

《Original》 Gamma-ray Dosimetry with Thin Plastic Film

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Abstract

Thirty two different kinds of domestic plastic films for use in measuring high gamma-ray dose have been collected and their dosimetric characteristics investigated with the help of a Co-60 gamma radiation source.

Among them a rigid polyvinyl chloride(PVC) film of 0.06mm in thickness which is manufactured by Lucky Chemical Co., Korea, seem to be the most suitable one for this purpose. The relation between optical density at 3100Å and radiation exposure in this PVC film was linear in the range of $0.6 \times 10^6 R$ to $1.3 \times 10^7 R$, and also the film showed a good reproducibility within 9% under the standard experimental condition. The effect of absorbed dose, oxygen content of surrounding atmosphere and irradiation temperature have also been studied for this film. It appeared to have a good property in the dosimetric point of view.

요 약

고준위 gamma 선량 측정에 사용할 목적으로 시중에서 판매되고 있는 32종의 국산 plastic film 을 수집하여 gamma 선량계로서의 효용성 판단기준이 되는 gamma 선에 대한 반응도 및 온도효과등을 조사하였다.

Co-60 gamma 선을 이용한 실험결과에 의하면 럭키화학회사 제품인 0.06mm 두께의 경질 PVC film 이 선량계로서 사용하기에 가장 적합한것 같았다. 3100Å 파장에서 측정된 이 PVC film 의 optical density 는 $0.6 \times 10^6 R$ 으로부터 $1.3 \times 10^7 R$ 까지의 구간에서 조사선량에 따라 비례하여 변했으며 표준실험조건하에서 9%이내의 재현성을 보여 주었다.

한편 흡수선량, 주위 대기중의 산소함량 및 방사선 조사시의 온도효과등을 조사한 결과 실용적인 관점에서 가장 좋은 특성을 나타냈다.

1. Introduction

The use of transparent plastic film for radiation dosimetry was proposed by Day¹⁾ and by Charelesby²⁾. At the present time, polyethylene³⁻⁶⁾, polyvinyl chloride⁷⁻⁸⁾, poly-

styrene⁹⁾, and some others¹⁰⁻¹⁴⁾ are well known materials usable in dosimetry. Various colors such as grey, green, or brown appear depending on the origin of film, and the developed color intensity is further dependent on the dose intensity. The intensity of color

is directly proportional to the absorbed dose in some range, usually in high-level doses. Even though no color appears in some plastics, there are ultraviolet absorptions. Therefore, it can be used for high-level dosimetry.

When the plastic films are irradiated to radiation, inherent properties such as external appearance, chemical state, physical state, and mechanical properties are usually changed. Chemical changes by irradiation include double-bond formation, dehydrochlorination, crosslinking, oxidative degradation, polymerization, depolymerization, and gas evolution¹⁵⁾. However, response of PVC film by irradiation is very different from each other in accordance with the component of film depending on its origin.

The PVC film is not sold commercially as the pure polyvinyl chloride, and usually a small percentage of monomer, plasticizer, stabilizer, etc. in the film remains. It is apparent that the radiation effect is strongly dependent on the amount of additives present. It is said that the monomer content is the most essential factor influencing the radiation response of plastic film¹⁶⁾. In commercial film the monomer content is said to be usually less than 0.1% and seldom above 0.5%.

The present paper deals with more detailed studies of plastic dosimetry using the domestic PVC film. Its dosimetric characteristics such

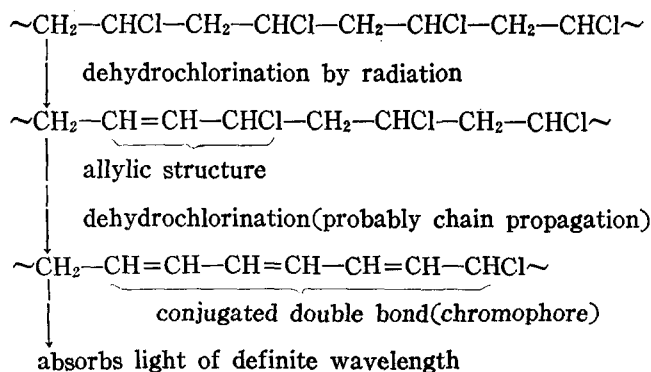
as radiation response, and temperature and oxygen effects have been investigated. The samples under investigation were exposed to Co-60 gamma-rays. Also a study has been carried out to look for well-standardized experimental condition for high-level gamma-ray dosimetry.

