# RADIOCHROMIC DYE DOSIMETRY OF NEUTRON AND GAMMA FIELDS

by

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19 February 1969
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ABSTRACT

The application of leuco malachite green carbinol to dosimetry of neutron and gamma fields was investigated. This radiochromic dye was dissolved in a poly (4-chlorostyrene) base. The basic sensitivities of this system were determined to be:

| | | |
|---|---|---|
| Gamma radiation | (70 r/min) | $1.44 \times 10^{-6}$ ΔOD/mil/r |
| | (660 r/min) | $7.65 \times 10^{-6}$ ΔOD/mil/r |
| Thermal neutron response | | None |
| Fast neutron response | | $5.5 \times 10^{-16}$ ΔOD/mil/(n/cm$^2$) |

Methods to improve these intrinsic sensitivities are discussed.

# CHAPTER 1

## INTRODUCTION

In the 1870's it was observed that light falling on certain materials would cause them to change color (1). These changes were reversible, the color formed reverting to the original hue after the light was removed. Marckwald (in (2)) studied them and introduced the term phototropy for these reactions. Since then many of these reactions both in solids and in solutions have been observed.

In 1919, Lifschitz and Joffé (3,4) using ultraviolet light as the exciting mechanism found they could form the colored dye derivatives from an originally colorless (leuco form) triphenylmethane dye derivative solution. These reactions were still reversible, the color disappearing after the ultraviolet light was removed. In 1935 Harris and Kaminsky (5) determined that the amount of photolysis was proportional to the amount of light absorbed, and later Sporer (6) determined that two photoreactions occurred in these systems; namely, cleavage occurs in solvents having a low dielectric constant, while in solvents with a high dielectric constant, dye formation is the prominent reaction.

Recently triphenylmethane dye derivative solutions have been subjected to ionizing radiation and the same color changes are apparent (7). The response has been found to be essentially proportional to the energy absorbed yet independent of the variation in the energy spectrum (8).

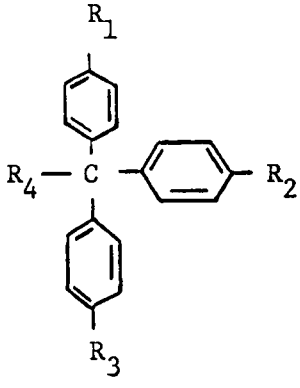
The introduction of activators into the solution has served to stabilize the color change produced and allowed the system to be used as a radiochromic dosimeter since the color change is proportional to the amount of ionizing radiation absorbed.

Work has been done to show the response of triphenylmethane dyes to gamma radiation (8), but neutron fields have not been investigated. This work extends the calibration of the triphenylmethane dye dosimeter when exposed to neutron as well as gamma fields. Techniques to increase this intrinsic color change are developed.

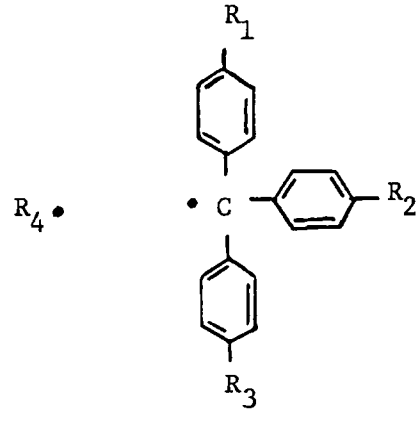
## CHAPTER 2

## THEORY

Exposing triphenylmethane dye derivative molecules to ionizing radiation results in excitation, ionization, and bond breakage. The formation of color centers in the species is produced as the leuco form is broken into the dye intermediate form (see Fig. 1) and is subsequently stabilized by electron rearrangement and ion pair formation. This was shown by Sauer (9) and group during electron pulse experiments; that is, a triphenylmethyl radical (the color forming radical) and a negative ion were formed by dissociative electron attachment from triphenylmethyl carbinol.




Basic Triphenylmethane Dye Derivative




Triphenylmethyl Dye Radical Formation Upon Exposure to Ionizing Radiation

Fig. 1. Result of Exposing Triphenylmethane Dye Derivatives to Ionizing Radiation.

$R_4$ is a group such as H, OH, CN, $OCH_3$ and $R_1$, $R_2$, $R_3$ can be H, $NH_2$, $N(CH_3)_2$.

Neutrons also produce bond rupture but the mechanism differs. For thermal neutrons, energy deposition within the dosimeters is due to 1) capture gammas having interactions within the dosimeter, 2) nuclei recoils from $(n,\gamma)$ interactions or 3) charged particles, by-products of thermal capture (10). Proton recoil is the primary energy deposition mechanism for fast neutrons. Again, the overall result of energy deposition within the dosimeter is the rupture of bonds in the leuco triphenylmethane dye species leading to the formation of color centers in the molecules.

In forming the color centers, the amount of energy transferred to the medium by a fast neutron can be calculated by the first collision dose formula (11).

$$D_f(E) = k\, E_n \sum_i N_i \sigma_i \varepsilon_i \qquad (1)$$

k, a conversion factor

$E_n$, the neutron energy

$N_i$, the number of atoms/gm of the $i^{th}$ species

$\varepsilon_i$, the average fraction of energy imparted to the $i^{th}$ interacting nucleus $\varepsilon_i = \dfrac{2A_i}{(A_i+1)^2}$

$A_i$, the atomic mass of the $i^{th}$ nuclei

$\sigma_i$, the elastic scattering cross section of the $i^{th}$ species.

However, these films are so small that not all of the energy imparted to the nucleus will be retained in the film therefore the expression for $\varepsilon_i$ cannot be used. In order to make use of the properties of the film a simplified expression of the form

$$\Delta OD = \Phi_n(E) \times F(E) \qquad (2)$$

is used. ΔOD is the change in density of color centers the dosimeter undergoes when subjected to a fast neutron fluence (ΔOD is measured with a spectrophotometer and known in terms of the optical density). $\Phi_n(E)$ is the neutron fluence of energy E to which the film is exposed. F(E), the sensitivity of the film, is the efficiency factor that will relate the color change to the neutron fluence. F(E) was determined experimentally by exposing a group of 3.2, 3.5 and 3.7 mil thick films to fast neutron fluxes. Then from this the sensitivity can be determined by

$$F(E) = \frac{\Delta OD}{\Phi_n(E)} \qquad (3)$$

The color centers that are formed are unstable since recombination can occur. To prevent this, activators, substances which are essentially sources of electrons, are introduced into the solution. Activators serve to stabilize the radicals formed and preserve the color change by ion pair formation. These activators are compounds such as aromatic amines, phosphoric acid esters, aromatic carbinols, halides, and some alcohols (12).

The color change formed during irradiation is defined in terms of the optical density, OD, and is read directly with a spectrophotometer. The optical density is defined by (13)

$$OD = \log \frac{I_o}{I} \qquad (4)$$

where $I_o$ is the intensity of light incident on the surface of the film and I is the intensity of light transmitted through the film.

With the symbols $I_i$ as the intensity of light transmitted through an initially unexposed film and $I_f$ as the intensity of light transmitted through a radiation colored film, the relationship between optical density and color change is

$$\Delta OD = OD_{final} - OD_{initial} \tag{5a}$$

$$\Delta OD = \log \frac{I_o}{I_f} - \log \frac{I_o}{I_i} . \tag{5b}$$

Beer-Lambert's law relates the optical density to the thickness of the film dosimeter by (13)

$$OD = \varepsilon c t \tag{6}$$

where $\varepsilon$ is the molecular extinction coefficient expressed in liters/mole/mil

$c$ is the concentration of the solute (dye) in moles/liter

$t$ is the thickness of the solution (film) in mils.

Since the color change is a function of the thickness of the dosimeter, the normalized optical density (optical density change per unit thickness) is used for the dose correlation. This modifies equation 5b to

$$\frac{\Delta OD}{t} = \frac{1}{t} \left( \log \frac{\frac{I_o}{I_f}}{\frac{I_o}{I_i}} \right) \tag{7a}$$

And since in all of the measurements the reference $I_o$ was air then the normalized change in optical density can be expressed as

$$\frac{\Delta OD}{t} = \frac{1}{t} \log \frac{I_i}{I_f} . \tag{7b}$$

Fig. 2 shows a typical absorption spectrum for the dosimetry film used in this work as a function of radiation exposure. $I_i$ and $I_f$ are determined from the spectrum; t is measured separately hence the normalized optical density change is known. Because internal reflections in the film can cause the intensity of light transmitted to be less than it would in an ideal system, the optical density change is compared to a non-absorbing wavelength region in the same dosimeter. The mathematical treatment of the absorption data is shown in Appendix A.

