. For example, an anion in an alkali-halide crystal loses an electron and is changed into a halogen atom or "positive hole". Such an electron immediately returns to its positive hole; therefore, there is no permanent effect on the crystal. The introduction of impurities or imperfections into the lattice structure disturb the periodicity of the lattice structure and produce localized energy levels which in ideal crystals would lie in the forbidden zone. The energy levels for ideal crystals and for real crystals having impurities are illustrated in Fig. 1. Electrons raised to the conduction band can fall back into these defect levels and become bound to the defect. Hence, an impurity bearing crystal may be changed by radiation and possess a changed absorption spectrum.

Schulman et al.⁴ describe the radiophotoluminescent system used in this research. They use Przibram's definition, "...that phenomenon where a material, originally non-luminescent under UV or visible

¹²K. Przibram, Z. Phys. 20, 196 (1923).

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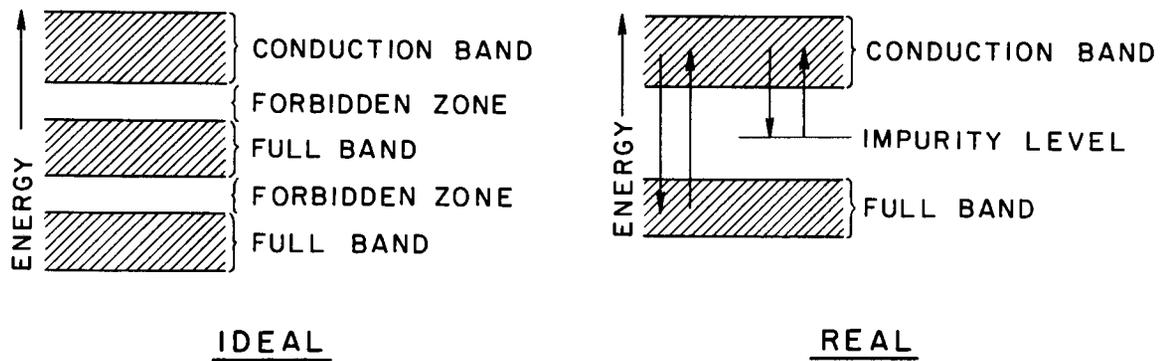


Fig. 1. Energy Level Diagrams for Real and Ideal Crystals.

light, is made responsive to such excitation by pre-treatment with gamma rays or X rays", with the further distinction that the newly created stable luminescent centers would not be destroyed by the UV radiation that is subsequently used to excite them. Silver is the impurity introduced into the metaphosphate glass. In the pre-irradiation state, it exists as Ag^+ ion, and there is evidence¹³ that from its interstitial lattice position its trapped orbital electrons extend over several interatomic distances. This is compared with the post-irradiation state of the silver which is atomic silver. Now the electron orbits are smaller and in their interstitial position a sufficient degree of energy isolation is achieved for luminescence. It is reasonable to expect, therefore, that the absorption spectrum of the pre- and post-irradiated silver metaphosphate glass will be different since the amount of energy required to release the trapped orbital electron of the Ag^+ ion is considerably different from the energy necessary to elevate electrons from atomic silver to the conduction band.

It is not definitely established that just any Ag^+ ion in the lattice is eligible to become a luminescence center upon irradiation, or that the Ag^+ ion must be located adjacent to an anion vacancy. In the latter case the atomic silver center would be closely analogous to the normal F-center, an electron hole adjacent to an anion vacancy in the lattice.

¹³W. A. Weyl, J. H. Schulman, R. J. Ginther, and L. W. Evans, Elec. Chem. Soc. J. 95, No. 2, 70 (1949).

X- or gamma-radiation induced changes in the absorption and emission spectra of silver phosphate glass have been noticed by Schulman et al.¹⁴ Prior to irradiation the absorption peak is 2400 Å and after irradiation it becomes 3300 Å. A similar change occurs in the emission spectrum. The peak shifts from 3700 Å to about 6400 Å. The intensity of the resulting orange luminescence is proportional to the number of atomic silver centers formed and hence to the radiation dose received by the glass.

Calculation of X- and Gamma-Ray Response

It is assumed that the glass response is proportional to the energy deposited in it by electrons. If other factors, such as the number of lattice defects and the density of these defects, are maintained constant, then, based on discussion in the previous section and because the spectrum for tertiary electrons is an insensitive function of photon energy,¹⁵ the number of tertiary electrons produced is directly proportional to the photon energy absorbed. Therefore, the number of atomic silver centers formed and the response of the glass is proportional to the energy flux of Compton secondary electrons.

To calculate the energy absorbed in the glass, assume a monoenergetic beam of photons of initial intensity I_0 in Mev/cm^2 , incident on the glass and a subsequent arbitrary transmitted monoenergetic

¹⁴J. H. Schulman, W. Schurcliff, R. J. Ginther, and F. H. Attix, *Nucleonics* 11, No. 10, 52 (1953).

¹⁵J. R. Greening, *Brit. J. Radiol.* 30, 254 (1957).

intensity I . Under narrow beam geometrical conditions, assuming that the presence of the small glass rod does not perturb the field significantly, the energy absorbed per roentgen exposure is inversely proportional to $[(\tau + \sigma_{\alpha})/\rho]_{\text{air}}$, the true mass absorption coefficient for air in cm^2/g at the energies discussed (τ is the photoelectric cross section, σ_{α} is the Compton absorption cross section, and ρ is the density).

From the following argument, illustrated in Fig. 2, an equation for the energy absorbed in a glass rod may be derived. E_a will represent the absorbed energy. Imagine an infinite number of strips of thickness ΔX_i and length L making up the glass rod. Then let I_i be the intensity transmitted from a given strip, and

$$I_0 - I_i = \Delta I_i \quad (1)$$

The change in intensity may be expressed as

$$\Delta I_i = I_0 \left(1 - e^{-[(\tau + \sigma_{\alpha})/\rho]2y} \right) \quad (2)$$

where an integration along the photon path has already been carried out by assuming that there is no net contribution of photons scattered into or out of a given strip of volume element $2yL\Delta X_i$. The glass density is given by ρ . Then, assuming that the Compton and photoelectrons lose all their energy in the volume element,

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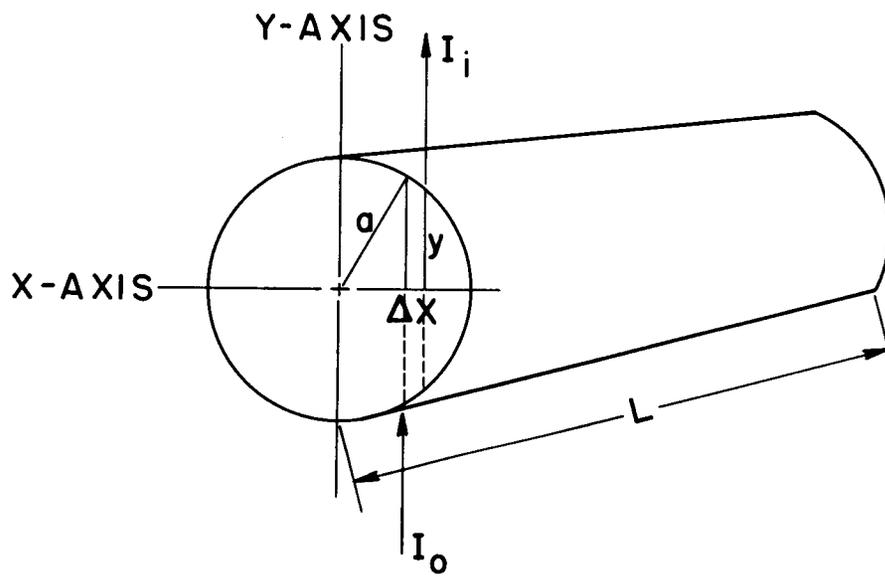


Fig. 2. Model for Energy Absorption Calculation.

$$E_a = L \sum_i \Delta I_i \Delta X_i \quad (3)$$

Using the representation in Fig. 2, an integral expression of the energy absorbed may be formulated.

$$E_a = 2LI_o \int_0^a \left(1 - e^{-(\tau + \sigma_\alpha)2y} \right) dx \quad (4)$$

Upon substituting, the integral becomes

$$E_a = 2I_o L \int_0^a \left(1 - e^{-(\tau + \sigma_\alpha) 2\sqrt{a^2 - x^2}} \right) dx \quad (5)$$

To solve, let

$$x = a\omega$$

$$dx = a d\omega$$

The change in variables gives

$$E_a = 2I_o La \int_0^1 \left(1 - e^{-2(\tau + \sigma_\alpha) a\sqrt{1 - \omega^2}} \right) d\omega \quad (6)$$

The integral may be evaluated only by an approximation method.

Table 1 shows numerical values for the increments of the integral of Eq. (6) for 1.0 Mev photons. In this sample calculation the following constants and equivalents were used:

Table 1
 SAMPLE CALCULATION OF FACTORS FOR EVALUATING
 THE INTEGRAL IN EQ. (6)

ω	ω^2	$1 - \omega^2$	$\sqrt{1 - \omega^2}$	K	$1 - e^{-K}$
0.0	0.00	1.00	1.000	0.00754	0.00754
0.1	0.01	0.99	0.996	0.00751	0.00751
0.2	0.04	0.96	0.981	0.00740	0.00740
0.3	0.09	0.91	0.955	0.00720	0.00720
0.4	0.16	0.84	0.918	0.00692	0.00692
0.5	0.25	0.75	0.867	0.00654	0.00654
0.6	0.36	0.64	0.801	0.00604	0.00604
0.7	0.49	0.51	0.715	0.00539	0.00539
0.8	0.64	0.36	0.601	0.00453	0.00453
0.9	0.81	0.19	0.436	0.00329	0.00329
1.0	1.00	0.00	0.000	0.00000	0.00000
$\sum_{\omega=0}^{\omega=1} 1 - e^{-K}$					0.06236

$$(\tau + \sigma_{\alpha})/\rho = 0.0275 \text{ cm}^2/\text{g}$$

$$a = 0.05 \text{ cm}$$

$$\rho = 0.74 \text{ g/cm}^3$$

$$2a(\tau + \sigma_{\alpha}) = 0.00754$$

$$K = 2(\tau + \sigma_{\alpha}) a \sqrt{1 - \omega^2} \quad (7)$$

If

$$d\omega = 0.1 \quad (8)$$

then

$$\int_{\omega=0}^{\omega=1} \left(1 - e^{-2(\tau + \sigma_{\alpha}) a \sqrt{1 - \omega^2}} \right) d\omega = \sum_{\omega=0}^{\omega=1} \left(1 - e^{-2(\tau + \sigma_{\alpha}) a \sqrt{1 - \omega^2}} \right) d\omega$$

$$= 0.006236 \quad (9)$$

Substituting this value into Eq. (6),

$$E_a = 2LI_0 a \times 0.006236 \text{ Mev}$$

$$= 0.0006236 LI_0 \text{ Mev}$$

which is the energy absorbed in the glass from a 1.0 Mev photon.

