Evaluation of polycarbonate films as detection materials for high-dose electron-beam radiation detection

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Abstract

In this study, the dosimetric characteristics (thickness applicability, preheating time, temperature and hu midity dependence, in-batch uniformity, readout reproducibility, dose linearity, self-decay, and electron energy response) of engineered polycarbonate films irradiated with an electron beam ($0--600$kGy) were investigated using photoluminescence spectroscopy. The results show a linear relationship between photoluminescence in tensity and radiation dose when the thickness of the polycarbonate film is 0.3mm. A higher fluorescence intensity can be obtained by preheating at $60\circ$C for 180min before photoluminescence-spectrum analysis. As the temperature during spectral testing and the ambient humidity (during and after irradiation) increased, the photoluminescence intensity of the polycarbonate films decreased. The photoluminescence-intensity deviation of the polycarbonate films produced within the same batch at 100kGy is 2.73%. After ten times of repeated excitations and readouts, the coefficients of variation in photoluminescence intensity are less than 8.6%, and the linear correlation coefficient between photoluminescence intensity and irradiation dose is 0.965 in the dose capture range of $20 - 600$kGy. Within 60 days of irradiation, the photoluminescence intensity of the polycar bonate film decreased to 60% of the initial value. The response of the 0.3mm polycarbonate films to electron beams with energies exceeding 3.5MeV does not differ significantly. This comprehensive analysis indicates the potential of polycarbonate films as a high-radiation dose detection material.

Full Text

Preamble

Evaluation of Polycarbonate Films as Detection Materials for High-Dose Electron-Beam Radiation Detection

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In this study, the dosimetric characteristics of engineered polycarbonate films irradiated with electron beams (0–600 kGy) were investigated using photoluminescence spectroscopy. The evaluated parameters included thickness applicability, preheating time, temperature and humidity dependence, in-batch uniformity, readout reproducibility, dose linearity, self-decay, and electron energy response. The results demonstrate a linear relationship between photoluminescence intensity and radiation dose for polycarbonate films with a thickness of 0.3 mm. Higher fluorescence intensity can be obtained by preheating at 60 °C for 180 min before photoluminescence-spectrum analysis. The photoluminescence intensity of the polycarbonate films decreased as the temperature during spectral testing and the ambient humidity (during and after irradiation) increased. The photoluminescence-intensity deviation of polycarbonate films produced within the same batch at 100 kGy was 2.73%. After ten repeated excitations and readouts, the coefficients of variation in photoluminescence intensity were less than 8.6%, and the linear correlation coefficient between photoluminescence intensity and irradiation dose was 0.965 in the dose capture range of 20–600 kGy. Within 60 days of irradiation, the photoluminescence intensity of the polycarbonate film decreased to 60% of the initial value. The response of the 0.3 mm polycarbonate films to electron beams with energies exceeding 3.5 MeV did not differ significantly. This comprehensive analysis indicates the potential of polycarbonate films as a high-radiation-dose detection material.

Keywords: Electron-beam irradiation, Polycarbonate, Dose detection, Radio-photoluminescence, Dosimetric characteristics

Introduction

In recent years, several types of thin-film polymer materials—including Gafchromic [**?**], clear Perspex [**?**], cellulose triacetate [**?**], and Sunna [**?**]—have been listed as conventional radiation dosimeters according to the ISO/ASTM 51261 (2002) standard. Compared with liquid or other types of dosimeters, organic-film dosimeters feature low economic cost, stable fluorescence signal response, wide range of sizes, high detection dose limit, and broad dose detection range [**?**, **?**]. They demonstrate significant potential in medical treatments, radiation processing control (such as food preservation and industrial material modification), nuclear track detection and visualization, dose monitoring at radiation sites, and nuclear decommissioning facilities \cite{5,7–9}.