2. Principle of the Plastic Dosimeter

The mechanisms of the radiation-induced optical density in plastic materials are very difficult to be described in brief. The reasons¹⁶⁾ are; (1) the additives of the plastics are unknown, (2) there are a number of possible radiation interactions, and (3) there are a multitude of possible resultant chemical reactions. Nevertheless, it is interesting to note that irradiation always causes some optical darkening in transparent plastics.

When PVC film is irradiated by radiation dehydrohalogenation reaction occurs. The resulting allylic structure has especially high reactivity, and thus dehydrohalogenation will be propagated to some extent bringing about conjugated double bond which is typical chromophore. Consequently, the radiation dosimetry is possible since the chromophore absorbs light of definite wavelength. The absorption intensity or coloration is a function of total dose only, but is independent of dose rate or source of radiation.

PVC film;



3. Experimental

(1) Selection of Material

Thirty two different kinds of domestic plastic films of 0.015~0.1mm in thickness have been collected to determine their response to gamma-radiation from a Co-60 source. A variety of chemically different plastic films is available in Korea. They are usually known as polyethylene, polyvinyl chloride, polypropylene, and so on.

The method of selection was based on the changes of optical density by radiation, which was measured by the use of spectrophotometer. Thus, optical density spectra were first obtained for each unirradiated sample in the visible and ultraviolet regions. This is considered background. The approximate wavelength covered was from 2000Å to 5000Å. Thereafter, these plastic thin films were exposed to Co-60 gamma-rays. The optical density spectra were again measured for the irradiated samples in the same way as described above, and the changes of optical density were compared with those of the unirradiated ones. Of the materials surveyed, rigid PVC film from Lucky Chemical Co., Korea was selected as a material suitable for dosimetry because it showed a good linearity between the optical density and the exposure.

We used different thicknesses of the film, but the results given here concern with only one of the batch of 0.06mm in thickness. Since the optical density is proportional to the thickness of the film, it is important to select film of uniform thickness. The uniformity of the thickness of film used in the present work is approximately $\pm 30\%$. Rigid PVC film is a homopolymer of vinyl chloride, and the name and amounts of stabilizer, plasticizer, and other additives are unknown.

Some exploratory works were also done with polyethylene films, but these appeared

to be less suitable than PVC film and were abandoned.

(2) Optical Density Measurement

The PVC samples were usually cut to a standard size of $3.5 \times 1.2 \text{ cm}^2$ so as to be firmly held into the specially made sample holder of the spectrophotometer. Before and after irradiation the optical density of each sample was measured relative to a control sample of the same size chopped from the same sheet batch. For these measurements a Beckman Model DU-2 spectrophotometer was used.

The PVC films were handled with tweezers and washed in alcohol and dried before use. In each measurement of optical density they were cleaned with brush to remove dust and finger grease from them.

(3) Irradiation Method

Irradiations of samples at dose rates equal to or less than $2.2 \times 10^6 \text{ R/hr}$ and $1.5 \times 10^5 \text{ R/hr}$ were carried out in the Shopboard Irradiator of BNL and Co-60 Gamma-ray Irradiation Facility. The former has been calibrated with a Fricke dosimeter, while the latter with a Victoreen Integrating Ratemeter. The exposure rate which is determined in such ways is known with the accuracy of $\pm 5\%$. In these facilities the samples were placed at a position of a given dose rate and the irradiation time was adjusted in order to give the desirable dosage. The PVC films were packed with polyethylene film to ensure against dust contaminations and held up by the aluminum supporter for irradiation.

