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TACTICAL GAMMA AND FAST NEUTRON DOSIMETRY WITH LEUKO DYE OPTICA--ETC(U)  
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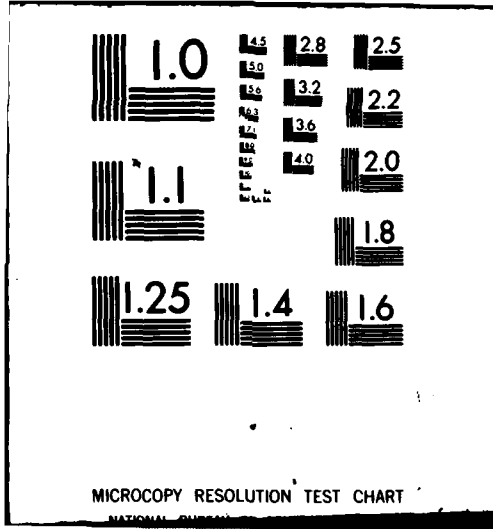
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TACTICAL GAMMA AND FAST NEUTRON DOSIMETRY  
WITH LEUKO DYE OPTICAL WAVEGUIDES (U)

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INTRODUCTION

The change of color induced by nuclear radiation in radiochromic leuko dyes can be used to measure radiation doses (1,2). To significantly increase the sensitivity of such dosimeters, the layer of the dye-containing solution must be made very thick. To solve this problem in a practical manner we constructed optical waveguides (fiber optics) whose core is a solution of radiochromic leuko dye and the outside layer a thin teflon or FEP tubing. Proper choice of solvents with a refractive index greater than the refractive index of the wall results in total reflection of the light traveling through the core, thus satisfying the waveguide condition. This approach is analogous to our earlier approach to increase resistivity of glass dosimeters by using fiber optics, (3).

RADIATION EFFECTS ON THE INDEX OF REFRACTION  
OF THE DYE SOLUTION

While experimenting with the radiochromic waveguide dosimeters we observed an effect which makes the leuko dye dosimetry in waveguides operationally different from leuko dye dosimetry with bulk solution and which can be exploited for construction of dosimeters with a very wide dynamic range.

For example, an irradiated solution of hexahydroxyethyl pararosaniline cyanide appears blue in a test tube but red after being incorporated in a waveguide and observed through its end. This effect can be understood from the theory of anomalous dispersion (4) illustrated qualitatively in Fig. 1. Exposure of the dye solution to radiation induces an absorption band. This absorption band spans the region between the red and the blue, and results in an increase in the value of the refractive index (n) of the solution at the longer wavelengths and a decrease in n at shorter wavelengths. At shorter wavelengths, where n of the irradiated core solution drops below the value of n of the cladding, the waveguide condition no longer applies

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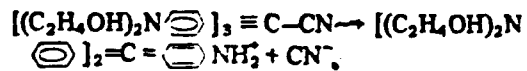
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and the device ceases to be a propagator of light through its core. In that wavelength region the light is not channeled through the waveguide, but either escapes axially or is absorbed by the cladding.

The result is that irradiation of the waveguide makes it virtually opaque for a certain optical frequency band whose width increases with increasing dose. The observed resulting dynamic range of such dosimeters covers at least six orders of magnitude.

### MATERIALS AND CONSTRUCTION OF DOSIMETERS

We have chosen hexahydroxyethyl pararosanile cyanide (molecular weight 578.7) from among commercially available triphenylmethane radiochromic leuko dyes.



This leuko dye changes to deep blue by exposure to ionizing radiation. It is also sensitive to ultraviolet light and must be shielded from it. In slightly acidic polar solvents containing oxygen there is no back reaction, thus ensuring stability (no fading) of the radiation-induced color change.

Polar solvents which can be used are dimethyl sulfoxide ((CH<sub>3</sub>)<sub>2</sub>SO), triethyl phosphate ((C<sub>2</sub>H<sub>5</sub>)<sub>3</sub>PO<sub>4</sub>) or N,N-dimethyl formamide (HCON(CH<sub>3</sub>)<sub>2</sub>). The later two solvents are liquid between -56.4 C and +153 C; the indices of refraction are 1.48, 1.41 and 1.43, respectively. A 5-20% solution of the dye is drawn into a tube of "FEP", a teflon-like flexible thermoplastic whose refractive index is 1.34. We used tubing with 3.0 mm outer diameter and 2.3 mm inner diameter.

Two approaches were used to terminate the ends of such waveguides. One approach by means of commercially available glass beads is shown in Fig. 2. The beads provide a hermetic seal as well as a lens effect for injecting and extracting light from the waveguide. The second approach uses no explicit termination. Mixing the dissolved dye with vinyl pyrrolidone (with the inhibitor removed) and letting the mixture polymerize results in a stable jelly-like substance which is sufficiently solid to prevent leakage through the ends of the waveguide. This very convenient approach makes it possible to fill a long piece of FEP tubing with the liquid mixture and, after partial polymerization takes place, to cut it up in pieces of desired length.

### READING THE DOSE

Dosimetry is accomplished by comparing ratios of transmissivity of the waveguide at two different wavelengths. The same calibration applies for all dosimeters having the same dimensional and chemical parameters.