However, the aforementioned film dosimeters present certain disadvantages in actual dose-detection processes, including low environmental adaptability, complex production processes, easy dose saturation, and short shelf life. Currently,

studies pertaining to organic-film dosimeters focus primarily on two aspects. The first involves investigating new polymer films with favorable dose responses as alternative materials, such as the development of new organic–inorganic composite materials. However, composite material processing involves issues of low material compatibility, complex manufacturing processes, unsatisfactory dose-response linearity, low durability, poor long-term signal stability, rapid aging, low environmental adaptability, and high cost [**?**, **?**]. The second aspect involves further analysis of the dosimetry characteristics of commercial or potential dosimeter materials, investigation into their applicable dosimetry scenarios, and proposal of schemes for structure optimization and performance improvement [**?**, **?**].

Polycarbonate (PC) is a thermoplastic engineering plastic known for its excellent light transmission, impact resistance, ultraviolet-radiation resistance, high mechanical strength, and ease of processing [**?**]. It is widely utilized in industrial manufacturing, space science, electronic instruments, and medical devices [**?**]. Further investigations into the radiation effects of PC revealed that the aromatic ring structure in PC exhibited greater susceptibility to energy absorption and deposition compared with aliphatic structures [**?**]. Irradiation can induce disorder in the internal structure of PC, resulting in intermolecular crosslinking (at 30 kGy), chain scission (at 200 kGy), and free-radical generation \cite{16–18}. Subsequent studies on radiation modification of PC revealed its radiophotoluminescence properties [**?**, **?**] and suggested PC as a promising material for radiation-dose detection [**?**, **?**]. During the initial period, relevant investigations on radiation-dose detection of PC films focused primarily on analyzing the measurable dose range, photoluminescence (PL) intensity, optimal excitation wavelength, fluorescence lifetime, optical quantum yield, light absorption, and transmission. However, few studies have investigated the dosimetry characteristics of PC films; consequently, PC films were primarily used for fluorescence-tracking detection in the early stages rather than as dosimeters.

The dosimetric characteristics of PC films must be investigated before they can be used for practical dose detection [**?**]. These include considerations such as thickness applicability, preheating time, temperature and humidity dependence, dose-rate response, dose linearity, in-batch uniformity, readout reproducibility, annealing, self-decay, and energy response. Soliman et al. investigated the PL spectra of different types of Makrofol PC materials irradiated with 60Co γ-rays (300 kGy) [**?**]. The findings revealed distinct PL characteristics for each film, enabling application of different PC film types in diverse scenarios based on their unique properties. Abdul-Kader et al. investigated a Makrofol LT 6-4 PC film irradiated with 60Co γ-rays and observed a decrease in PL intensity with increasing irradiation dose within the range of 150–950 kGy, with a linear correlation coefficient of 0.96 [**?**]. Posavec et al. reported that the PL intensity of irradiated PC films decreased with increasing temperature, and the ratio of PL peak intensity at 77 K to 293 K ($I_{77}K/I_{193}K$) was 1.93, demonstrating clear temperature dependence during testing [**?**]. Galante et al. discussed commercial PC detection films employed for monitoring radiation fields inside cylindrical

container products, reporting a correlation coefficient of 0.99 between absorbed dose and response value, indicating favorable linear dose response [**?**]. Resta et al. irradiated Makrofol-KG PC films with 28Si$^+$ ions at 0.5, 1, and 2 MeV and discovered that PL intensity varied by two orders of magnitude at the same radiation dose of 1 and 2 MeV 28Si$^+$ ions, highlighting significant differences in energy response [**?**]. However, reports regarding the effects of PC thickness adaptability, preheating time before PL testing, and ambient humidity on dose detection—all of which can affect detection accuracy—are scarce. These factors have been considered in studies involving other dosimeter types. Kattan et al. observed that polyvinyl-chloride films of varying thicknesses irradiated with 0–125 kGy γ-rays exhibited different degrees of radiation sensitivity [**?**]. Bhata et al. investigated a 250 µm diethyl terephthalate film (Gar film-EM) irradiated with equal doses of 60Co γ-rays under various relative humidity (RH) conditions and discovered that net absorbance remained stable after 14 days, except for a rapid decline on the first day, indicating that dose response was affected by humidity during irradiation [**?**]. Mejri et al. discussed a commercial inorganic glass dose tablet exposed to different RH environments after γ-irradiation; the net absorbance showed a distinct trend of reverse proportional decline within 22 days, indicating significant humidity response during post-irradiation storage [**?**]. In summary, only a few dosimetric characteristics have been briefly mentioned in existing studies pertaining to detection performance of various PC film types, and no comprehensive discussion of a specific PC film has been provided.

