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COBALT BOROSILICATE GLASS GAMMA DOSIMETRY AND ITS USE AT THE US ARMY NUCLEAR DEFENSE LABORATORY

Max Stuetzer

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SEPTEMBER 1968

US ARMY
NUCLEAR DEFENSE LABORATORY
EDGEWOOD ARSENAL, MARYLAND

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ABSTRACT

This report presents a description of the cobalt borosilicate glass gamma dosimetry system used by the US Army Nuclear Defense Laboratory. Cobalt borosilicate glass is an inexpensive, efficient, and reliable passive dosimeter for gamma exposures from approximately 5×10^{3} to 5×10^{8} R. The use of energy discrimination shields provides the system with an almost flat photon-energy response from 60 keV to 10 MeV. When exposed to mixed neutron and gamma radiation fields, thermal neutron shields eliminate 99.9 percent of the thermal neutron fluence. The system is rate independent to 5.5×10^{11} R s^{-1}.

The main problems associated with the system are fast neutron corrections, fading, saturation effects, and variations in plate parameters.

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1. INTRODUCTION

Cobalt borosilicate glass plates have been used since the mid 1950's for the measurement of large gamma radiation exposures. Kreidl and Blair (Reference 1) were among the first to investigate their utility. More recently, Johnson (Reference 2) made an extensive study of the system.

Since 1956, part of the mission of the US Army Nuclear Defense Laboratory (USANDL) has been to provide radiation measurements at nuclear weapon tests and weapon simulation facilities. In the early 1960's, cobalt borosilicate glass was incorporated as an integral part of the gamma dosimetry system used to accomplish this effort. The object of this report is to present, as a reference, a description of the system as currently used by this Laboratory to measure gamma radiation.

2. THEORY

2.1 The Ideal Dosimeter.

Any object exposed to radiation that undergoes a physical change proportional to its exposure may be used as a dosimeter. Some characteristics of an ideal dosimeter are:

(1) Response to a wide range of exposures.

(2) Response independent of exposure rate.

(3) Response per unit of exposure independent of photon energy.

(4) Response unaffected by time, environment, and readout process.

(5) Readout technique simple, rapid, and reproducible.

(6) Dosimeter inexpensive, of good quality, and readily available in quantity.

Cobalt borosilicate glass, while not an ideal dosimeter, has many of these properties.

2.2 Cobalt Glass.

The property of cobalt glass which enables it to be used for dosimetry from 5×10^3 to 5×10^6 R is that radiation produces changes in

its light absorbance spectrum. Changes in absorbance or optical density are attributed to the trapping of electrons produced by the energy deposition of the radiation at or near positively charged impurities, which in this case are cobalt ions (Reference 3).

A glass is considered to be a physical system with local order and general disorder, that is, crystal domains extending over several lattice constants with random orientation of domains (Reference 4). Accordingly, it is reasonable to apply some concepts from the theory of color centers in crystals to glasses with special consideration for the structure of the glass. Color centers are defined as the special electronic configurations in a solid that give rise to optical absorption in a normally transparent spectral region (Reference 5). Coloration may be produced by the trapping of electrons and/or holes at any of a multiplicity of lattice defects.

The absorbance spectrum of cobalt glass (Figure 2.1) peaks strongly at 300 mμ. This peak is probably due to the cobalt doping. The small diffuse peak at 720 mμ may be due to aluminum-oxygen defects (Reference 5).

2.3 Response of Cobalt Glass to Radiation Fields.

If the probability of forming a color center per exposure unit remains constant, the absorbance of the dosimeter will increase linearly with exposure until a significant number of the color centers have been occupied. This has been observed for monoenergetic gamma-rays, a gamma-ray spectrum, and mixed neutron-gamma radiation environments by Friddell (Reference 6) using silver metaphosphate glass rods as monitors. He found the radiation response to be linear (as in the ^{60}Co calibration curve in Figure 2.2).

2.4 Transmittance and absorbance.

A Beckman DU 2 spectrophotometer is used by this Laboratory for readout and since this instrument does not measure absorbance directly, the transmittancy of each irradiated plate is measured.

The transmittance of a sample is the fraction of the light intensity incident on the sample which passes through the sample. The transmittancy, T, of an irradiated glass plate is measured relative to an unirradiated plate:

$$T = \frac{T_s}{T_o} \qquad (2.1)$$

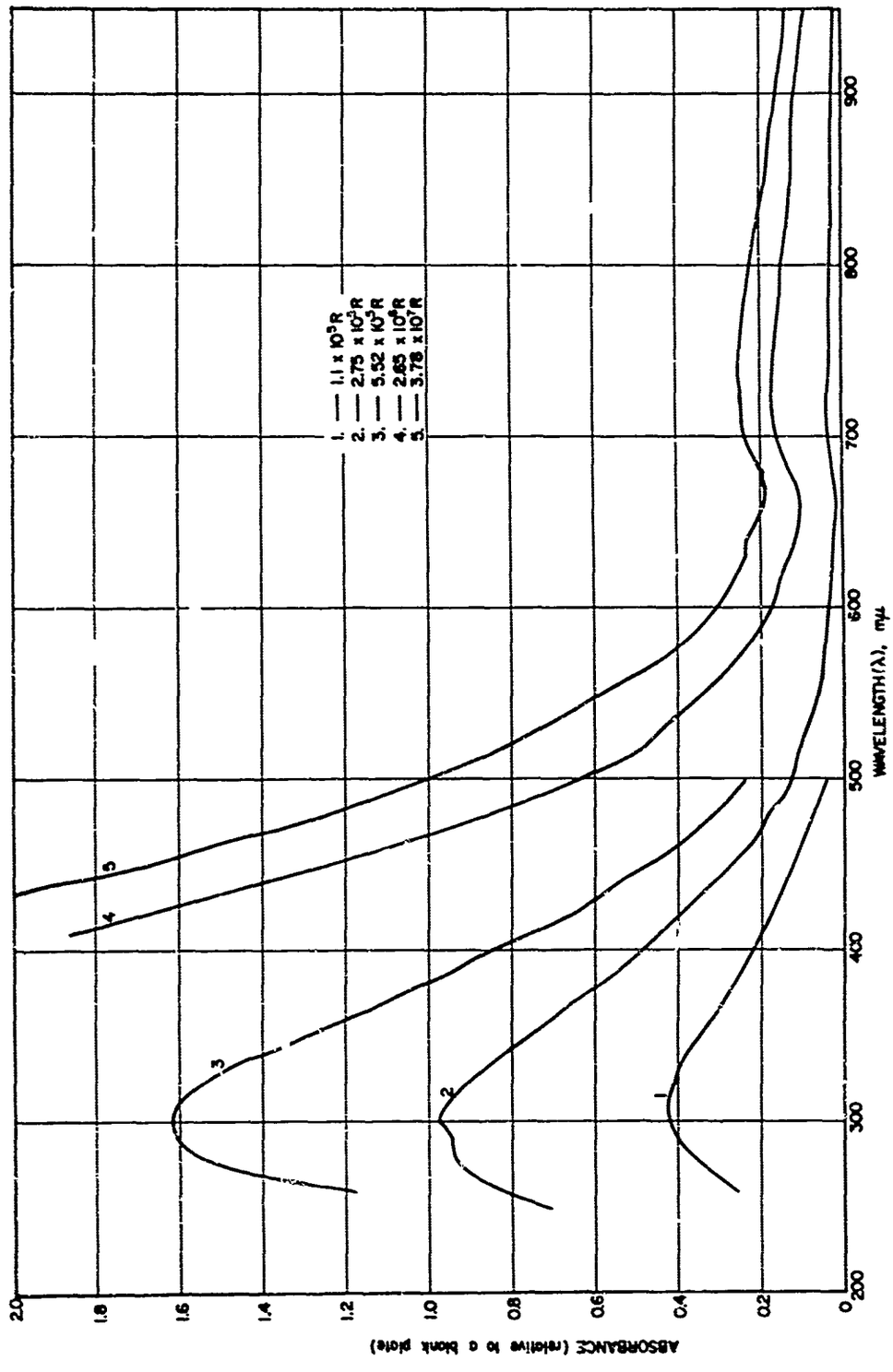

Figure 2.1 Absorbance as a function of wavelength, with exposure as the parameter (1.5 mm cobalt glass plate).

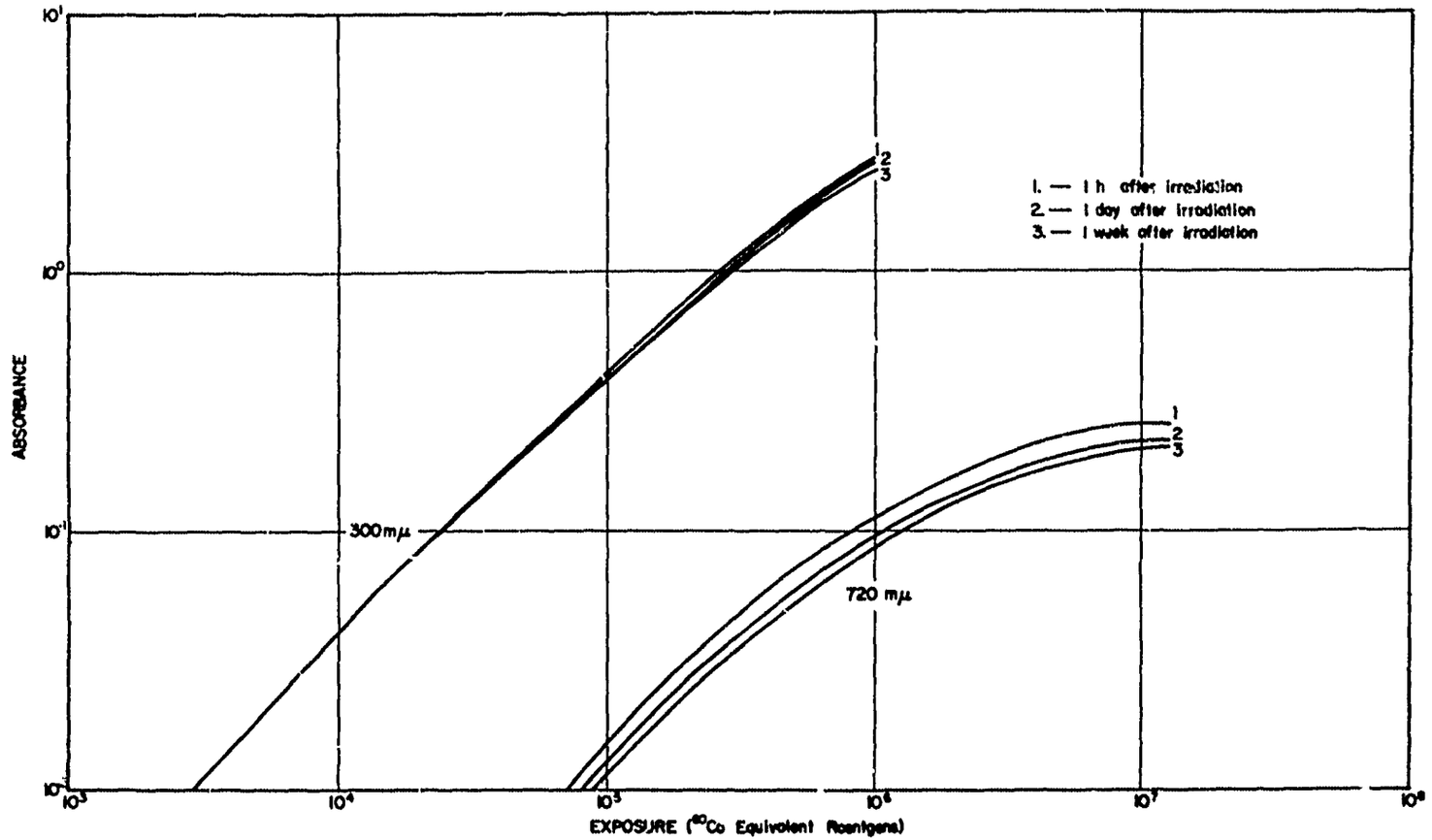

Figure 2.2 Typical calibration curves, (cobalt borosilicate glass plates, 3 mm thick).

where T = transmittancy,

T_s = transmittance of the irradiated plate,

and T_o = transmittance of an unirradiated plate.