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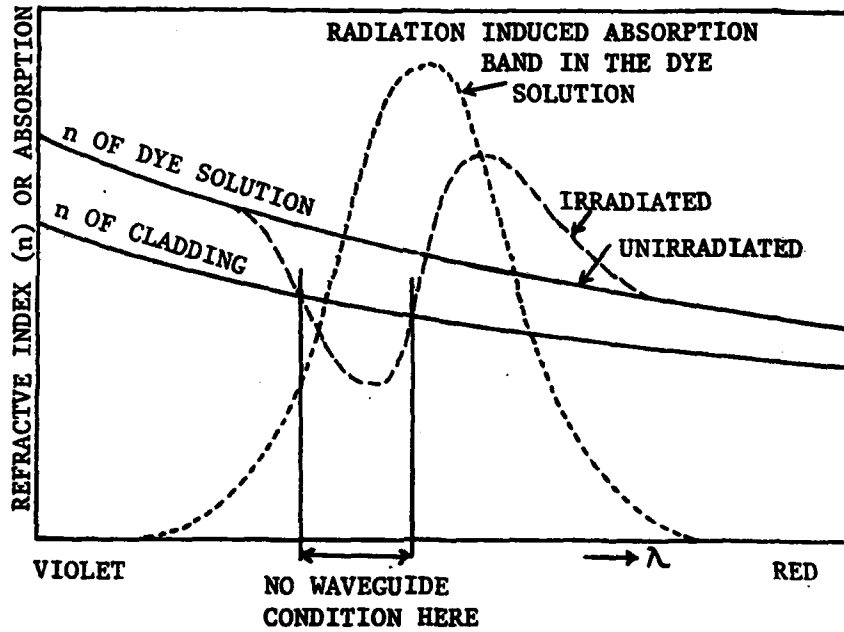


Figure 1. Illustration of radiation-induced anomalous dispersion and its effect on the optical waveguide.

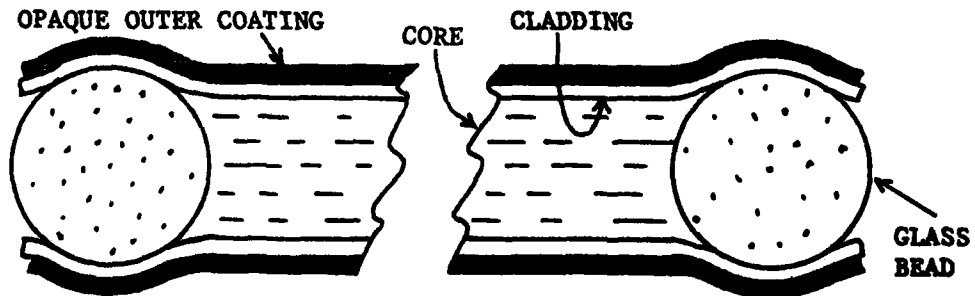


Figure 2. Construction of an optical waveguide whose core is a solution of radiochromic dye.

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Figure 3 illustrates a laboratory dosimeter reader. Because the critical measurement is a ratio of optical transmittance values, reproducibility of the waveguide mounting and stability of the instrumentation are not essential. The two parameters which are required to be fixed are the spectral distribution of the light source and the response characteristics of the light detector.

A different approach to dose reading is the use of different spectral bands instead of monochromatic light sources. Figure 4 shows the schematics of such a dosimeter reader which fits in a box 10 x 15 x 5 cm. The spectral output of the three light-emitting diodes used in this device is shown in Fig. 5. To read the dose to which the 11 cm long waveguide was exposed, place it between one of the three LED's and the photo diode and then compare the digital readouts with the light on and the light off. Unfolding the difference between the two readings on a calibration curve yields the dose. The choice of which one of the three LED's should be used depends on the dose to which the dosimeter was exposed.

### GAMMA RAY DOSE MEASUREMENTS AND GAMMA ENERGY DEPENDENCE

The absorbed dose is measured most accurately by taking the ratios of transmitted intensities before and after exposure at two different wavelengths, calculating the ratio of these ratios and reading the dose from calibration curves or tables. As an example, Fig. 6 shows such a calibration curve for a 14.5 cm long waveguide containing hexahydroxyethyl parosaniline cyanide in N,N-dimethylformamide (concentration by weight: 0.09651). Electron equilibrium was obtained by surrounding the sample during the irradiation with tissue-equivalent material so that 1 roentgen exposure corresponds to an absorbed dose of 1.54 Rad (tissue). The photocathode was 110 (S-20) and the light source of the scanning monochromator was a quartz halogen lamp.

The dose received by any dosimeter of the same construction can be read by means of this calibration over a dose range of six orders of magnitude by selecting appropriate wavelengths for analysis. All data shown in Fig. 6 was obtained from a single waveguide by measuring its transmittance after consecutive exposures to gamma rays. The effect of bleaching by radiation, which is visible in Figure 6 above 2 megaroentgen, exists only in liquid dye solutions; it does not occur in solid solutions.