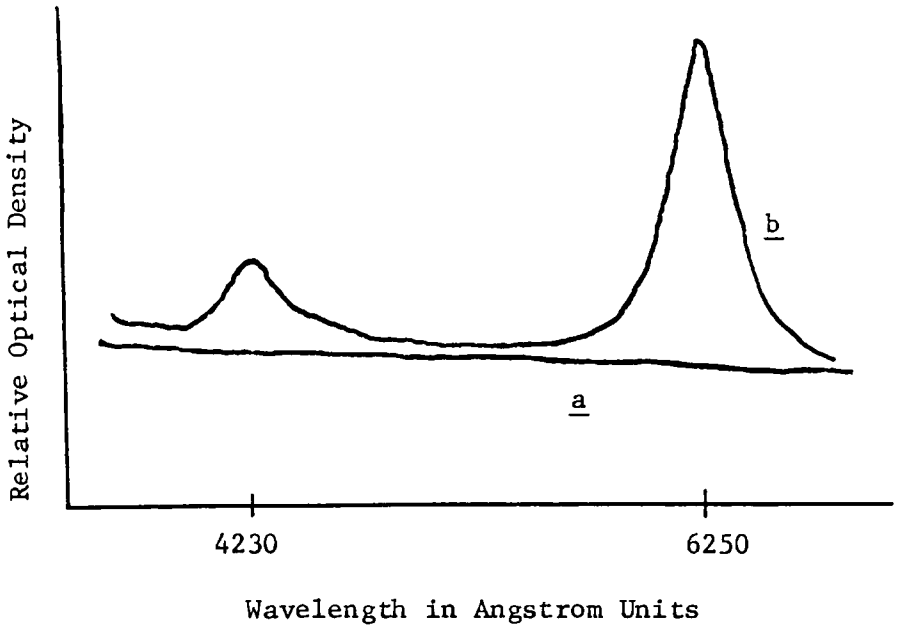



Fig. 2. Typical Absorption Spectrum for a Dosimeter.
curve a is the unexposed film
curve b is the absorption spectrum after irradiation.

The dosimetric system used is one per cent malachite green carbinol in a poly (4-chlorostyrene) plastic base. The activators are the chlorine atoms in the base, and the organic groupings with reference to Fig. 1 are $R_1 = R_3 = N(CH_3)_2$, $R_4 = OH$, and $R_2 = H$. This film was supplied by Dr. Larry Harrah of the Sandia Corporation, Albuquerque, New Mexico. When unexposed, the films have a water white color but take on progressively deeper shades of blue as they are exposed to radiation. The films are also sensitive to ultraviolet radiation and have to be shielded from light to prevent erroneous results. Upon irradiation two peaks appear: one at 6250 Å and the other at 4230 Å. The peak at 6250 Å is used as a measure of radiation response.

There is also a reaction related to trace amounts of chloroform present in these films when irradiated in an oxygen environment. The mechanism involves formation of HCl and depletion of oxygen at the higher dose rates. The result is that irradiation in the presence of oxygen increases the response at a given dose rate and, in addition, introduces a dose rate dependence which decreases this enhancement as the dose rate increases.

# CHAPTER 3

## EXPERIMENTAL

It was necessary to expose the dosimeters to as pure sources as possible, in order to determine and calibrate the response of this dosimeter system to gamma and neutron fields. Following is a description of the facilities and procedures used to accomplish this.

### Gamma Irradiation Facility and Procedures

The gamma irradiation facility is located in the Engineering Building on The University of Arizona campus. The source is essentially a Co-60 point source (approximately 450 curies) hermetically sealed in a cylinder 1 1/2" in diameter and 1 1/2" long positioned with the axis directed flush onto the face of an irradiation wheel (Fig. 3). Rotating the wheel insures an equal dose to all positions on a fixed radius of the wheel.

To determine the average exposure dose rate on the irradiation wheel at some fixed distance from the source the dose rate was measured at intervals along the face of the wheel by inserting a Victoreen Radacon probe into a hole in the wheel and turning the wheel to the desired position. With the use of a polar planimeter to accomplish the integration, the average dose rate can be determined from the relationship:

$$\bar{\dot{R}} = \frac{A}{2\pi} + \dot{R}_o . \qquad (8)$$

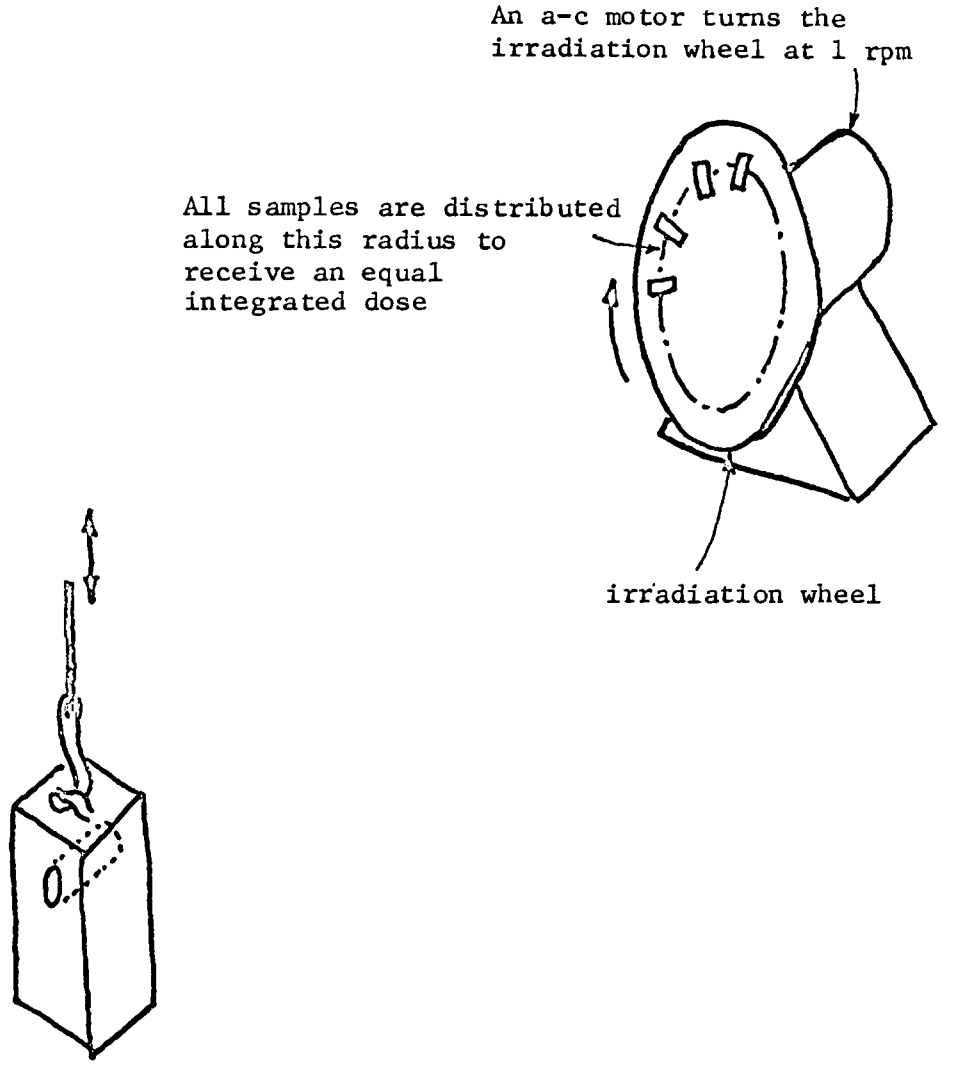



Fig. 3. The Gamma Irradiation Facility.

where $\bar{\dot{R}}$ is the average exposure dose rate in air at the wheel face in r/min, and

$$A = K \int_{o}^{2\pi} \dot{R}(\theta) d\theta \qquad (9)$$

$K = 12.80$ radians - r/min/in$^2$ and is a conversion factor dependent on the particular planimeter used. The planimeter integrated area is in units of in$^2$. $\dot{R}(\theta)$ is the variation of the exposure dose rate along the fixed radius line of the wheel (see Fig. 4). $\dot{R}_o$ is a reference exposure dose rate and is equal to 55 r/min. The variation in dose rate along the wheel was thus determined to be 94.2 $\frac{\text{radians-r}}{\text{min}}$ corresponding to an average dose rate,

$$\bar{\dot{R}} = \frac{94.2}{2\pi} + 55 = 70 \text{ r/min.} \qquad (10)$$

The dose rate can be varied by positioning the irradiation wheel closer or farther away from the source and the same analysis used in determining the average dose rate at the new position.

The dosimeters, cut into 1 cm x 3 cm rectangles, a convenient size to handle and read, were placed on the wheel. Since there is no light in the gamma irradiation chamber when the irradiation is in progress there is no need to protect the dosimeters. However, a light shield was necessary for placing and removing them from the chamber. This was accomplished by putting a hood over the samples.

The dosimeters were exposed at two dose rates, 70 r/min and 660 r/min and given total exposure doses of 100 Kr in increments of 20 Kr. At the 70 r/min dose rate this was 4.76 hours for every 20 Kr and at 660 r/min this was 0.505 hour for every 20 Kr interval.