In this study, PL spectroscopy was employed to investigate the dosimetric characteristics of engineered PC films after electron beam (EB) irradiation (0–600 kGy). The effects of PC-film thickness, preheating time after irradiation, temperature during PL spectrum testing, ambient humidity during irradiation, and ambient humidity of the PC storage environment after irradiation on PL spectral intensity were investigated. Additionally, in-batch uniformity, readout reproducibility, dose linearity, PL-intensity attenuation characteristics after irradiation, and PC response to different EB energies were evaluated. This study aims to provide detailed data indicators and analysis results for each dosimetric characteristic, comprehensively evaluate the feasibility of using PC films as dosimeters in practical radiation detection, and provide references for dosimetry studies involving other PC materials.

II. Materials and Methods

A. Sample Preparation

The PC film used in this study was fabricated by Covestro Polymer Co., Ltd., Shanghai, China. It is a nontoxic, odorless, colorless, and transparent glassy amorphous organic polymer [**?**]. The film exhibited a density of 1.25 g/cm^3, light transmittance of 90% $\pm$ 1%, refractive index of 1.585 $\pm$ 0.001, linear expansion rate of 3.8 $\times$ 10^{-5} cm/°C, and thermal deformation temperature of 140 °C. PC films of various thicknesses (0.2, 0.3, 0.5, 0.8, 1.0, 1.5, and 2.0 mm) were prepared. The surface of the PC film was flat and the thickness uniform. The

films were cut into 20 mm × 20 mm squares, and protective layers were removed from both sides for further use.

B. EB Irradiation

Electron accelerators with different energies (3.5, 4, 4.5, 10, and 20 MeV) were used for PC irradiation in this study. A DD-type electron accelerator (Jinwo Technology Co., Ltd., CGN, China) operating in an energy range of 3.5–5 MeV and beam power range of 30–200 kW was used. Additionally, an IS1020 back-wave electron linear accelerator (Xianghua Huada Biotechnology Co., Ltd., Hunan, China) operating at 10 MeV and rated beam power of 20 kW was employed. The IS1020 high-energy electron accelerator (Huada Biotechnology Co., Ltd., Guangzhou, China) was also operated at 20 MeV with maximum beam power of 20 kW. The PC film was placed on a conveyor belt and passed through the central area of the irradiation window multiple times to receive the required dose, with 10 kGy per pass. Irradiation was performed at room temperature, and radiation dose was traced using dichromate. Samples were uniformly irradiated using an EB with dose deviation of 1%.

C. PL Spectrum

An FLS1000 steady-state/transient fluorescence spectrometer (including conventional and variable temperature types) manufactured by Edinburgh Instruments, UK, was used to analyze the PL spectra of irradiated PCs. The conventional type operated under the following parameters: excitation-spectrum scanning wavelength range, 250–450 nm; emission-spectrum scanning wavelength range, 340–800 nm; scanning rate, 5°/min; step size, 1 nm; and integration time, 0.2 s. A xenon lamp served as the excitation source, and tests were performed at ambient temperature. The variable-temperature type featured a built-in Oxford Optistat thermostat with temperature range of 273–373 K and insulation accuracy of ± 1 K. The emission-spectrum scanning wavelength range was 395–800 nm, and other parameters were set based on the conventional type. The irradiated PC film was placed at the same position on the PL spectrum test platform to ensure consistency of the PL measurement path.