The transparent PVC film absorbs ultraviolet rays even before exposure to radiation. This background of optical density must be subtracted from that after irradiation. Since it is difficult to achieve constant value for the background of PVC film, the optical density of each sample should be measured in each run.

4. Results and Discussion

(1) Absorption Spectra of PVC Film

PVC film of 0.06mm in thickness is transparent at the visible region of spectrum and near the ultraviolet, but its absorption increases rapidly at wavelengths below 3000Å. As shown

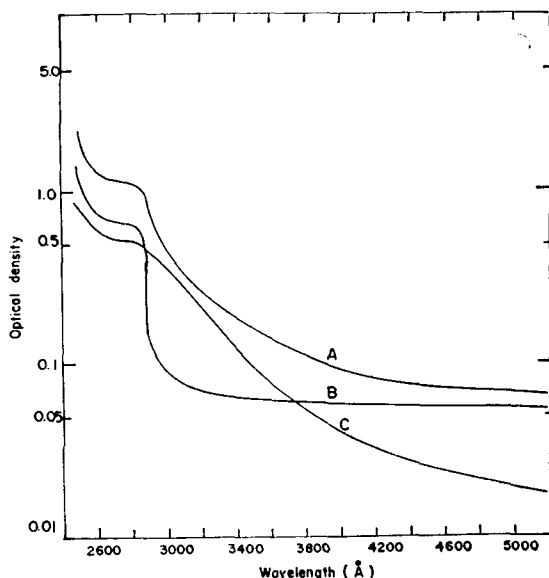


Fig. 1. Ultraviolet absorption of PVC film from Lucky Chemical Co:

A, irradiated; B, unirradiated; C, difference between A and B

in Fig. 1, there is a plateau near the wavelengths of 2800Å, and then the optical density suddenly increases with decreasing wavelength. The typical absorption curves of unirradiated PVC film(B) and of the irradiated film(A) show the changes in optical density as a function of wavelength. The curve C is obtained by subtracting B from A.

The absorption induced by irradiation does not increase simply toward shorter wavelengths, and the greatest change in absorption occurs around 2800Å. However, in practice, one must realize that the natural absorption of PVC film increases steeply as the wavelength of light is decreased. If increase in absorption is to be used as a measure of radiation dose, it is convenient to take the optical density at the wavelength corresponding to the peak of the curve as a measure of this induced absorption. This will not only give maximum effect for a given dose but also minimize the effect of wavelength or slit-width errors. In this region, the measured optical density was found to be slightly greater than 1.0, and at this optical density the resolution of the spectrophotometer was so poor that we could

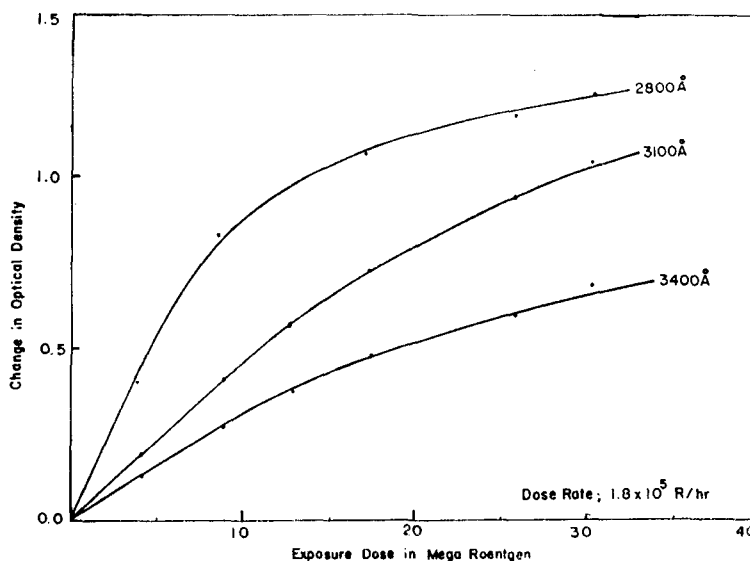


Fig. 2. Linearity of induced optical density of PVC film (0.06mm thick) from Lucky Chemical Co

not apply to the radiation measurements.