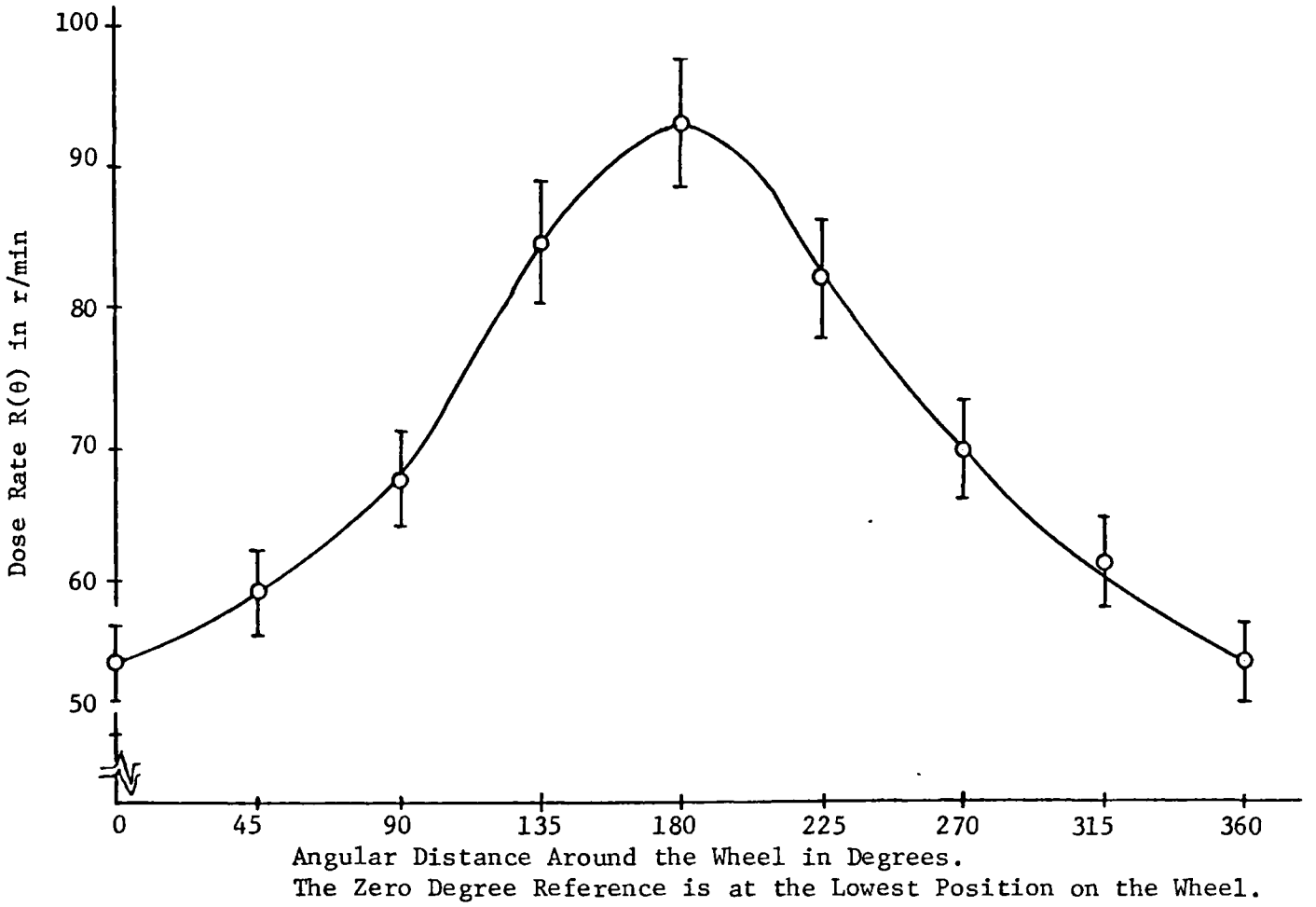



Fig. 4. Variation In Dose Rate On the Irradiation Wheel.

In an attempt to increase the gamma ray response uranium foils of 2.45 mil thickness were also put around the lower half of the dosimeters during the irradiation at 660 r/min. Therefore, for the exposure at 660 r/min two distinct sets of information were obtained, the intrinsic response at 660 r/min and the increase in sensitivity due to a high atomic number material present.

## Fast Irradiation Facility (FIR) and Procedures

The Fast Irradiation Facility is an aluminum tube that is inserted directly into the F ring of the TRIGA reactor core. The end of the tube in the core has a cadmium and gold shield over it so that the thermal flux is minimized while the fast flux is essentially retained. The gamma dose rate measured with a probe was determined to be 1475 r/min/KW.

In the test a polyethylene rabbit containing the dosimeters was exposed to the reactor environment. The dosimeters were covered with a 4 mil thick layer of lead that stopped any charged particles from depositing their energy in the film and also acted as a light shield. It will be shown later that a threshold for detectable effects from fast neutrons on the film exists at $10^{12}$ n/cm$^2$ while thermal neutrons have no measureable effect on the film at these fluences. In this test the fast neutron fluence was below the detection threshold so that the only effects would be from the gamma radiation in the core.

## ARGONAUT Reactor and Procedures

The ARGONAUT is a 200 watt enriched uranium water moderated reactor located in building 25 at the Argonne National Laboratory, Argonne, Illinois. The fuel is in an annulus and is surrounded by a

graphite cube, see Fig. 5. The film samples to be irradiated are inserted into the number 4 hole of a graphite stringer (J-10) which when inserted into the reactor is part of the graphite cube structure (14). In this instance light shielding of the dosimeters was accomplished by surrounding them with a sheet of brown paper.

The flux, measured by gold foil activation, is quite thermalized at the number 4 hole (cadmium ratio = 47.27) and at 200 watts the thermal flux is $1.08 \times 10^8 n/cm^2/sec$. The gamma dose in the core is 50 r/hr/watt (15) and an order of magnitude less than this at the number 4 hole. The limitations on the gamma field can be deduced from the thermal neutron film calibration. Since there was no perceptible film color change in seven hours of irradiation at 200 watts and the minimum film sensitivity for a 3.9 mil thick film is approximately 10 Kr, the gamma dose at the number 4 hole is certainly less than 7 r/hr/watt.

In the test for the thermal neutron sensitivity of the dosimeters a seven hour irradiation in the number 4 hole was used. Since the flux was nearly thermalized, the total fluence of $2.8 \times 10^{12}$ $n/cm^2$ was assurance that any fast neutron effects were below their threshold and also the gamma dose was below its threshold.

The same hole and the same irradiation procedure was used to check a technique to create a thermal neutron response. Here a layer of boron-10 powder was sprinkled onto a piece of Scotch tape. The average boron concentration with this method was determined to be $1.92$ $mg/cm^2$. Two of these boron tapes are used around the dosimeter

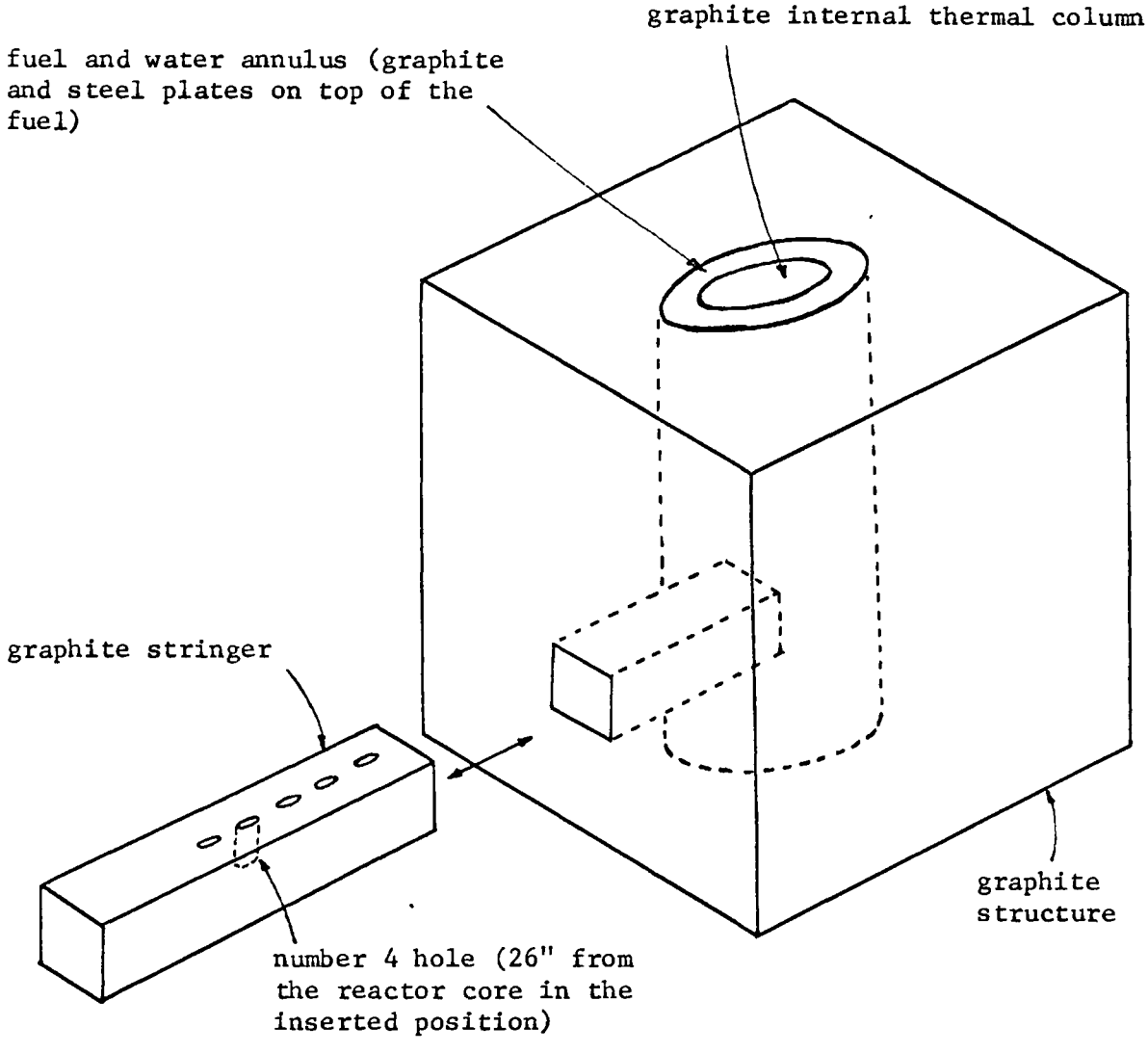



Fig. 5. Schematic of the ARGONAUT Reactor.