Similar calculations have been made for other energies.

Table 2 is a compilation of $(\tau + \sigma_{\alpha})/\rho$ for glass and $(\tau + \sigma_{\alpha})/\rho$ for air as a function of energy. Table 3 shows the per cent by weight of each element in the glass, and Table 4 shows the per cent by weight of each element for air.

Table 2
 MASS ABSORPTION CROSS SECTIONS FOR GLASS AND AIR

$h\nu$ (Mev)	$[(\tau + \sigma_{\alpha})/\rho]_{\text{glass}}$ (cm^2/g)	$[(\tau + \sigma_{\alpha})/\rho]_{\text{air}}$ (cm^2/g)
0.020	5.7264	0.5136
0.030	2.0900	0.1464
0.040	3.5103	0.0637
0.050	1.9242	0.0388
0.060	1.1865	0.0289
0.080	0.5443	0.0242
0.100	0.3011	0.0234
0.150	0.1103	0.0252
0.200	0.0643	0.0270
1.000	0.0275	0.0279

Table 3
COMPOSITION OF GLASS

Element	Per Cent by Weight ^a
Ba	10.760
K	07.650
Al	04.734
Ag	04.278
P	28.480
O	44.101

^aBased on the composition advertised by Bausch and Lomb.

Table 4
COMPOSITION OF AIR

Element	Per Cent by Weight ¹⁶
O	23.15
N	75.51
A	1.28

¹⁶F. A. Berry, Jr., E. Bollay, and N. R. Beers, Handbook of Meterology (McGraw-Hill Book Co., Inc., New York, 1945), p. 351.

After E_a has been calculated, $R_b(E)$, the response relative to the 1.0 Mev response, is given by

$$R_b(E) = \left(\frac{E_a}{[(\tau + \sigma_\alpha)/\rho]_{\text{air}}} \right)_{E_\gamma} \div \left(\frac{E_a}{[(\tau + \sigma_\alpha)/\rho]_{\text{air}}} \right)_{E_\gamma = 1.0 \text{ Mev}} \quad (10)$$

The response calculated using this method is shown in Fig. 3.

The preceding method was also followed in calculating the response to radiation of the glass with filters surrounding it to reduce the response in the photoelectric region. It is assumed, in calculating the response of the shielded glass, that the X-ray spectrum does not change upon passing through the shield. The geometry considerations are simplified by calculating the energy absorbed in an infinite slab of metal of a given thickness rather than for the actual case of the cylindrical can in which the glass was exposed. A 0.015-inch teflon liner was used to absorb the photoelectrons from the metal cylinder and was not taken into account in the calculation of gamma absorption in the filter since its absorption of gamma rays was negligible.

With these simplifying assumptions, the relative response of the shielded glass per roentgen, $R_s(E)$, at photon energy, E_γ , is given by

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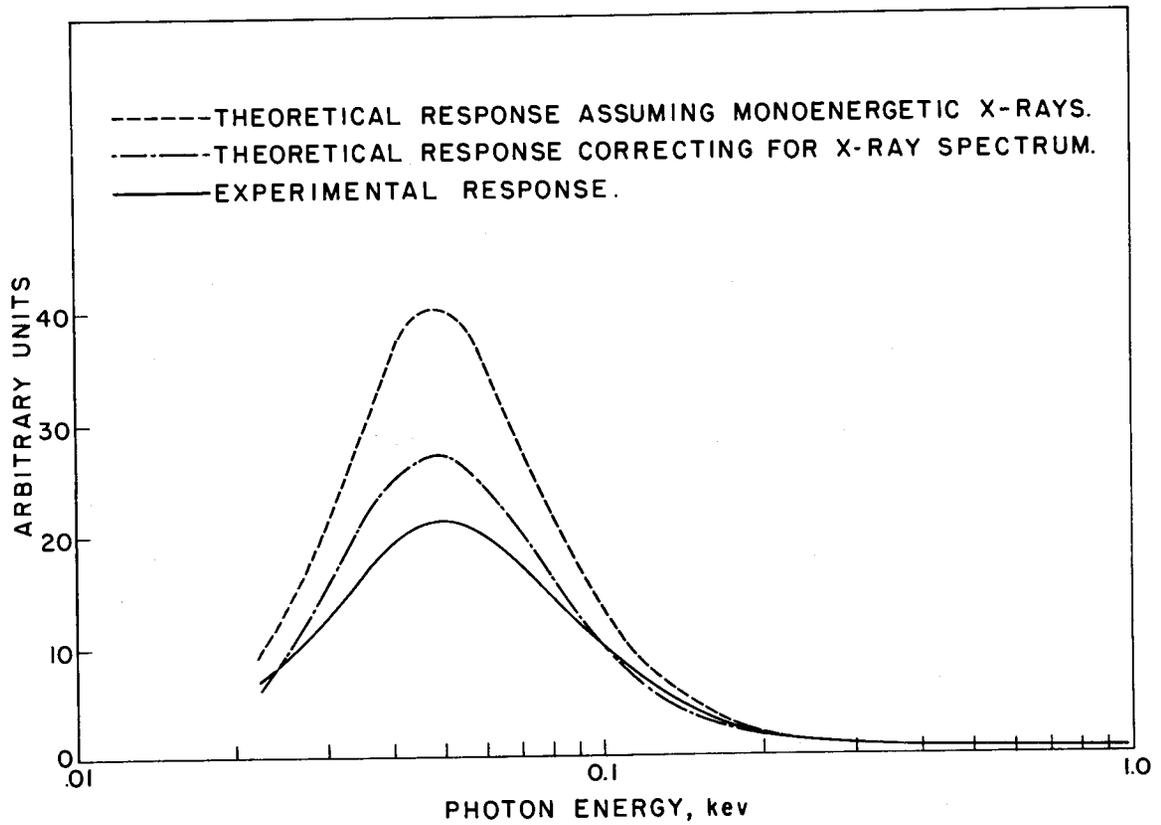


Fig. 3. Comparison of the Theoretical and Experimental Photon Response of the Unshielded Glass.

$$R_s(E) = \left[\left(\frac{E_a}{[(\tau + \sigma_\alpha)/\rho]_{air}} \right) e^{-(\tau + \sigma_\alpha)_f X_f} \right]_{E_\gamma}$$

$$\div \left[\left(\frac{E_a}{[(\tau + \sigma_\alpha)/\rho]_{air}} \right) e^{-(\tau + \sigma_\alpha)_f X_f} \right]_{E_\gamma = 1.0 \text{ Mev}} \quad (11)$$

where

$(\tau + \sigma_\alpha)_f$ = true absorption coefficient of filter, and

X_f = filter thickness in cm^2/g .

The calculated response as a function of photon energy, neglecting the effect of geometry and scattering, is given in Table 5.

Calculation of Neutron Energy Absorption by Glass Rods

It is assumed that the absorbed energy (ergs per gram of glass) is proportional to the effective total neutron cross section, $\sigma_{T_{eff}}$, of the glass. Also, the assumption is made that $\sigma_{T_{eff}}$ is due to the elastic scattering cross section; this cross section is predominant in the 0.5 to 1.5 Mev region. At this point an explanation of the term "effective total neutron cross section" is necessary. It is the summation of the total neutron cross section of each element in the glass weighted by the relative number of atoms of the element and the fraction of the average neutron energy imparted to the recoil nucleus.

Table 5

CALCULATED RESPONSE OF THE GLASS RODS AS A FUNCTION OF PHOTON ENERGY FOR A TIN-TANTALUM SHIELD

$h\nu$ (Mev)	$(\tau + \sigma_{\alpha})_{Sn}$ (cm^2/g)	$(\tau + \sigma_{\alpha})_{Sn}^{(a)}$	$(\tau + \sigma_{\alpha})_{Ta}$ (cm^2/g)	$(\tau + \sigma_{\alpha})_{Ta}^{(b)}$	$\sum_{Sn, Ta} (\tau + \sigma_{\alpha}) \times$	$\sum_{e, Sn, Ta} (\tau + \sigma_{\alpha}) \times$	$R_b(E)$	$R_s(E)$
0.020	1.63×10	9.05	5.06×10	42.7	52	0.0000	6.21	0.00
0.029	5.33	2.96		(15) ^c	18	0.0000	(14)	0.00
	4.38×10	24.3			39	0.0000		0.00
0.030	4.14×10	22.9	1.59×10	13.4	36	0.0000	15.15	0.00
0.040	1.88×10	10	7.04	5.93	16	0.0000	37.96	0.00
0.050	1.01×10	5.61	3.64	3.07	8.68	0.0002	40.46	0.01
0.060	6.15	3.41	2.14	1.80	5.21	0.0054	36.18	0.204
0.0674		(2.1)	1.52	1.28	3.38	0.0340	(30)	1.07
			1.12×10	9.44	11.54	0.0000		0.00
0.080	2.74	1.52	7.17	6.04	7.56	0.0005	21.7	0.012
0.100	1.46	0.81	3.98	3.36	4.17	0.0155	12.5	0.203
0.150	4.72×10^{-1}	0.26	1.31	1.10	1.36	0.2567	4.41	1.18
0.200	2.22×10^{-1}	0.12	6.08×10^{-1}	0.51	0.63	0.5326	2.40	1.34
1.000	2.69×10^{-2}	0.015	3.46×10^{-2}	0.03	0.045	0.9560	1.0	1.00

^a x_{Sn} = thickness of Sn = 0.030 inch = $0.555 g/cm^2$ ($\rho = 7.3 g/cm^3$).