The absorbance of an irradiated plate is directly proportional to the gamma exposure. Transmittancy is converted to absorbance by:

$$A = \log_{10} \frac{1}{T} \tag{2.2}$$

where A = absorbance of a sample,

and T = transmittancy of the same sample.

3. SYSTEM DESCRIPTION

3.1 Readout Instrumentation.

3.1.1 Spectrophotometer. The glass plates are read on a Beckman DU 2 single beam, null balancing, ultraviolet spectrophotometer (Figure 3.1). Transmittancy measurements are precise to one or two parts in a thousand on the 0 to 110 percent T scale, which is normally used. There are also 90 to 101 percent and 0 to 11 percent T scales that can be read to within 0.01 percent T and on which successive readings have a standard deviation of approximately 0.08 percent T. Readings on either of these scales have to be converted to readings on the 0 to 110 percent T scale by addition of a wavelength independent constant.

Making transmittancy measurements with a specially prepared blank plate as standard is more convenient and accurate than making transmittance measurements with air as the standard. An unirradiated plate has an absorbance of approximately 0.469 at 300 mμ relative to air (stp and 50 percent humidity). A reading of 0.1 percent T (the smallest division on the linear 0 to 110 percent T scale) corresponds to an absorbance of 0.002 relative to air. Relative to an unirradiated plate, 0.1 percent T corresponds to an absorbance of 0.0004. Thus, some sensitivity and a large range of spectrophotometer response is lost if one measures transmittance relative to air.

The DU 2 Spectrophotometer has two light sources, a hydrogen lamp and a tungsten lamp. It also has a red sensitive phototube and a

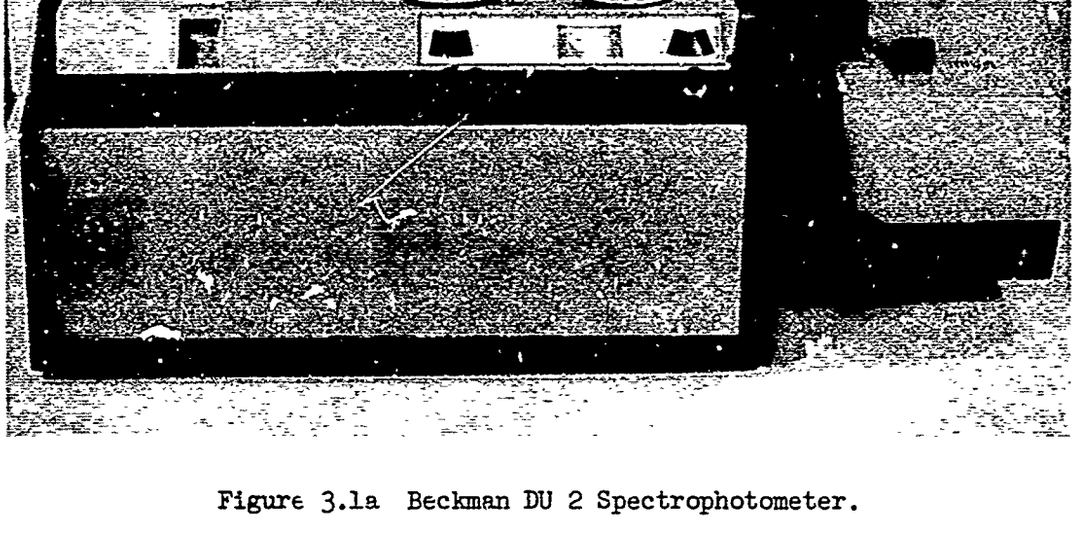

Figure 3.1a Beckman DU 2 Spectrophotometer.

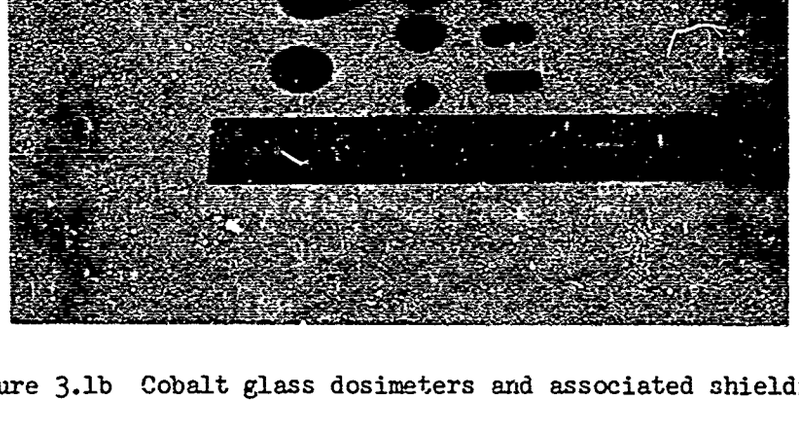

Figure 3.1b Cobalt glass dosimeters and associated shielding.

PM tube. The wavelength ranges for these are:

H_2 lamp	190 to 375 mμ
W lamp	320 to 1000 mμ
PM tube	$\lambda < 700$ mμ
Phototube	$\lambda > 600$ mμ

3.1.2 Plate Holder. An aluminum plate holder, with four matched apertures 4.5 x 13.5 mm, is used to position the plates in the sample compartment. The apertures are not quite perfectly matched in that a wavelength-dependent correction must be made for precise work. These corrections are small and depend on the relative lamp and detector positions. Whenever the optical path is changed, new corrections should be determined. At the present time, corrections are less than 0.01 percent T at 300 mμ for all four apertures. Errors of 0.5 percent T may result from improper positioning of plates in the holder.