sandwiching the dosimeter between them with the boron side toward the film. This package was also irradiated at 200 watts and received a total fluence of $2.8 \times 10^{12}$ n/cm$^2$.

## Neutron Generator and Procedures

The neutron generator used was made by Texas Nuclear and is located in the Chemistry Building at the Argonne National Laboratory, Argonne, Illinois. The target used was tritium absorbed on a layer of titanium which is evaporated onto a 0.010 inch copper backing. The accelerating potential was 150 KV and a $T(d,n)He^4$ reaction employed with $E_n$ the neutron energy equal to 14.7 MeV (16).

The dosimeters were put into a sample holder for the neutron generator equipment. They were surrounded on each side by 15 mils of polyethylene. Previous experiments with a Van de Graaff generator at Argonne utilizing a $Li^7(d,n)\ Be^8$ reaction had determined that the fast neutron threshold was above $10^8$n/cm$^2$ in the energy range of 1 to 10 MeV. A layer of hydrogeneous material surrounding the dosimeter was used to enhance its reponse. The dosimeters were then enclosed inside a glass microscope slide to stabilize the assembly allowing it to fit into the sample holder and covered with brown paper to mask any light effects. Finally an aluminum foil was put on each package to later determine the flux to which it was exposed.

The packages were given exposures of 15 min at an average fast flux of $4.3 \times 10^8$ n/cm$^2$/sec 90 min at a average fast flux of $6.6 \times 10^8$ n/cm$^2$/sec and 3 hr. and 5 min at a average fast flux of $1.0 \times 10^9$ n/cm$^2$/sec. The intrinsic response of the film was also determined using this same

procedure with the exception that no polyethylene surrounded it during this test.

## Optical Density Measurements

In all cases the optical density of the film before and after the irradiation were measured with a Cary Model 14 Recording Spectrophotometer. The change in optical density after irradiation was normalized to unit film thickness (one mil) and is reported as the change in optical density (see Appendix A) per mil of film thickness per roentgen of dose received ($\Delta$OD/mil/r).

## Gamma Field Measurements

The gamma dose rates were measured with a Victoreen Radacon ionization chamber model 575. This instrument was calibrated by the manufacturer to $\pm$ 2% of the full scale meter reading which corresponds to a maximum gamma dose deviation of $\pm$ 20 r/min.

# CHAPTER 4

## RESULTS AND DISCUSSION

### Intrinsic Gamma Dose Response of the Dosimeter

Dosimeters of 3.9 mil thickness were positioned on the irradiation wheel in the gamma facility (as previously described). At a dose rate of 70 r/min the sensitivity of the film to gamma radiation was found to be $1.44 \times 10^{-6}$ $\Delta$OD/mil/r. The results are tabulated in Appendix B and shown in Fig. 6.

There was no reason to expect a dose rate dependence of this film; however, subjecting these films to a reactor environment showed conflicting results. In this experiment the Fast Irradiation Facility in conjunction with The University of Arizona TRIGA reactor was used. In the procedure described previously, the fast neutron fluence was below the detection threshold so that the only effects would be from gamma radiation in the core. The results showed a much lower $\Delta$OD than anticipated. Since the gamma dose rate in the core was much greater than the calibration dose rate, a dose rate dependence was suggested.

Further information on the response as a function of dose rate was obtained by exposing a group of 4.1 mil thick films to a gamma field of 660 r/min in the gamma irradiation facility. Again a 100 Kr total exposure with increments of 20 Kr was used. Fig. 6 shows the sensitivity of the 6250 Å peak to be $7.65 \times 10^{-7}$ $\Delta$OD/mil/r which further illustrates this dose rate dependence. The data are tabulated in Appendix C.

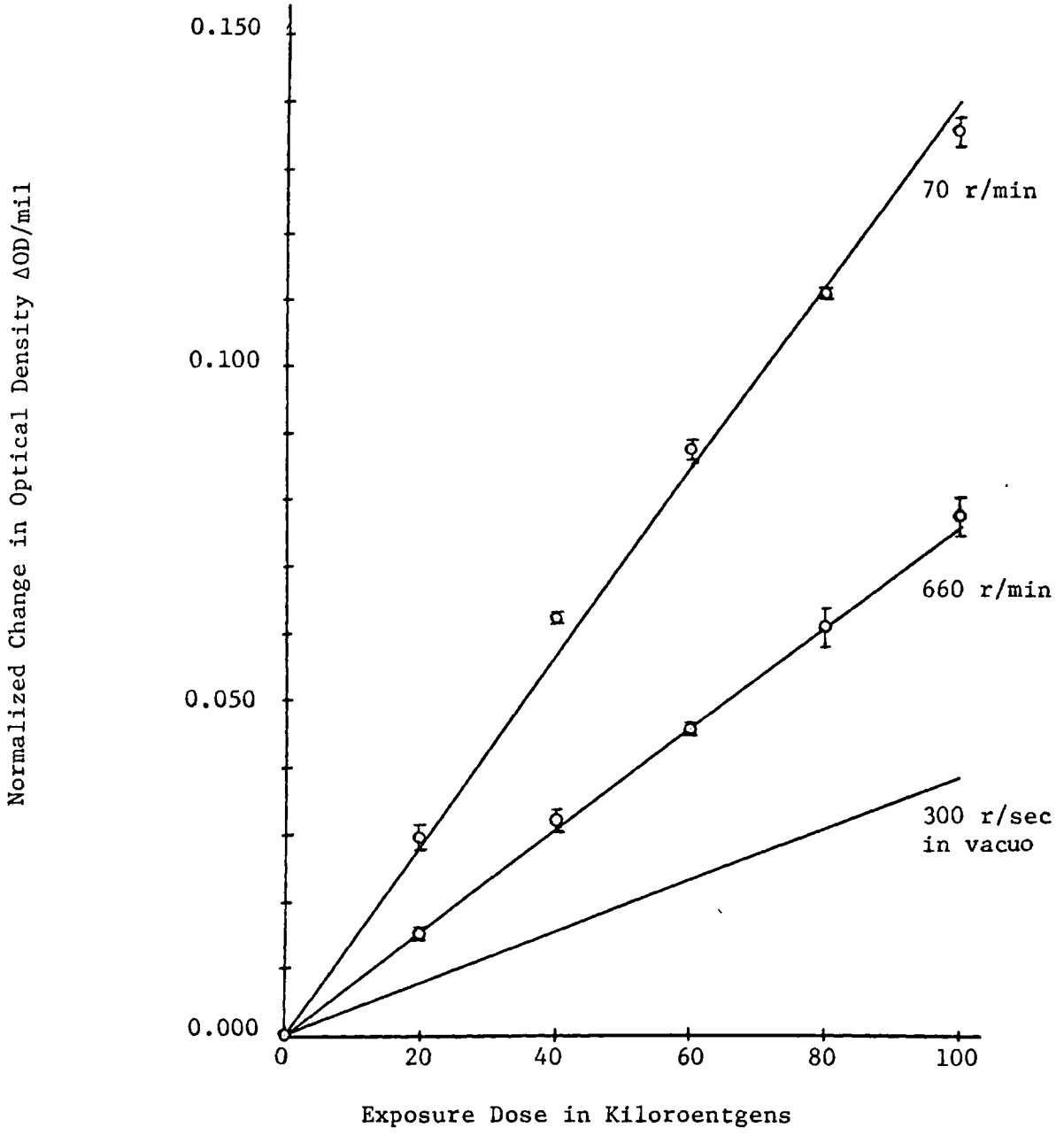



Fig. 6 Variation of the Dosimeter Response as a Function of Dose Rate

Information supplied by Dr. Harrah at a dose rate of 300 r/sec (in vacuo) shows a sensitivity to gamma radiation of $4.27 \times 10^{-7}$ ΔOD/mil/rad. The exposure-dose correlation, measured to absorbed, can be determined with the relationship (17):

$$D_{abs} = .877 \frac{\left(\frac{\mu_a}{\rho}\right)_{film}}{\left(\frac{\mu_a}{\rho}\right)_{air}} D_{air} \qquad (11)$$

where $D_{abs}$ is the absorbed dose in the film, in rads

$D_{air}$ is the exposure dose in air, in roentgens

$\left(\frac{\mu_a}{\rho}\right)_n$ is the mass energy absorption coefficient for medium n

$$\left(\frac{\mu_a}{\rho}\right)_{air} = 0.0268 \text{ cm}^2/\text{gm}$$

$$\left(\frac{\mu_a}{\rho}\right)_{film} = W_{C_8H_7Cl} \times \left(\frac{\mu_a}{\rho}\right)_{C_8H_7Cl} + W_{dye} \times \left(\frac{\mu_a}{\rho}\right)_{dye}$$

