

Electron Dose Measurement with Polycarbonate Film Dosimeter

Young Soo Yoo

Korea Atomic Energy Research Institute, Seoul, Korea

(Recd. Dec. 10, 1975)

Abstract

Dosimetrical properties of polycarbonate film for high-level dosimetry of electrons have been examined. Polycarbonate film of 0.1mm in thickness was chosen for this purpose. It can cover the dose range of 1.0-130 Mrad and the measurable range can be extended up to 200 Mrad by using calibration curve. The measurement error was within 3.5%.

The radiation induced optical density at 330nm shows rapid initial fading of 7-13% for one day after irradiation at room temperature and subsequent fading rate is very small, about 0.6% per day. The fading depends on the absorbed dose, storage temperature, and wavelengths. The effects of storage time and temperature during and after irradiation of this film are presented.

For practical dosimetry, it is necessary to stabilize the induced optical density by storing the irradiated film for a day or by heat treatment at 100°C for an hour.

요 약

고준위 전자선량을 측정하기 위하여 두께 0.1mm인 polycarbonate 필름을 선정하여 선량계 특성을 조사하였다. 선량범위는 1.0-130 Mrad이며 보정곡선을 이용하여 200 Mrad까지도 측정할 수 있었고 측정오차는 3.5% 이내였다. 330nm에서 측정할 농도는 실온에서는 조사후 1일 동안에 약 7-13% 감소하였으며 그 후의 감소율은 매우 적어 약 0.6%/일 였다. 조사후의 농도변화는 흡수선량, 보존온도 및 파장에 따라 상이하였기 때문에 조사후 경과시간 및 보존온도와 조사시의 온도에 관한 효과를 검토하였다. 이 선량계를 사용할때에는 농도 변화의 오차를 줄이기 위해서는 1일후에 농도를 측정하거나 혹은 100°C에서 1시간 열 처리가 필요하였다.

1. Introduction

For radiation dose measurement, many materials, such as polyvinyl chloride,¹⁻³⁾

polymethylmetacrylate,⁴⁻⁵⁾ cellulose nitrate⁶⁾ and polyethylene terephthalate⁷⁾, etc,⁸⁻¹⁰⁾ have been developed for the dose measurement in the megarad ranges. Some of them are widely used as reliable dosimeters in the

fields of radiosterilization and radiation chemistry, etc. In these applications the dose range of 1~100 Mrad is required.

Recently, polycarbonate film has been examined for the use in the field of high dose measurement. A simple dosimeter using polycarbonate film has been designed by Douglas et al¹¹⁾. for the measurement of dose from 0.1 to 100 Mrad of ⁶⁰Co gamma-rays and also Endo¹²⁾ reported the use of thin polycarbonate film as a dosimeter covering a range from 1 Mrad up to 100 Mrad for electrons.

Unirradiated polycarbonate film is transparent to visible light. After irradiation it shows an absorption in the ultraviolet range; the optical density is related to the amount of absorbed radiation in the same range. It was observed that on irradiation polycarbonate turned yellow, and the developed color intensity was dependent on the absorbed dose. Since the polycarbonate film seems to be a suitable material for higher dose measurement, this paper outlines the dosimetric characteristics of polycarbonate film as a cheap, accurate, simple, and reliable secondary dosimeter in the megarad range. The experiments have been performed with electron beam. The effects of dose and storage time and temperature after irradiation have been investigated.

2. Experimental

2.1 Material

A commercially available polycarbonate film known as Teijin Pan Light Film in Japan, have been used as of secondary dosimeter. The film thickness chosen is 0.1mm and the density is 1.2g/cm³. The uniformity of the film thickness was approximately

±1.5%. All specimens were cut to a standard size of 4.5cm×1.2cm for convenience in spectrophotometric measurements. The films were washed in ethanol detergent solution, wiped with soft tissue paper, and sealed in a black plastic envelope to protect it from scratches and light before and after use.