3.2 Calibration.

The system is calibrated with radiation from 26,100 Ci of ^{60}Co in a Gammacell 220 (Atomic Energy of Canada, Limited). The gamma cell position at which the cobalt plates were exposed for known lengths of time was calibrated with a Fricke ferrous sulfate chemical dosimeter. A typical calibration curve is reproduced in Figure 2.2.

Transmittancy measurements are made at two wavelengths, 300 and 720 mμ. These wavelengths correspond to the absorbance maxima (Figure 2.1) easily available on the DU 2 Spectrophotometer. The more sensitive 300 mμ curve extends from 10^4 to 10^6 R. Above 10^6 R, the 300 mμ absorbance of the plates is greater than 2 and difficult to read accurately. Exposures greater than 10^5 R can be measured at 720 mμ. However, at 720 mμ saturation effects (Figure 2.2) and fading (Section 3.7) cause considerable loss of precision.

3.3 Dosimeter Description.

The cobalt borosilicate glass dosimeter is a 1.5 x 6 x 15 mm glass plate (Figure 3.1). A typical melt composition by weight is: 57.75% SiO_2; 10.13% Na_2O; 22.33% B_2O_3; 9.42% Al_2O_3, and 0.37% Co_3O_4. The absorbance as a function of wavelength for several gamma exposures is given in Figure 3.2 for air at 16° C and 50 percent humidity. Exposures greater than 10^3 R produce changes in the absorbance and the transmittancy of the cobalt glass plates as shown in Figures 2.1 and 3.3.

The dosimeters, manufactured by Bausch and Lomb, are sealed in individual plastic compartments, and packaged 25 to a batch, and 100 to a box. The standard deviation (σ) of thickness, as well as melt concentration variations of the plates within a batch, are typically 0.4 percent.

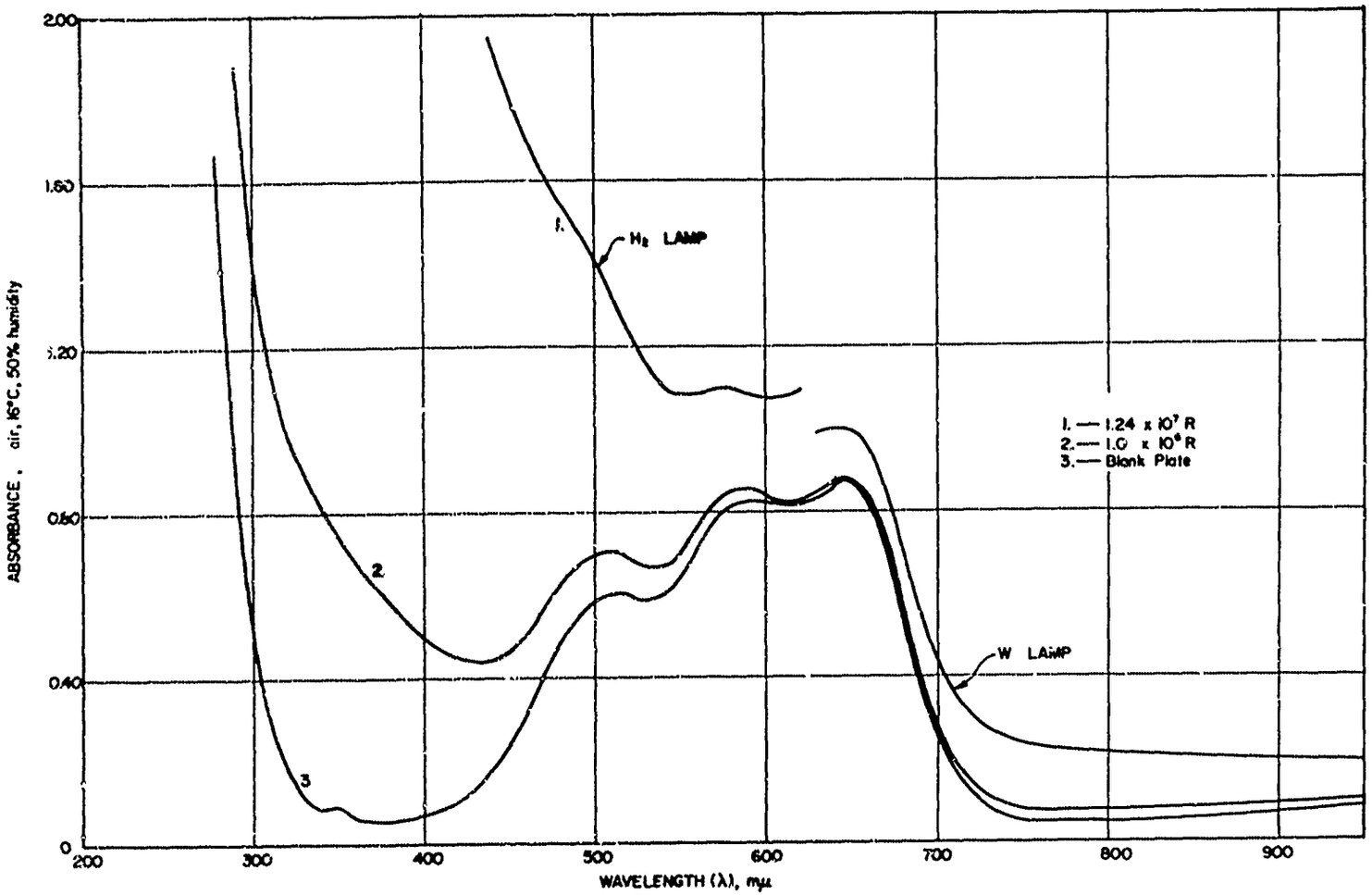

Figure 3.2 Absorbance as a Function of Wavelength, Exposure as a Parameter.