^b x_{Ta} = thickness of Ta = 0.020 inch = $0.843 g/cm^2$ ($\rho = 16.6 g/cm^3$).

^cNumbers in parentheses are interpolated values.

$$\sigma_{T_{\text{eff}}} = \sum_A f_A \frac{2A}{(A+1)^2} \sigma_{T_A} \quad (12)$$

$$A = 137, 108, 39, 31, 27, 16$$

$$= \sum_A \Delta \sigma_{T_{\text{eff}}}$$

where

A = atomic mass of element in glass,

f_A = fraction of atoms in glass of given A ,

$\frac{2A}{(A+1)^2}$ = average fraction of neutron energy received by the atom recoil in the elastic collision, and

σ_{T_A} = total cross section for given element.¹⁷

Therefore, the energy absorbed, E_{an} , due to neutron exposure is

$$E_{\text{an}} = \phi_n E_n N \sigma_{T_{\text{eff}}} \quad (13)$$

where

ϕ_n = fast neutron flux (i.e. the number of neutrons passing through 1 square centimeter),

E_n = energy of neutrons in Mev, and

$N = 2.52 \times 10^{22}$ atoms per gram of glass.

In Table 6, $\sigma_{T_{\text{eff}}}$ for glass is tabulated for the neutron energies

¹⁷ D. J. Hughes and J. A. Harvey, "Neutron Cross Sections," Brookhaven National Laboratory Report BNL-325 (1955).

Table 6
EFFECTIVE NEUTRON CROSS SECTIONS FOR GLASS

Element	f_A	$\frac{2A}{(A+1)^2}$	$E_n = 1.5176$		$E_n = 1.3745$		$E_n = 0.9989$		$E_n = 0.5852$	
			σ_{TA}	$\Delta\sigma_{T_{eff}}$	σ_{TA}	$\Delta\sigma_{T_{eff}}$	σ_{TA}	$\Delta\sigma_{T_{eff}}$	σ_{TA}	$\Delta\sigma_{T_{eff}}$
Ba	0.0187	0.0143	7.5 ^a	0.0020 ^a	7.5 ^a	0.0020 ^a	7 ^a	0.0019 ^a	6 ^a	0.0016 ^a
Al	0.0422	0.0688	0.003	0.0000	0.004	0.0000	0.003	0.0000	0.008	0.0000
K	0.0470	0.0488	(3) ^b	0.0069	(3)	0.0069	(3)	0.0069	2	0.0046
Ag	0.0095	0.0182	6	0.0010	6	0.0010	6.6	0.0011	7	0.0012
P	0.2260	0.0603	(3)	0.0409	(3)	0.0409	(3)	0.0409	3.2	0.0436
O	0.6566	0.1110	2.1	0.1530	3.2	0.2210	7.8	0.5520	3.2	0.2355
$\sigma_{T_{eff}}$				0.2038		0.2718		0.6028		0.2865

^aTimes 10^{-24} cm².

^bNumbers in parentheses are extrapolated values.

used in the experiment (see section entitled "Van de Graaff Experiment"). Table 7 gives the energy absorbed in the glass at these energies for typical flux values obtained in the experiment.

Table 7
 ENERGY ABSORBED IN TISSUE AND GLASS AS A FUNCTION OF NEUTRON ENERGY

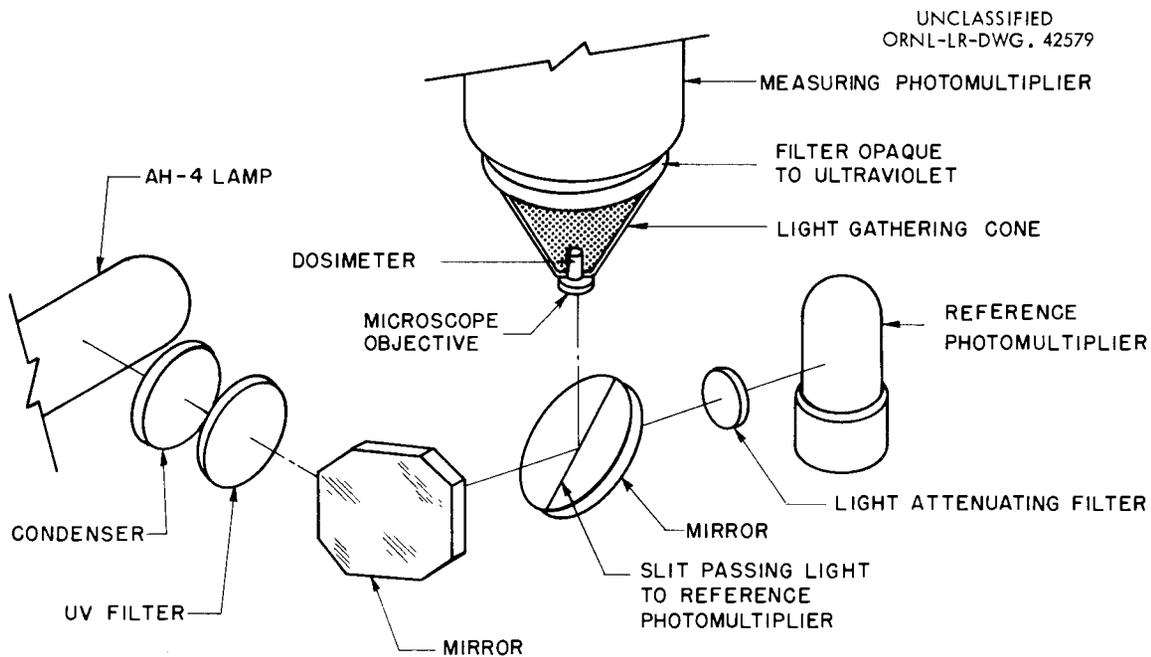
E_n Mev	ϕ_n $\times 10^{-12}$ n/cm ²	$E_{an}(\text{glass})$ $\times 10^{-5}$ ergs/g	$E_{an}(\text{tissue})$ $\times 10^{-5}$ ergs/g	$\frac{E_{an}(\text{glass})}{E_{an}(\text{tissue})}$
1.5176	1.306	0.163	3.53	0.046
1.3745	1.030	0.155	2.66	0.058
0.9899	0.646	0.157	1.58	0.099
0.5852	0.658	0.044	1.12	0.025

III. EXPERIMENTAL APPARATUS AND PROCEDURES

Fluorimeter

The Bausch and Lomb Microdosimeter Reader used in this experiment was an experimental model or prototype. It employs a refined optical system and a balancing or "null" type electronic circuit.

For a given dose, maximum fluorescence from the glass is obtained by excitation with ultraviolet radiation of wavelength 3200 Å, and the undesirable pre-dose luminescence or "noise" background results when the glass is exposed to a shorter wavelength, 2400 Å, radiation. It is necessary to filter the ultraviolet source to discriminate against the shorter wavelengths, i.e. the filters must pass 3200 Å but not 2400 Å radiation. The reader, shown in Fig. 4, uses a mercury lamp (AH-4) as an ultraviolet light source (3650 Å). It is followed in the light path by an optical condenser, an ultraviolet filter to remove wavelengths less than about 3000 Å, and two mirrors. A slit in the second mirror allows a small part of the ultraviolet light to pass through to a light attenuating filter and on to a reference photomultiplier. The main part of the light is reflected from mirror No. 2 to a microscope objective focussed on the end of the glass rod. The rod is positioned in a miniature three-jaw chuck in a sliding drawer. The drawer provides a convenient method of moving the rod in and out of the ultraviolet light beam. The chuck is part of an efficient conical reflector which collects the fluorescence radiation from the glass and directs it to the measuring photomultiplier. The emission spectrum of the dosed glass has its peak at 6400 Å. Therefore, to eliminate stray



(FROM BAUSCH AND LOMB MICRODOSIMETER CATALOG)

Fig. 4. Microdosimeter Reader - Optical Layout.

ultraviolet from reaching the photo tube, an "orange-pass" filter is inserted over the face of the photo tube. This last filter will also discriminate against the 3700 Å pre-dose luminescence and reduce the inherent background of the glass.

The electronic system consists of a stabilized 1000 volt power supply for the photomultipliers with a means of adjusting the potential and thus the sensitivity of the measuring photomultiplier, a measuring and a reference photomultiplier, and a difference amplifier (null indication type) for comparing signals from the two photomultipliers. Thus the ratio of the intensity of fluorescent light to the intensity of the exciting light is the measure of the dosimeter fluorescence and is proportional to the dose. By balancing the signals from the two photomultipliers a value proportional to dose may be read from a graduated scale.

The absolute stability of the fluorimeter over a period of days is uncertain due to the lack of a stable standard. For a few hours, however, the machine seems to remain quite stable. For most accurate work, the uncertainty of prolonged stability requires that an exposure to Co^{60} radiation be made every time an exposure to radiation in the energy dependent region is made. This eliminates the factor of fluorimeter instability from the energy dependence part of the experiment by relating all experimental measurements to those resulting from Co^{60} radiation.

Glass Techniques

(a) Cleaning and Handling

For the highest degree of sensitivity it is necessary to minimize and measure accurately the pre-dose reading of the glass rod. Since many substances fluoresce to some extent, the rods must be freed of dust and any oily film that may be on the surface. This "extra" fluorescence can be reduced by a cleaning procedure which will not leave a residue. The following procedure was used. First, the rods were rinsed in acetone, then in distilled water, and finally in methyl alcohol. The acetone dissolved the surface oils; the water removed the residue which would have been left when the acetone evaporated, and the alcohol eliminated the water film and facilitated the drying process. The rods were dried in a stream of dry filtered air.

This process minimized the pre-dose reading, but accurate measurements require consistency in application of the process, i.e. pre-irradiation and post-irradiation techniques must be identical. It is advisable to recheck frequently to be sure that the reading is not a function of the cleaning process. The need for cleaning is most acute on the ends of the rods because they are exposed to the exciting ultraviolet radiation in an end-on position.

Erratic and non-reproducible readings are incurred when a rod has been chipped or etched. Etching of the glass can occur during the acetone rinse if a rod is allowed to stay in the rinse too long. Facility in handling the small rods is attained by the use of stainless steel reverse action forceps.

