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DYE FILM DOSIMETRY FOR RADIATION PROCESSING

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Summary

Commercially available plastic films containing dyes or dye precursors are convenient dosimeters and imaging media for electron beams or photons used for industrial radiation processing. As "grainless" imaging systems having thicknesses down to a few micrometers, they provide high spatial resolution for determining detailed absorbed dose distributions through microdensitometric analysis. The radiation absorption properties of these systems are adjusted by changing film composition so that the dosimeter materials can be made to simulate the material of interest undergoing irradiation (e.g., tissue, bone, various insulating materials, semiconductor devices, and a wide assortment of polymers). Other advantages include long-term stability, dose-rate independence, and ease of use and calibration. Radiochromic dye films with thicknesses varying from 0.005 to 1 mm are presently used to monitor electron-beam or gamma-ray doses from 10 to 10^5 Gy (10^3 to 10^7 rad), typical of those encountered in medical applications, radiation curing of polymeric composites, wire and cable insulation, shrinkable plastic tubing and film, as well as sterilization of medical supplies and treatment of municipal and industrial wastes. An NBS calibration service to industry involves the traceability of standard ^{60}Co gamma ray absorbed dose measurements by means of these films employed as transfer standards.

Applications of Radiation Dosimetry

The wide range of applications of radiation processing, either commercially successful or potentially viable, is illustrated in Figure 1. Almost all of these applications can utilize radiochromic dye film dosimetry[1] as a means of verifying that the product has received the desired absorbed dose.* For certain biological applications, such as sterilization of medical supplies, water purification, and pasteurization, a specified value of minimum absorbed dose in a given product must be achieved within required statistical confidence limits.[2] Maximum acceptable absorbed dose values are generally determined by product degradation effects such as loss of mechanical strength, discoloration, etc. The energies of the electron beams and photon fields used in these applications mainly range from a few hundred keV to about 10 MeV.

Any dosimeter that is employed in determining absorbed dose in an irradiated product should be calibrated in terms of national radiation standards. This is especially true for processes that involve public health, such as the sterilization of medical goods. In addition, improvements in measurement assurance of the dosimetry system increase quality control and energy savings in the radiation processing procedures.

Traceability of Absorbed Dose Measurements

The means of achieving traceability of routine dosimetry measurements performed in intense fields of electrons and photons to NBS calibration standards are shown in Figure 2. The approximate uncertainty of each level of measurement is given on the left. Each step of these routes of traceability will be discussed separately.

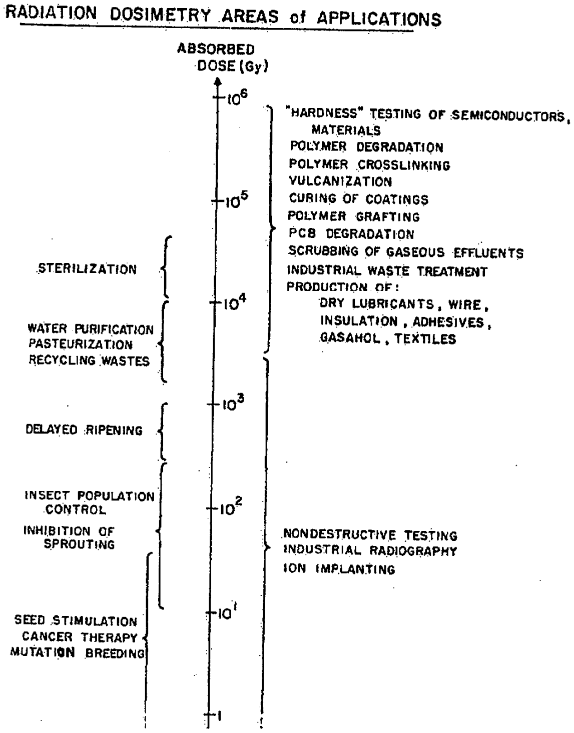

Fig. 1. Areas of application for radiation processing that are already commercially successful or potentially viable.

Primary Measurements

One of the primary methods of making absolute measurements of absorbed dose in intense radiation fields is by calorimetry.[3] Relatively simple calorimeter designs may be used in high intensity electron beams (either pulsed or continuous) if the specific heat and the thermal decay constant of the system are known with sufficient accuracy. Calorimeters made of thin foils of carbon and metals (such as aluminum, copper, and tantalum) have been successfully utilized.[4] Water calorimeters have also been employed in measuring such radiation sources.[5] The electron energy range covered by these types of calorimeters is about 0.1 to 10 MeV.