D. Analysis of Dosimetric Characteristics

1. Thickness Applicability A typical feature of thin-film dosimeters is availability at different thicknesses [**?**]. To evaluate the response of PC films with different thicknesses to EB irradiation, seven groups of PC films (0.2, 0.3, 0.5, 0.8, 1.0, 1.5, and 2.0 mm) were prepared. Each group was irradiated with different doses (0, 100, 300, and 600 kGy) under a 10 MeV EB at ambient temperature in air atmosphere. Subsequently, PL intensity was analyzed, and the relationship between PL peak intensity and irradiation dose for PC films of different thicknesses was obtained through PL spectral testing at the same ambient temperature.

2. Preheating Time   Generally, the non-luminous center effect occurs in irradiated polymer films. When irradiated PC film is preheated prior to PL testing, its PL intensity changes accordingly. Therefore, optimal preheating conditions must be determined to achieve higher PL intensity values. In this study, eight groups of 0.3 mm PC films (three parallel samples per group) were irradiated at 100 kGy by a 10 MeV EB at ambient temperature in air atmosphere, then preheated at 60 °C for 10, 30, 40, 60, 90, 120, 180, and 300 min separately. Finally, PL peak intensities of PC films preheated for different durations were measured.

3. Temperature Dependence   Temperature dependence refers to the effect of ambient temperature on PL intensity of PC film during PL-spectrum testing [**?**]. To investigate this relationship, 0.3 mm PC films were irradiated at 100 kGy using a 10 MeV EB, and three parallel samples were prepared. PL tests were conducted at eight temperature points: 273, 278, 283, 293, 303, 323, 343, and 373 K. The variation in PL intensity with ambient temperature during PL testing was analyzed.

4. Humidity Dependence   Environmental humidity affects the dose response of polymer films. Therefore, the effect of relative humidity (RH) in air (both during irradiation and post-irradiation storage) on PL intensity of PC film was evaluated [**?, ?**]. To establish different RH gradients in the range of 12.4–97.2%, various saturated salt solutions were prepared using techniques reported by Wexler and Hasegawa (1954) and Levine (1979). Table 1 lists the selected saturated salt solution types and their corresponding RH values under sealed conditions. The corresponding saturated salt solution (25 ml) was added to a borosilicate glass bottle to establish different humidity gradients.

Table 1.   Different types of saturated salt solutions and the corresponding humidity.

Salt solution type	$LiCl \cdot$ H_2O	$MgCl_2 \cdot$ $6H_2O$	$Mg(NO_3)_2 \cdot$ $6H_2O$	K_2SO_4
Relative humidity (%)	12.4	33.6	54.9	75.5

In the study of humidity dependence during irradiation, 0.3 mm PC film was suspended in sealed glass bottles under different humidity environments (12.4, 33.6, 54.9, 75.5, and 97.2%) and stored at room temperature for seven days to equilibrate ambient humidity. Subsequently, the glass bottles were irradiated at 100 kGy using a 10 MeV EB, after which the five groups of PC films were subjected to PL tests. In the study of humidity dependence during post-irradiation storage, each group of PC films was first irradiated using a 10 MeV EB (100 kGy) and then encapsulated in bottles under different humidity conditions for

seven days under environmental regulation. Finally, PL tests were performed and the effect of humidity on PL intensity of PC films was analyzed.

5. In-Batch Uniformity   In-batch uniformity refers to PL-intensity uniformity of PC films produced in the same batch under the same irradiation dose. It is a key index for describing dosimetric characteristics of RPL materials [**?**]. Three groups of 0.3 mm PC films (15 parallel samples per group) were irradiated (10 MeV EB) at 100, 300, and 600 kGy separately. Irradiation treatment was performed at ambient temperature in air atmosphere. The PL peak intensity of each sample was measured. Relative average deviation (RAD), expressed in Eq. (1), was used to evaluate uniformity degree.

$$\text{RAD} = \frac{\sum_{i=1}^{n} |x_i - \bar{x}|}{n \times \bar{x}} \times 100\%,$$

where x_i denotes the measured value, $\bar{x}$ the average value, and n the number of parallel samples in the same dose group. After recording PL peak intensities of parallel samples in each dose group, the RAD value was obtained to analyze in-batch uniformity of irradiated PC film.