(b) Reading

For reading, the glass is inserted end-on into the three-jaw chuck of the fluorimeter which is opened by rotating the knob on the sliding drawer. Both ends of the rod are read, and the average is taken as the response. This is done before and after the rod is irradiated. The difference in pre-dose and post-dose readings is proportional to the dose the glass has received.

The maximum effect of varying the rod orientation was found to be about 8% of the pre-dose reading. This deviation is caused by a lack of roundness in a rod. However, it becomes insignificant within the accuracy limits of the glass when the glass has been exposed and readings of both ends of a rod are averaged.

The greatest sensitivity is obtained if the rods are read not less than 24 hours after exposure; the response of the glass continues to increase for about 24 hours after exposure. At 4 hours 95% of the peak response has been attained, and the subsequent buildup would be unaffected if the rod were read at this time.

Co⁶⁰ Calibration Source

A Co⁶⁰ source¹⁸ of approximately 150 curies provided the high energy gamma radiation for exposure of the glass dosimeter. Having in cascade 1.7 Mev and 1.34 Mev gamma rays, this source allowed a determination of the response in the region where the glass is free of

¹⁸J. A. Ghormley and C. J. Hochanadel, Rev. Sci. Instr. 22, 473 (1951).

energy dependence. The exposure was made at a rate of about 1400 r per minute. This is based on a source calibration utilizing the ferric-ferrous ion technique.¹⁹ The reproducibility of the dose is determined by the consistency with which the operator raises and lowers the source. In the sample holder the field is essentially uniform with the source in position. The dose measurement error caused by variation in time to raise and lower the source was found to be less than 1% for a 1000-r exposure. With the source vertically in position but held in the uppermost position, the dose rate at the sample position is about 0.2% of the dose rate when the source is in the exposure position. The consistency of operation required for 1% accuracy can be achieved by practicing lowering the source into exposure position, timing, and raising it again.

X-Ray Equipment and Techniques

The response of the dosimeter to X rays in the 20 to 200 kev range was determined using a modified Westinghouse Quadrocondex X-ray machine. By using standard National Bureau of Standards filters, the energy spectrum of the X rays was narrowed and the exposure energies reported in the data are the effective energies²⁰ as determined by

¹⁹C. J. Hochanadel and J. A. Ghormley, J. Chem. Phys. 21, 880 (1953).

²⁰The effective energy of a heterochromatic X-ray beam is the energy of a monochromatic beam which has the same absorption coefficient as the given beam in an incremental thickness of standard filter material.

Villforth et al.²¹ who described the filtered spectrum from this machine. It was impractical to narrow the X-ray beam sufficiently to produce ideal conditions because of the exposure time that would be required, but a collimator, 1.27 cm inside diameter, located approximately 10 cm from the target was used. The uncertainty in the dose for given machine settings was determined to be not greater than 1%.

The samples to be irradiated were mounted on a polyethylene slab 0.050 inch in thickness. To insure electronic equilibrium at the dosimeter, a polyethylene slab was placed in the beam path ahead of the sample. For photon energies below 100 kev a 0.050-inch slab was used and for photon energies between 100 and 200 kev a 0.100-inch slab is sufficient.

The exposure doses were determined with a 100-r Victoreen condenser r-meter which had been compared with a standard air chamber. For each effective X-ray energy at specific machine settings, the dose was calibrated in a 5-minute exposure prior to inserting the dosimeter into the field. The exposures were then made for 15 to 20 minutes, the dose being usually between 70 and 90 r. The probable error from all effects in any exposure was not more than 2%.

Van de Graaff Experiment

Fast neutron exposures were made using the ORNL 3 Mev Van de

²¹J. C. Villforth, R. D. Birkhoff, and H. H. Hubbell, Jr., Oak Ridge National Laboratory Report ORNL-2529, July 1, 1958.

Graaff. A tritium gas target,²² defined by R. Lamphere, was used to produce the neutrons by the ${}_1\text{H}^3(p,n){}_2\text{He}^3$ reaction. Standard laboratory techniques were employed in determining the neutron flux and dose.²³⁻²⁵ The neutron field was contaminated by a 20 Mev gamma ray once in 10,000 events because of a ${}_1\text{H}^3(p,\gamma){}_2\text{He}^4$ reaction. Also, the contribution to the field by thermal neutrons due to a 2-foot water wall in the target room is not known quantitatively, but it should contribute thermal neutrons from elastic scattering processes and gamma rays from the ${}_1\text{H}^1(n,\gamma){}_1\text{H}^2$ reaction.

The exposures were made for energies from 0.5 to 1.5 Mev by distributing the dosimeters angularly on an aluminum wire hemisphere, the radii of which converge at the target. They were located 15 cm from the target and the angle with the direction of the proton beam varied from 7.5° to 90° .

²²C. H. Johnson and H. E. Banta, Rev. Sci. Instr. 27, 132 (1956).

²³J. L. Fowler and J. E. Brolley, Jr., Rev. Mod. Phys. 28, 103 (1956).

²⁴E. B. Wagner and G. S. Hurst, Health Physics 2, 57 (1959).

²⁵A. O. Hanson and J. L. McKibben, Phys. Rev. 72, 673 (1947).

IV. PRESENTATION OF DATA

X-Ray Response

The response of the unshielded glass is shown in Fig. 3. The response per unit exposure dose at 50 kev is about 21 times the response at 1.25 Mev. Since the "active ingredient" in the glass is the same as that for film, and the energy dependence curves are similar, and since cadmium had been used successfully by Gupton²⁶ to correct the energy dependence of film, the cadmium was fabricated into cylinders of 0.3 inch in length and 0.070-inch inside diameter with wall thicknesses of 0.040 inch and 0.080 inch. Each container had a 0.015-inch teflon liner and 0.020-inch end pieces. The results of exposing the glass in these containers are shown in Fig. 5. There is a peak in the response at 150 kev of about 2:1, and a rapid decrease below approximately 100 kev for the thicker filter.

In an attempt to improve further, similarly shaped tantalum containers were made with wall thicknesses of 0.020 inch, 0.025 inch, and 0.030 inch (all with 0.015-inch teflon liners and 0.010-inch end pieces). The choice of these thicknesses was based on preliminary calculations similar to those discussed in the section Theory, and reduced the energy dependence significantly as indicated in Fig. 6. However, the response peak is shifted to a higher energy. This shift is also apparent in the response of glass exposed in platinum filters of 0.014-inch, 0.0175-inch, and 0.0245-inch wall thicknesses, Fig. 7.

²⁶E. D. Gupton, Radiol. 66, 253 (1956).

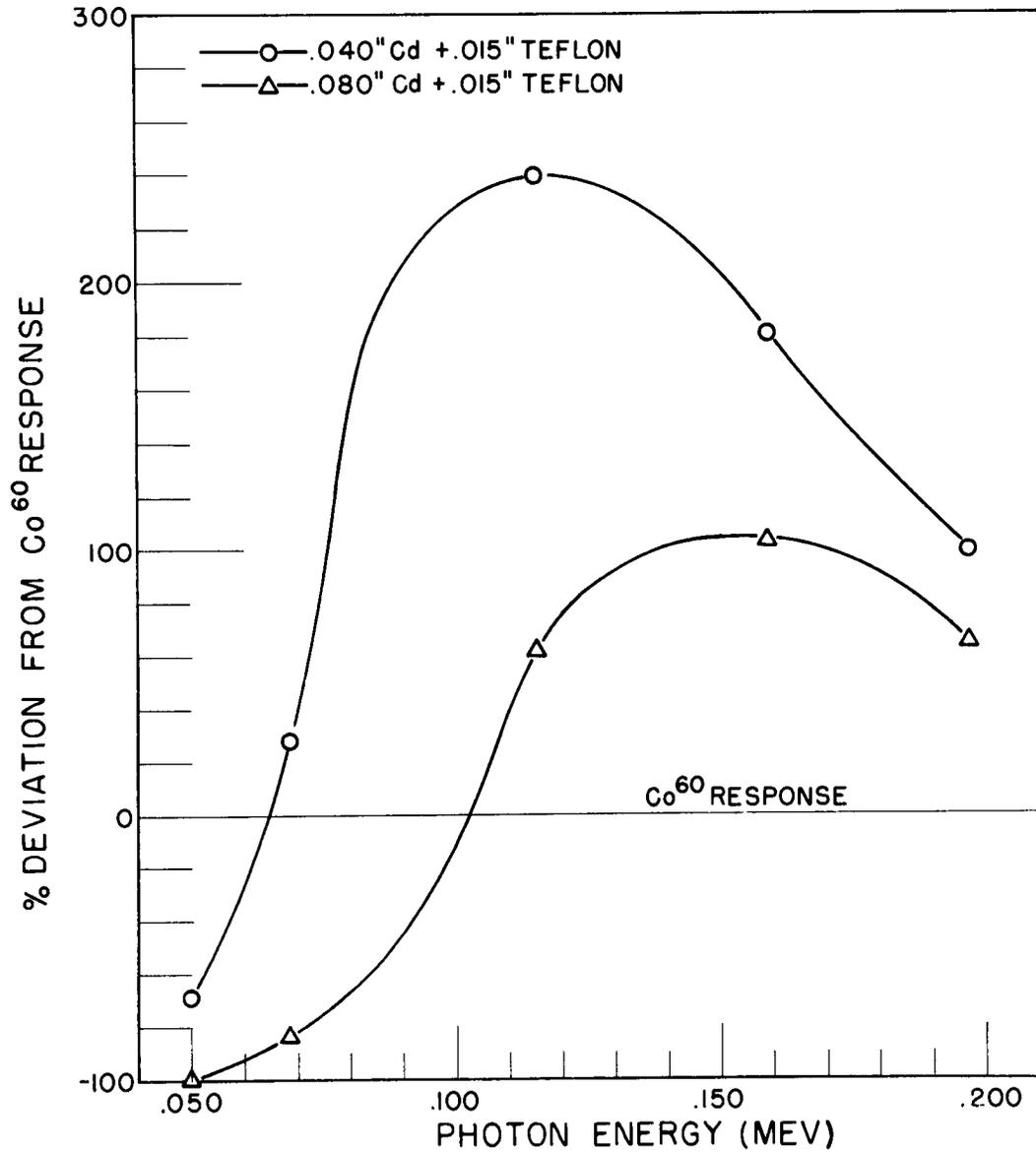
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Fig. 5. Response of Glass with Cadmium and Teflon Shield.