Measurements at lower dose rates for either electrons or photons require more elaborate adiabatic or heat-loss-compensated calorimeter designs. These types of calorimeters have been used to measure absorbed dose rates from ^{60}Co sources, 50-MeV photon and electron beams, and 20-GeV electron beams.[6]

An NBS water-shielded ^{60}Co source is used for most calibrations of dosimeters for radiation processing users. This source consists of twelve sealed source rods, each about 14 cm long, with a total activity of about ten kilocuries, in a cylindrically isotropic array located at the bottom of a three-meter deep water pool. Primary absorbed-dose-rate measurements were

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*The new SI derived unit for absorbed dose in the gray, symbol Gy. The old unit is rad. 1 Gy = 1 J · kg^{-1} = 10^2 rad.

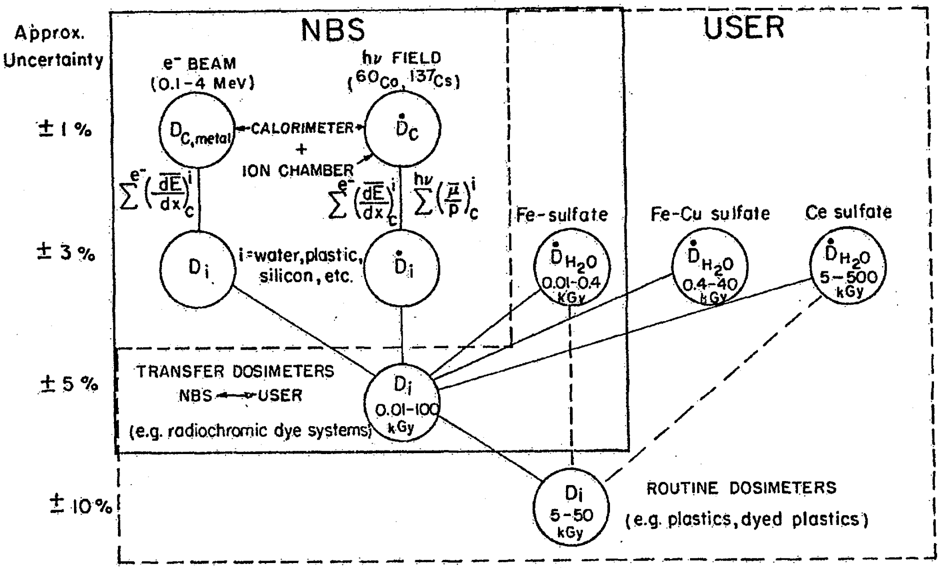

Fig. 2. Traceability routes of NBS dosimetry calibration services.

performed in this source with an adiabatic graphite (carbon) calorimeter of spherical shape.[7] The dose rate in this source is checked periodically with a graphite ionization chamber of the same size and geometry as the graphite calorimeter and with ferrous sulfate solution dosimeters of similar dimensions.

Although not used in this NBS program, certain types of chemical dosimetry systems may be considered to be primary measurement methods. One system involves acidic aqueous solutions of ferrous sulfate or ferrous ammonium sulfate, the "Fricke dosimeter".[3,8] The solution changes its optical absorption properties in the uv region of the spectrum in proportion to the absorbed dose received. It may be considered a primary system in that the values of radiation-chemical yield of the ferric ion (G-value), and molar extinction coefficient ($\varepsilon_{Fe^{3+}}$) at 305 nm, are well documented and measurable with high precision (±1%) with good spectrophotometers. There are difficulties in using this system for routine measurements since any organic impurities in the solutions must be avoided and optically clear fused-quartz containers that are immaculately clean must be used. The dose measurable by this system is limited to a maximum of about 400 Gy.

Other liquid chemical systems that can be employed at higher absorbed dose levels than the Fricke dosimeter include ferrous-cupric sulfate[8] and ceric-cerous sulfate.[9] These systems are also considered to be primary methods, since the G-values and molar extinction coefficients are known, but impurities may be even more troublesome than in the Fricke system.

Absorbed Dose Determination in Other Materials

In order to convert the absorbed dose measured in graphite by the calorimeter system in the NBS ^{60}Co source to absorbed dose in another material, it is necessary to know the approximate photon spectrum of the ^{60}Co source. A recent Monte Carlo spectrum calculation by Frederickson[10] for a source of similar geometry to the NBS source is shown in Figure 3. The scattered photon component amounts to about 22% of the total. This spectrum can be used to weight the energy absorption of various materials of interest such as water, silicon, aluminum, polystyrene, etc. Thus the absorbed dose in these materials can be estimated from the calorimeter response to an uncertainty of approximately ±2% by using the spectrum-weighted ratio of the mass energy absorption coefficient of each material relative to carbon.