6. Readout Reproducibility   To investigate whether multiple PL tests affect dose information stored in PC film, three groups of 0.3 mm PC films were irradiated (10 MeV EB) at 100, 300, and 600 kGy separately. Irradiation treatment was performed at ambient temperature in air atmosphere. Each film was excited repeatedly and measured 10 times to obtain PL peak intensity. To assess PL intensity dispersion, coefficient of variation was estimated using Eq. (2).

$$C_v = \frac{\sigma}{\bar{x}} \times 100\%,$$

where C_v is the coefficient of variation, σ the standard deviation of the 10 measurements, and $\bar{x}$ the average value. Obtaining the standard deviation and coefficient of variation allowed evaluation of readout reproducibility of PC films.

7. Dose Linearity   Dose linearity, which refers to the linear relationship between measured signal intensity and radiation dose, is a crucial parameter in dose detection [**?**]. In this study, 0.3 mm PC films were irradiated with different doses (0, 20, 50, 100, 200, 400, and 600 kGy) using a 10 MeV EB at ambient temperature in air atmosphere. The relationship between irradiation dose and PL intensity after irradiation was analyzed via PL-spectrum tests, and the dose capture range of PC film was determined. The correlation coefficient (R^2) was used as a statistical indicator to reflect linear correlation degree.

8. Self-Decay   Self-decay refers to the process in which signal intensity of an irradiated sample declines with time [**?**]. To analyze change in PL peak intensity of irradiated PC film after a certain duration, 0.3 mm PC film was irradiated (10 MeV EB) at 300 kGy at ambient temperature in air atmosphere and then stored in a dark, dust-free environment. PL intensities were measured on days 0, 1, 3, 5, 10, 20, 30, and 60 after irradiation. The decay characteristic curve was obtained by performing appropriate function fitting.

9. Electron Energy Response   Electron energy response refers to the difference in PL peak intensity when PC films are irradiated with the same dose under different EB energies, typically depicted as the relationship between PL intensity and energy [**?**]. In this study, five EB energies (3.5, 4, 4.5, 10, and 20 MeV) were selected to irradiate 0.3 mm PC films at 100 kGy dose in air atmosphere at room temperature. The PL peak intensity of PC film irradiated with 10 MeV EB was used as a normalized basis to calculate deviation in PL peak intensity of PC films irradiated with EBs of other energy levels. Positive and negative deviations were used, where values higher and lower than the reference value were designated as positive and negative, respectively.

III. Results and Discussion

The color of PC film transformed to yellow and darkened gradually as irradiation dose increased. This yellowing is associated with formation of color centers during irradiation, such as phenoxyl and phenyl radicals. Radiation induces increased internal structural disorder in PC, creating conditions conducive to color center formation [**?**]. However, fading of irradiated samples after a certain duration is attributable to oxygen reaction of free radicals within PC film [**?**]. Based on excitation spectra tests, the optimal excitation wavelength for PL spectrum of PC film was determined to be 320 nm.

A. Thickness Applicability

Variation in mean PL peak intensity detected by PC films of different thicknesses with irradiation dose is shown in Fig. 1 [Figure 1: see original paper]. Results show that PL intensity of PC films with thickness $ $0.3 mm decreased as dose increased, whereas PL intensity of 0.2 mm PC films showed the opposite trend. The color of PC film with the same thickness darkened as irradiation dose increased. At the same irradiation dose, color of PC films darkened as film thickness increased. Darker film color indicated greater internal radiation damage. When PC film thickness exceeded 0.5 mm, PL peak intensity under the same irradiation dose decreased as thickness increased. Thicker PC film experienced higher deposited energy under the same irradiation dose, facilitating formation of a two-layer structure composed of a top carbonization layer and bottom cross-linked layer [**?**]. The formed carbonization layer affected transmission of excitation and emission light during PL spectral testing.