As shown in Fig. 2, the 2800Å curve does not show the linearity at about 6×10^6 R, and there is a large fluctuation in optical density. The 3100Å curve is linear up to about 1.3×10^7 R, and that of the 3400Å up to about 10^7 R. With the 3100Å curve, dose estimation can roughly be made up to about 3×10^7 R by reading from the nonlinear portion of the curve. The most suitable wavelength for the PVC film of 0.06mm in thickness seemed to be 3100Å.

(2) Optical Density Variation of Irradiated PVC Film

The optical density produced in PVC film by irradiation was not stable since like many other plastics it was continuously increased. It was measured immediately after irradiation. A change of optical densities in the film irradiated was repeatedly measured for a

period of one week. The optical densities were reached to saturation after 50~100 hours, as can be seen in Fig. 3, and after that there was no considerable change. The curve representing the lower doses becomes nearly flat sooner than those representing the higher doses. This phenomenon is expected since an increase in exposure results in increased damage to the PVC film. The increase in optical density was observed within the first 40 hours, and then reached to the final value for all the samples after sufficient time had elapsed. The maximum increasing rate in optical density at 3100Å was about 6.0~12.0% depending on the initial optical density.

For calibration, the value of the optical density measured about two hours after irradiation has been used. To reduce errors in measurements of the optical density, a delay time for color development must be allowed after exposure. One should consider for such

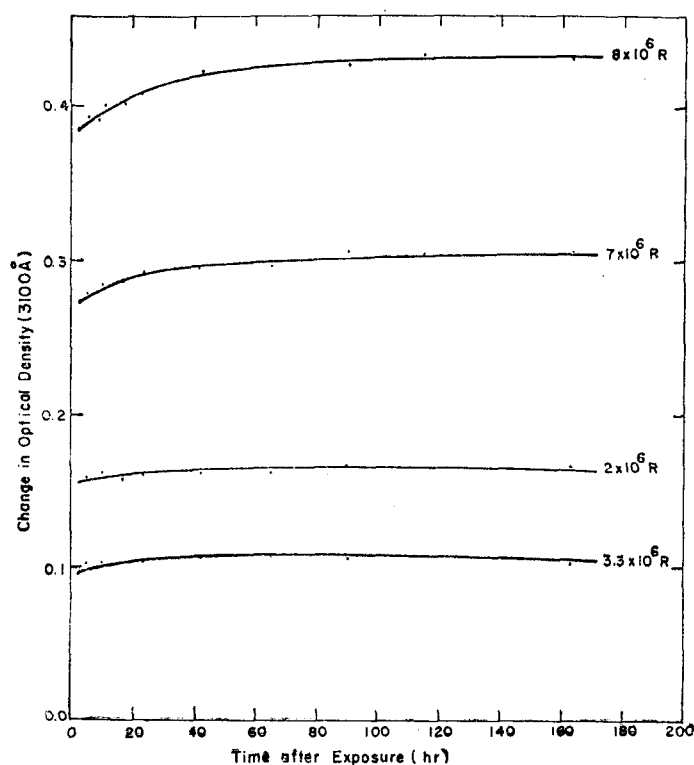


Fig. 3. Post-irradiation density variation in PVC film from Lucky Chemical Co for given dosages