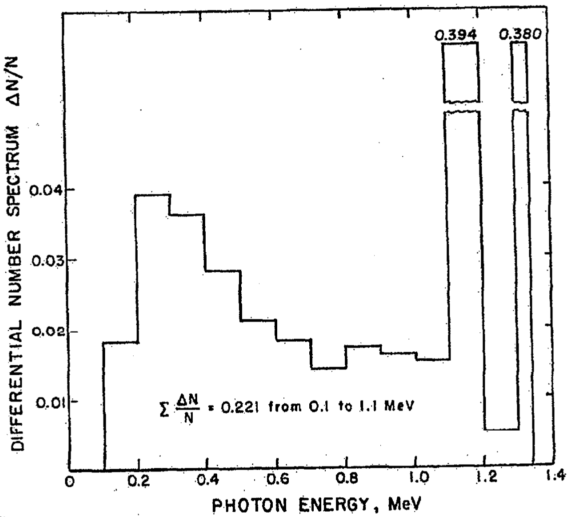

Fig. 3. Scattered ^{60}Co photon spectrum calculated by Frederickson for RADC ten-rod source array.[10]

A similar technique may be used for electron beam irradiations by using ratios of mass collision stopping powers. These ratios change rather slowly as a function of electron energy, so precise knowledge of the spectrum is generally not required.

Transfer Dosimeters

A number of different dosimetry systems have been considered as candidates for use as transfer dosimeters to provide traceability between the primary standards of a national laboratory and routine field dosimeters. Some of the candidates were:

1. Liquid solutions:
   (a) ceric sulfate plus cerous-sulfate
   (b) radiochromic dyes.

2. Undyed plastics:
   (a) cellulose triacetate
   (b) polymethyl methacrylate (clear Perspex).

3. Dyed plastics:
   (a) nylon or polyvinyl butyral
   (b) polymethyl methacrylate (red or amber Perspex)

Of those listed, radiochromic dye films(a clear nylon or polyvinyl butyral matrix with a colorless leucocyanide dye derivative dissolved in it) were chosen by NBS as having the best combination of desirable properties.[11,12] These films are available in thicknesses from about 8 µm to 700 µm. They can cover an absorbed dose range of 10 to 10^5 Gy. Some of the desirable characteristics of these films are:

1. Rugged and easy to handle; readily mailed.
2. Small dependence on LET; basically the same response to photons, electrons, or protons.
3. Low average atomic number; i.e., similar to water.
4. Stable in storage, long shelf life.
5. Simple readout by use of spectrophotometer at visible wavelengths.
6. Have reasonably linear response as a function of absorbed dose with reproducible response characteristics.
7. Stable change in optical density after irradiation; no fading.
8. Response independent of dose rate up to 10^{12} Gy/s. However, dose rate dependence at relatively low dose rates (<1 Gy/s) has been observed when the films are irradiated under low humidity conditions.[13]
9. Fairly small temperature dependence of response.
10. Commercially available in large batches of reasonably uniform sensitivity and thicknesses.
11. Relatively inexpensive.

Certain precautions must be used with these dosimeters:

1. They should not be irradiated in extremes of relative humidity, i.e. below 20% or above 80%. If such conditions can not be avoided during irradiation, then the dosimeters should be sealed in polyethylene pouches with the relative humidity between 50 and 70% before irradiation.

2.. Since the films are read optically, they should be kept free of dirt, scratches, and finger prints.

3. To maintain traceability to national standards, representative samples of each batch should be calibrated.

4. The films must be protected from uv light at all times.

5. In general, the films should not be read until approximately 24 hours or more after irradiation since it requires several hours for full dye development. However, it has been shown that if the films are kept at moderately high relative humidity, it is possible to make accurate dose interpretations immediately after irradiation.[14]

Figure 2 indicates that the ferrous sulfate (Fricke) dosimeter might be used as a transfer dosimeter. While the ferrous sulfate system has good accuracy, it is inconvenient for use as a transfer dosimeter because of the low maximum dose that can be measured. It also is fragile and difficult to read (it requires a constant temperature during readout in a spectrophotometer).

Routine dosimeters

The requirements for dosimeters used in routine dosimetry measurements are basically the same as those used as transfer dosimeters. The most widely and successfully used in radiation processing are plastic dosimeters such as red and amber Perspex, radiochromic dye film, and cellulose triacetate. As indicated in Figure 2, the liquid-glass systems of ceric-cerous sulfate and ferrous-cupric sulfate are also used. These latter dosimeters have the expected disadvantages of liquids: fragile vials that are easily broken in transit as well as being less convenient to handle and analyze. One advantage of the ceric-cerous sulfate system is the extended maximum dose range (up to 500 kGy) available.