The quantum energy response to gamma rays of these dosimeters is determined mainly by the composition of the solvent and of the cladding. For example, if the dosimeter is kept in a container made of tissue-equivalent material during exposure to gamma radiation under conditions approximating electron equilibrium, the response will be close to that of tissue over a broad range of photon energies.

To obtain rad readings in materials other than tissue it is practicable to expose the solution not in a waveguide but in a container made of the material in question. The solution should be placed in a layer sufficiently thin to assure the applicable electron equilibrium according to

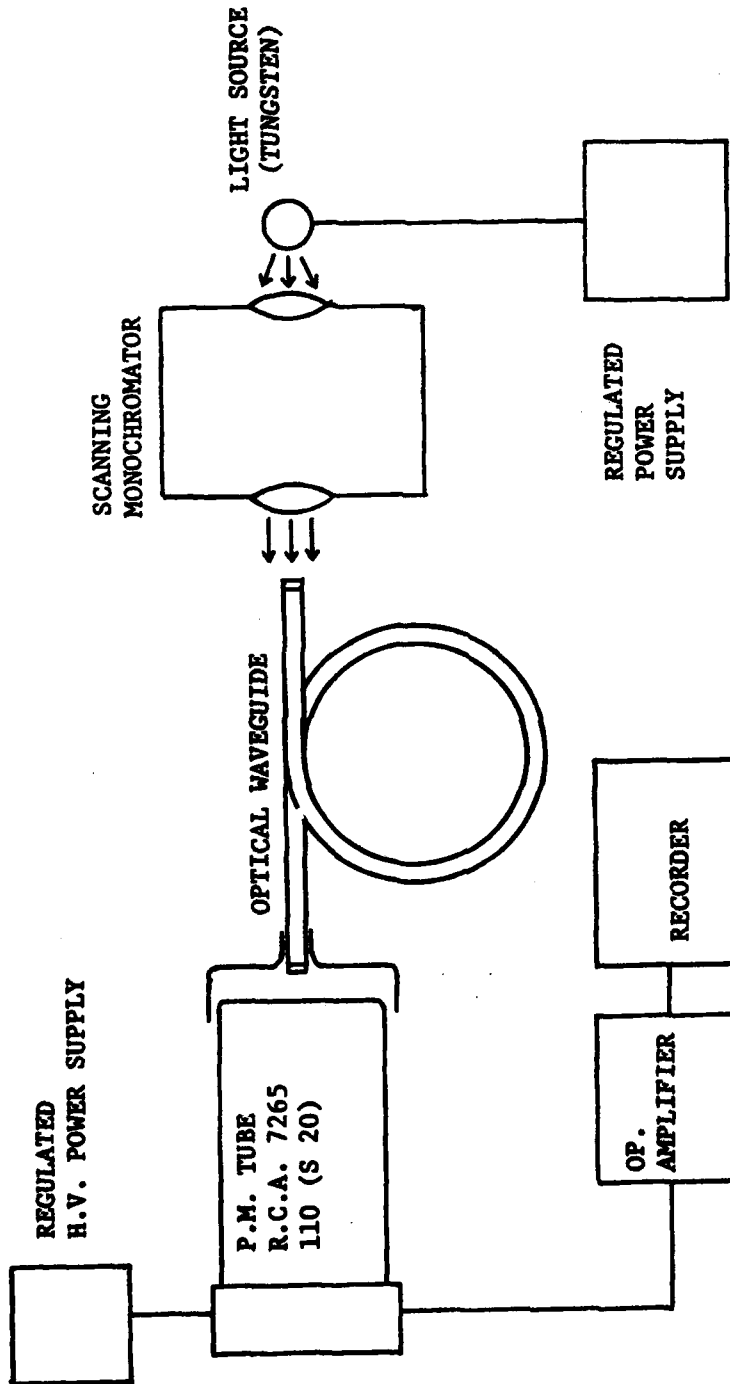


Figure 3. Apparatus for recording relative transparency of waveguides versus wavelength for visible and near infrared light.