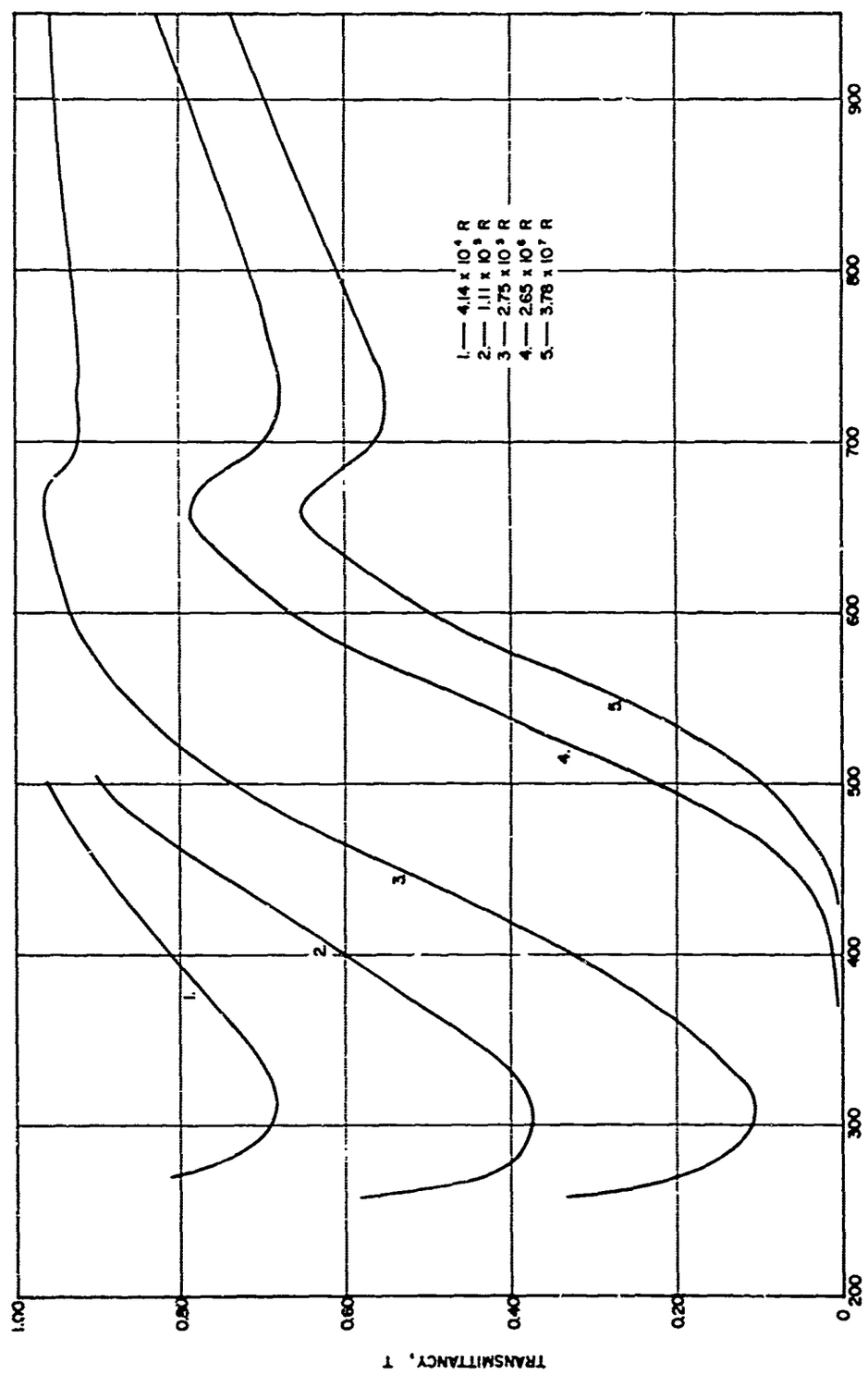

Figure 3.3 Transmittancy as a Function of Wavelength.

Between boxes and sometimes between batches there is a larger variation, typically: $\sigma(\Delta T) = 0.011$, $\sigma(\Delta t) = \pm 1.0\%$ where t is the thickness of a given plate.

3.4 Energy Dependence.

The photon energy response of the unshielded cobalt borosilicate glass plates is reproduced in Figure 3.4 (Reference 7). Due to the low-energy overresponse, energy discrimination shields have been developed by EG&G. They are 1/2 inch in diameter, 3/4 inch long aluminum, lead, and graphite cylinders (Figure 3.1). Energy response for shielded plates is presented in Figure 3.5.

Since exposure environment has a significant effect on the dosimeter's response, the dosimeters are exposed and calibrated in energy discrimination shields.

3.5 Exposure Rate Dependence.

Exposure rate has been investigated by EG&G (Reference 8) who found that the plates are exposure-rate independent to 10^{9} R s^{-1}. Results of work at this Laboratory indicate that the rate independence extends to 5.5×10^{11} R s^{-1}.

3.6 Neutron Sensitivity.

The cobalt glass plates are known to respond to thermal neutrons. Therefore, they are usually exposed to mixed radiation fields in 2 mm thick pressed and sintered ^{6}LiF thermal neutron shields. These shields have thermal neutron attenuation factors of 1,000. If the shields are not used and the thermal neutron fluence is known, a numerical correction can be applied. The thermal neutron sensitivity is 3.6×10^{-9} R/(n cm^{-2}).

The fast neutron response is not well known. EG&G estimate the sensitivity to be within ±50% of 5.8×10^{-11} R/(n cm^{-2}) (Reference 9). Experiments conducted at USANDL indicate the 14 MeV neutron sensitivity is $2.1\ (\pm 0.8) \times 10^{-10}$ R/(n cm^{-2}).