2.2 Irradiation

Irradiation of polycarbonate dosimeter was carried out using a Cockcroft-Walton type electron accelerator in the Takasaki Radiation Chemistry Research Establishment, JAERI, Japan. This accelerator has the maximum output 3 MeV in accelerating voltage and 5 mA in beam current. Irradiation of the dosimeter, placed on the polyethylene plate, 1.0cm thick, and 28cm from the exit window, was carried out with a conveyor speed of 3.1m/min and beam energy 1.5 MeV, beam current 3 mA. Under these conditions the absorbed dose was 1.67 Mrad per pass and the average dose rate in the center of the scanner was 5.17×10^5 rad/sec. All irradiations were performed at the center of the scanned beam. Absorbed dose was determined by the graphite calorimeter¹³⁾. Experimental arrangements for electron

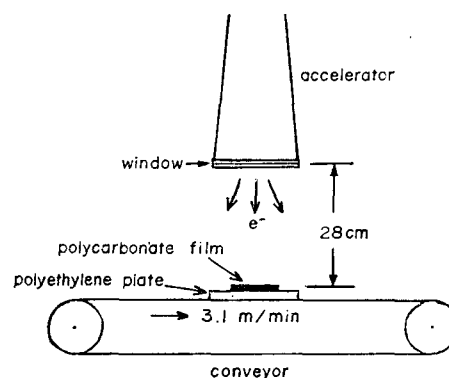


Fig. 1. Geometrical arrangement of conveyor irradiation

on irradiation are shown in Fig. 1.

2.3 Optical density measurement

The ultraviolet absorption was measured with a Japan Spectroscopic Model UVIDEC-2 digital spectrophotometer at a wavelength range of 285nm to 500nm. In most cases complete spectra were plotted for each film and some were spot checked at selected wavelength. Since the transparent polycarbonate film absorbs ultraviolet rays before irradiation, the initial optical density of each film must be subtracted from the optical density after irradiation. Since the values of initial optical density are vary, the initial optical density of each sample should be measured in each run. And the sample which

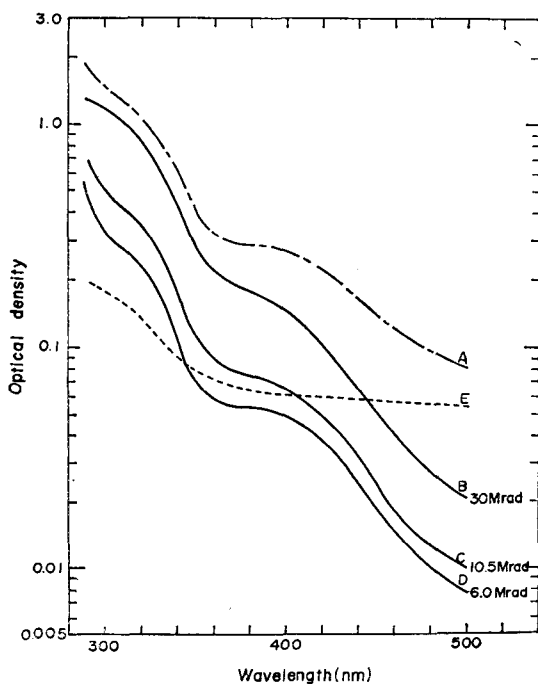


Fig. 2. Absorption spectra of the polycarbonate film (0.1mm thick). A; irradiated to 30 Mrad, B, C, D; net optical density for given dose, E; unirradiated.

has large initial optical density was rejected.

3. Result and discussions

3.1 Absorption spectra

Typical absorption spectra for irradiated and unirradiated polycarbonate sample are shown in Fig. 2. The radiation induced spectra are obtained by subtracting optical density of unirradiated sample from that of irradiated sample. The optical density of unirradiated sample shows rapidly decreasing absorption with increasing wavelength until 350nm, and beyond this wavelength the optical density decreases slightly.

The radiation induced absorption curves for different doses were used for selection of suitable wavelength. The absorption does not increase simply toward shorter wavelength, and a shoulder in absorption appears around 400nm. The spectra of the sample for three dose levels are shown in Fig. 2. The absorption spectra show almost the same shape except for around 300nm. However,