As shown in the fitting curve, only PL intensities of 0.3 mm and 0.2 mm PC films were linearly related to irradiation dose (0–600 kGy), with linear correlation coefficients of 0.962 and 0.834, respectively. However, the 0.2 mm PC film exhibited low mechanical strength (implying susceptibility to mechanical damage and deformation), low dose linear response values (potentially unable to absorb sufficient radiation energy, resulting in inaccurate measurements), and dose-saturation tendencies. Therefore, 0.3 mm-thick PC film was selected as the analysis sample, aligning with thicknesses of existing organic-film dose-detection materials such as low-density polyethylene films (0.3 mm) [**?**] and Indian Garfilm-EM films (0.25 mm) [**?**].

B. Preheating Time

Figure 2 [Figure 2: see original paper] shows PL emission spectrum of PC film irradiated at 100 kGy after preheating at 60 °C for various durations. In the preheating-time range of 0–300 min, PL peak intensity initially increased then decreased as preheating time increased, reaching maximum at approximately 180 min. As shown in the upper-right corner of Fig. 2, PL peak intensities at 10, 30, 40, 60, 90, 120, and 300 min represent 68.9%, 76.4%, 78.4%, 79.2%, 83.7%, 88.4%, and 67.2% of the PL peak intensity at 180 min, respectively. The initial increase in PL peak intensity is attributable to secondary electrons inside PC film after irradiation being captured by non-luminescent centers; these electrons escape after absorbing heat energy and are recaptured by luminescent centers. However, when preheating time exceeds 180 min, the fluorescence signal is more easily quenched by heat, resulting in decreased fluorescence intensity. These results suggest that PL intensity reaches saturation after preheating for a certain period, with optimal preheating time at 60 °C being 180 min. Based on existing literature, such as optimal preheating conditions for silver-doped inorganic glass RPL dosimeters being 40 min at 90 °C [**?**], future studies may investigate reducing preheating time by appropriately increasing preheating temperature.

C. Temperature Dependence

After irradiation, PL spectra of PC films were recorded at various ambient temperatures. Variations in PL spectra of irradiated (100 kGy) PC films at different ambient temperatures are shown in Fig. 3 [Figure 3: see original paper]. The emission spectrum peak was located at 470 nm. Additionally, a weak peak at 410 nm emerged and disappeared upon excitation with 350 nm UV light, indicating it was a scattering peak. Within the temperature range of 273–373 K, PL intensity decreased as ambient temperature increased. PL peak intensity at 373 K decreased to 13.3% of its initial intensity at 273 K, consistent with temperature effects on PL intensity of fluorescent materials [**?**]. This may result from inhibition of non-radiative recombination processes at low temperatures [**?**], with fluorescence thermal quenching occurring as temperature increases, thus reducing PL intensity [**?**]. The relationship between PL peak intensity and temperature is shown in the upper-right illustration of Fig. 3,

exhibiting the relationship $y = -0.2609x + 99.2735$ (where y is PL peak intensity and x is ambient temperature), with correlation coefficient of 0.964. These results indicate that PC film maintained favorable temperature-dependent linear relationship within 273–373 K, suggesting utility as a temperature sensor in future applications.