3.7 Fading.

Figures 3.6 and 3.7 show the change in absorbance as a function of time for exposures measured at 300 and 720 mμ, respectively. Friddell (Reference 6) found an empirical function for fading versus time and absorbance with wavelength as a parameter:

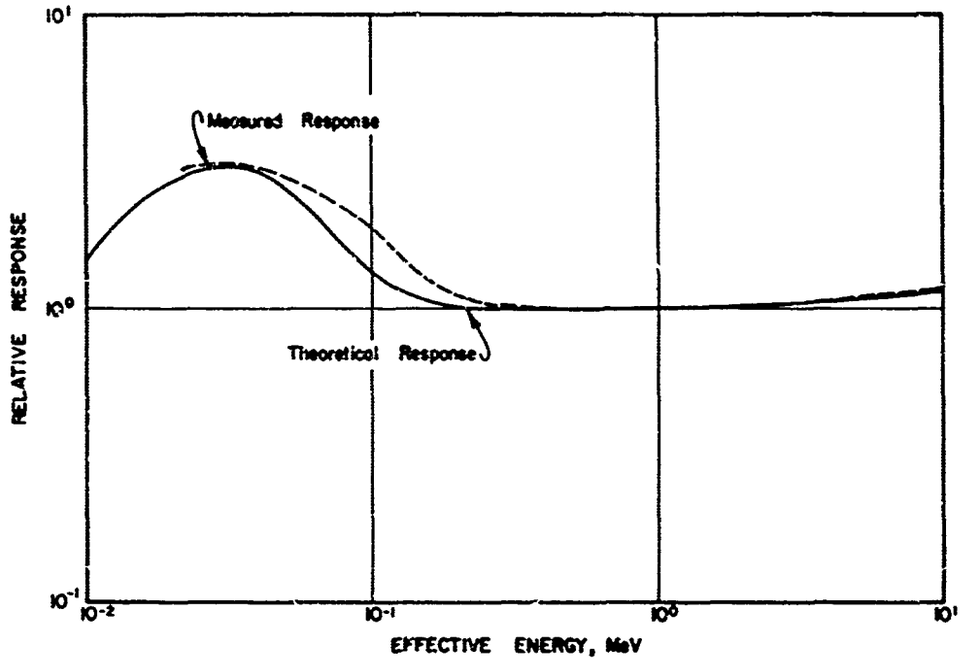

Figure 3.4 Energy Response of Unshielded Cobalt Glass Dosimeters.

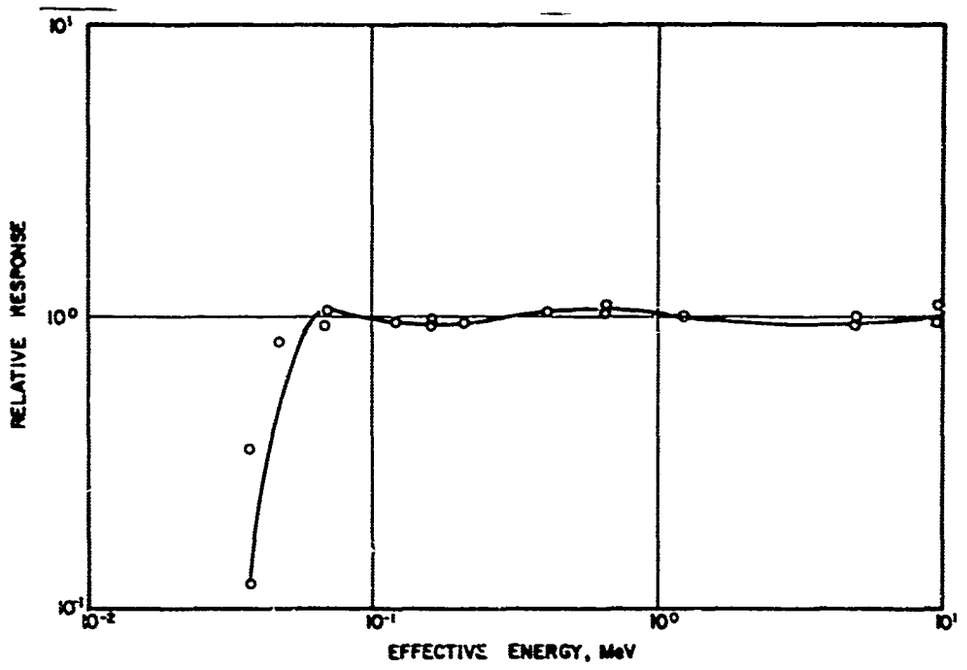

Figure 3.5 Energy Response of Shielded Cobalt Glass Dosimeters

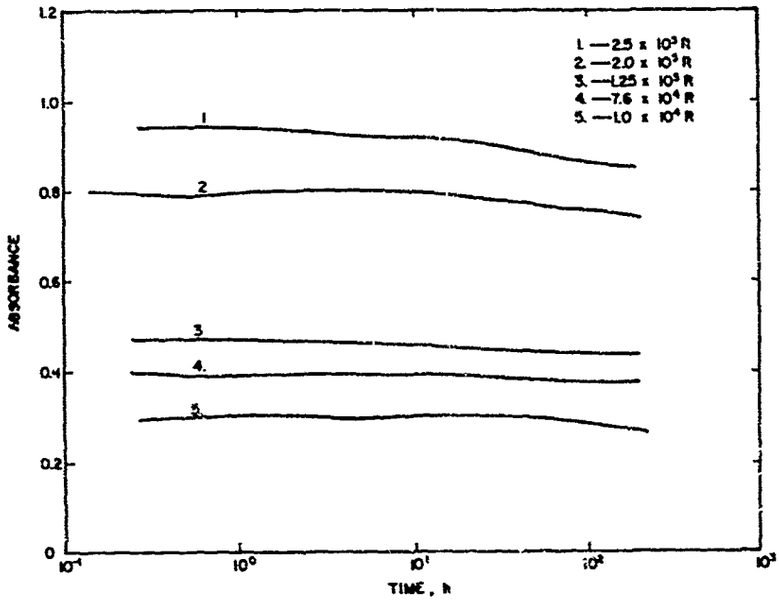

Figure 3.6 Absorbance Fading of Glass at 300 mμ with Exposure as Parameter.

