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THE MEASUREMENT OF MEGARAD DOSES
OF ⁶⁰COBALT-60 GAMMA RAYS
WITH ULTRAVIOLET TRANSMITTING LUCITE

A.C. MULLER, F.X. RIZZO, AND R.B. MOGK

March 1966

BROOKHAVEN NATIONAL LABORATORY
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ABSTRACT

Commercially manufactured ultraviolet transmitting Lucite was evaluated as an integrating gamma dosimeter. Cobalt-60 gamma irradiation induces an ultraviolet absorbance in Lucite and this absorbance is directly proportional to absorbed dose in the range from 10^4 to 6×10^6 rads. After the proper corrections are made for bleaching and thickness, dosimetric measurements can be made with an expected precision of $\pm 3\%$.

THE MEASUREMENT OF MEGARAD DOSES OF COBALT-60 GAMMA RAYS WITH ULTRAVIOLET TRANSMITTING LUCITE

INTRODUCTION

The objectives of the dosimetry program at the BNL High Intensity Radiation Development Laboratory (HIRDL) are oriented toward the development of methods and systems suitable for measuring absorbed gamma dose in various targets for the radiation processing industry. Since most of the target materials used in these processes are of low density and atomic number, dosimeters that resemble these materials have been of great interest. Such a dosimeter is the Ultraviolet Transmitting (UVT) Lucite dosimeter developed recently at Brookhaven National Laboratory.

When commercially produced UVT Lucite is exposed to cobalt-60 gamma radiation it develops an ultraviolet (uv) absorbance. The absorbance, which can be easily and accurately measured with a Beckman DU spectrophotometer, is proportional to total dose over a range of several megarads. The absorbance fades slowly, and at certain wavelengths bleaching corrections may be neglected. If corrections are made for thickness variations, Co⁶⁰ gamma doses may be measured with a precision of $\pm 3\%$.

THEORY

Commercially produced polymethylmethacrylate is sold under the trade names of Plexiglas, Perspex, and Lucite. Lucite, the most generally used term, was adopted in describing and reporting this work.

The density of Lucite is between 1.1 and 1.2 g/cc. It is manufactured in two general varieties, ultraviolet absorbing (UVA) and ultraviolet transmitting. UVA Lucite, the more common, contains sun-screening additives designed to absorb short-wavelength light. These agents make UVA material unsuitable for dosimetric use. Because UVT Lucite is free of these impurities, it has a much greater range of light transmission. Its lower wavelength limit of transmission is below 2000 Å.

When ionizing radiation interacts with a plastic, such as Lucite, part of the energy deposited is

utilized to produce chemical and physical changes; the remainder is degraded to thermal energy.

The primary chemical effects involve the production of free radicals, excited and ionized atoms, and molecules and electrons. The end result is main-chain scission of the polymer, together with the production of considerable quantities of gas.

Concurrent with these chemical changes, irradiated UVT Lucite assumes a yellow-brown coloration. Spectrophotometric measurements indicate that the visible coloring is accompanied by an optical absorption band located in the uv region of the spectrum. The occurrence of this uv absorption band in the commercially manufactured product is attributed to the interaction of the radiation with one or more of the standard additives used in the manufacture of Lucite. The additive responsible for the coloring phenomenon is probably a catalyst, plasticizer, or antioxidant.

Irradiation of thermally polymerized, 99.9% pure Lucite proves that the uv absorption cannot be induced in Lucite that is free of these additives. Presumably the absorbance results from the generation of optical absorbing centers located at the impurities within the volume of the Lucite. Because the induced absorbance is proportional to the number of absorbing centers produced within the volume of the Lucite, it will also be proportional to the thickness of this material.

Lucite was first tested as a dosimeter several years ago. In 1958, Boag¹ successfully employed UVT Lucite for electron dosimetry studies. In this work he tested UVT Lucite as a dosimeter for high-intensity pulsed electron beams, and he found that the uv absorption at 292 mμ increased linearly from 10⁵ to 3 × 10⁶ rads, independent of dose rate.