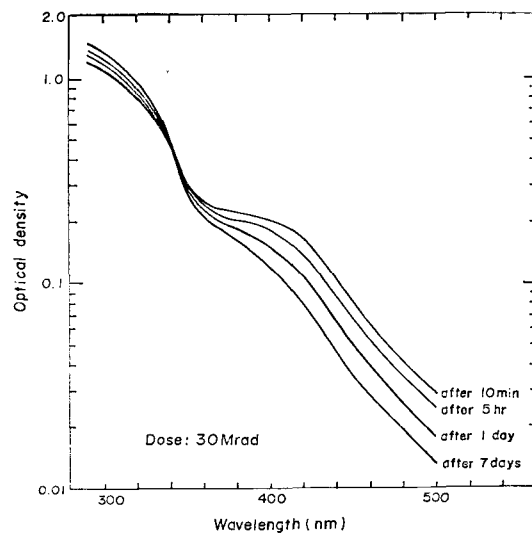


Fig. 3. Radiation induced spectral change for 0.1mm thick film kept at room temperature.

it is apparent that the shoulder around 400nm was reproduced at all dose levels. But there was small difference with absorbed dose, namely, the shoulder around 400nm was predominant at 6 Mrad but it was not apparent in high dose region. In general, all samples exhibited substantially the same spectral changes. The fading characteristics and dose dependence were examined at 300, 325, 330 and 350nm for given doses.

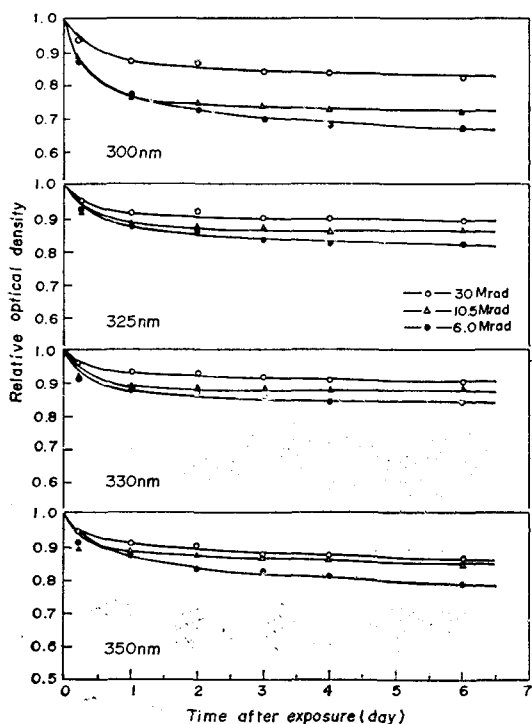


Fig. 4. Post-irradiation fading at room temperature at given wavelength.

3.2 Fading of the induced absorption

The optical density of polycarbonate film produced by irradiation is unstable as in many other plastics dosimeters, and it was gradually faded. Effects of absorbed dose, wavelength, and storage conditions on the fading were investigated. The changes of the spectra were observed at various interv-

als, keeping them in the dark at room temperature, for a period of two months or more.

As shown in Fig.3 the shoulder around 400nm gradually vanished as the time elapsed, and minimum changes were observed around 330-340nm. Since the spectrum after 7 days storage showed almost no further fading, this curve can be reliably used for the analysis of the radiation induced absorption spectra.

Relative change of optical density with time after irradiation at given dose levels are shown in Fig.4 for the selected wavelengths. Radiation induced optical density decreased rapidly during the first one day after irradiation for samples kept at room temperature, and subsequent changes were very slow (less than 0.6% per day at 330nm). The decrease during the first one day at 330nm was about 10-16% of the initial optical density. Therefore, a storage of the sample after irradiation is necessary to reduce the dosimetric error due to stabilization process. The dependence of fading on the absorbed dose was also observed. The fading at the lower dosage was greater than in the higher one. Fading observed at 330nm was the smallest of all selected wavelengths. In the case of 330nm it showed about 35% decrease in optical density after one week.

The long-term fading over a period of three months at room temperature are shown in Fig. 5. The fading occurs within the first one week after irradiation, and thereafter the optical density becomes nearly constant. Decrease of the optical density for 6 days after an initial storage period of 24hr at room temperature was about 2.3%. If all measurements are made at 330nm very little fading takes place even up three months