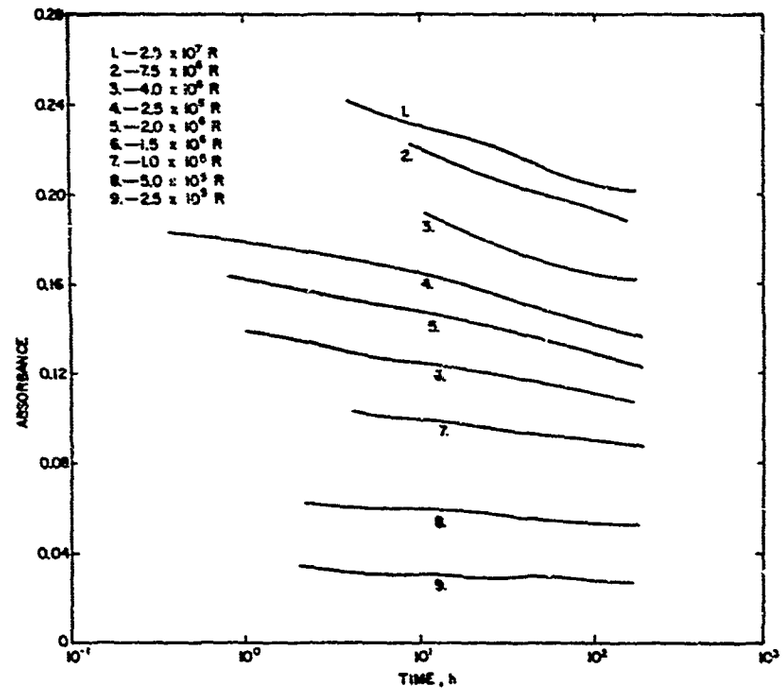

Figure 3.7 Absorbance Fading of Glass at 720 mμ with Exposure as Parameter.

$$A\,(t,\lambda) = A\,(t_0,\lambda) - n\log_{10}\,(t/t_0)\ , \qquad (3.1)$$

where $$n = k\,[A(t_0,\lambda)]^{4/3}\ , \qquad (3.2)$$

A is the absorbance ($\log_{10}\frac{1}{T}$, T = transmittancy),

t is the time since exposure, in hours,

λ is the wavelength in millimicrons,

t_0 is the unit of time, 1 hour,

and k is an empirical constant depending on wavelength:

$k_{300} = 0.06 \pm 0.02$ when $0.1 \leq A \leq 1.5$,

$k_{720} = 0.20 \pm 0.02$ when $0.05 \leq A \leq 0.2$.

Figure 3.8 shows the absorbance fading of the cobalt glass plates as a function of time at 300 mμ with temperature as a parameter. Fading may also be corrected by using a calibration curve made with plates which have been permitted to fade the same length of time and under similar storage conditions as the experimental plates.

3.8 Annealing and Reuse of Dosimeters.

The cobalt borosilicate glass plates can be annealed and reused. Heating from 5 to 60 minutes at 550°C will erase nearly all the absorbance of the plates. The average annealed plate shows a transmittancy of 101 percent compared with an unused plate. Results obtained when the annealed plates were reexposed indicated an increase in sensitivity. However, this sensitivity decreased after repeated annealing and exposures.

When differential annealing was performed on the plates, it was found that the response was affected slightly by the length and temperature of annealing as well as the magnitude of the original exposure. The plates begin to melt at 620°C.

The reuse of annealed plates is not recommended because a loss of precision (approximately 3 percent) results from the fluctuations in sensitivity.

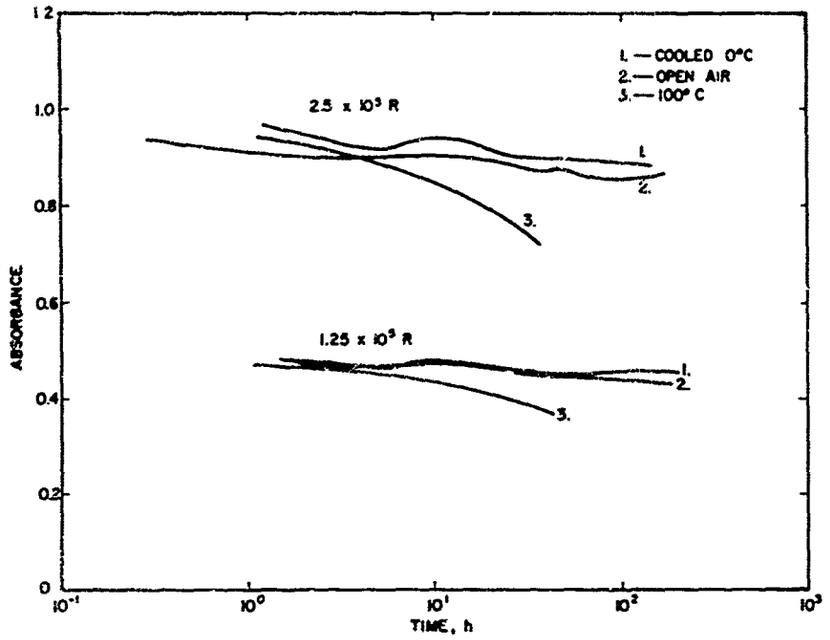

Figure 3.8 Absorbance fading of glass at 300 mμ, with temperature as parameter.

3.9 Effect of Pre-Irradiation Exposure.

In an attempt to improve the accuracy of response to small exposures, a number of plates were exposed to 5,000 R and then exposed to increments of 500 R. No change in accuracy was observed; that is, the standard deviation of the readings of exposed plates was not significantly different from that of virgin plates with comparable exposures.