EXPERIMENTAL RESULTS

Apparatus and Materials

The UVT Lucite used in these experiments was purchased under the Rohm and Haas trade name of Plexiglas II UVT. This plastic is a water-white, colorless, transparent material, highly resistant to chemicals, acids, and physical shock. UVT Lucite

is manufactured in 3-ft-sq sheets in a variety of thicknesses. It can be sawed, drilled, and machined like wood or soft metal.

Samples from three separately manufactured batches of material were tested in this study. These batches are referred to in this report as batches *A*, *B*, and *C*. The results reported in the following sections were obtained mainly with batch *A* material.

All spectral measurements were performed with a Beckman Model DK-1 Ratio Recording Spectrophotometer. Absorbance measurements, requiring high accuracy, were made with a standard Beckman Model DU Quartz Spectrophotometer. Thermal bleaching experiments were performed in an atmosphere of dry, prepurified nitrogen.

Spectral Characteristics

The uv absorption induced in UVT Lucite by Co^{60} gamma radiation is illustrated in Figure 1. Lucite samples fabricated from different batches of mix show variations in the shape and position

Figure 1. Absorption spectra in UVT Lucite irradiated to a Co^{60} gamma dose of 2×10^6 rads.

Figure 2. Absorption spectra of batch *A* Lucite irradiated to a Co^{60} gamma dose of 2×10^6 rads. Spectra were taken 5 min (—), 3 hr (---), and 10 hr (- - -) after irradiation.

of their induced absorption spectra. The spectra illustrated in this figure correspond to samples of batches *A*, *B*, and *C*. The main absorption maxima of the *A*, *B*, and *C* material are located at 250, 278, and 350 $\text{m}\mu$, respectively.

The radiation-induced absorption spectrum is usually asymmetrical, which indicates that Lucite contains two or more sensitizers. For example, in the spectrum of batch *A* Lucite (Figure 2), three separate peaks are discernible.

Linearity

The linear relationship between induced uv absorbance and total dose is illustrated in Figure 3. Duplicate samples of 1-mm-thick batch *A* UVT Lucite were irradiated with a series of 10^5 -rad doses to a total dose of 10^6 rads, and the induced optical density was plotted against total dose at a wavelength of 265 $\text{m}\mu$. These data indicate that the induced optical density is directly proportional to dose above 10^5 rads.

Figure 3. Induced optical density vs total Co^{60} gamma dose in 1-mm-thick batch *A* Lucite.

Figure 4. Induced optical density vs total Co^{60} gamma dose in threshold range of batch *A* Lucite.

Figure 5. Induced optical density vs total Co^{60} gamma dose in 1-mm-thick batch B Lucite.

Below total doses of 10^5 rads, the linear relationship between total dose and induced absorbance does not hold. This threshold effect is shown in greater detail in Figure 4. An additive, such as an antioxidant, probably competes with the coloring process in this dose range and inhibits the coloring until the additive is depleted. A linear relationship between absorbance and total dose can be guaranteed by irradiating all dosimeters with a dose greater than the threshold dose. All Lucite dosimeters used at BNL receive a predose of 200,000 rads before use.

The linear relationship between induced absorbance and total dose (Figure 3) is found only within a small wavelength range. This region is located on the long-wavelength side of the peak, and is centered $\approx 20 \text{ m}\mu$ from the peak position. When the total dose is < 1 megarad, the optical density is directly proportional to dose over a range of $\approx 30 \text{ m}\mu$. If the total dose is > 3 megarads, this range is considerably smaller. For example, when batch B Lucite is irradiated to a total dose of > 6 megarads, the linear relationship between induced optical density and total dose is found only between 295 and $300 \text{ m}\mu$. This is illustrated in Figure 5.