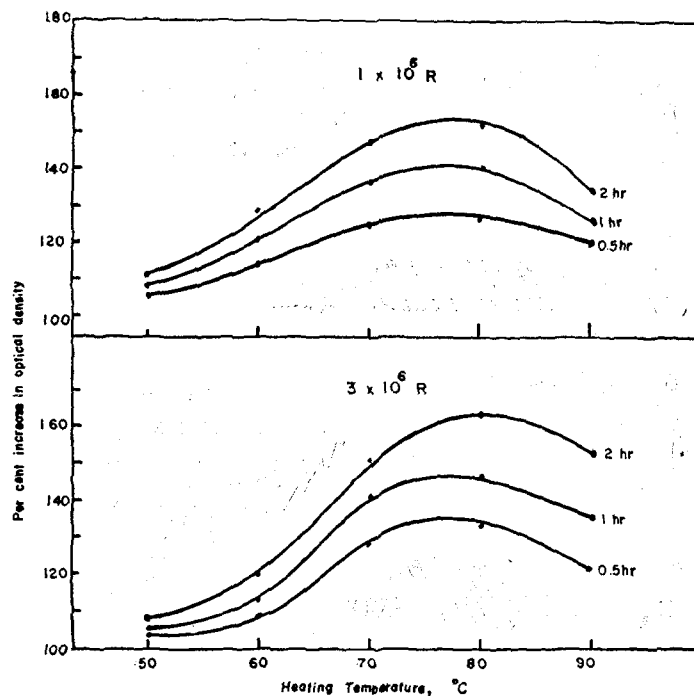


Fig. 4. Influence of given heat treatments on the optical density of irradiated PVC film

post-effects when starting with a new sheet of PVC film.

(3) The Effects of Heat Treatment

To stabilize the optical density after irradiation, and to shorten the time needed for saturation of the optical density, heat treatments suggested by Artandi and Stonehill⁷⁾ had been applied. The optical density of PVC film at room temperature reached to the maximum in 2 to 4 days. The optical density increase can be accelerated by heating. The color of PVC film was changed from light brown to brown depending on heating periods. The optical density growth due to the heat treatment was examined as functions of parameters such as heating period and heating temperature.

The color development was examined by heating samples after irradiation ($1 \times 10^6 R$ and $3 \times 10^6 R$) in the range of temperature from 50°C to 90°C. For heat treatment, we used an ordinary drying oven which is installed

with a thermometer and a system for forced air circulation by electrical motor fan. The temperature was controlled with the accuracy of $\pm 1.2^\circ C$. The optical density measurements of PVC film were carried out after heating for a period of 30, 60, and 120 min. As shown in Fig. 4, the effect appeared to be maximum at 80°C. Heat treatment at 80°C is to be suitable for routine work.

The intensity of color developed by heat increases with temperature up to 80°C. Above this temperature it decreases gradually. This reduced intensity may be due to reactions of trapped radicals with chromophore grouping⁷⁾. In order to determine the optimum time required at 80°C for making the optical density develop sufficiently, a variation of the optical densities was observed with changing the period of the heat treatment. It rises rapidly during the first 30 min as shown in Fig. 5, and then slowly increases until 120 min. It was found that the variation of optical density with heat treatment is also dependent on the

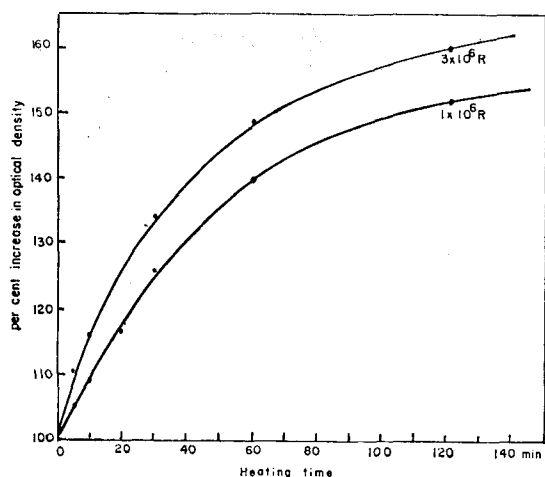


Fig. 5. Optical density increase for different heating periods at 80°C

radiation dose. The increasing ratios of optical densities by heat treatment are much higher for the film exposed to higher doses than those exposed to lower doses. As no maximum intensity of the optical density reaches within a suitable time, determination of the optimum heating time is a matter of choice to some extent. We have chosen 30 min as a reasonable time for routine work because of fading characteristics after irradiation of PVC film, as will be seen later.