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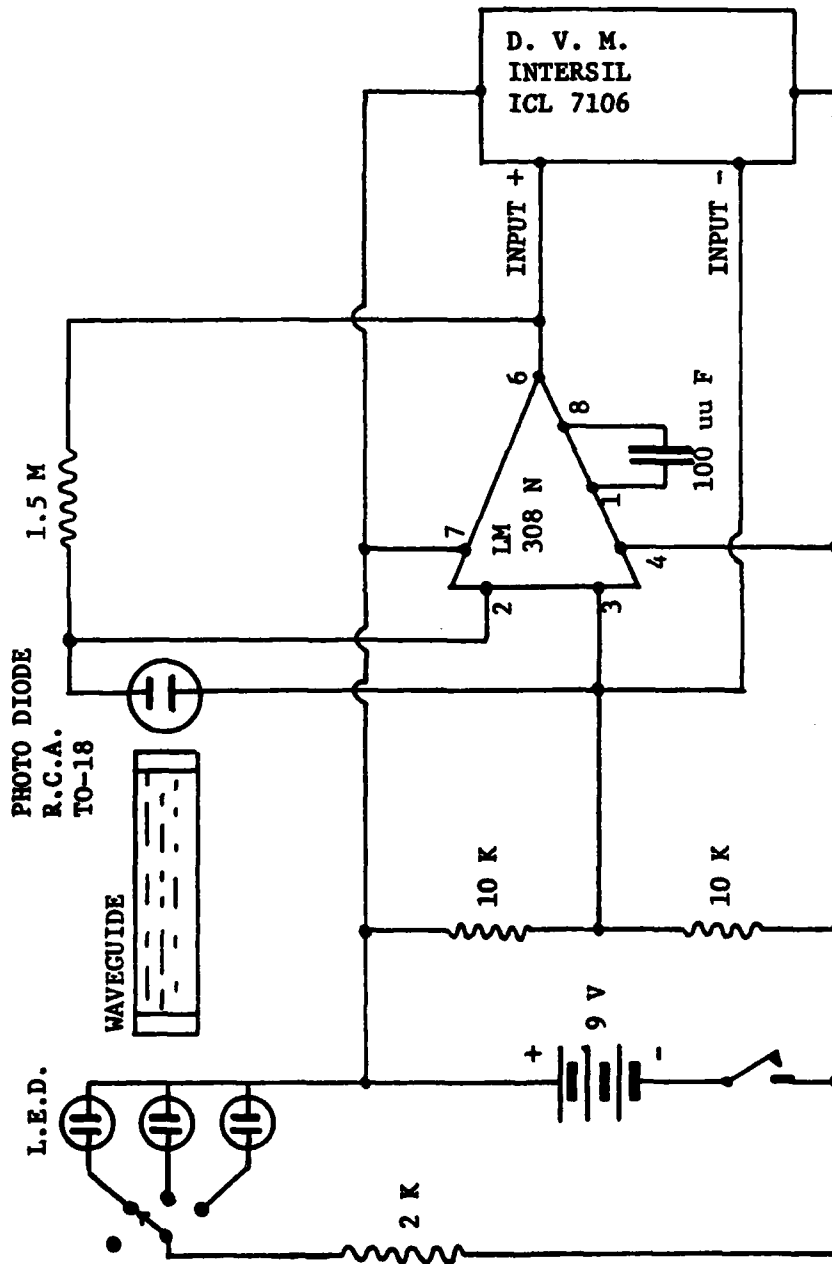


Figure 4. Waveguide dosimeter reader.