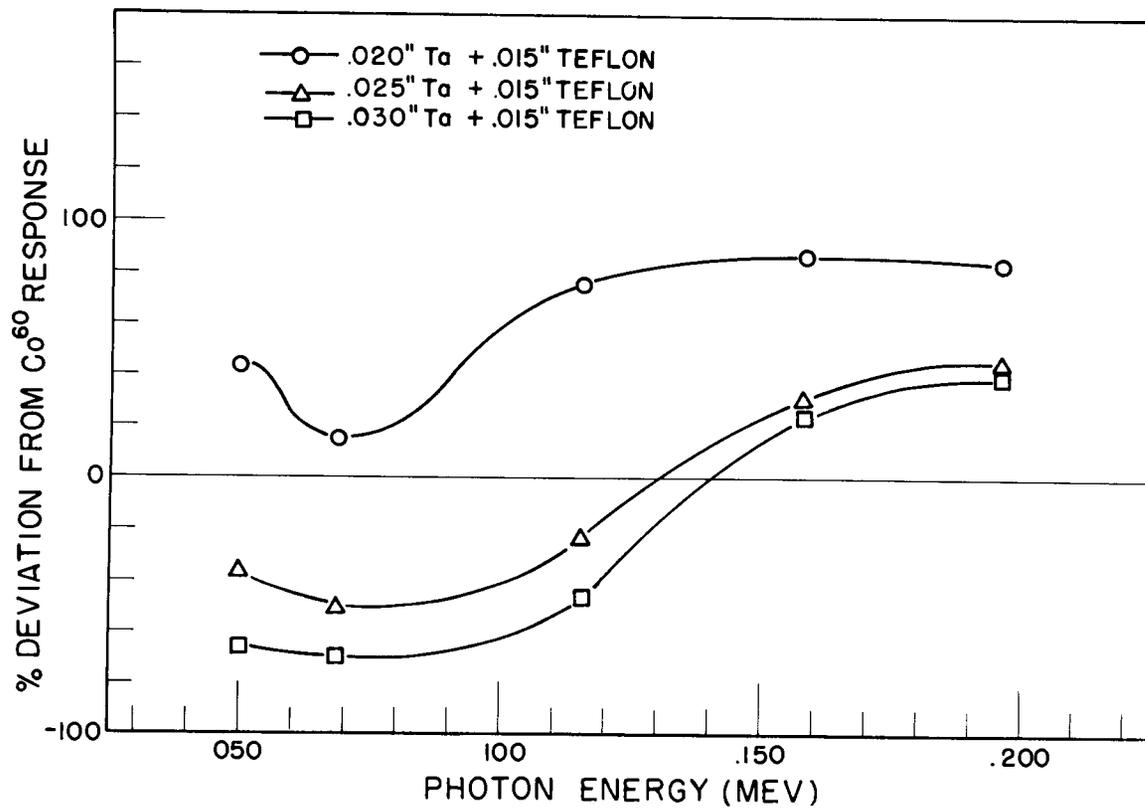
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Fig. 6. Response of Glass with Tantalum and Teflon Shield.

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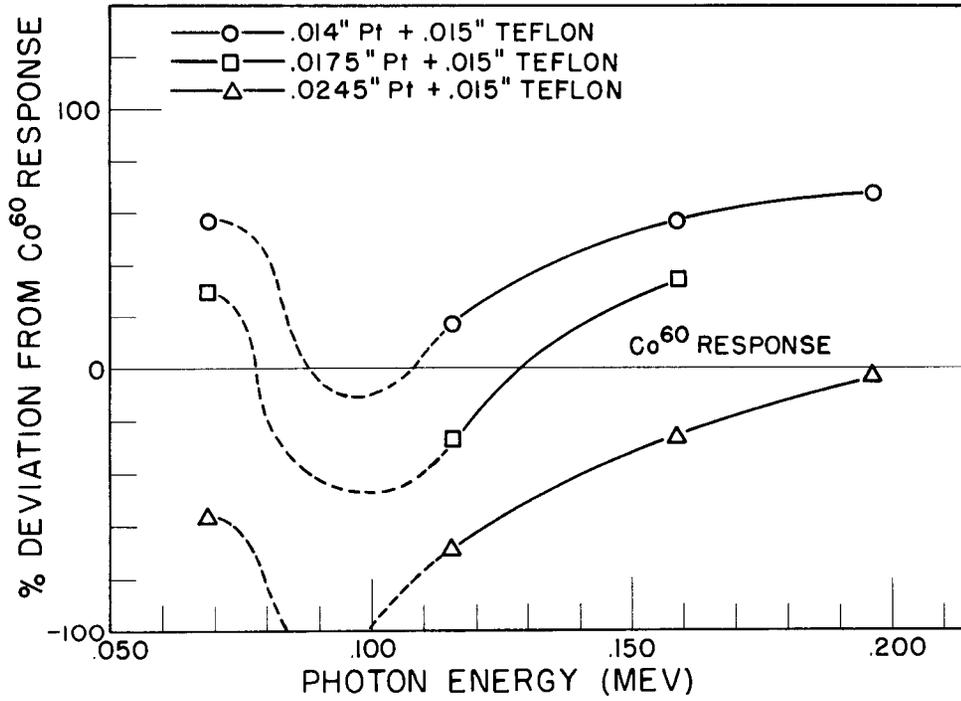


Fig. 7. Response of Glass with Platinum and Teflon Shield.

The broken line portion of the curve is uncertain since there was no effective X-ray energy available near the K-absorption edge of platinum. This interpolation was made based on the response measured using 0.020-inch tantalum filters. The peak response with the platinum filter has been reduced to 1.7:1 with a dip in response at 90 to 100 kev. Tin sleeves were inserted between the tantalum and the teflon liners and the measurements repeated (Figs. 8 and 9). For 0.020-inch tantalum plus 0.030-inch tin the relative response is unity within $\pm 30\%$ above about 115 kev.

Fast Neutron Response

The neutron exposures were made with several different containers as indicated in Fig. 10. The curves indicate an energy dependence. However, the low response of the glass to fast neutrons does not allow the accuracy necessary to determine the energy dependence. The fluorothene container was included because it was not hydrogenous. In Fig. 11 the response of the glass to fast neutrons relative to Co^{60} gamma rays as a function of neutron energy is shown to vary between 0.5 and 0.7% per tissue rad. This curve is uncorrected for any gamma radiation in the target room.

Determination of Sensitivity to Gamma Radiation

The response of the glass as a function of dose is as indicated in Fig. 12. At 5 r the probable error is 11%; this decreased to 3% at 50 r, and remained nearly constant at 3% at greater exposures doses.

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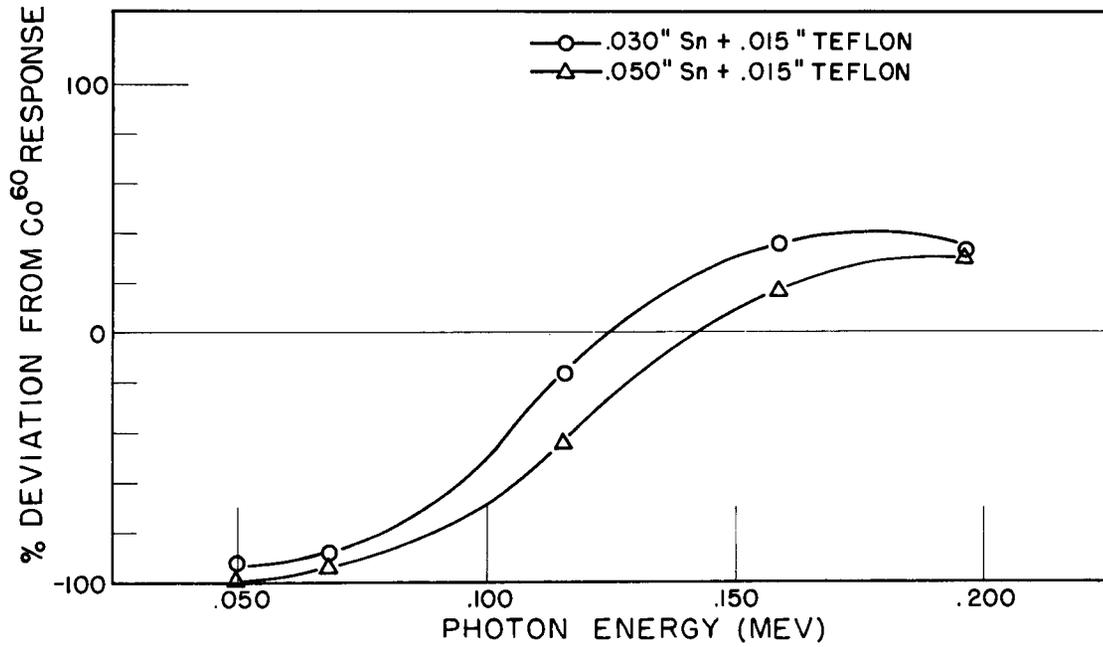


Fig. 8. Response of Glass with 0.015 inch Tantalum Plus Tin Plus Teflon Shield.