Because the uv absorption is caused by the presence of absorbing centers in the volume of the Lucite, the induced optical absorption per unit exposure is directly proportional to thickness. The thickness tolerance on commercial, 0.040-in.-thick Lucite is $+0.006 \text{ in.}$ to -0.010 in. Because of this large variation in nominal thickness, corrections must be made if a reproducibility of $> \pm 20\%$ is desired.

Figure 6. Postirradiation stability of batch A Lucite irradiated to total Co^{60} gamma dose of 10^6 rads and maintained at 25°C .

Energy Dependence

Lucite has been compared with the Fricke dosimeter at distances up to 12 in. from a source in an infinite water medium. No relative energy dependence effects were found in this range.

Bleaching

The stability of the induced uv absorbance of irradiated batch A Lucite is illustrated in Figure 2. In this experiment the irradiated sample was maintained at 25°C and the absorption spectra were measured on termination of irradiation and again at 3 and 10 hr after irradiation. The second and third peaks of the absorption spectrum, mentioned previously, are more pronounced in these curves. Each of the three absorption peaks bleaches at a different rate, which causes the bleaching of the absorbance curve to be wavelength dependent.

The effect of wavelength on bleaching rate was studied in detail and the results are shown in Figure 6. In this experiment batch A Lucite was irradiated to a Co^{60} gamma dose of 6×10^5 rads. Following irradiation the uv absorption was monitored continuously for 10 hr. The induced absorbance is plotted at wavelengths of 250, 275, and $300 \text{ m}\mu$ as a function of total time after irradiation. The absorbance bleaches very rapidly ($\approx 15\%/hr$) at $250 \text{ m}\mu$. At $300 \text{ m}\mu$ the bleaching rate is slower and the absorbance increases with time. At $275 \text{ m}\mu$, after a small rise, the absorbance

remains essentially constant for a period of 5 hr. Bleaching corrections are unnecessary when Lucite dosimeters are monitored at this wavelength.

Reproducibility

A determination of the reproducibility of the dosimeter was made using batch *A* Lucite. In this test 100 samples 1 mm thick were randomly selected from one 9-sq-ft sheet. They were stabilized with an initial dose of 200,000 rads and then irradiated to a Co^{60} dose of 10^6 rads. The dose was administered in the HIRDL gamma irradiation facility and standardized against the Fricke dosimeter.

Two hours after irradiation was completed, the optical densities of the 100 samples were measured at a wavelength of 275 m μ . The induced optical density of each dosimeter was then normalized to a nominal thickness of 1 mm (0.040 in.), and the arithmetic mean of the group was computed.

The distribution of optical densities resulting from this experiment appears in Figure 7. The mean value determined was 0.385 per megarad per millimeter, and the standard deviation was $\pm 3\%$. This value was unchanged 2 hr later (4 hr after irradiation).

Miniaturization Studies

Initial experimental data on UVT Lucite were obtained with samples measuring 1 cm $\times$ 2 cm $\times$ 1 mm thick. Minor modification of the DU spectrophotometer, in conjunction with installation of a precision positioning sample holder, made it possible to reduce the size to 5 $\times$ 12 $\times$ 1 mm. These two dosimeters are already in use in the characterization experiments at HIRDL and are illustrated in

Figure 7. Distribution of induced optical density in sample of 100 batch *A* dosimeters irradiated to Co^{60} gamma dose of 10^6 rads.

Figure 8. A 2 $\times$ 2 $\times$ 1-mm dosimeter now being developed will make possible the accurate placement of dosimeters at target interfaces.

Additional miniaturization studies now under way indicate that with slightly more sophisticated spectrophotometric techniques, a "Lucite rod dosimeter" is feasible. This dosimeter, shown at the left in Figure 8, measures $\approx 1 \times 1 \times 10$ mm and will further increase the possible applications of the UVT Lucite dosimeter. One such application would be the calibration of prototype grain irradiators. For example, Lucite dosimeters approaching the size of grain particles would be mixed in with the product and processed along with it. Dose measurements obtained in this manner should yield an excellent "dynamic" calibration of bulk-flow-type irradiators.