(4) The Fading Effect after Heat Treatment

When the irradiated samples were stored at

room temperature without heat treatment, the optical density increased gradually for a few days. However, when these samples were stored at higher temperature, color development proceeded more rapidly and full intensity was completed after sufficient heat treatment. The samples stored at room temperature showed a decrease in optical density depending on the heating periods. The decreases are more predominant in case of shorter heating period as shown in Fig. 6. The results given in Fig. 6 are for the sample exposed to radiation dose as high as $3 \times 10^6 R$, and a similar behaviour is noted for other doses.

It is evident that the optical density of the film treated with heat for a period of 30 min or more at 80°C is practically stable for at least 10 days. Significant fading appeared when the sample was exposed to lower dose or its heat treatment was insufficient. However, the decrease in optical density did not exceed above 15% during the given period. In view of the practical application it may be advantageous to adopt the heat treatment technique. According to the present work, the heat treatment for 30 min at 80°C gave no fading effect.

(5) The Influence of the Irradiation Temperature and of the Oxygen Content of the Surrounding Atmosphere

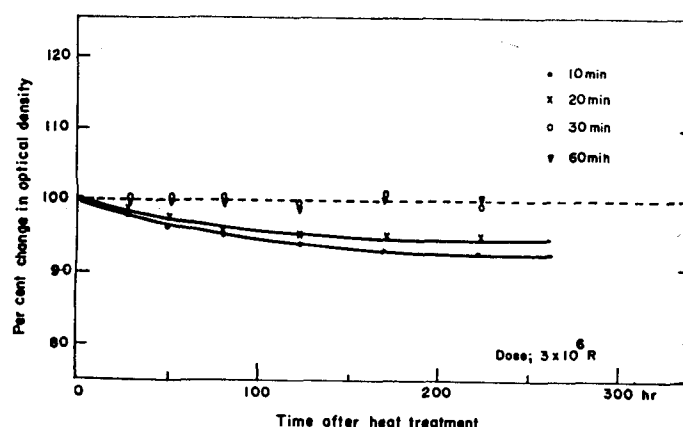


Fig. 6. Fading effect after given heating periods at 80°C

The optical density increase by heat treatment of irradiated PVC slides indicates that it has been influenced strongly by the irradiation temperature. According to Miller¹⁷⁾, the HCl-yield in the irradiated PVC powder is dependent on the irradiation temperature.

We studied the influence of this factor on the optical density by irradiating PVC film at a given temperature. The PVC sample was heated during irradiation in the ordinary dry oven in the temperature range 20°C~90°C. The results are shown in Fig. 7. Although the higher temperature over 40°C in routine work is not expectable, it will be a reference for

understanding on the effects of surrounding temperature of PVC slides when irradiation is made. The rate of variation of the optical density on the part of lower temperature is not serious compared with other additional errors. It is clear that the temperature influence cannot be disregarded, and a correction must be made if the PVC dosimeter is to be used at a given temperature.

The effects of oxygen content of the surrounding atmosphere have also been studied. It is found that the oxygen can play an essential role in a reaction mechanism when plastics are exposed to radiation. We have examined at different circumstances of irradiation procedures using a glass joint where the PVC slides were put in pure nitrogen, in pure oxygen, and in air. The results indicated in Fig. 8 show that the presence of oxygen hastens the increase of optical density of the PVC film. When the optical density in air was normalized, the values in nitrogen and in oxygen were found to be 140% and 76.8%, respectively, but it does not seem to be important for routine work where irradiations are made in air.