$W_n$ is the mass fraction of the $n^{th}$ component of a mixture. Since the dye comprises 1% of the film it can be neglected so that

$$\left(\frac{\mu_a}{\rho}\right)_{film} \simeq W_C \times \left(\frac{\mu_a}{\rho}\right)_C + W_H \times \left(\frac{\mu_a}{\rho}\right)_H + W_{Cl} \times \left(\frac{\mu_a}{\rho}\right)_{Cl}$$

$$= 0.02769 \text{ cm}^2/\text{gm}.$$

From Eq. 11 this yields $D_{abs} = 0.906\ D_{air}$. Thus the sensitivity at 300 r/sec in vacuo is

$4.27 \times 10^{-7}$ ΔOD/mil/rad x 0.906 rad/r or $3.87 \times 10^{-7}$ ΔOD/mil/r.

This is also plotted on Fig. 6 for comparison.

The explanation of this dose rate dependence is related to the oxygen effect and accentuated by the presence of a small amount of chloroform (.25%) which was used as a solvent in the film preparation. The net result is that a decrease in sensitivity is apparent at the higher dose rates due to the depletion of the oxygen supply which reduces the production of the color developing HCl from the chloroform in the film.

## Enhancement of the Gamma Response

With a 4 mil thick film the minimum measurable exposure dose is approximately 10 Kr to 20 Kr at 70 r/min in air. In order to measure lower doses a technique to increase the film sensitivity was investigated.

The intrinsic gamma ray response of the film is due primarily to gamma interactions within the film causing compton scattered electrons. These electrons in turn deposit a major portion of their energy within the film. The gamma sensitivity of the film should be increased by taking advantage of the photoelectric effect. To do this a layer of high atomic number material can be put around the film (the photoelectric effect is proportional to $Z^5$) while attempting to keep the layer thickness small so as not to attenuate the original gamma flux to a negligible value before reaching the film.

For this experiment the gamma irradiation facility was used as previously described. The dose rate was 660 r/min and uranium foils of 2.45 mil thickness were put on each side of a 4.1 mil thick dosimeter film. The attenuation of the gamma flux at the front face of the dosimeter

due to the uranium was 0.7%. The films were exposed to doses of 20 Kr to 100 Kr in increments of 20 Kr and this procedure gave a sensitivity of $1.83 \times 10^{-6}$ $\Delta$OD/mil/r or an increase of 2.38 times over that of the films without the uranium foils. The data are tabulated in Appendix C and the curve is shown in Fig. 7. Thus with uranium foils sandwiched around the dosimeter the minimum measurable dose is about 5 to 10 Kr.

Thermal Neutron Response of the Dosimeter

Determination of the dosimeter's neutron response involves consideration of the fast and thermal neutron effects. These effects were isolated in reactor and accelerator environments and investigated separately.

For the thermal neutron response the ARGONAUT reactor was used. Dosimeters of 3.2 mil thickness were given an integrated fluence of $1.05 \times 10^{12}$ n/cm$^2$ and dosimeters of 3.9 mil thickness were given an integrated fluence of $2.72 \times 10^{12}$ n/cm$^2$ and no change in optical density occurred. This was expected as the cross sections for the energy depositing processes are small hence the amount of energy deposited within the film is not sufficient to produce a response.

Thermal Neutron Response Utilizing the (n,$\alpha$) Reaction of Boron-10

Since there is essentially no intrinsic thermal neutron response of the dosimeter an attempt to create one using the high (n,$\alpha$) cross section of boron-10 was attempted. As described previously, a layer of boron-10 powder is sprinkled onto Scotch tape. The boron layer must be

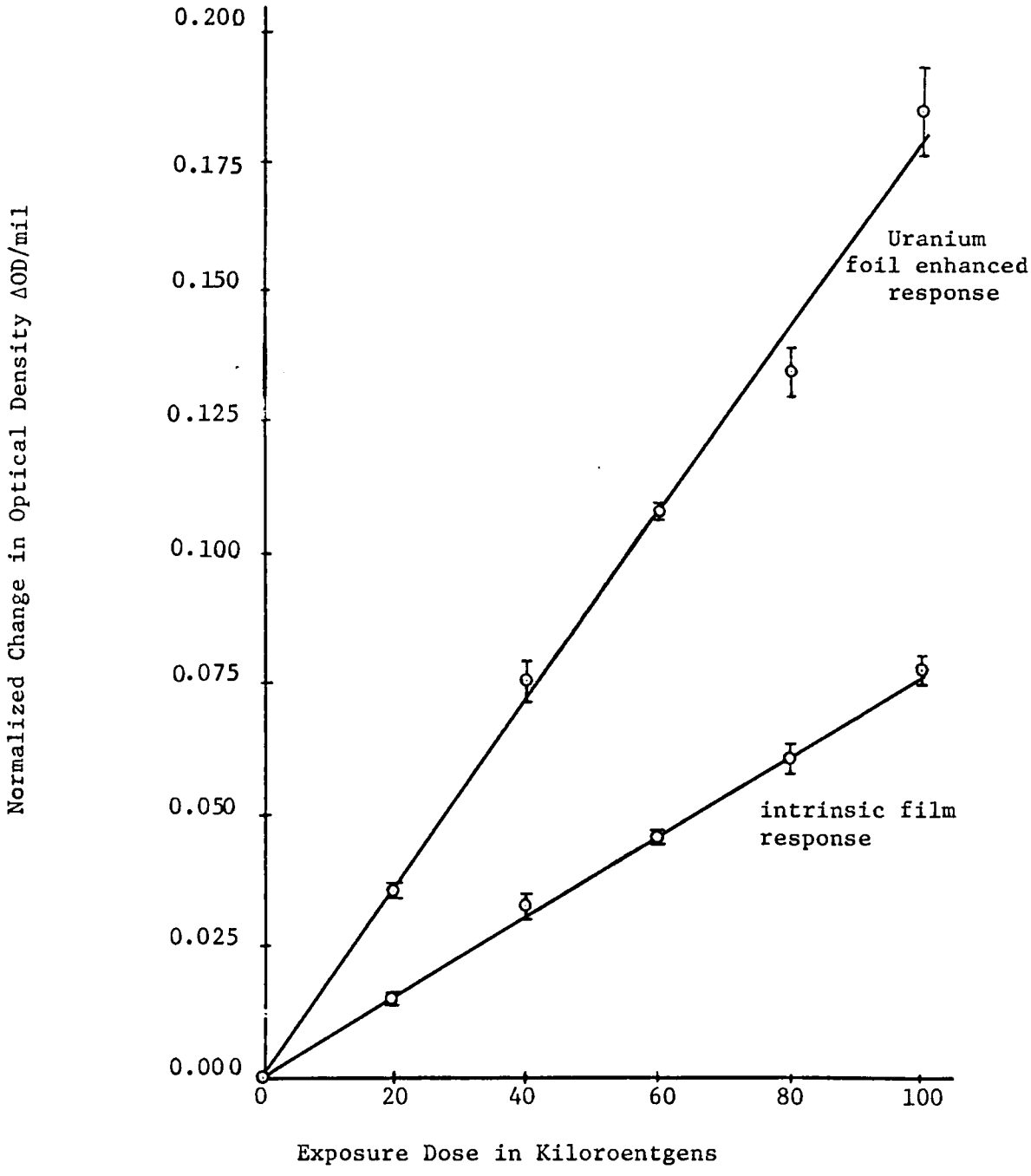



Fig. 7 Use of a Uranium Foil to Increase the Gamma Response of the Dosimeter.
Dose rate is 660 r/min.

kept thin so that the neutron flux incident on the front face of the tape is not significantly attenuated before emerging from the package and so that the alpha particles are not appreciably stopped before reaching the film. One boron tape strip is put on each side of the dosimeter and the package exposed to a thermal neutron flux. The $n,\alpha$ reaction, cross section of 3840 barns, produces alpha particles of 2.3 MeV energy. These alpha particles are of extremely short range (0.017 mm in the film) and essentially deposit their energy around their point of formation. With a thin boron layer, the film will receive a portion of this alpha particle energy resulting in a color change.