D. Humidity Dependence

Effects of ambient humidity on PL intensity of PC film during and after irradiation are shown in Figs. 4(a) and 4(b) [Figure 4: see original paper], respectively. Comparison of results shows that PL intensity decreased as RH increased (during irradiation and post-irradiation storage), while peak position remained unchanged. However, when ambient humidity increased from 12.4% to 97.2% during irradiation, PL peak intensity decreased to 74.3% of initial value, whereas when ambient humidity increased from 12.4% to 97.2% after irradiation, PL peak intensity decreased to 53.3% of initial value. Thus, storage-environment humidity after irradiation has more significant effect on PL intensity than humidity during irradiation, and PL response value decreases with increasing ambient humidity. This phenomenon is attributable to radiation generating corresponding free radical sites on the PC main chain, and interaction between radiation and water molecules generates free radicals and active particles that can interact with PC film. Therefore, increased environmental humidity during irradiation generates more free radicals that further interact with PC film. Increased humidity in storage environment after irradiation accelerates oxygen diffusion into PC matrix and further oxidizes PC, thus reducing PL intensity [**?**]. Humidity-dependence results of PC films are similar to those of FWT, Mylar, Melinex, and PET film dosimeters \cite{45–49}. Further discussion is warranted regarding declining trend of PL intensity over time under different ambient humidities.

E. In-Batch Uniformity

PL peak intensities detected for different PC films produced in the same batch after irradiation under the same dose are shown in Fig. 5 [Figure 5: see original paper]. At radiation doses of 100, 300, and 600 kGy, variation ranges of PL peak intensity counts were 5.1×10^5–5.76×10^5, 3.18×10^5–4.2×10^5, and 1.76×10^5–2.92×10^5, respectively. As irradiation dose increased from 100 to 600 kGy, fluctuation range of PL peak intensity increased. Relative average deviations corresponding to the three doses were 2.73%, 8.4%, and 12.8%, respectively. These results show that in-batch uniformity of PC film under doses <100 kGy is ideal, with signal uniformity similar to that of GD-300 dosimeter (deviation values of GD-300 at doses of 0.2, 20, and 200 mGy are $\pm$1.7%, $\pm$1.3%, and $\pm$1.1%, respectively) [**?**, **?**]. However, when dose exceeded 100 kGy, relative average deviation of in-batch uniformity increased, and its effect on dose detection accuracy should be considered [**?**].

F. Readout Reproducibility

Under three different radiation doses, PC film was repeatedly excited and measured 10 times; corresponding PL peak intensity values are shown in Fig. 6 [Figure 6: see original paper]. Results show that variation ranges of PL peak intensity counts at irradiation doses of 100, 300, and 600 kGy were 5.04×10^5– 5.56×10^5, 3.54×10^5–3.7×10^5, and 2.16×10^5–2.81×10^5, respectively, while mean values were 5.277×10^5, 3.616×10^5, and 2.547×10^5, respectively. Dose response is consistent with data shown in Fig. 5. Additionally, standard deviations of the three data groups were 0.158, 0.044, and 0.23, respectively, and coefficients of variation of PL intensity values were calculated to be between 1.2% and 8.6%. Compared with GD-351 dosimeter (coefficient of variation for readout reproducibility in dose range of 0.2–200 mGy is 15%–19%) [?], PC film exhibited smaller standard deviation and coefficient of variation, indicating favorable readout reproducibility of irradiated PC film.

G. Dose Linearity

PL emission spectra of PC films irradiated at different doses (0–600 kGy) are presented in Fig. 7 [Figure 7: see original paper]. A broad emission band was observed within 400–600 nm. PL spectral peak position of irradiated PC films was redshifted compared with unirradiated (pristine) films, attributed to defect formation after irradiation or partial release of hydrogen molecules, resulting in generation of carbon-rich clusters and subsequent reduction in optical bandgap energy [?]. Additionally, PL intensity decreased as irradiation dose increased. PL peak intensity of PC irradiated with 600 kGy decreased by 63.2% compared with pristine samples (0 kGy). During PL process, energy is transferred to chromophore sites via UV excitation, and radiative recombination occurs in thermalized electron–hole pairs, resulting in fluorescence [?]. The decrease in PL intensity may be associated with formation of internal defects caused by radiation [?]. Radiation-induced disturbance in PC internal structure resulted in emergence of defect states, including chain scission and intermolecular crosslinking [?]. Defect formation created a new radiative-recombination level for electrons and holes in PC [?].

