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Research Article

Br-LMG/PVB Film: A Novel UVC Dosimeter for Process Monitoring in Chemical and Environmental Engineering

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Abstract

In chemical engineering processes such as photocatalytic wastewater treatment, cooling water biofouling control, and filling line surface sterilization, confirming UVC dose is critical for process efficiency and safety. However, existing UVC dosimeters rely on complex equipment, acid generators, or high cost, limiting field deployment. This study fabricated a UVC dosimeter based on polyvinyl butyral (PVB) film containing bromo-leuco malachite green (Br-LMG) without any acid generator, and evaluated its dosimetric characteristics for 253.7 nm dose monitoring. Br-LMG/PVB films were prepared by spin-coating and irradiated with UVC doses of 0, 25, 50, 75, 100, 200, and 300 mJ/cm². Changes in optical density, dose-response linearity, post-irradiation stability (30 min and 30 days), and colour difference (ΔE) based on CIE L^*, a^*, b^* were quantified using UV-VIS spectrophotometry and reflectance spectroscopy. Upon UVC irradiation, the film exhibited a maximum absorption peak at 630 nm corresponding to the oxidized form of malachite green. The dose-response curve showed excellent linearity ($R^2 \approx 1$) over 0-300 mJ/cm² with sensitivity of 0.0031 (mJ/cm²)$^{-1}$ at 630 nm. After 30 days of room-temperature storage, optical density increased by only 5.2-9.2% without fading. ΔE exceeded 10 at 25 mJ/cm² (perceptible) and reached 18 at 50 mJ/cm² (clearly distinguishable by naked eye). The Br-LMG/PVB film serves as a simple, low-cost, field-deployable UVC dosimeter requiring no additional equipment for dose assessment in chemical and environmental engineering applications.

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Keywords: UV disinfection; UVC dosimeter; bromo-leuco malachite green; polyvinyl butyral; colour difference; dose–response linearity

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1. Introduction

Ultraviolet (UV) disinfection is widely recognised as an effective method for inactivating a broad range of microorganisms, including viruses and bacteria. Among the UV spectrum, the UVC region (200–280 nm) exhibits the highest germicidal efficacy, primarily due to its direct absorption by nucleic acids, leading to the formation of photodimers that inhibit replication. Consequently, UVC irradiation has been extensively employed for disinfection of air, water, surfaces, and medical equipment. For effective UVC disinfection, it is essential to deliver an appropriate dose, as insufficient exposure may result in incomplete microbial inactivation, whereas excessive exposure can cause unnecessary energy consumption and potential material degradation. Therefore, a simple, inexpensive, and reliable dosimeter capable of confirming the delivered UVC dose is highly desirable [1,2].

Over the past decades, various types of sensitive films have been developed for UV dose measurement. Among them, radiochromic films, which undergo a distinct colour change upon exposure to radiation, have been widely utilised for dosimetry applications [3,4]. ICRU Report 80 systematically summarises various dosimetry

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systems, including radiochromic films [3]. Miller and colleagues first reported a new radiochromic thin-film dosimeter based on a radiation-sensitive dye [4], and later Miller and McLaughlin presented a broad overview of dosimetry techniques for radiation processing [14]. Sidney *et al*. [18] subsequently reported a radiochromic film using acid-sensitive leuco dyes in a chlorine-containing polymer matrix with improved stability.

The application of radiochromic films has since expanded from primarily medical uses to the food industry and environmental monitoring. Butson *et al.* analysed the characteristics of radiochromic films in medical radiation dosimetry, focusing on energy dependence, dose rate dependence, and post-irradiation stability [5]. Devic *et al*. reviewed reference radiochromic film dosimetry in clinical radiotherapy [6]. Casolaro *et al.* reported real-time dosimetry techniques using radiochromic films [7].