Exposing a 3.9 mil thick dosimeter to a total integrated fluence of $2.72 \times 10^{12}$ n/cm$^2$ resulted in an optical density change of 0.089. These data are reproduced in Appendix E. From this the film sensitivity to thermal neutrons with a 1.92 mg/cm$^2$ layer of boron-10 surrounding it is $8.39 \times 10^{-15}$ $\Delta$OD/mil/(n/cm$^2$) showing that this technique can be successfully applied to yield a thermal neutron response of the film.

Another technique which may be usable is to wrap gadolinium around the dosimeter. The large cross section for the $(n,\gamma)$ reaction of $^{64}$Gd (240,000 barns, 15.68% of naturally occurring Gd) would use the gamma response of the film to provide a thermal neutron sensitivity.

## Fast Neutron Response of the Dosimeter

The films were exposed to fluences of 3.25 to $9.34 \times 10^7$ n/cm$^2$ in the energy range of 1 to 10 MeV with no noticeable change in the film optical density. The sensitivity threshold was above the $10^8$ n/cm$^2$ level. Since this intrinsic threshold is so high a method was developed

to try to improve the sensitivity. The technique was to place a layer of hydrogenous material on each side of the film. When the film is exposed to a neutron flux the response will consist of two parts, one the intrinsic neutron response of the film, and two, an enhancement due to fast neutron interactions in the hydrogenous layers creating recoil protons which also deposit energy within the film. To maximize the response several factors should be considered; one of which is that excessive layers of hydrogenous material should be avoided since they will just serve to attenuate the fast flux, another is that the layer of hydrogenous material should not be so thick that recoil protons created on the forward layers will not penetrate to the film and deposit their energy there.

With these considerations in mind packets were prepared with 3.5 mil films sandwiched between three polyethylene films each 5 mils thick (proton range in film and polyethylene is approximately 32 mils). Neutrons of 14.7 MeV were produced by a T (d,n) reaction and monitored by aluminum foil activation. A three and one-half hour irradiation resulted in an integrated fluence of $10^{13}$ n/cm$^2$. The efficiency factor using this enhancement technique is $2.5 \times 10^{-15}$ $\Delta$OD/mil/(n/cm$^2$) while the intrinsic neutron ,efficiency factor established by a reference point at $7.0 \times 10^{12}$ n/cm$^2$ is $5.5 \times 10^{-16}$ $\Delta$OD/mil/(n/cm$^2$). The data are tabulated in Appendix D and shown in Fig. 8.

## Proposed Mixed Field Dosimeter

Combining the previous results a very compact dosimeter could be fabricated for mixed field applications. Consider the configurations

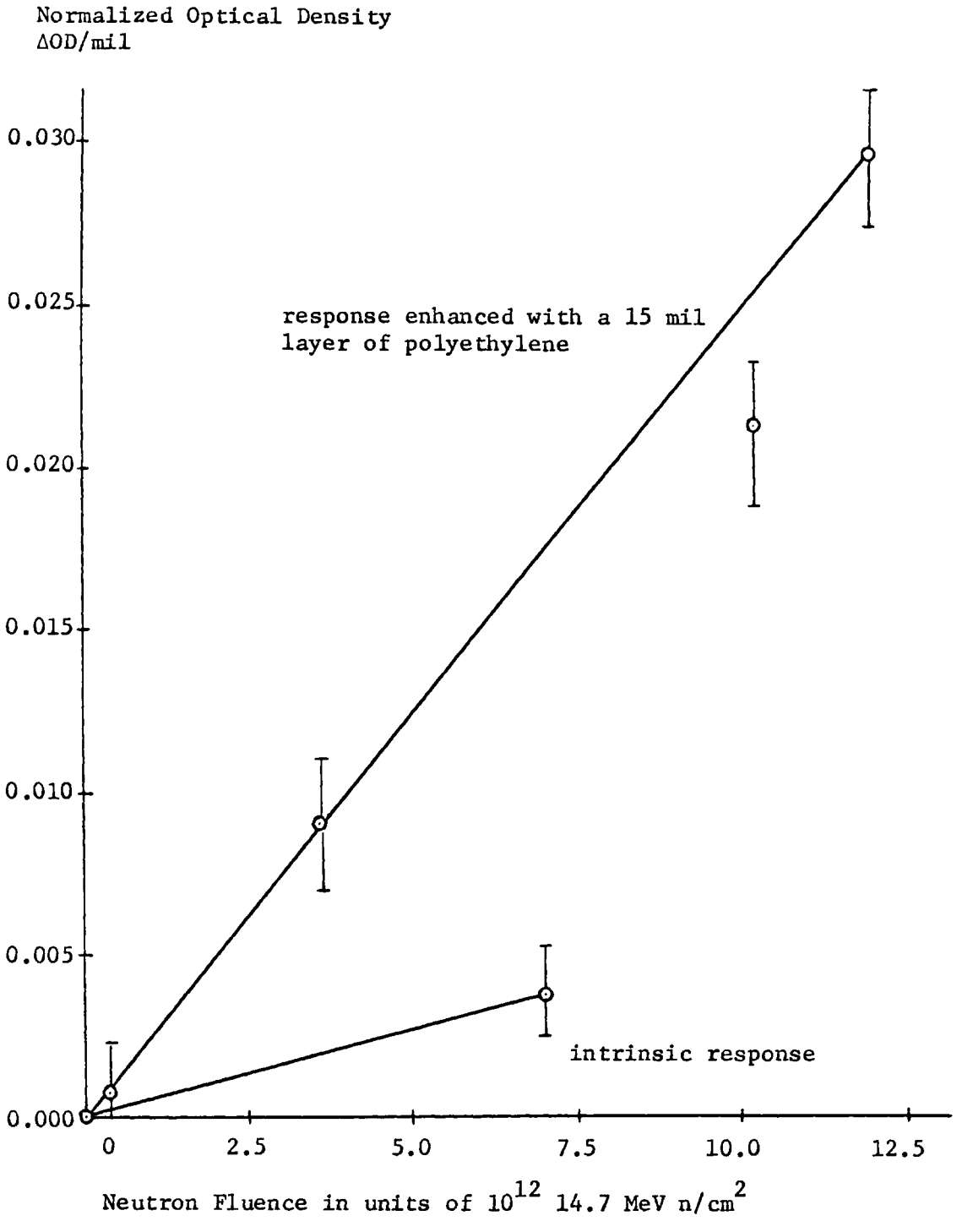



Fig. 8. Dosimeter Response to Fast Neutrons

shown in Fig. 9 and exposed to an environment of gamma rays and fast and thermal neutrons. The entire dosimeter would be approximately

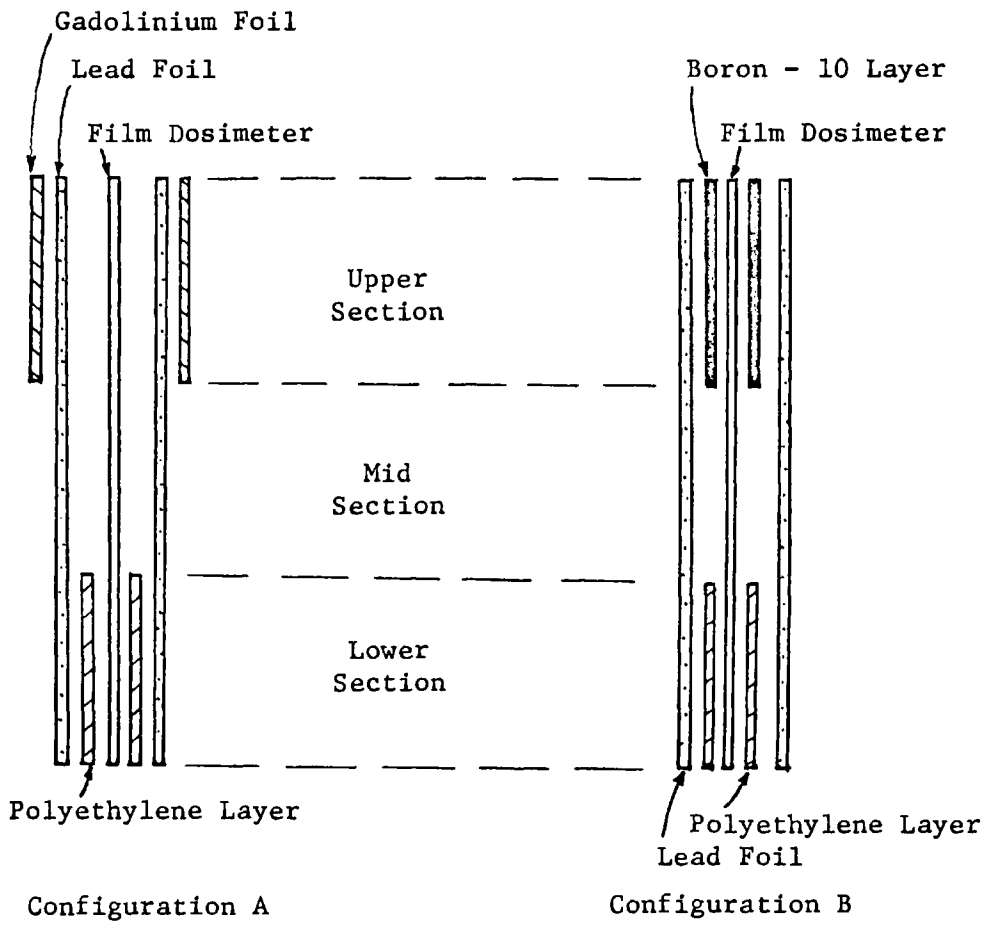



Fig. 9 Possible Configurations of Mixed Field Dosimeters.