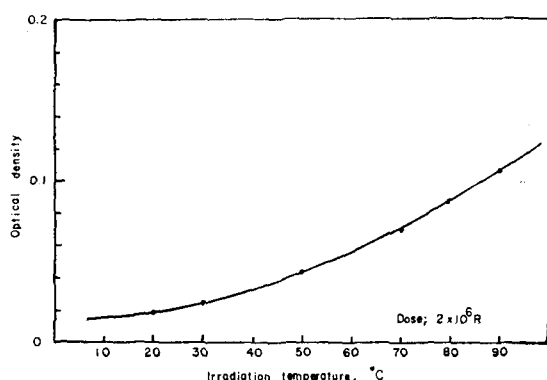


Fig. 7. Influence of irradiation temperature on the optical density of irradiated PVC film

(6) Dose Measurement

In dose determination by using PVC film as

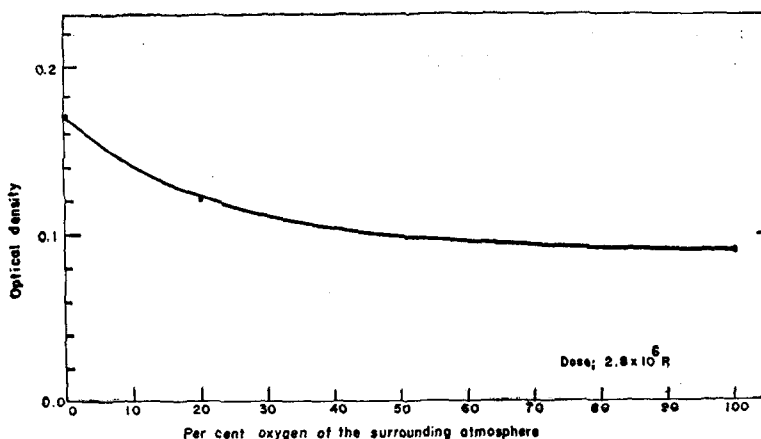


Fig. 8. Influence of the oxygen content of the surrounding atmosphere on the optical density of PVC film

a dosimeter, it is very important that measurements of the irradiated PVC slides should be performed under the standard experimental condition that is defined as 3100Å in measuring wavelength by the spectrophotometer and a duration of 30min at 80°C in heat treatment here in this work. Otherwise, a correction must be made in case of different circumstances. A calibration curve was obtained at 3100Å in wavelength after heating the PVC films exposed to Co-60 gamma-rays for 30 min at 80°C

The usefulness of PVC film as a dosimeter is shown in Fig. 9. The radiation-induced

optical density measured at 3100Å after heat treatment, when plotting against exposure on linear graph paper, gives straight line over the range from $6 \times 10^5 \text{R}$ to $1.3 \times 10^7 \text{R}$. The relation between dose and optical density in the straight line can be expressed by the formula

$$R = 21.530 \Delta OD + 0.064$$

where ΔOD is the change in optical density and R is the exposure in mega roentgen.

It is seen that the linearity of the calibration curve ceases at about $1.3 \times 10^7 \text{R}$ although it is still usable up to $3 \times 10^7 \text{R}$. Each point in Fig. 9 is an average of ten different samples

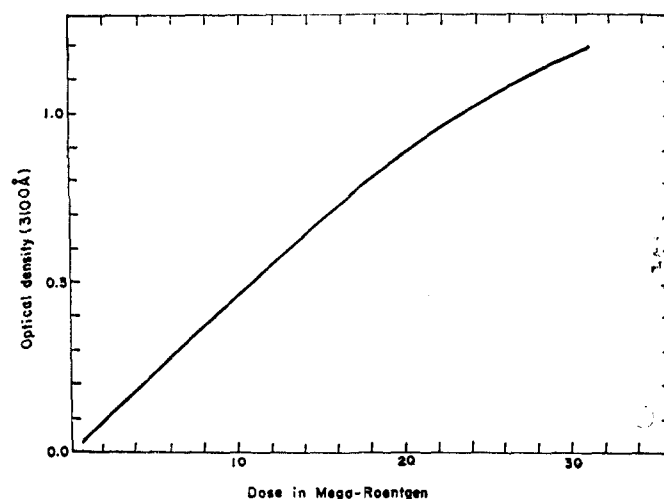


Fig. 9. Typical calibration curve for PVC film(0.06mm thick) from Lucky Chemical Co