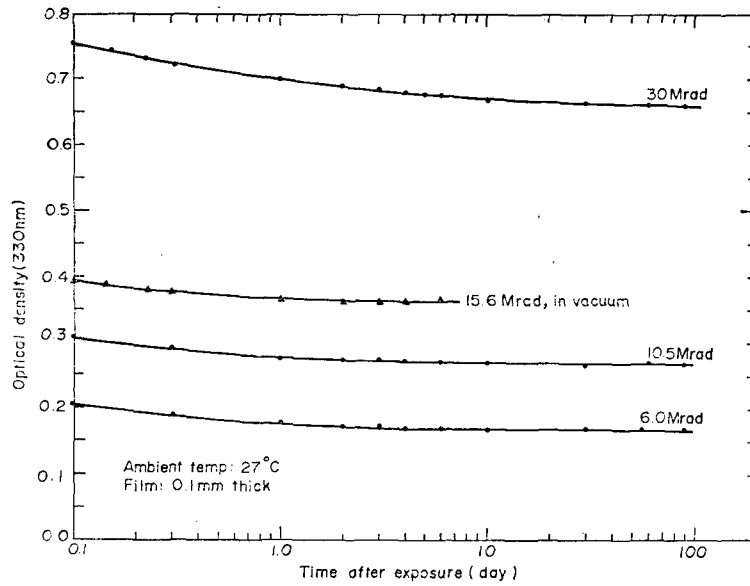


Fig. 5 Fading of optical density for various doses at room temperature.

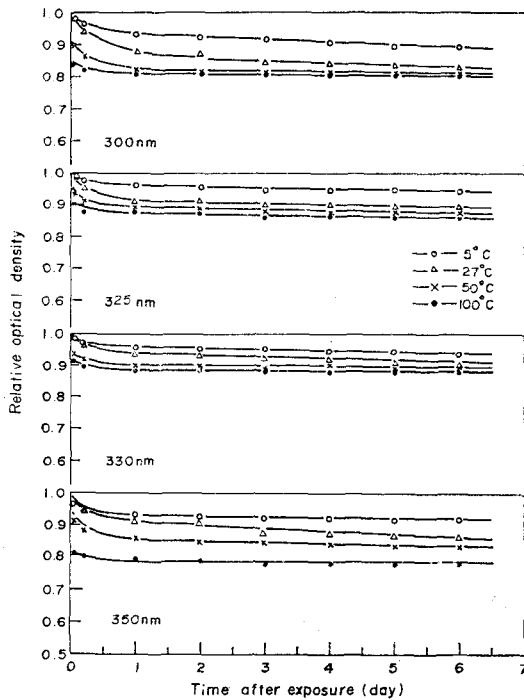


Fig. 6. Fading of optical density at various ambient temperatures (Dose; 30 Mrad, film; 0.1mm thick).

Because of this, 330nm was chosen as the measurement wavelength.

The effect of keeping the irradiated samples in vacuum, approximately 0.01mmHg, at room temperature was also shown in Fig. 5. It is found that fading is not critically affected by the surrounding atmosphere.

3.3 Effects of heat treatment

The fading of optical density in irradiated polycarbonate film depends on the storage temperature. Figure 6 shows the effect of storage temperature on the fading in the 0.1 mm film thick; the samples were kept at 5°, 50°, 75° and 100°C after electron irradiation of 30 Mrad. It is evident that the fading rate is higher at higher temperatures. At 100°C the optical density decreases within an hour and reaches almost the stable value. The decrease of optical density after six days storage at 100°C is 10–22% of its initial value depending on the wavelength, while the decrease at 5°C is only 6–8%.

Figure 7 shows the long-term fading measured at the wavelength of 330nm under various conditions. The absorption spectrum

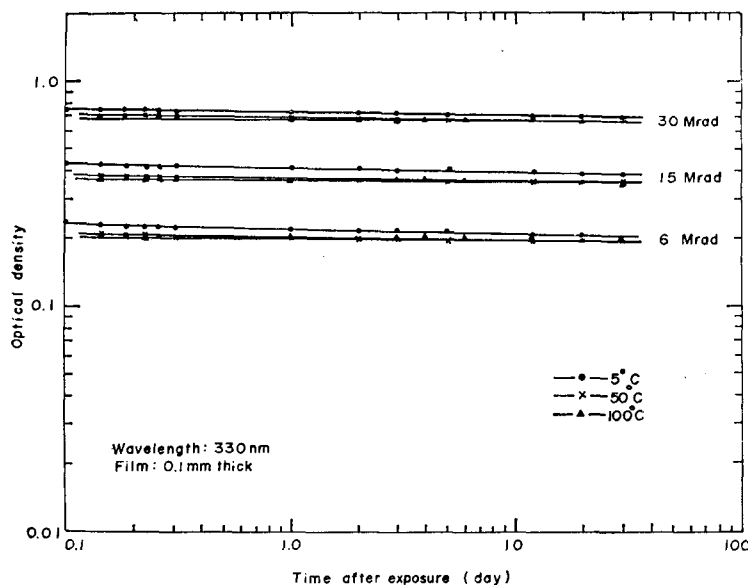


Fig. 7. Fading of optical density for various doses at given ambient temperatures.