6 cm x 1 cm. For configuration A the gamma dose would be obtained by measuring the change in optical density of the middle section of the dosimeter. The lead foil would enhance the response (high atomic number effect) of this section as well as acting to shield the dosimeter from light and charged particles. The intrinsic fast neutron response is so much less than the gamma response that it would be masked by it. The thermal neutron response would be nil.

The upper-most portion of the dosimeter would measure the gamma plus the thermal neutron dose. That is the intrinsic gamma response would be coupled with Gadolinium's $(n,\gamma)$ thermal cross section to yield the gamma and thermal neutron response. In this section the fast neutron contribution would be nil. Note that by placing the gadolinium outside the lead the thermal neutron response is enhanced somewhat more since the high atomic number of lead will promote the photoelectric effect on the gadolinium produced gamma rays causing more energy to be deposited in the film.

In the lowest section of the dosimeter the gamma plus fast neutron dose would be obtained. The intrinsic gamma response of the film would be complemented by a fast neutron contribution due to the polyethylene layer. Thermal neutrons would have no effect in this section. The average fast neutron energy would need to be known to unambiguously interpret their dose contribution.

For configuration B the same information would be obtained; however, the method of obtaining the gamma plus thermal neutron dose would be different. Instead of gadolinium, the boron-10 technique of

producing a thermal neutron response would be substituted. The lead in this instance must be outside the film so that it does not eliminate the alpha particles produced in the boron-10 layer from reaching the dosimeter to deposit their energy therein.

# CHAPTER 5

## SUMMARY AND CONCLUSION

The use of leuco malachite green carbinol dye with a poly (4-chlorostyrene) base as a dosimeter would seem to require some *a priori* knowledge of the dose rate before meaningful results are obtained unless used in vacuo. This results from the dose rate dependence of the film in air. In applications where the dose rate varies very slowly with time (Co-60 irradiation) or can be controlled (reactor or accelerator environments) then useful dosimetric correlations can be established. This dosimeter would have a threshold (for 4 mil thick films) on the order of 10 Kr for gamma radiation and integrated fluences of $10^{12}$ n/cm$^2$ for neutron responses.

For convenience, the results of the sensitivities of leuco malachite green carbinol dye in a poly (4-chlorostyrene) base are summarized in Tables I and II. All sensitivities are with reference to the peak at 6250 Å.

TABLE I

GAMMA RAY SENSITIVITY

| | |
|---|---|
| at 70 r/min | $1.44 \times 10^{-6}$ ΔOD/mil/r |
| at 660 r/min | $7.65 \times 10^{-7}$ ΔOD/mil/r |
| at 300 r/sec in vacuum (Dr. Harrah's work) | $3.87 \times 10^{-7}$ ΔOD/mil/r |
| with uranium foils over the film (at 660 r/min) | $1.83 \times 10^{-6}$ ΔOD/mil/r |

TABLE II

NEUTRON SENSITIVITY

| | |
|---|---|
| thermal neutron | none(up to $2.7 \times 10^{12}$ $n/cm^2$) |
| thermal neutron sensitivity using boron-10 tapes | $8.39 \times 10^{-15}$ ΔOD/mil/$(n/cm^2)$ |
| 14.7 MeV neutrons | $5.5 \times 10^{-16}$ ΔOD/mil/$(n/cm^2)$ |
| 14.7 MeV neutrons with 15 mils of polyethylene surrounding the film | $2.5 \times 10^{-15}$ ΔOD/mil/$(n/cm^2)$ |

Several qualitative techniques have been demonstrated for improving the intrinsic sensitivity of the film. For gamma radiation high Z materials surrounding the film enhance the response by increasing the photoelectric contribution without attenuating the initial gamma intensity very markedly. For thermal neutrons, boron-10 sandwiched around the film forms the basis of a technique to get a thermal neutron response by utilizing the large $(n,\alpha)$ cross section of boron-10 to provide the $\alpha$ particle field which can deposit energy within the film. A technique to improve the response for fast neutrons consists of surrounding the film with a hydrogeneous layer. Neutron interactions with hydrogen atoms will create recoil protons which in turn can deposit a portion of their energy within the film contributing to the film response.

Using the above mentioned techniques, a compact (1 x 6 cm) mixed field dosimeter has been proposed. This dosimeter would separate the total dose into its components: gamma dose, fast and thermal neutron dose.

## APPENDIX A

## USE OF THE 6900 Å REFERENCE IN THE OPTICAL DENSITY READINGS

A characteristic of the film is that it becomes slightly embrittled upon irradiation. For this reason, unless one is careful in handling these films there is the possibility that the films will be damaged particularly if the films are thin (< 3 mils). This bruising occurs when the film is bent or stressed in such a manner that internal cracks or tears are formed. These internal tears are particularly bothersome since light from the spectrophotometer is reflected from them resulting in erroneous optical density readings. To alleviate this a reference OD at 6900 Å is established. Consider the spectra shown in Fig. 10.

i represents the initial unexposed film OD trace. A typical value for it is drawn in.

f is an ideal irradiated film trace (no bruising).

$f^*$ is an irradiated film trace showing the effects of bruising. It was made by just bending the film that gave the previous (f) trace but not exposing it to any more radiation.

Now $\Delta OD = OD_{6250f} - OD_{6250i}$ (12)

Adding and subtracting $OD_{6900i}$ yields

$$\Delta OD = OD_{6250f} - OD_{6900i} - OD_{6250i} + OD_{6900i} \quad (13a)$$

$$= (OD_{6250f} - OD_{6900i}) - (OD_{6250i} - OD_{6900i}) \quad (13b)$$

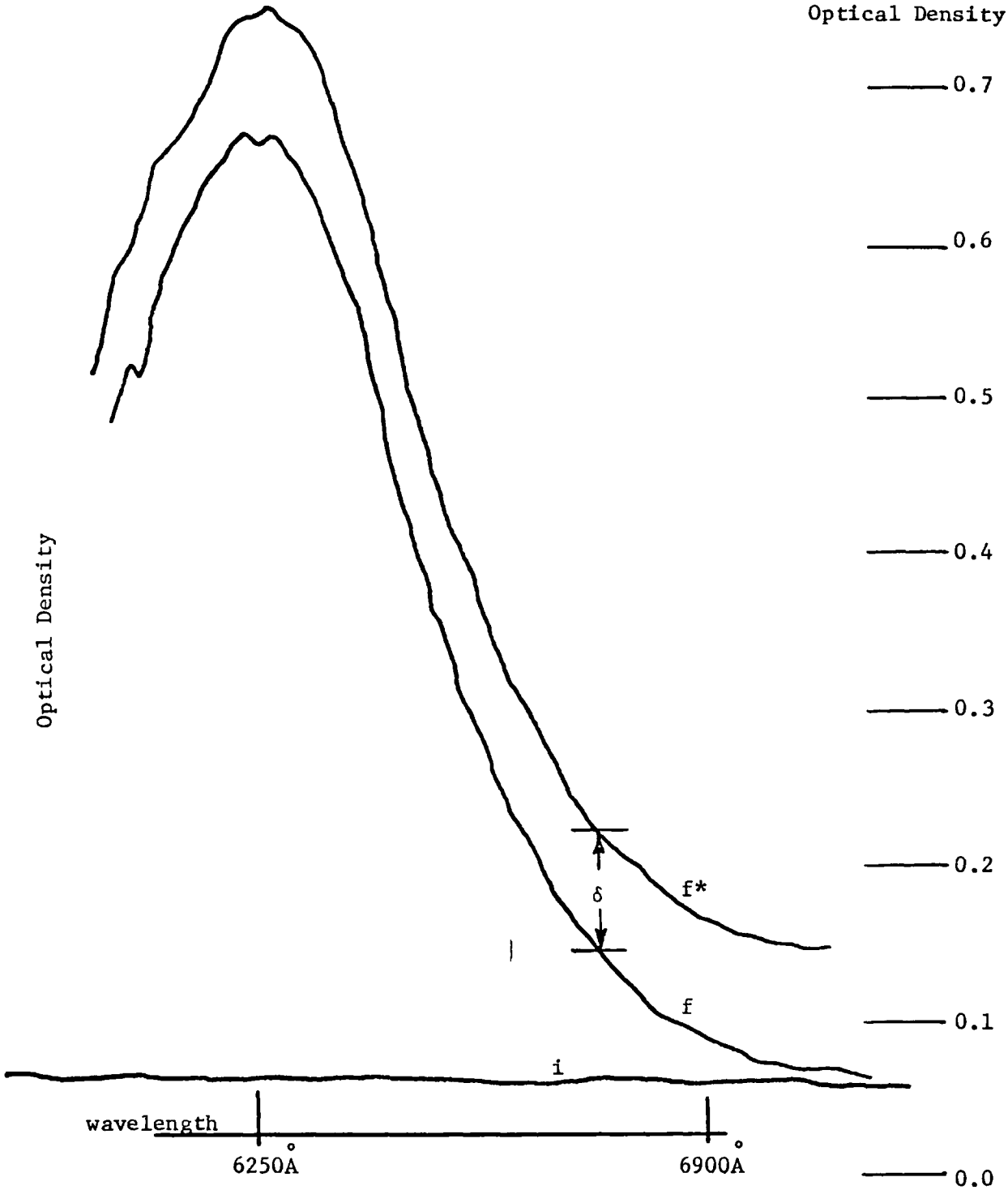



Fig. 10. The Effect of Bruising on the Optical Density.