More recently, Seyfi Cankal *et al*. developed an innovative method for evaluating UVC dose distribution at 254 nm on food surfaces using radiochromic film dosimetry integrated with image processing and a convolutional neural network (CNN) [8]. According to their findings, a linear relationship exists between colour difference and UVC dose in the range of 2.5–125 mJ/cm², with a maximum measurable dose of approximately 60 mJ/cm² [8]. Furthermore, the colour of the irradiated film remained stable for up to 15 days when stored in the dark [8,9]. In a follow-up study, Seyfi Cankal and Unluturk [19] optimized leucomalachite green-based radiochromic films in a PVB matrix, achieving tunable sensitivity for different UVC dose ranges.

Polyvinyl butyral (PVB) has attracted increasing attention as a matrix material for dosimeters owing to its excellent film-forming ability, optical transparency, mechanical flexibility, and low moisture sensitivity. Rabaeh et al. developed a film dosimeter by combining MMT dye with PVB, which changes colour upon gamma irradiation and reduces humidity dependence [9,10]. The Defence Laboratory in Jodhpur also developed a radiochromic film using a diacetylene monomer dispersed in a PVB-like matrix, exhibiting a main absorption peak at 675 nm [10]. Saad *et al.* [16] demonstrated that PADC polymer films exhibit dose-dependent optical property changes upon UVC exposure over 0.446–4.458 kJ/cm². Kozicki *et al.* [17] developed a leuco crystal violet-based dosimeter in a Pluronic F-127 matrix that responds to all UV subranges (UVA/UVB/UVC).

Leuco-dye-based dosimeters exploit the colour change from colourless to coloured upon exposure to radiation or UV light. Malachite green derivatives are representative photo-oxidizable dyes that develop colour under UV irradiation. The UVC sensitivity of malachite green derivatives was quantitatively analysed by Kovács *et al*. [11]. Ebraheem *et al*. established general principles for UV-measuring films based on tetrazolium dyes dispersed in a polymer matrix [12]. Jaszczak-Kuligowska *et al.* [20] extended this approach by developing an elastic TTC-PVA gel dosimeter capable of measuring UVA, UVB, and UVC doses separately. Nivi *et al.* evaluated the low-dose (≤50 mSv) response of a commercial radiochromic film for rapid in-field monitoring [13].

Despite these significant advances, several important limitations remain. First, most studies have focused on high-dose ionising radiation [9,13], whereas UVC disinfection requires low doses of tens to hundreds of mJ/cm². Second, existing PVB-based dosimeter films generally require additional sensitisers or acid generators, complicating fabrication and increasing cost [9]. Third, systematic evaluation of long-term stability and quantitative visual discrimination criteria using the CIE L^*, a^*, b^* colour space has not been sufficiently addressed [8]. These gaps limit practical deployment in real-world disinfection settings. To overcome these limitations, the present study fabricates a PVB film incorporating bromo-leuco malachite green (Br-LMG) alone, without any acid generator. This approach eliminates the need for additional chemical reagents, simplifying fabrication and reducing costs. By quantitatively analysing optical properties, dose–response linearity, post-irradiation stability, and colour difference (ΔE) of Br-LMG/PVB films upon UVC irradiation, this study systematically verifies the film as a simple, low-cost, field-deployable, immediate-readout UVC dosimeter.

2. Materials and Methods

2.1. Reagents and Equipment

Reagents: Bromo-leuco malachite green (Br-LMG), polyvinyl butyral (PVB, Pioloform BR18, Mw ≈ 50–60 ×10³, Wacker Co., USA), ethyl alcohol, and butyl acetate (POCh, Poland). All reagents were purchased from Wako Pure Chemicals, Japan, and used without further purification. Equipment: Magnetic stirrer, low-pressure mercury lamp (G8T5, 254 nm, Sankyo Denki, Japan), UV-VIS spectrophotometer (H.SWX 420BS), UV radiometer (UVR-2), digital thickness gauge.

2.2. Preparation of Br-LMG/PVB Film

All operations were performed under light-shielded conditions. PVB (100 g) was uniformly dispersed in ethyl alcohol (500 g) and butyl

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acetate (100 g) at room temperature with stirring for 48 h. Subsequently, Br-LMG (2 g) was added and stirring was continued. The mixed suspension was spin-coated onto a horizontally maintained glass plate (225 cm²) to form a thin film, which was then dried at 40 °C for 24 h. The resulting film was colourless and had a thickness of 0.2 ± 0.03 mm, measured at five different positions using a digital thickness gauge. From the prepared film, seven specimens of size 1.5 cm × 1.5 cm were cut, and each was irradiated with a different UV dose.