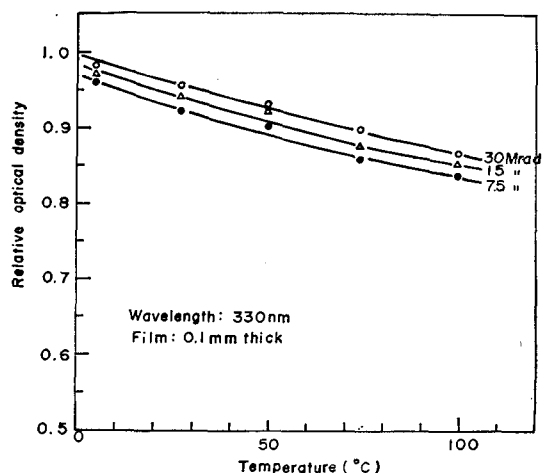


Fig. 8. Changes in optical density after storage of 1 hr at various temperature.

obtained after long-term storage at room temperature is similar to obtained after the storage at 100°C for a short time. The result suggests that the absorption spectrum of irradiated polycarbonate consists of two components, namely the unstable and stable components. By warming the sample up to 100°C the unstable component is almost eliminated, while the stable component still

remains.

Temperature dependence of the stability of the optical density during the storage is shown in Fig. 8. Obviously the optical density decreases with increasing storage temperature. In this experiment the optical density measurements were made after storing the sample at given temperatures for an hour. The results of the temperature effects indicate that heat treatment of the irradiated sample is effective for stabilizing the radiation-induced absorption. In order to determine the optimum condition of heat treatment, variation of the optical density with the heating duration was observed. The optical density is independent of the heating duration at 100°C, while it slowly decreases with heating duration at 80°C. Consequently, the heating at 100°C for 30min or more was the optimum condition for stabilizing the induced absorption.

3.4 Calibration of polycarbonate film dosimeter

The absorbed dose calibration of polycar-

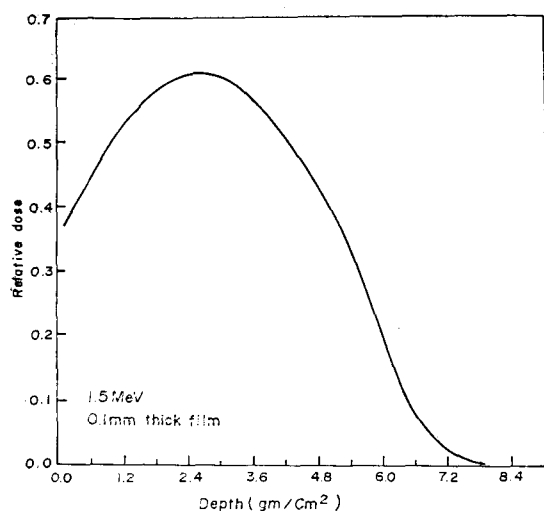


Fig. 9. Depth-dose curve for 1.5 MeV electron irradiation.

bonate dosimeter for the electron beam was carried out by normalizing the integration value of a relative depth dose curve by the energy fluence determined from the graphite calorimeter. A polyethylene block with a rectangular depression of 4.5cm long, 1.3cm wide, and about 0.7cm deep was used as a sample holder. A stack of polycarbonate films 0.7cm high was set in the depression. The electrostatic equilibrium at the outside of polycarbonate film stack is approximately established by this arrangement. The sample was irradiated with electron of 1.5 MeV and 1 mA at a distance of 30cm from the center of beam window. Irradiation time was controlled by beam shutter which operates with high speed. The optical density change at 330nm in each film was converted into relative absorbed dose, which was read from the relation between optical density change and irradiation time. The relative depth dose curve is given in Fig. 9. The energy fluence of incident electron beam was measured by setting the calorimeter at the same position of the polycarbonate stack. The energy fluence was determined from initial increasing

rate of graphite temperature after opening the shutter and irradiation time.¹³⁾

The optical density obtained with each film was converted to relative absorbed dose per unit fluence by means of the following formula:¹⁴⁾