With the level of exposures made, $OD_{6900f} \simeq OD_{6900i}$ so that

$$\Delta OD \simeq (OD_{6250f} - OD_{6900f}) - (OD_{6250i} - OD_{6900i}) \tag{14}$$

From experiments and also in the trace, $\delta$ the difference in optical density due to the bruising, is a constant in the wavelengths under consideration. Then $OD_{6250f} - OD_{6900f} = OD_{6250f*} - OD_{6900f*}$ where the optical densities actually obtained from the spectrophotometer are the starred quantities. Hence

$$\Delta OD \simeq (OD_{6250f*} - OD_{6900f*}) - (OD_{6250i} - OD_{6900i}) \tag{15}$$

and using the notation

$$\Delta_f = OD_{6250f*} - OD_{6900f*}$$

$$\Delta_i = OD_{6250i} - OD_{6900i}$$

then $\Delta OD = \Delta_f - \Delta_i$. (16)

$\Delta_f$ and $\Delta_i$ are the quantities used in the presentation of the data. The data has a much better correlation with the expression just derived than $\Delta OD = OD_{6250f} - OD_{6250i}$ because internal surface reflections are not taken into account in it.

## APPENDIX B

### GAMMA EXPOSURE DOSE DATA AT 70 r/min (3.9 MIL THICK FILMS)

In presenting the data in Appendices B through F the following notation is used:

$$\Delta_i = \text{OD } 6250\ \mathring{A}_i - \text{OD } 6900\ \mathring{A}_i$$

$$\Delta_f = \text{OD } 6250\ \mathring{A}_f - \text{OD } 6900\ \mathring{A}_f$$

$$\Delta = \Delta_f - \Delta_i$$

| Exposure Dose | Optical Densities at | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| | $6250_i$ | $6900_i$ | $\Delta_i$ | $6250_f$ | $6900_f$ | $\Delta_f$ | $\Delta$ | $\Delta$OD/mil |
| 20Kr | .070 | .059 | .011 | .182 | .062 | .120 | .109 | .0280 |
| | .072 | .061 | .011 | .205 | .072 | .133 | .122 | .0313 |
| 40Kr | .072 | .061 | .011 | .330 | .076 | .254 | .243 | .0624 |
| | .075 | .062 | .013 | .332 | .075 | .257 | .244 | .0626 |
| 60Kr | .071 | .061 | .010 | .443 | .085 | .358 | .348 | .0893 |
| | .073 | .061 | .012 | .424 | .075 | .349 | .337 | .0865 |
| 80Kr | .073 | .061 | .012 | .516 | .075 | .441 | .429 | .1110 |
| | .073 | .061 | .012 | .545 | .085 | .460 | .448 | .1150 |
| 100Kr | .094 | .080 | .014 | .639 | .100 | .535 | .521 | .1338 |
| | .074 | .063 | .011 | .629 | .080 | .549 | .538 | .1380 |

APPENDIX C

GAMMA EXPOSURE DOSE DATA AT 660 r/min (4.1 MIL THICK FILMS)

| Exposure Dose | Optical Densities at | | | | | | | ΔOD/mil |
|---|---|---|---|---|---|---|---|---|
| | $6250_i$ | $6900_i$ | $\Delta_i$ | $6250_f$ | $6900_f$ | $\Delta_f$ | $\Delta$ | |
| 20Kr | .067 | .061 | .006 | .137 | .072 | .065 | .059 | .0144 |
| | .068 | .060 | .008 | .148 | .078 | .070 | .062 | .01512 |
| 40Kr | .065 | .059 | .006 | .219 | .074 | .145 | .139 | .0339 |
| | .072 | .068 | .004 | .202 | .070 | .132 | .128 | .0313 |
| 60Kr | .076 | .068 | .008 | .274 | .076 | .198 | .190 | .0464 |
| | .088 | .078 | .010 | .281 | .082 | .199 | .189 | .0461 |
| 80Kr | .068 | .060 | .008 | .346 | .075 | .271 | .263 | .0641 |
| | .070 | .065 | .005 | .322 | .080 | .242 | .237 | .0579 |
| 100Kr | .068 | .060 | .008 | .421 | .082 | .339 | .331 | .0809 |
| | .090 | .084 | .006 | .420 | .105 | .315 | .309 | .0754 |

## APPENDIX D

## GAMMA EXPOSURE DOSE RESPONSE ENHANCED WITH URANIUM FOILS

Uranium Thickness is 2.45 mils

Dose Rate is 660 r/min

Film Thickness is 4.1 mil

| Exposure Dose | Optical Densities at | | | | | | | ΔOD/mil |
|---|---|---|---|---|---|---|---|---|
| | $6250_i$ | $6900_i$ | $\Delta_i$ | $6250_f$ | $6900_f$ | $\Delta_f$ | $\Delta$ | |
| 20Kr | .067 | .061 | .006 | .219 | .070 | .149 | .143 | .0349 |
| | .068 | .060 | .008 | .238 | .078 | .160 | .152 | .0371 |
| 40Kr | .065 | .059 | .006 | .400 | .070 | .330 | .326 | .0795 |
| | .072 | .068 | .004 | .370 | .070 | .300 | .296 | .0722 |
| 60Kr | .076 | .068 | .008 | .546 | .099 | .447 | .439 | .1070 |
| | .088 | .078 | .010 | .556 | .091 | .456 | .446 | .1089 |
| 80Kr | .068 | .060 | .008 | .674 | .094 | .580 | .572 | .1394 |
| | .070 | .065 | .005 | .632 | .092 | .540 | .535 | .1304 |
| 100Kr | .068 | .060 | .008 | .910 | .109 | .801 | .793 | .1931 |
| | .090 | .084 | .006 | .849 | .120 | .729 | .723 | .1761 |

## APPENDIX E

### 14.7 MeV NEUTRON EXPOSURES (3.5 MIL THICK FILMS)

With 15 Mils of Polyethylene Surrounding Each Film

| Total Neutron Fluences in $n/cm^2$ | Optical Densities at | | | | | | | ΔOD/mil |
|---|---|---|---|---|---|---|---|---|
| | $6250_i$ | $6900_i$ | $\Delta_i$ | $6250_f$ | $6900_f$ | $\Delta_f$ | $\Delta$ | |
| $.387 \times 10^{12}$ | .071 | .060 | .011 | .106 | .092 | .014 | .003 | .000856 |
| $3.57 \times 10^{12}$ | .068 | .055 | .013 | .147 | .108 | .039 | .026 | .00744 |
| | .072 | .055 | .017 | .175 | .123 | .052 | .035 | .00999 |
| $10.28 \times 10^{12}$ | .147 | .108 | .039 | .177 | .070 | .102 | .063 | .01798 |
| | .175 | .123 | .052 | .182 | .069 | .113 | .061 | .01741 |
| $11.89 \times 10^{12}$ | .106 | .092 | .014 | .169 | .084 | .087 | .071 | .02025 |

Intrinsic Response (i.e. Without Polyethylene)

| | | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| $7.00 \times 10^{12}$ | .070 | .050 | .020 | .101 | .068 | .033 | .013 | .00323 |
| | .070 | .050 | .020 | .116 | .082 | .034 | .014 | .00325 |

## APPENDIX F

## THERMAL NEUTRON RESPONSE WITH THE BORON-10 TAPES

Concentration of $^{10}B$ is 1.92 $mg/cm^2$

Films are 3.9 Mils Thick

Total Thermal Neutron Fluence is $2.72 \times 10^{12}$ $n/cm^2$

| | Optical Densities at | | | | | | | ΔOD/mil |
|---|---|---|---|---|---|---|---|---|
| | $6250_i$ | $6900_i$ | $\Delta_i$ | $6250_f$ | $6900_f$ | $\Delta_f$ | $\Delta$ | |
| Boron Tape Films | .084 | .076 | .008 | .204 | .110 | .094 | .086 | .0221 |
| | .072 | .064 | .008 | .174 | .090 | .084 | .076 | .0195 |
| Reference Film | .082 | .073 | .009 | .093 | .080 | .013 | .004 | .001025 |
| | .078 | .068 | .010 | .090 | .070 | .020 | .010* | .00256 |

*This film was scratched during handling and Δ is higher than it should be.

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