2.3. UVC Irradiation

Irradiation was performed at 25 °C and 50% relative humidity. A 254 nm low-pressure mercury lamp (G8T5, Sankyo Denki, Japan) was used as the light source, and the distance between the film specimen and the lamp was fixed at 10 cm. The irradiation intensity measured with the UVR-2 radiometer was 0.5 mW/cm². Irradiation times of 0, 5, 10, 15, 20, 40 and 60 s were used to obtain doses of 0, 25, 50, 75, 100, 200 and 300 mJ/cm², respectively. The dose was calculated as (intensity × time) and confirmed in real time with the UVR-2 radiometer. A total of seven dose conditions, including the non-irradiated control (0 mJ/cm²), were prepared. The colour changes of the irradiated specimens are shown in Figure 1.

2.4. Measurements and Analysis

UV-VIS absorption spectra (200–800 nm) were measured before and after irradiation. All measurements were performed in triplicate for each dose condition. The change in optical density (ΔOD) was calculated using the following equation:

$$\Delta OD_i = OD_i - OD_0 \quad (1)$$

where OD_i is the optical density of the Br-LMG/PVB film specimen irradiated with a given UVC dose, and OD_0 is that of the non-irradiated control specimen.

For reflectance spectrum measurements, chromatography filter paper was attached to the back of the film. Reflectance spectra were recorded from 400 to 700 nm. Based on Kubelka–Munk theory, the reflectance was converted to an absorption coefficient, from which the tristimulus values were calculated. Subsequently, CIE 1976 L^*, a^*, b^* values were derived. The total colour difference (ΔE) was calculated using the following equation:

$$\Delta E = \sqrt{(L_i^* - L^*)^2 + (a_i^* - a^*)^2 + (b_i^* - b^*)^2} \quad (2)$$

Stability evaluation: Short-term stability was assessed by measuring the optical density at 630 nm at 0, 1, 2, 5, 10, 15, 20, 25, and 30 min after irradiation. Long-term stability was evaluated by storing the irradiated specimens in the dark at room temperature for 30 days, with optical density measurements performed at regular intervals (0, 1, 2, 5, 10, 15, 20, 25, and 30 days).

3. Results and Discussion

3.1. Optical Density Characteristics in the UVC Wavelength Region

The spectral response of the Br-LMG/PVB film in the UVC region (200–400 nm) was examined to determine the optimal excitation wavelength. As shown in Figure 2, the maximum optical density change occurred between 250 and 260 nm, which coincides well with the emission peak (254 nm) of the low-pressure mercury lamp

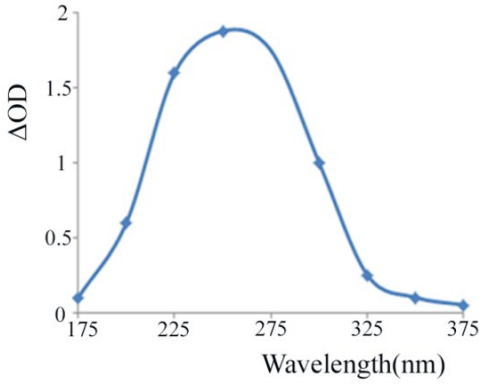


Figure 1. Colour change of Br-LMG/PVB films upon UVC irradiation at different fluences (0, 25, 50, 75, 100, 200, 300 mJ/cm²).