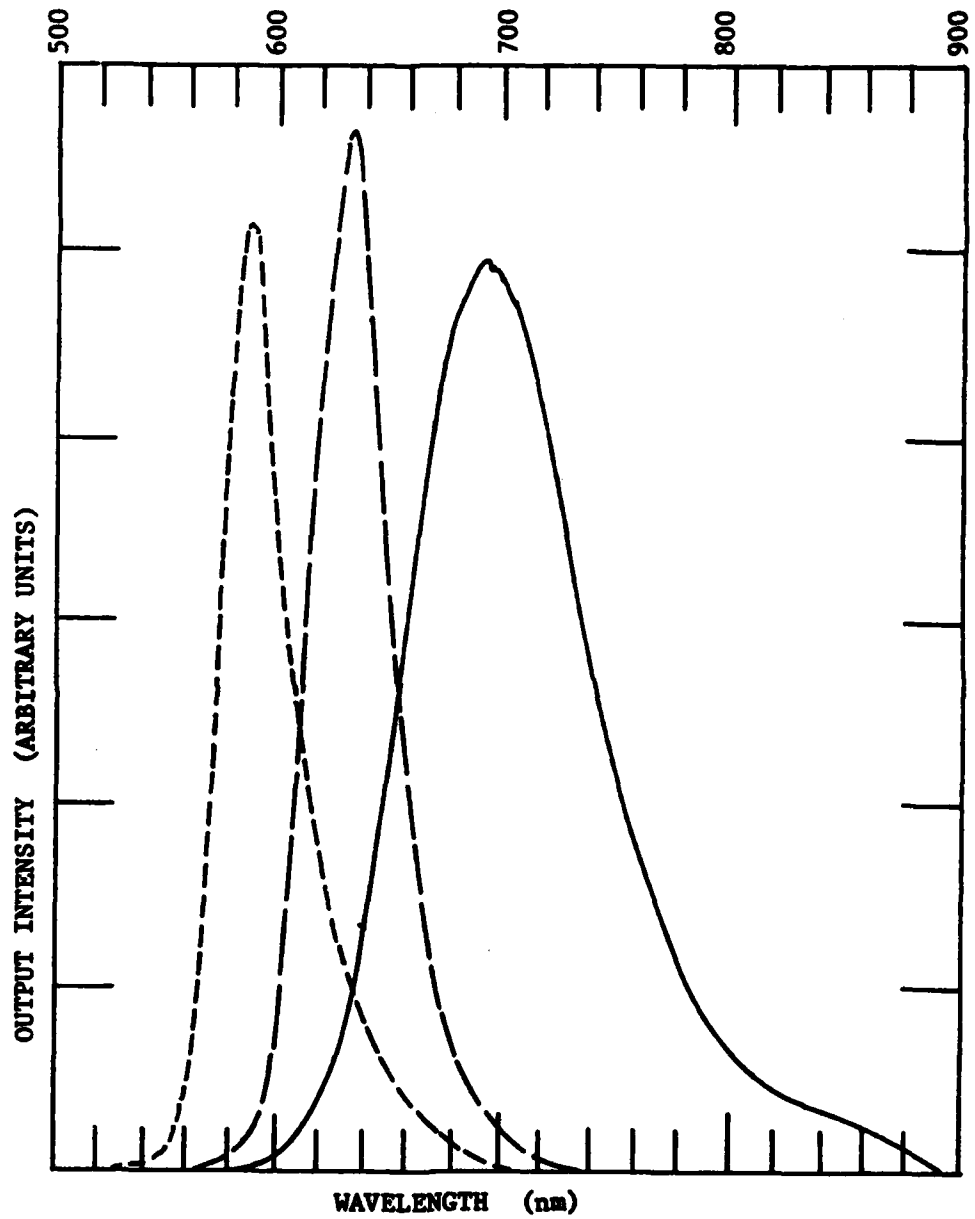


Figure 5. Spectra of the outputs of the three LEDs used in the dosimeter reader illustrated in figure 4.

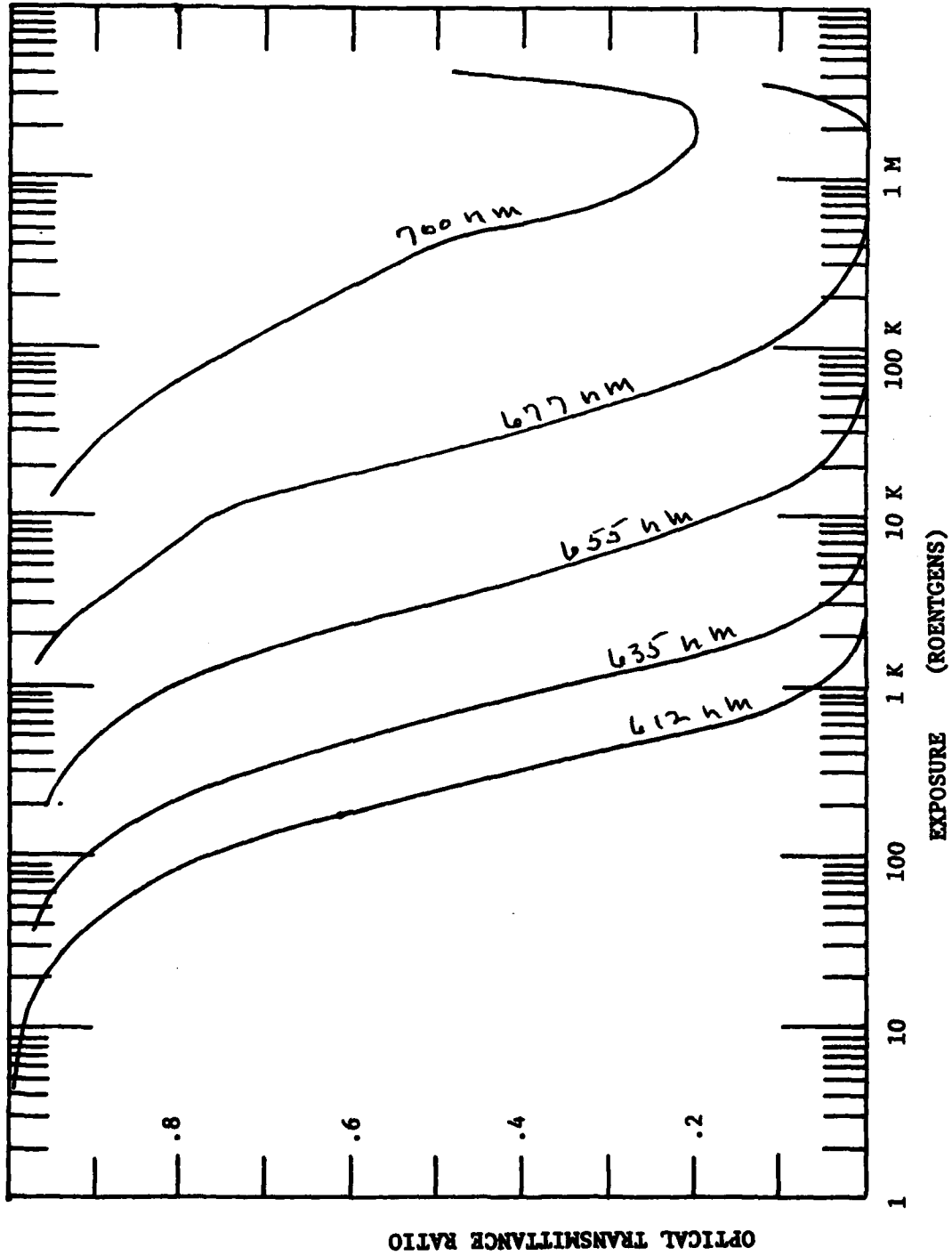


Figure 6. Optical transmittance ratio of irradiated to unirradiated waveguide vs exposure at several different wavelengths.