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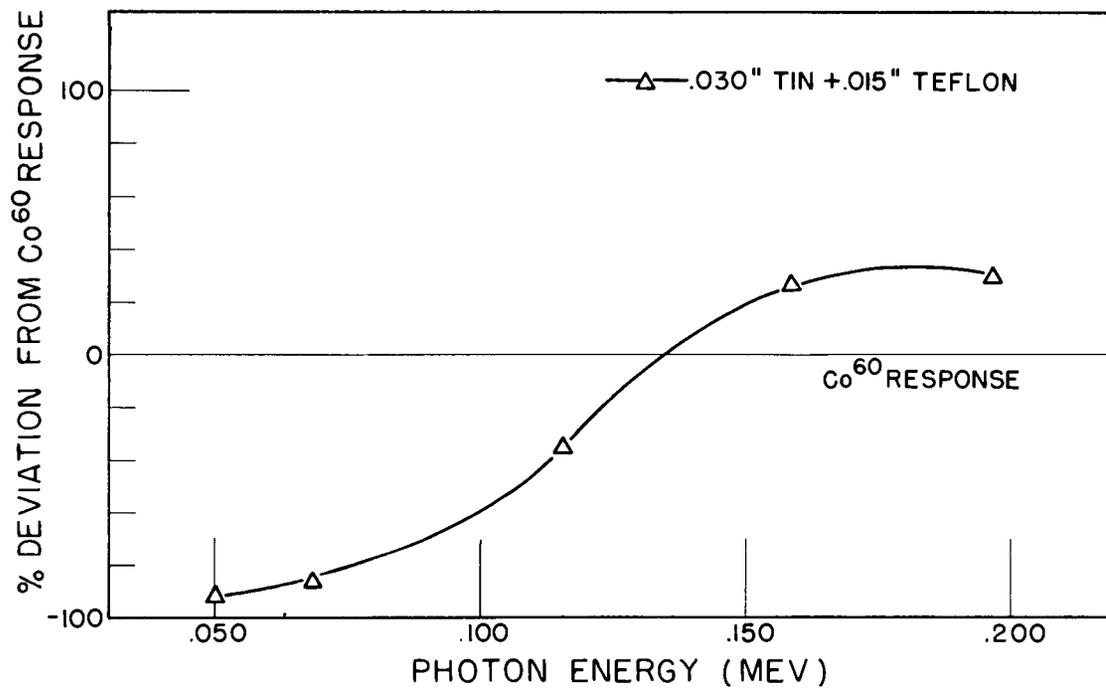


Fig. 9. Response of Glass with 0.020 inch Tantalum Plus Tin Plus Teflon Shield.

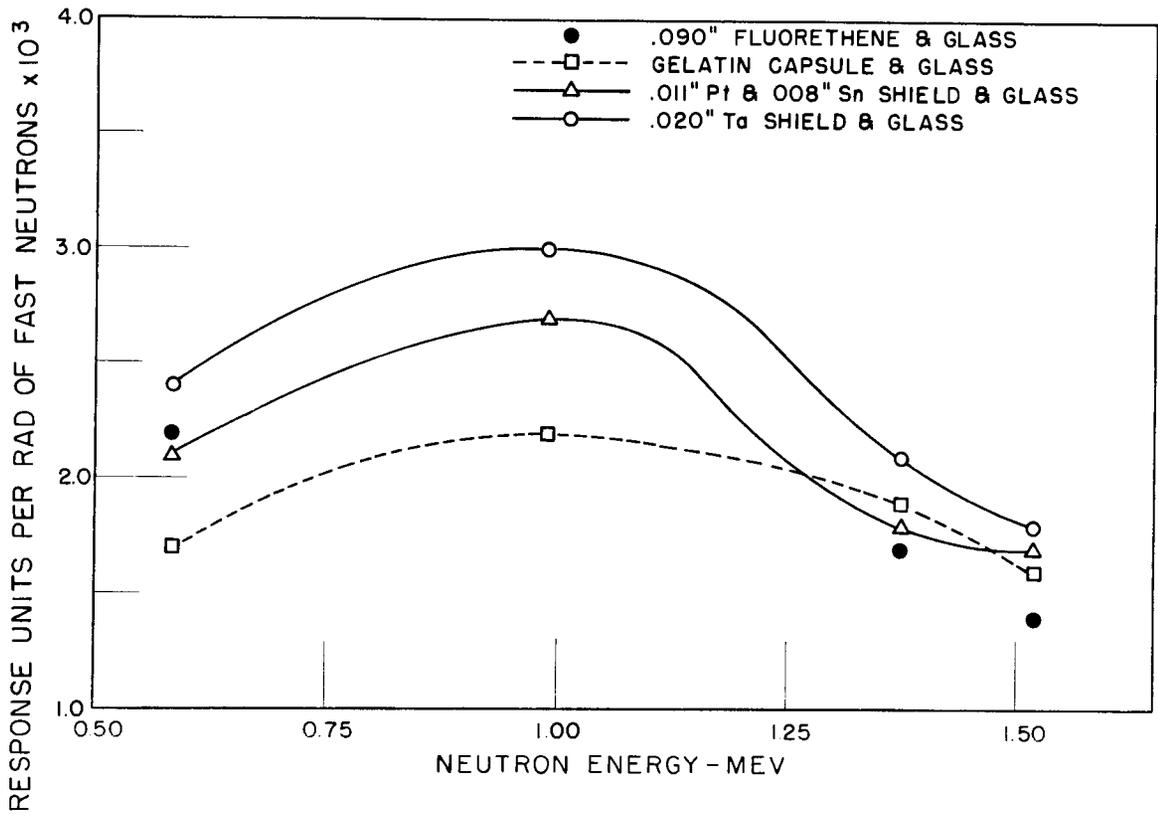
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Fig. 10. Neutron Response of Glass in Various Containers.

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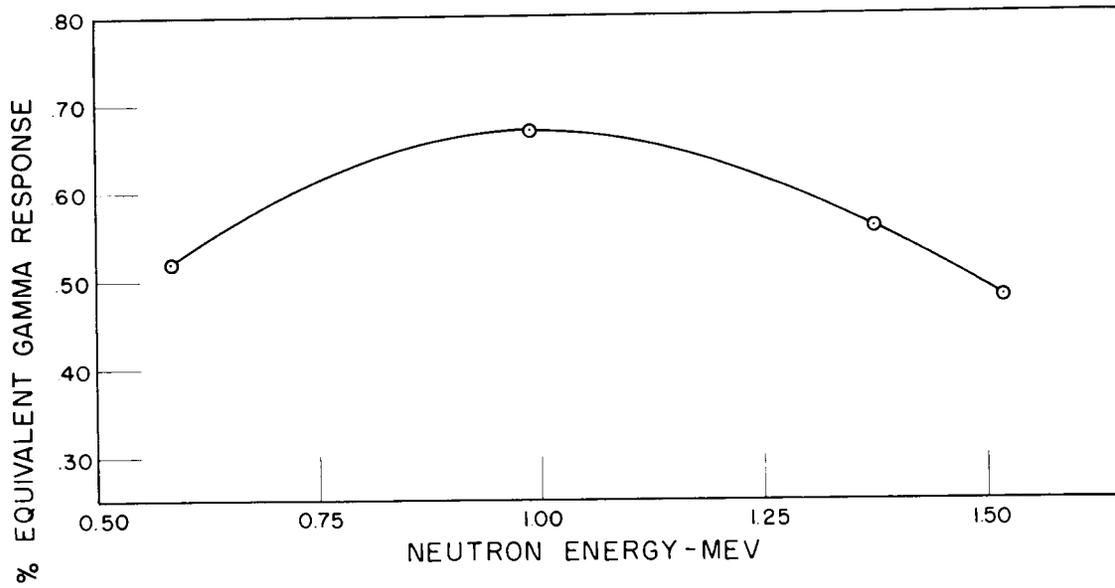


Fig. 11. Neutron Response of Glass Relative to Co^{60} Gamma Response.

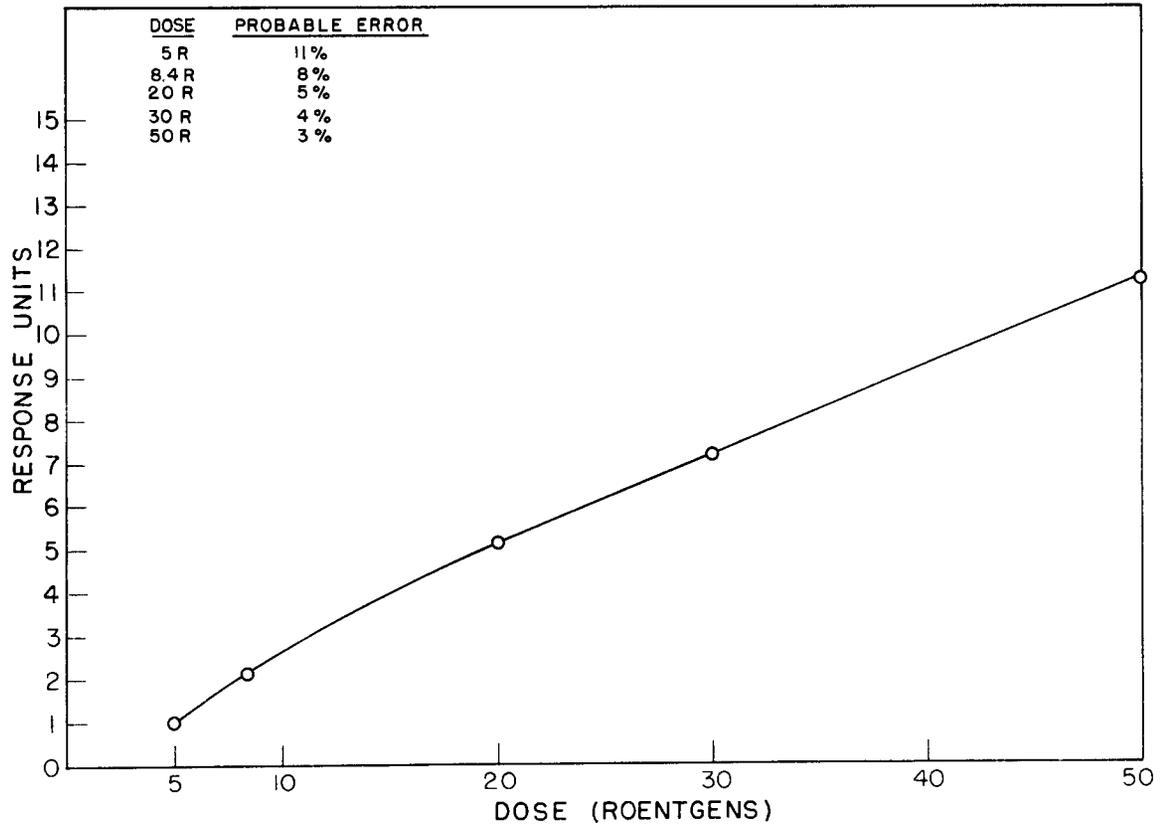
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Fig. 12. Sensitivity of Glass to Gamma Radiation.