Fluorescence peak positions of all samples were concentrated at 470 nm. The relationship between PL peak intensity value (y) and irradiation dose (x) within 20–600 kGy is shown in the upper right of Fig. 7. Linear-regression fitting yielded the equation $y = -0.00695x + 6.32303$, with correlation coefficient of 0.965 (standard error of this correlation coefficient is 1.72%). This correlation coefficient closely matches that of Makrofol LT 6-4 PC film in the range of 150–950 kGy [?], demonstrating that PC film maintained favorable linear relationship in the dose capture range of 20–600 kGy.

H. Self-Decay

Attenuation characteristics of PL peak intensity in irradiated PC film (300 kGy) within 60 days are shown in Fig. 8 [Figure 8: see original paper]. PL peak intensity decreased continually with time, possibly due to ambient temperature or other factors resulting in reduction of fluorescence centers inside irradiated PC film. Results indicate significant decrease in PL peak intensity within the first 20 days after irradiation, with peak intensity decreasing to 64% of initial value. Subsequently, the declining trend became gentler over the following 40 days, with peak intensity decreasing to 60% of initial intensity within 60 days. Fitting with the ExpGro1 function yielded the decay equation:

$$y = 2.1483 + 1.1850 \times e^{-5.7560x}.$$

The correlation coefficient between PL peak intensity (y) and time (x) was 0.956. Thus, the fitting function reflects decline in fluorescence intensity to a certain extent. This fluorescence signal decay after irradiation is commonly observed in commercial thin-film dosimeters such as GD-300 RPL [?] and CTA dosimeters [?], with decay characteristics depending on material properties [?]. Considering significant decay in PL signal during initial days after irradiation, PL spectral testing should be conducted as soon as possible after irradiation, or corresponding dose compensation should be performed based on decay characteristic law.

I. Electron Energy Response

Figure 9 [Figure 9: see original paper] shows PL peak intensity of PC film irradiated by EBs of different energies (3.5, 4, 4.5, 10, and 20 MeV) at the same dose (100 kGy). Results show that PL peak response values at these energy points were 3.499×10^5, 3.422×10^5, 3.393×10^5, 3.352×10^5, and 3.236×10^5, respectively. Using PL peak intensity at 10 MeV as normalized basis, deviations at 3.5, 4, 4.5, and 20 MeV were +4.38%, +2.09%, +1.22%, and -3.46%, respectively. Compared with energy-response characteristics of GD-351M RPL and LiF:Mg,Ti dosimeters [?], signal intensity of PC film exhibited similar trend, although amplitude of change was smaller. This is due to low density and small thickness of PC film as well as marginal difference in electron energy response under irradiation with high-energy EB ($ $3.5 MeV).

IV. Conclusion

In this study, thickness applicability, preheating time, temperature and humidity dependence, in-batch uniformity, readout reproducibility, dose linearity, self-decay, and electron energy response of engineered PC films after EB irradiation were discussed based on PL-spectrum analysis. Upon excitation with 320 nm UV light, fluorescence peak of PC film appeared at emission wavelength of 470 nm. Results show that optimal thickness for dose detection using PC film was 0.3 mm and optimal fluorescence value can be obtained by preheating at 60 °C

for 180 min. However, environmental factors such as temperature (during PL spectral testing) and humidity (both during irradiation and post-irradiation storage) can affect PL intensity. At low irradiation dose (100 kGy), dose-response uniformity of PC film was ideal. In the dose-capture range of 20–600 kGy, PL spectral peak intensity showed favorable dose linearity ($R^2 = 0.965$) and readout reproducibility ($C_v \leq 8.6\%$). Additionally, when PC film is used for radiation-dose measurement, PL-spectrum testing should be performed promptly after irradiation to prevent PL-signal decline. Based on comprehensive analysis of these dosimetric parameters, this PC film can be proposed as a promising RPL material with potential application in high-radiation-dose detection.

Acknowledgement

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