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the Brag-Gray principle. After exposure the solution should be filled into a FEP tubing to form a waveguide and to be read.

### FAST NEUTRON DOSE MEASUREMENTS AND NEUTRON ENERGY DEPENDENCE

For readout wavelengths below 620 nm, fast neutrons and gammas produce virtually the same effects in radiochromic waveguide dosimeters. Above  $\lambda = 620$  nm, fast neutrons produce an additional effect illustrated in Fig. 7. This effect may be exploitable for differentiation between gamma and neutron doses and thus may lead to a future mixed radiation, rem dosimetry. It is currently being investigated and is not a subject of this paper whose scope for mixed dose measurements is limited to readouts at wavelengths below 620 nm.

The response of these dosimeters to fast neutrons is defined mainly by the abundance of fast neutron-generated ionizing recoil particles, particularly the abundance of recoil protons, in the solvent. As the teflon-like cladding contains negligible recoil-producing hydrogen, it contributes little to the neutron dose reading.

So far it has not been possible to find a suitable cladding material which contains hydrogen. Therefore, all currently available waveguide dosimeters have a variable fast neutron response which depends on the fast neutron spectrum and on the diameter of the waveguide; a waveguide with an inside diameter much larger than the range of an average energy recoil proton being more sensitive to fast neutrons than one with a small inside diameter. The proton contribution to the dose was calculated theoretically. Figure 8 shows the specific response of a waveguide dosimeter to fast neutrons of the energy  $E_n$  versus the waveguide diameter. This curve applies to an isotropic distribution of fast neutrons. The diameter of the waveguide core ( $\phi$ ) is expressed in range of maximum energy of recoil protons ( $E_{pr} = E_n$ ) in the waveguide core material. Figure 9 shows ranges of recoil protons (in  $\text{mg cm}^{-2}$ ) versus proton energy for interpretation of Fig. 8.

Analogous to what was said above, in the case of gamma rays the neutron response problem can be simplified by exposing the dye solution not in a waveguide but in a container whose walls are made of a hydrogenous material and by transferring it later into the waveguide to read the dose.

### UTILIZATION IN ARMY TACTICAL DOSIMETRY

The purpose of tactical dosimetry (as opposed to administrative dosimetry) is to provide the troop commander with current data on the radiological status of his troops. The very low-cost leuko dye optical waveguide dosimeter is an eligible candidate to replace the current IM 185, which is a reliable tactical dosimeter but costly and not easily maintained. The leuko dye optical waveguide dosimeter may also be used as the integrated dose component of the Army Miniature Multipurpose Radiac (MMR).