V. ANALYSIS AND DISCUSSION

Discussion of X-Ray Response

A comparison of the theoretical and experimental response curves shown in Fig. 3 shows agreement as to the energy at which the peak occurs but disagreement on the relative magnitude of that peak. It was suspected that this is largely due to the heterogeneity of the X-ray beam used in the experimental exposures. Making use of Villforth's calculation of Kramer's distribution of filtered X rays, an attempt was made to correct the theoretical response for the effect of the X-ray spectrum. Figure 13 shows the X-ray spectra (Kramer's theoretical distribution) normalized to unit dose. This spectrum for a given effective energy was weighted point by point by the relative response of the glass (calculated curve, Fig. 3), and the products plotted, Fig. 14. The area under each curve may be represented by

$R_{E_{eff}}$.

$$R_{E_{eff}} = \int R_{theo}(E) D(E) dE \quad (14)$$

where $R_{theo}(E)$ is the theoretical response as a function of energy and $D(E)$ is the dose as a function of energy. This integration is made over the energy range of the X-ray spectrum for a given effective X-ray energy. $R_{E_{eff}}$ is proportional to the corrected response at a given effective energy. A plot of $R_{E_{eff}}$ normalized to 200 kev is shown in Fig. 3. This curve and the experimental curve compare to within $\pm 25\%$, which is within the limits of error inherent in the

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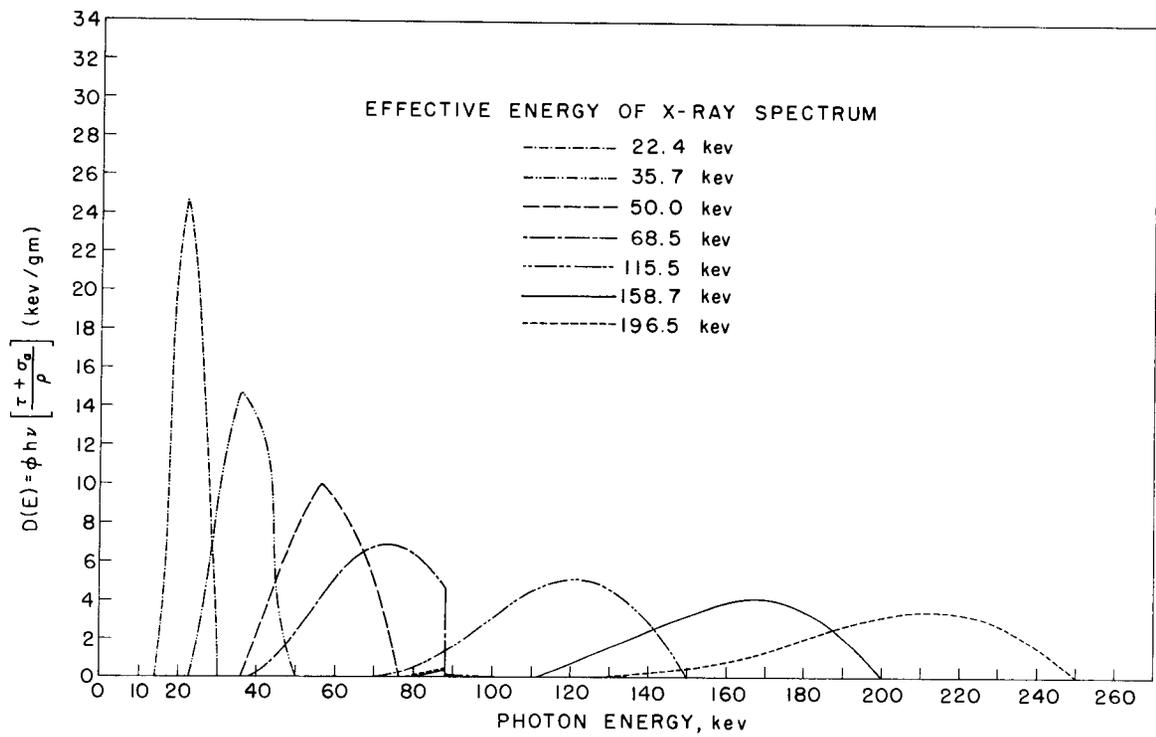


Fig. 13. Filtered X-Ray Spectra Normalized to Unit Dose.

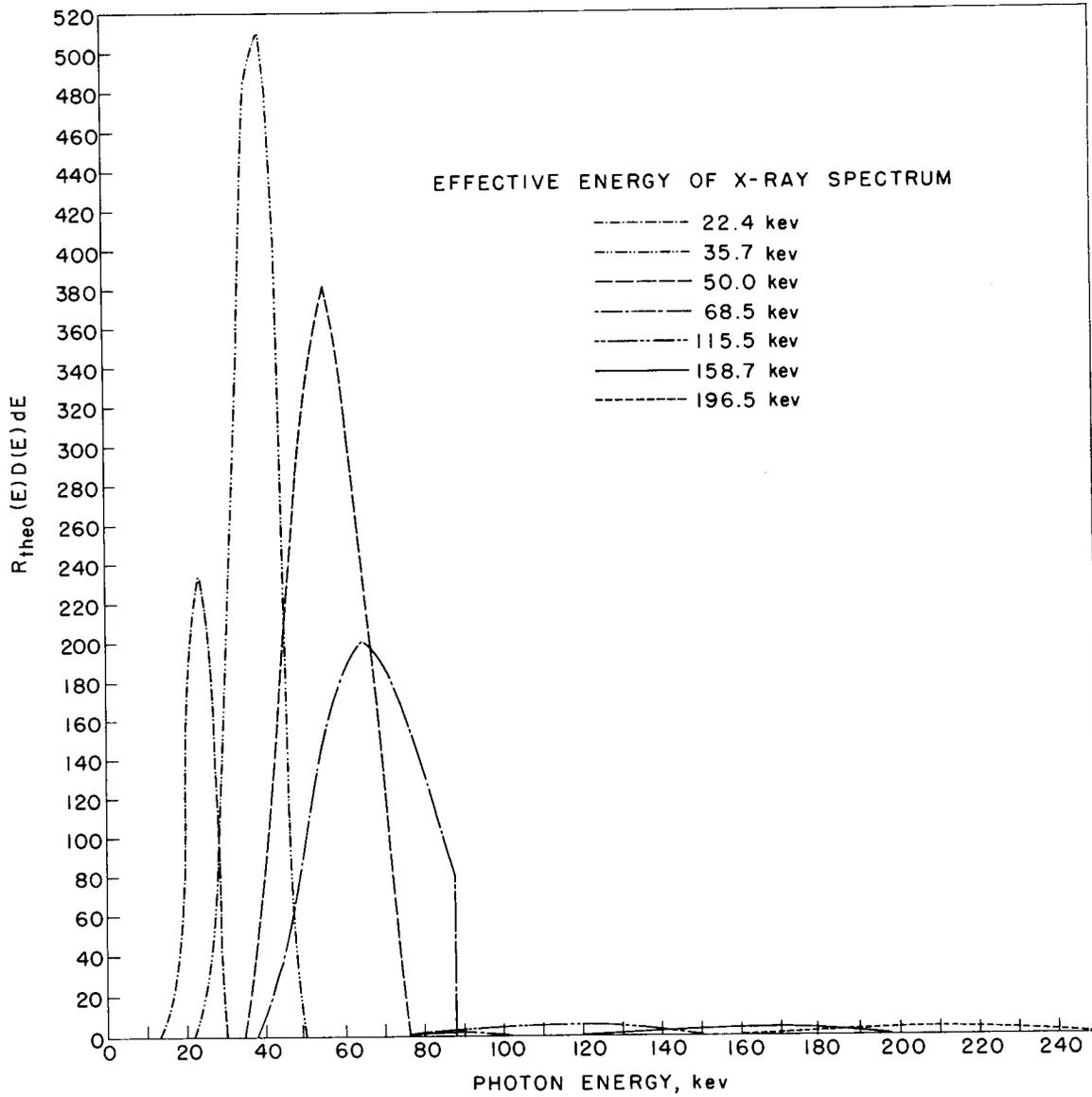
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Fig. 14. Representation of $R_{E_{eff}}$ as an Area.

calculations due to the assumptions made in setting up the equations.

Discussion of Experimental and Calculated Deviations in Response of the Shielded Dosimeter as a Function of Photon Energy

The lack of coincidence of the theoretical and experimental curves, particularly in the energy region from 55 to 70 kev, is largely attributable to the width of the X-ray energy spectrum. For 0.020-inch tantalum plus 0.030-inch tin filters (Fig. 13), the per cent discrepancy is a maximum at 70 kev because the relation is made to a theoretical response which is essentially zero due to the K-edge. (All experimental measurements were made with a 0.015-inch teflon line inserted between the glass and the filter). The glass response at the K-absorption edge of tantalum (67.46 kev) would be nil, as the calculated curve predicts, if the X-ray source had not contributed a significant amount of radiation of energy other than the effective energy of the given spectrum. The nearest effective energy experimentally available was 68.5 kev. The effect of the undesired photons is greatly magnified by the selective absorption by the glass in this region.

Alteration of the X-ray spectrum by added filtration has the effect of reducing the number of photons whose energies are below the effective energy of the given spectrum and thus increases the effective energy. The additional filtration is particularly noticeable at the given effective energy of 68.5 kev because the photons are in the resonance absorption region of the dosimeter. This effect would tend

to decrease the dosimeter response at this energy more than could be attributable to the simple filter absorption.

Analysis of Fast Neutron Response

The response of the metaphosphate glass to neutrons in the 0.5 to 1.5 Mev range as determined by this experiment must be considered an upper limit due to the following factors. There is an uncertainty contributed by a 20 Mev gamma ray produced by the ${}^1_1\text{H}^3(p,\gamma){}^4_2\text{He}$ reaction which occurs about once per 10,000 neutrons produced. A 2-foot thick water wall, at one side of the target room, contributes 2.2 Mev gamma rays from the reaction $\text{H}^1(n,\gamma)\text{H}^2$. The significance of this gamma contamination may be slight but it is a positive effect; it is safe to assume that the 0.7% equivalent gamma response is an upper limit.