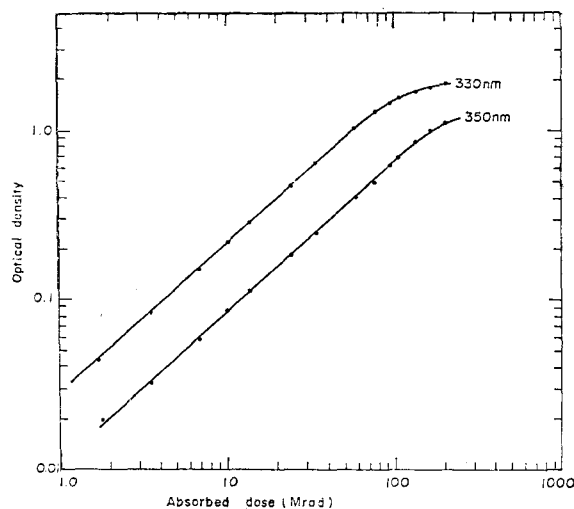


Fig. 10. Absorbed dose calibration curves after heat treatment for 1 hr at 100°C.

$$De = \frac{\rho \cdot t \cdot \sum_i J_i}{F} \cdot 10^8 \quad (1)$$

Where

De ; relative dose per unit dose (Mrad^{-1})

ρ ; density of polycarbonate (1.2 g/cm^3)

t ; film thickness (0.1mm)

J_i ; relative absorbed dose of i_{th} film

F ; incident energy fluence ($0.75 \times 10^7 \text{ erg/cm}^2$).

The resulting De is 1.113. By using De the absorbed dose in Fig. 10 was scaled as absolute value as shown in next paragraph.

3.5 Absorbed dose measurement

For the determination of absorbed dose by using polycarbonate film the 0.1mm thick film was selected as the standard sample. Calibration curves for electron dosimetry after heat treatment for 1 hr at 100°C is shown in Fig. 10. It is noticed with the

300nm curve that a linear range is shorter than other curves and there is a large fluctuation for measured optical density. As can be seen in Fig. 10 the calibration curve in log-log plot is linear in the ranges of about 1 Mrad to 60 Mrad at 330nm, and about 2 Mrad to 130 Mrad at 350nm. The empirical formula was obtained by least squares fit on the linear part;

$$D_{330} = 5.52 \cdot 10^7 \cdot (OD)^{1.127} \quad (1 \leq D \leq 60) \quad (2)$$

$$D_{350} = 1.52 \cdot 10^8 \cdot (OD)^{1.104} \quad (2 \leq D \leq 130) \quad (3)$$

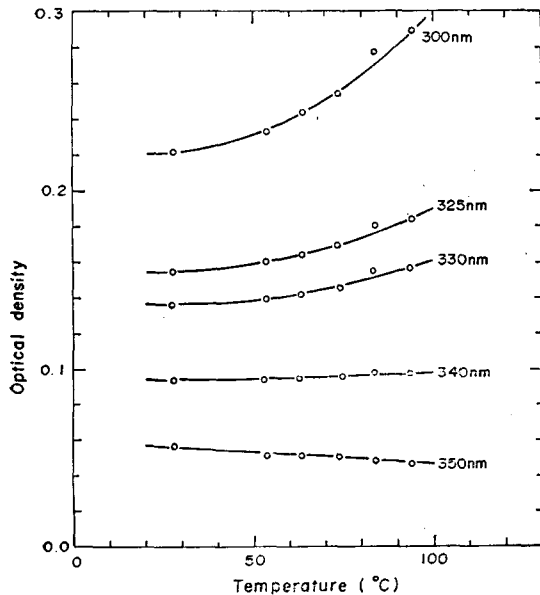


Fig. 11. Effects of irradiation temperature on optical density response (gamma-rays).

where OD is optical density increase and D the absorbed dose in rad. In practically, dose measurements can be made up to 200 Mrad by using the calibration curves measured at 350nm. All calibration points in Fig. 10 are obtained by means of the readings for three dosimeters irradiated simultaneously. The standard deviation for the range of measuring dose was with 0.1-3.5%. Decrease of the optical density for six days after and initial storage period of 24 hr at room temperature is about 2.3%. Also the dust and

finger prints on the film bring about a little increase in optical density, and this can be eliminated by brushing the samples.