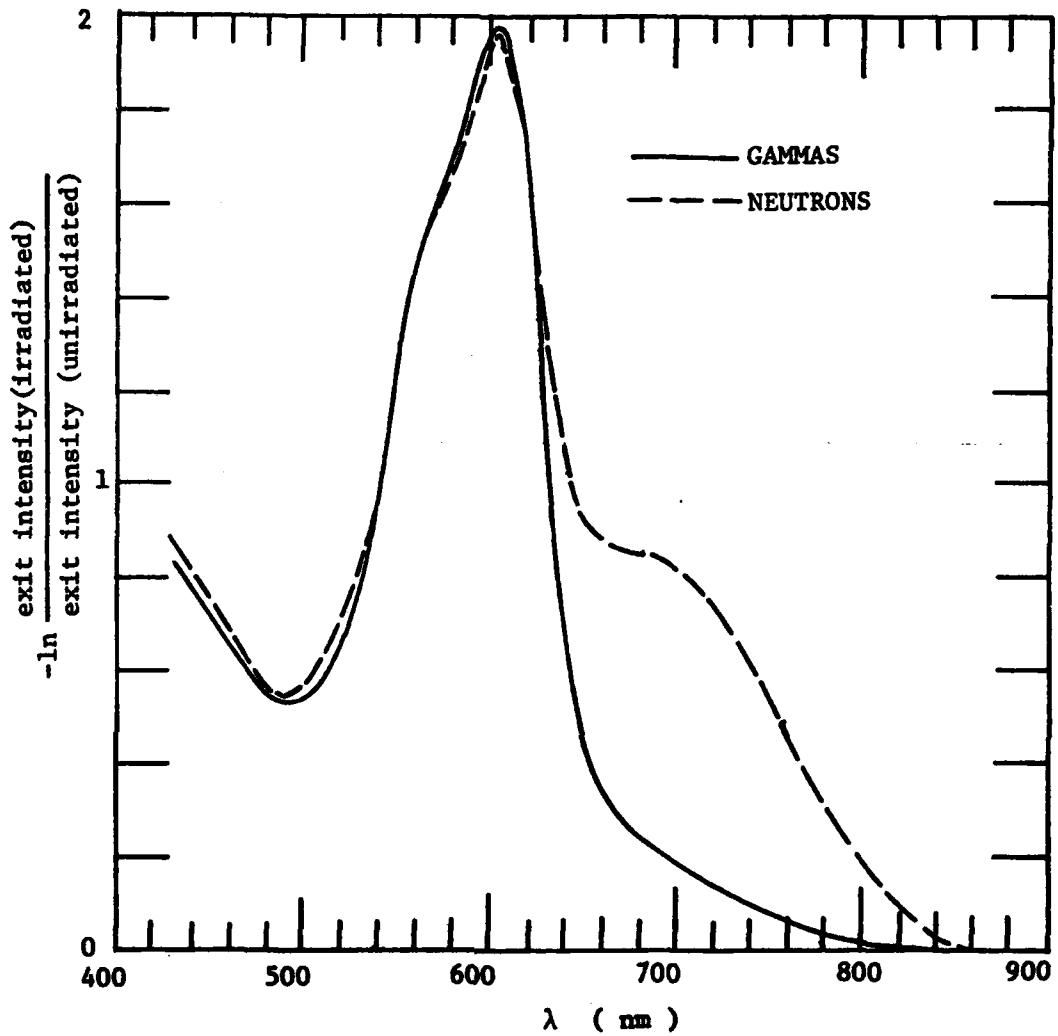


Figure 7. Negative natural logarithm of exit intensity ratio versus wavelength for identically constructed waveguides, 103 mm long, 2.3 mm inner diameter. The core solution (6.7 % dye in DMF) was exposed in two polyethylene capsules, one to 1 k rad (tissue) 14 MeV neutrons + 70 rad (tissue) associated gammas, the other capsule to 1 k rad (tissue)  $^{60}\text{Co}$  gammas.

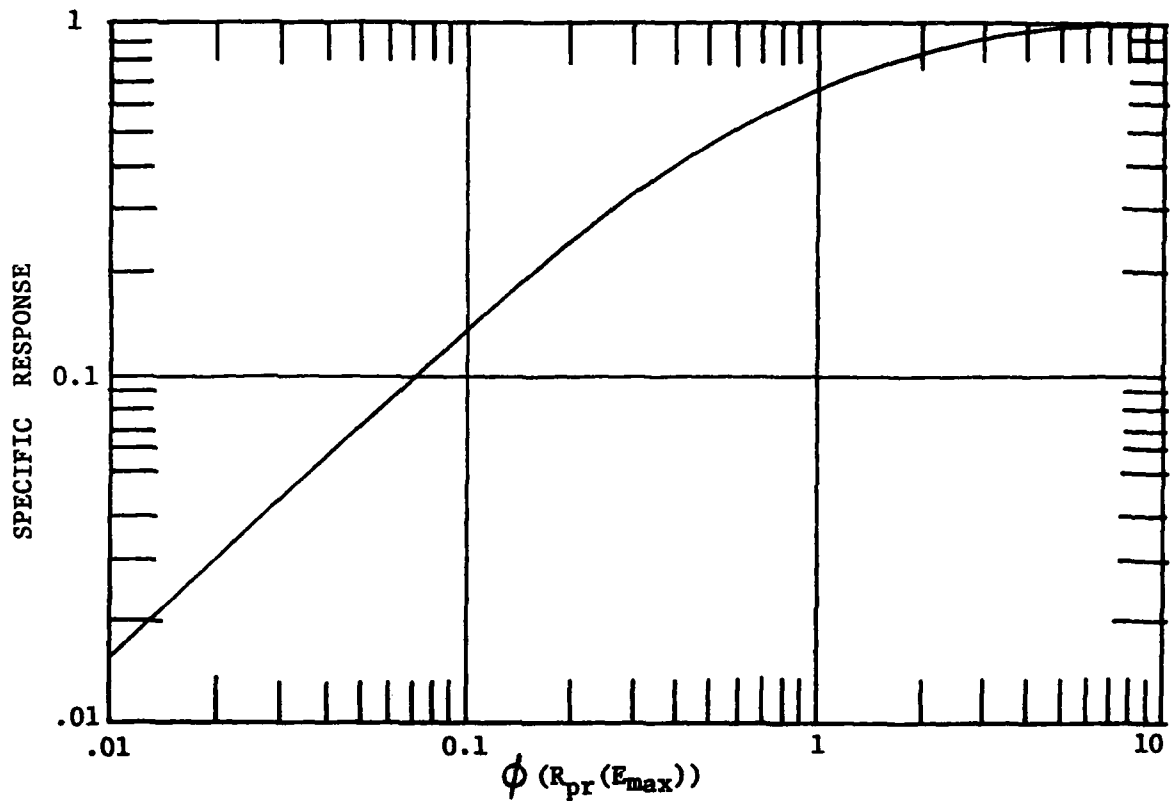


Figure 8. Calculated specific response of a waveguide dosimeter to fast neutrons of the energy  $E_n$  vs the waveguide diameter which is expressed in range of recoil protons of maximum energy ( $E_{pr} = E_n$ ) in the waveguide core material.