APPLICATIONS

UVT Lucite dosimeters are regularly used in characterization studies in the HIRDL facility. In these studies dose distribution measurements are made in a variety of homogeneous and heterogeneous targets.

Lucite dosimeters are placed at desired locations within these targets with the aid of holders specifically designed for each application. Two such holders are illustrated in Figure 9. The circular holder, a paraffin block, is a cylinder 1 in. high and 4 in. in diameter. Lucite dosimeters are placed at any desired radius or height within this holder. The paraffin block is then stacked with other similar sections to fabricate a density-one cylindrical target. This arrangement makes it possible to monitor dose rates in any standard size food can.

The rectangular holder is a 9 $\times$ 15 $\times$ 0.050-in. phenolic sheet with 0.040-in.-deep grooves milled along the diagonals and in the center of this plane. Once loaded with dosimeters, this sheet is placed in any desired plane of the dosimetry package.

Dose distribution data obtained from a prototype shipboard irradiator with this holder appear in Figure 10. In this determination, 5 $\times$ 12 $\times$ 1-mm dosimeters were placed at $\frac{1}{2}$ -in. intervals along the diagonals of the phenolic dosimetry sheet. This holder was then placed in the center plane of a 4 $\frac{1}{2}$ -in.-thick dosimetry package and exposed in the irradiator. The average dose rate measured was plotted as a function of distance along the diagonal from the outside corner to the center in inches. These data yield precise information about

Figure 8. Experimental Lucite dosimeters used at Brookhaven National Laboratory. Paper clip at right is shown for size comparison.

Figure 9. Holders used to position Lucite dosimeters in homogeneous and heterogeneous targets.

Figure 10. Dose distribution in density-one package irradiated in shipboard fish irradiator. Dose measured with $5 \times 12 \times 1$ -mm Lucite dosimeters.

dose rates at selected points in the target as well as maximum to minimum dose ratios.

CONCLUSION

Commercially manufactured UVT Lucite was evaluated as an integrating gamma dosimeter. Lucite has excellent machining characteristics and can be fabricated with standard machine-shop techniques. It is highly resistant to moisture, acids, and physical shock. Commercial manufacturers of plastics now produce homogeneous 3-ft² sheets of 1-mm-thick Lucite. Several thousand dosimeters can be produced from one 9-sq-ft sheet at a cost of <2¢ each.

Co^{60} gamma irradiation induces a uv absorbance in Lucite. This absorbance is directly propor-

tional to absorbed dose. Since the absorption of light takes place in the volume of the Lucite, the optical density is also directly proportional to sample thickness. The absorbance is reproducible among all the dosimeters fabricated from one batch of polymethylmethacrylate, and, if a 200,000-rad predose is employed, Lucite is linear over the range from 10^4 to 6×10^6 rads.

Lucite has a high postirradiation stability, and bleaching effects can be minimized at certain wavelengths. After the proper corrections have been made for bleaching and thickness, dosimetric measurements can be made with an expected precision of $\pm 3\%$. Although the calibration constants and bleaching characteristics must be redetermined for each new batch of Lucite, the dosimetric characteristics of the 60,000 to 70,000 Lucite dosimeters fabricated from one mix of materials are identical.

A plastic dosimeter of more universal characteristics is being studied at BNL. In this program pure samples of polymethylmethacrylate and polyethylene will be doped with a series of known additives. These experimental dosimeters will be irradiated and their uv absorbances studied. Successful doping of a plastic vehicle will lead to a dosimeter consistent in its radiation response properties and possessing a calibration constant reproducible from batch to batch.

REFERENCE

1. J.W. BOAG, G.W. DOLPHIN, AND J. ROTBIAT, *Radiation Res.* **9**, 589 (1958).