3.10 Cleaning.

To obtain accurate, reproducible readings, a fast and effective method of cleaning the plates is of great importance. Five methods were tried:

1. Rubbing with optical paper.
2. Washing in warm water and sparkleen, rinsing with hot water.
3. Immersing in acetone, washing with water, rinsing with isopropyl alcohol.
4. Immersing in 1 M hydrochloric acid, rinsing with water.
5. Immersing in 0.05 N sodium dichromate and sulfuric acid solution, rinsing with distilled water.

The plates were cleaned and read. They were then soiled with skin oil, graphite, and mineral oil, and read again. The standard deviations of the differences in readings before and after soiling were:

Method	Standard Deviation
1	0.2 percent T
2	0.3 percent T
3	0.2 percent T
4	0.1 percent T
5	0.2 percent T

Methods Nos. 4 and 5 have the disadvantage of etching the plates when left in the bath too long. Since method No. 1 is the simplest and is reasonably effective, this technique was adopted.

4. STANDARD OPERATING PROCEDURE

1. A "standard" unirradiated plate is prepared by heating an unirradiated plate for several minutes at 500°C and then bleaching it for approximately 24 hours at 300 mμ in the spectrophotometer. This procedure is necessary because absorbance of an untreated unirradiated plate seems to change spontaneously despite frequent cleaning. In extreme cases, changes of 3.0 to 5.0 percent T have been observed.

2. Two or three plates are selected at random from each batch of 25 and reread at 300 mμ. If there is a difference of 1.0 percent between two of these plates, all the plates in the batch are preread. If this is not the case, 1 plate is kept as a background and the other 24 plates are loaded into 8 energy discrimination shields. For calibration and precision work, every plate is read prior to irradiation.

3. The calibration curve, (Figure 2.2) is checked by exposing a set of plates in the gamma cell. If the newly determined points disagree with the calibration curve within 5 percent, a new calibration curve is constructed.

4. After exposure, the plates are removed from their shields and cleaned with optical paper.

5. The plates are placed into the plate holder and their transmittancy is measured relative to the standard plate; at 300 mμ, 10^3R ≤ Exposure ≤ 10^5R and at 720 mμ, 10^6R ≤ Exposure ≤ 10^7R. Care must be taken in placing the dosimeter in the holder because faulty positioning can cause errors up to 0.5 percent T.

6. The transmittancy is converted to absorbance by standard conversion tables or by Equation 2.2. The absorbance of the background plate is measured and subtracted from the absorbances of the irradiated plates in its batch.

7. The absorbance readings of the three dosimeters in each energy discrimination shield are averaged. The exposure corresponding to the average absorbance is read from a calibration curve made with plates stored similarly to the experimental plates. Alternately, the absorbance 1 day after irradiation may be calculated according to Equation 3.1 and then read from the 1-day calibration curve (Figure 2.2).

8. Thermal and fast neutron corrections are made where neutron fluence values are available. If this is the case, usually both the corrected and uncorrected exposures are reported.

5. PRECISION AND ACCURACY

5.1 Precision.

The limit of precision of the spectrophotometer as currently used is approximately 0.08 percent T. The null balancing scale is linear in T so that 0.08 percent T varies from an absorbance of 0.0003 at 100 percent T to 0.05 at 1 percent T. Thus, the precision of the spectrophotometer is a monotonically decreasing function until, at absorbances of 3.2, the expected error is as large as the reading.

The dosimeters show within-batch variations of typically ± 0.4 percent T, which correspond to an exposure of about 500 R at 300 mμ. Between boxes, the transmittancy variations are typically ±1.1 percent T. Three physical situations are distinguishable: (a) variations in plate thickness, (b) variations in color center concentration, (c) variations in degree of bleaching. Variations a and b are typically ±1 percent, and affect all exposures similarly. The effect of c is negligible at 10^5 R and is important only at 5×10^3 R or below.

At exposures of 5×10^5 R and above, nonlinearity and fading become important factors in the precision of the system. The system is routinely used without special precautions and the precision in pure gamma fields is approximately:

±25 percent when 10^3 R ≤ E ≤ 5 × 10^3 R,
±5 percent when 5 × 10^3 R ≤ E ≤ 2 × 10^4 R,
±3 percent when 2 × 10^4 R ≤ E ≤ 5 × 10^5 R,
±10 percent when 5 × 10^5 R ≤ E ≤ 5 × 10^6 R.

In mixed radiation fields, the variations range from 10 to 20 percent between the three plates exposed in each energy discrimination shield.

5.2 Accuracy.

Accuracy and precision are directly related by the uncertainty of the ^{60}Co calibration procedure. This uncertainty is ±2 percent, which is the standard deviation of several Fricke ferrous sulfate dosimeter determinations. For other monoenergetic gamma fields, the uncertainty in the relative photon energy response function is important. For an unknown spectrum, this is less than 10 percent.

In mixed neutron and gamma fields, it is difficult to define accuracy. In the past this Laboratory has observed 25 to 50 percent variations in results from dosimeter systems exposed in close proximity. The exposure environment is quite different from the calibration conditions, and results from the compared dosimetry systems are not necessarily within the recommended exposure ranges. Finally, the neutron sensitivities of the cobalt plates are not well known.

6. CONCLUSIONS

Cobalt borosilicate glass is an inexpensive, efficient, and reliable passive dosimeter for gamma exposures from approximately 5 × 10^3 to 5 × 10^8 R. The use of energy discrimination shields provides the system with an almost flat photon-energy response from 60 keV to 10 MeV, and thermal neutron shields eliminate 99.9 percent of the thermal neutron fluence. The system is rate independent to 5.5 × 10^{11} R s^{-1}.

The main problems associated with the system are fast neutron corrections, fading, saturation effects, and variations in plate parameters.

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13. ABSTRACT

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