It is of interest to compare the calculated glass absorbed dose with the calculated tissue absorbed dose. A calculation for tissue similar to the calculation for glass is made using Eq. (13) by substituting for N, 9.50×10^{22} atoms per gram of tissue, and

$$\begin{aligned}\sigma_{T_{\text{eff}}} &= \sum_A f_A \frac{2A}{(A+1)^2} \sigma_{T_A} & (15) \\ A &= 16, 14, 12, 1 \\ &= \sum_A \Delta \sigma_{T_{\text{eff}}}\end{aligned}$$

The symbols in the summation are the same as those in Eq. (12). E_{an} for tissue is shown in Table 7 and the ratio of $E_{\text{an}}(\text{glass})$ to

E_{an} (tissue) is also shown. Table 8 lists the effective neutron cross sections for tissue as a function of energy.

The average response of the glass rod to 1.5176 Mev neutrons for 1.306×10^{12} n/cm² is 6.2 units. By multiplying E_{an} , as calculated from Eq. (13) and shown in Table 7, by the weight of the rod, 0.013 g, the energy absorbed per glass rod may be determined. This is 212 ergs per glass rod. A measure of the sensitivity of the glass rod to fast neutrons is given by the energy absorbed per response unit.

$$\frac{212 \text{ ergs/glass rod}}{6.2 \text{ units/glass rod}} = 34 \text{ ergs/unit} \quad (16)$$

The following is a calculation of the energy absorbed in glass upon exposure to 1 Mev gamma rays. In this energy range, the absorption is principally due to τ , the photoelectric, and σ_{α} , the Compton absorption cross sections. An exposure of 1 r is equal to an absorbed dose in air of 87.7 ergs per gram. Therefore,

$$\frac{\text{Energy absorbed/g of air}}{\text{Energy absorbed/g of glass}} = \frac{[(\tau + \sigma_{\alpha})/\rho]_{\text{air}}}{[(\tau + \sigma_{\alpha})/\rho]_{\text{glass}}} \quad (17)$$

At 1 Mev

$$[(\tau + \sigma_{\alpha})/\rho]_{\text{air}} = 0.0279 \text{ cm}^2/\text{g} \quad (18)$$

$$[(\tau + \sigma_{\alpha})/\rho]_{\text{glass}} = 0.0275 \text{ cm}^2/\text{g} \quad (19)$$

Then the energy absorbed per gram of glass is approximately 86.4 ergs per gram. Multiplying this value by the weight of a glass rod,

Table 8

EFFECTIVE NEUTRON CROSS SECTIONS FOR TISSUE

Element	f_A	$\frac{2A}{(A+1)^2}$	$E_n = 1.5176$		$E_n = 1.3745$		$E_n = 0.9899$		$E_n = 0.5852$	
			σ_{TA}	$\Delta\sigma_{Teff}$	σ_{TA}	$\Delta\sigma_{Teff}$	σ_{TA}	$\Delta\sigma_{Teff}$	σ_{TA}	$\Delta\sigma_{Teff}$
H	0.6340	0.500	3.4 ^a	1.078 ^a	3.5 ^a	1.110 ^a	4.3 ^a	1.365 ^a	5.6 ^a	1.776 ^a
C	0.0951	0.142	2.1	0.029	2.2	0.030	2.6	0.035	3.2	0.043
N	0.0137	0.124	1.8	0.003	2.5	0.004	1.5	0.003	1.8	0.003
O	0.2580	0.111	2.1	0.060	3.2	0.092	7.8	0.223	3.2	0.092
σ_{Teff}				1.170		1.236		1.626		1.914

^aTimes 10^{-24} cm².

0.013 g, the energy absorbed per glass rod may be calculated as follows:

$$86.4 \text{ ergs/g glass} \times \frac{0.013 \text{ g glass}}{\text{glass rod}} \cong 1.1 \text{ ergs/glass rod/r} \quad (20)$$

The response produced in a glass rod per roentgen exposure is 0.287 response unit. Therefore, a measure of the sensitivity of a glass rod to gamma rays is given by the energy absorbed per unit response or

$$\frac{1.1 \text{ ergs/rod/r}}{0.287 \text{ unit/rod/r}} \cong 3.9 \text{ ergs/unit} \quad (21)$$

A comparison of the energy absorbed per unit response for fast neutron exposure with the energy absorbed per unit response for 1 Mev gamma exposure indicates a small heavy ion effectiveness relative to gamma rays. The average relative effectiveness of neutrons in the 0.5 to 1.5 Mev range is about 0.12 of that for Co^{60} gamma rays. The relative effectiveness of 14 Mev neutrons is between 0.21 and 0.11, according to Kondo.²⁷

²⁷S. Kondo, "Neutron Response of Silver-Activated Phosphate Glass," (to be published).

VI. CONCLUSIONS AND RECOMMENDATIONS

With the Bausch and Lomb Microdosimeter Reader adjusted for maximum sensitivity the glass will measure doses of 5 r with probable error of 11%. For a 50-r dose the maximum probable error is decreased to 3%. Comparison of the calculated photon energy dependence with the experimentally determined energy response for the unshielded glass shows the former to have a 40:1 peak at 50 kev and the latter a 21:1 peak at the same effective X-ray energy. The difference is attributed to the heterogeneous beam from the X-ray source used.

The assumption that the response of the glass to photons is a function of the amount of energy absorbed permits a theoretical prediction of its energy dependence. When the glass is shielded as shown in Fig. 15, agreement is within 5% except in the region of the K-absorption edge of the shield. Here the effect of the energy spectrum of the source is most pronounced and results in the largest discrepancy between theoretical and experimental responses. This and the fact that any shielding would harden the X-ray beam, i.e. cause its effective energy to be greater, would cause deviation particularly in the K-absorption region.

The energy dependence was corrected to within $\pm 30\%$ for photon energies greater than 115 kev using a 0.020-inch tantalum cylinder around 0.030-inch tin which enclosed a 0.015-inch teflon sleeve next to the dosimeter.

Dosimeter response to neutrons in the 0.5 to 1.5 Mev range varies between 0.7% and 0.5% of the equivalent gamma response. This low

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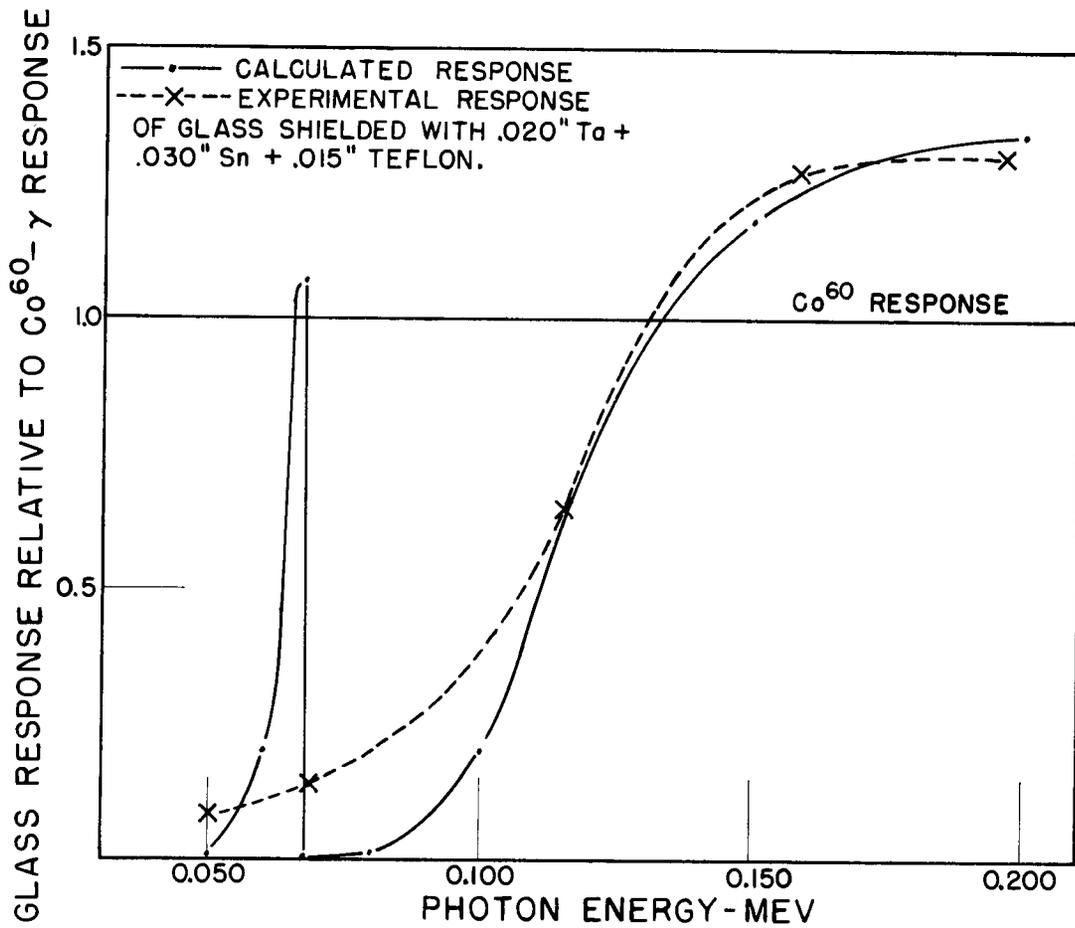


Fig. 15. Comparison of Theoretical and Experimental Gamma Responses of the Shielded Glass.

response makes silver metaphosphate glass a good gamma dosimeter in co-existent fast neutron and gamma radiation fields, especially those resulting from fission since fission neutrons are principally in this energy range.

It has been shown previously that the glass has a high sensitivity to thermal neutrons.²⁸ However, Li^6 encapsulation permits gamma measurements with the glass in fields containing thermal neutrons. The optimum capsule thickness must be determined. Further work in refinement of the filtering technique for energy dependence correction is necessary. Some work has been done by F. H. Attix²⁹ of the Naval Research Laboratory using a high Z shield to cover only part of the dosimeter, and this approach has shown promise.

The size of the dosimeter and reproducibility of results make this system a useful addition to gamma-ray dosimetry, particularly for gamma rays coexistent with neutrons. In some experiments the greatly increased sensitivity to low energy photons permits accurate measurement of exposure doses as small as a few hundred milliroentgens.

²⁸C. H. Bernard (Agricultural and Mechanical College of Texas), private communication.

²⁹F. H. Attix, private communication.

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