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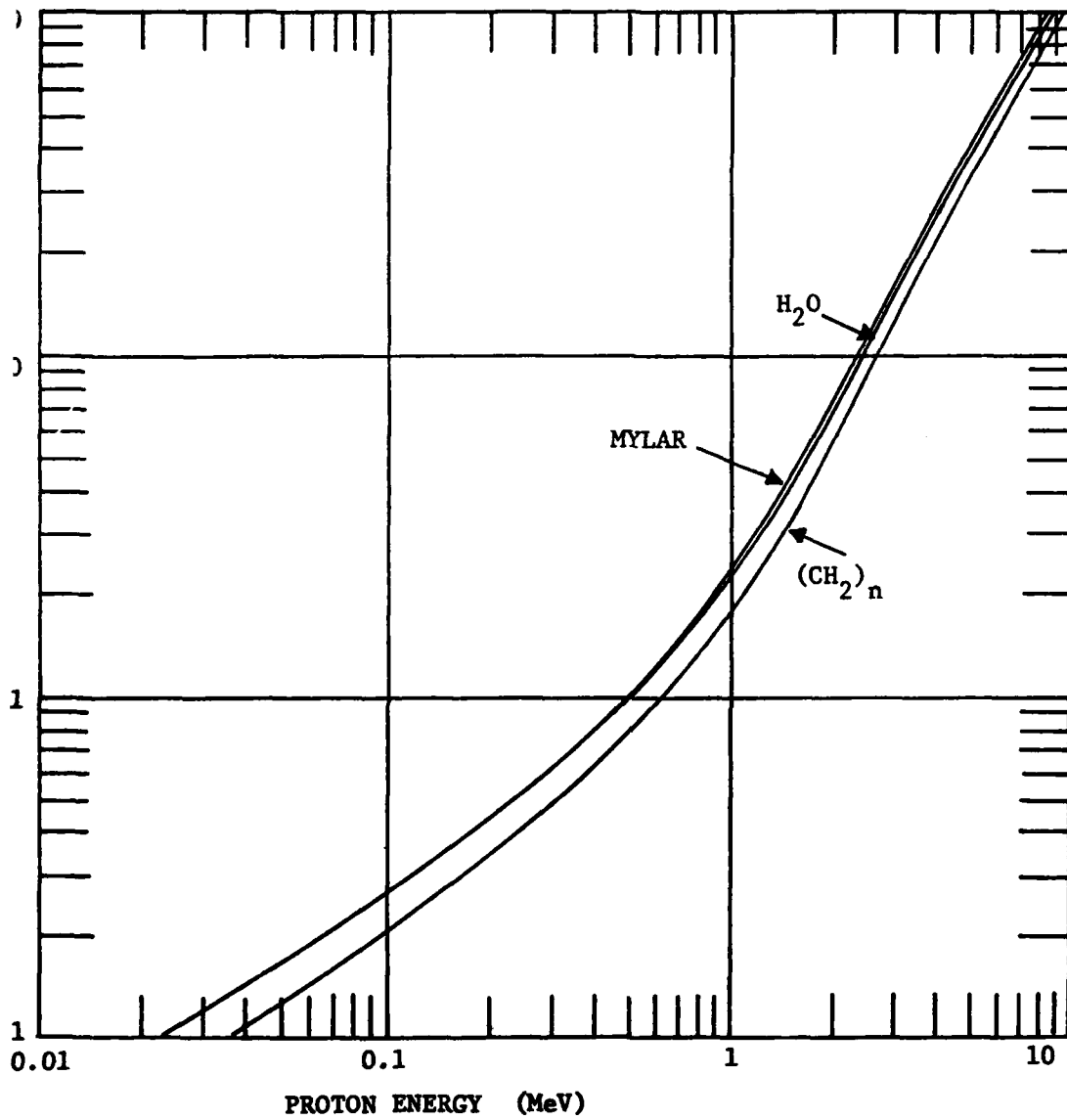


figure 9. Proton ranges versus proton energy needed for the interpretation of figure 7.

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The dosimeter does satisfy the main criteria required for a tactical dosimeter:

a. Radiochromic dyes are dose rate independent up to  $10^{12}$  rad/sec and are thus capable of responding to the prompt initial radiation from an exploding nuclear weapon as well as to the low dose rate fallout radiation (5).

b. The prompt initial radiation of a nuclear weapon consists of gammas and fast neutrons, the fallout radiation of gammas only. This dosimeter can be constructed to give mixed radiation readings as described above. The neutron radiobiological effectiveness (RBE) for Army tactical applications is defined as 1 (1 rad = 1 rem).

c. The radiation-induced changes remain fixed within required limits (e.g., lack of fading) provided that the solution is prepared correctly. A waveguide dosimeter was exposed to 1 K rad (tissue)  $^{60}\text{Co}$  gamma rays and its transmittance displayed at room temperature on the reader shown in Fig. 4 for 18 days. The observed output remained constant within 2% which was the readout accuracy of this experiment.

d. The sensitivity and the dynamic range of this system can be adjusted for the required 10 - 1000 rad (tissue) using a wavelength band which peaks at approximately 620 nm for the readout.

e. This system can be made compact by using waveguides made of available FEP tubing 0.8 mm O.D. and 0.25 mm I.D. which can be coiled in a 3 cm diameter spool.

In summary, ionizing radiation-induced changes in the refractive index of radiochromic dye solutions results in a novel dosimetry system with a very wide dynamic range. This approach is adaptable to personnel dosimetry and to Army tactical dosimetry.

### ACKNOWLEDGMENTS

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