The effect of irradiation temperature on the response was also observed as shown in Fig. 11. In the shorter wavelength the response becomes critically sensitive to the temperature and in contrast to the less effect at the longer wavelength. In case of electron irradiation, temperature rise may be negligible because the irradiation time is very short compared with gamma-ray irradiation. However, temperature rise during irradiation also should be taken into account for high dose electron irradiation.

Dose rate dependence on the optical density changes was examined in the range of Mrad per sec but the effect of dose rate was negligible within the experimental errors.

4. Conclusions

The polycarbonate film 0.1mm thick was found to be useful as a simple method of high-level dosimetry. Optical density change at the wavelengths of 330nm and 350nm were chosen for dosimetry by considering dose range and fading. The measurable dose range is 1-60 Mrad at 330nm and 2-130 Mrad at 350nm. Measurement can be extended up to 200 Mrad by using a calibration curve. The standard deviation derived from three dosimeters is less than 3.5%.

The radiation induced optical density rapidly decreases during the first one day after irradiation and thereafter the subsequent fading is small amounting to about 0.6% and 1.1% per day at 330nm and 350nm, respectively, at room temperature. To reduce the error in measurements of optical density without heat treatment a delay time of at

least one day must be taken after irradiation.

The decrease of optical density is sensitive to storage temperature. Fading rate is larger at higher temperature, and it depends on the wavelength and is the smallest at 330nm. Therefore, if all the measurements were made at 330nm very little fading takes place even up to several days after irradiation. Fading is a complex function of dose, initial density, and measured wavelength. Therefore, it is very difficult to give a simple formula on the decrease of the optical density.

Fading rate is accelerated by warming the irradiated film during storage, which indicates that optical density can be stabilized by the heat treatment. For practical dosimetry a heat-treatment for 1 hr at 100°C was found to be the most convenient.

Disadvantages of polycarbonate dosimeter are the increased response at irradiation temperature above 60°C at lower wavelength though these problems would not occur under normal irradiation. However, the dosimeter requires further investigation to determine the dose rate dependence and the precision under operation conditions.

To achieve the reasonable precision in routine use the sample should be carefully cleaned before use and the pre-irradiation optical density should be determined to obtain the net optical density change. The polycarbonate film dosimeter has proved to be an useful dosimeter for megarad dose measurements.

Acknowledgments

The author is indebted to Dr. N. Tamura and R. Tanaka, Irradiation Service Section, Takasaki Radiation Chemistry Establishment, JAERI, for their helpful discussions and encouragement during the course of this work, and also wish to thank the member of Irradiation Service Section for their assistance in irradiating the samples.

References

- 1) E. J. Henly and A. Miller. *Nucleonics* **4**(6), 62 (1951).
- 2) J. E. Maul, et al., *Risø Rep. No. 131. At. Energy Comm. Roskilde, Denmark.*
- 3) C. Whipple, *Nucleonics*. **8**(8), 68(1960).
- 4) J. W. Boag, et al., *Radiation Res.* **9**, 598(1958).
- 5) C. G. Orton, *Phys. Med. Biol.* **11**(3), 377(1966).
- 6) D. A. Young, *Nature* **182**, 375(1958).
- 7) V. H. Ritz, *Radiation Res.* **15**, 460(1961).
- 8) D. M. Bodilt and M. Dole, *J. Chem. Phys.* **45**, 1428(1966).
- 9) J. F. Kirchner, et al., *J. Phys. Chem.* **69**, 189 (1965).
- 10) R. L. Birdsall, et al., *IEEE Trans. Nucl. Sci.* **NS-15**, 3460(1968).
- 11) J. A. Douglas, et al., *AERE-M 2481*(1972).
- 12) H. Endo, et al., *Report of Osaka Industrial Promotion (Japaneses)* **41**, 88(1967).
- 13) N. W. Holm and R. J. Berry, *Manual on Radiation Dosimetry*, 45(1970), Marcell Dekker, Inc. New York.
- 14) R. Tanaka, et al., *JAERI-M 5608*(1974).