Another dosimetry system not given in Figure 2 that recently has been shown to be capable of measuring a very large dose range is lithium fluoride crystals. In this application the LiF is not used as a thermoluminescence dosimeter. Instead, it is read by measuring the change in optical transmission characteristics as a function of absorbed dose at certain wavelengths in the same manner as the plastic dosimeters are utilized. By employing different wavelengths for different dose levels, a dynamic range of absorbed dose of 10^2 to 10^7 Gy may be covered with the LiF system.[15]

Calibration Services

Several dosimetry calibration services are now offered by NBS to users of radiation processing facilities employing high-intensity γ-ray sources or electron beams.[16] The purpose of these services is to provide the industrial radiation processor with traceability of his routine dosimetry system to primary measurement standards at NBS based on calorimetry. The services offered include:

1. Irradiate the user's dosimeters to a known absorbed dose level (1 to 600 kGy) of ^{60}Co γ rays.

2. Provide dose interpretations for the user's irradiation facility with NBS supplied packaged radiochromic dye films. The observed dose can be interpreted in terms of dose in water or silicon (or aluminum).

3. Make intercomparisons of routine dosimeter performance between laboratories.

4. Analyze the response of various chemical and plastic dosimetry systems by means of spectrophotometry.

5. Determine the dependence of the response of routine dosimeters on dose rate, temperature, and humidity.

6. Carry out special measurements (such as dose profile mapping) at the user's facility.

The results of such calibration services are provided to the user in the form of an NBS Calibration Report.

The ^{60}Co source used for most of the calibrations discussed in this section is a water-shielded cylindrical array of source rods described previously. To make sure that the absorbed dose gradient over the volume of dosimeters being calibrated was within ±2%, detailed dose distributions were measured within the available irradiation volume. This was accomplished by irradiating large sheets of thin radiochromic dye films held between plates of polymethyl methacrylate. The sheets were analyzed with a scanning microdensitometer and the results are shown in Figures 4 and 5.

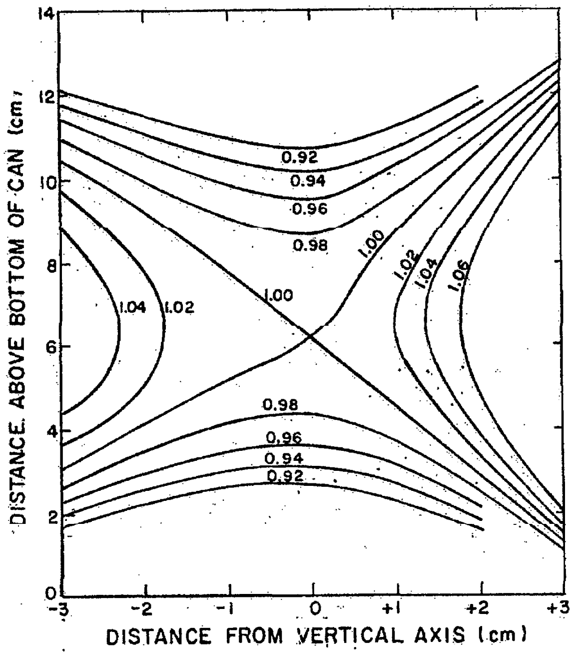

Fig. 4. Measured isodose curves for NBS twelve-rod cylindrical-array ^{60}Co source.

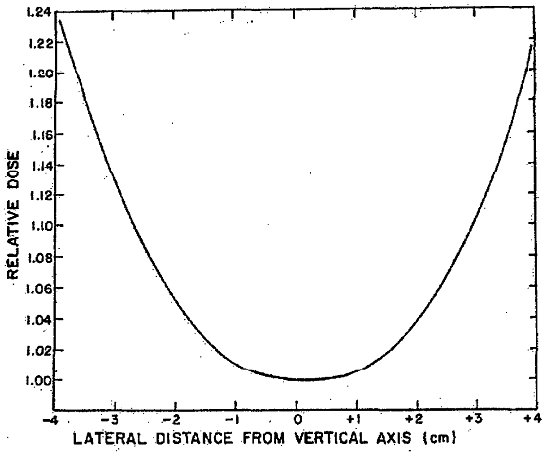

Fig. 5. Absorbed dose variation in NBS ^{60}Co source array in lateral direction 6 cm above bottom of irradiation can.

The assymetry of the distribution in the lateral direction from the vertical axis is probably due to a slight variation in dye film thickness. It can be seen that within 1.5 cm of the calibration position, centered 6 cm above the bottom of the irradiation can, the absorbed dose is uniform to within ±2%.

Conclusions

The NBS calibration services discussed in this paper are providing the industrial radiation processor with traceability of his dosimetry measurements to national standards. However, there are needs in other areas that have not been satisfied at present. These needs include;

1. More international dosimetry standards and international laboratory intercomparisons.

2. Development of transfer calorimeters to decrease uncertainties and reduce the number of transfer levels.

3. Development of improved routine dosimeters, particularly for use in areas not addressed at present, such as flash x-ray fields.

4. Improved knowledge of radiation spectra of both calibration sources and processing sources.

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