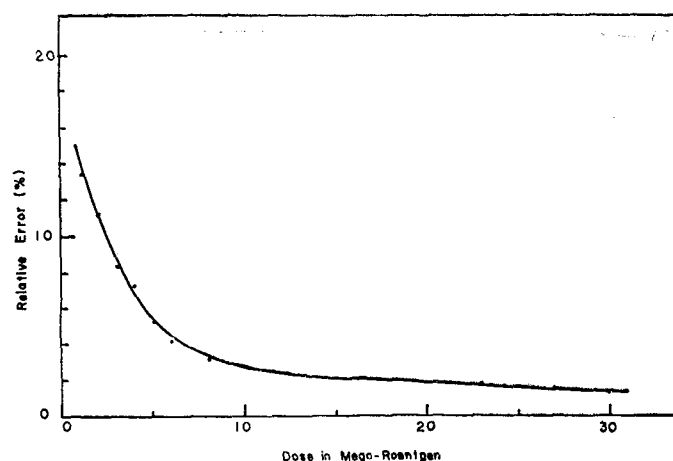


Fig. 10. Statistical errors estimated from irradiations of 10 samples in each point in PVC film

exposed to the same dose. To extend the upper limit of measurable dose thinner plastic film is preferably used, because the optical density is directly proportional to film thickness. In addition, the measurement in the longer wavelength is further desirable. By the way, the upper limit of measurable range of dose can be extended, on the contrary, the lower detectable limits are shortened.

It is important to keep the same batch of film throughout the experiments and to treat the sample under the standard experimental condition. Otherwise the accurate dose determination can not be made. Even though the composition differences within the same batch are negligible, it is always necessary to work out again a standard curve when different batch of film is to be used.

The standard error inherent in the method itself has been determined experimentally at several dose levels. The standard deviations derived from ten or less samples in each point for a given dose are indicated in Fig. 10. In the measurable range of dose the greatest percentage error in optical density is about 15% at low dose and in the case of higher doses the results are very satisfactory. Moreover, the reproducibility of this dosimeter is better than 9% although it is varied depending on the exposed dose.

It also appears that the PVC dosimeter is independent of the dose rate in a range from 1.8×10^5 R/hr to 10^6 R/hr under consideration in this study. On the other hand, there are important errors caused by a contamination, *i. e.*, dust and finger prints. It was observed that the dust particles on the film bring about a little increase in optical density, and this problem was eliminated by brushing the samples. Finger prints on the film also showed a little effect on the optical density. The PVC samples were, therefore, always washed with

a detergent and dried before any optical density measurements were made.

5. Conclusions

Rigid PVC film of 0.06mm in thickness can be used as a dosimeter in the range from 0.6×10^6 R to 3.6×10^7 R with the accuracy of 10%. The calibration curve in this case is linear up to about 1.3×10^7 R. The PVC dosimeter is dose rate independent and the reproducibility is better than 9%. It appears in PVC film that a post-irradiation has influence on increasing optical density. The time needed for this process to reach saturation was of the order of 100 hours for dose of 8×10^6 R, and for small doses the time is reduced.

Since the calibration curve is made under the condition of heat treatment for 30 min at 80°C and the measurement of optical density at 3100\AA , every dosimetrical measurement should meet this standardized condition.

Domestic PVC film has proved to be a useful dosimeter. In addition, the PVC dosimeter allows very small pieces to be placed throughout the radiation field and a measurement may be done simultaneously at a large number of points.

Summing up the above, it can be concluded that the PVC film dosimeter is very suitable for high level gamma-ray dosimetry.

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