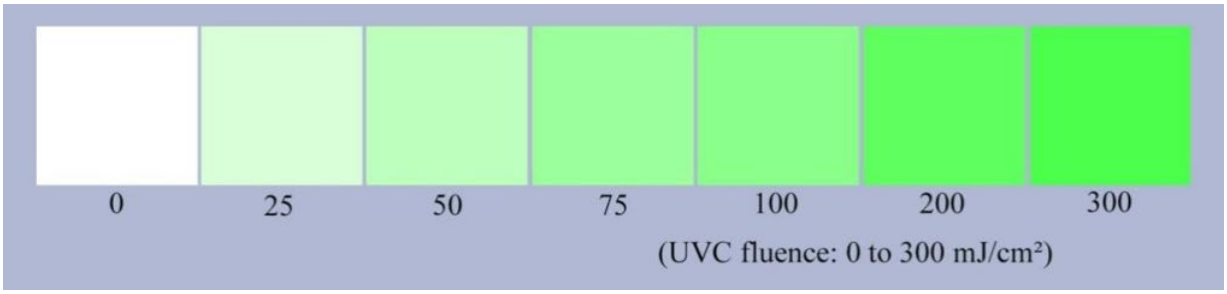


Figure 1. Colour change of Br-LMG/PVB films upon UVC irradiation at different fluences (0, 25, 50, 75, 100, 200, 300 mJ/cm²).

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used in this study. This agreement indicates that the film is effectively sensitive to the germicidal UVC wavelength, making it suitable for UVC dose monitoring without the need for spectral correction. Compared to tetrazolium-based films requiring specific UV wavelengths for activation [12], this film exhibits a broader sensitivity window around 254 nm without spectral correction filters.

3.2. Change in Visible Absorption Spectrum with UVC Dose

Upon UVC irradiation, the colourless Br-LMG/PVB film gradually turned bluish-green. The UV-VIS absorption spectra (Figure 3) reveal the appearance of a strong absorption peak at 630 nm and a weaker peak at 430 nm, both increasing in intensity with increasing UVC dose. These peaks are characteristic of the oxidised form of malachite green (MG^+), confirming that UVC irradiation induces the oxidation of the leuco form (LMG) to the coloured form. Notably, no additional absorption bands appeared, indicating a clean photochemical conversion without detectable side products.

The absence of an acid generator in the film composition suggests that reactive oxygen species (ROS), such as hydroxyl radicals or superoxide anions, generated within the PVB matrix upon UVC exposure, are likely responsible for the oxidation of LMG. This proposed mechanism is illustrated in Figure 4. The dose-dependent increase in peak intensity implies that the film can serve as a direct visual indicator of accumulated UVC dose.

The observed absorption peaks at 630 nm and 430 nm are consistent with the oxidation of LMG to MG^+ previously characterized by Kovács *et al.* [11], while the 630 nm peak is similar to the 675 nm peak of diacetylene-based films [10] but achieved without gamma pre-irradiation. After irradiation with 0, 25, 50, 75, 100, 200, and 300 mJ/cm², the 630 nm peak increases dose-dependently in the 300–800 nm region. Reactive oxygen species generated by UVC irradiation oxidise leuco malachite green (LMG) to the coloured form (MG^+).

3.3. Dose–response Curve

The change in optical density (ΔOD) at 430 nm and 630 nm was plotted against UVC dose (Figure 5). Both wavelengths exhibited excellent linearity ($R^2 \approx 1$) over the entire 0–300 mJ/cm²

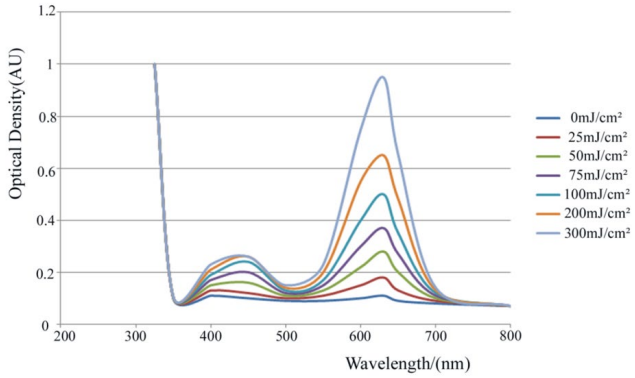


Figure 3. UV-VIS absorption spectra of Br-LMG/PVB films as a function of UVC dose.

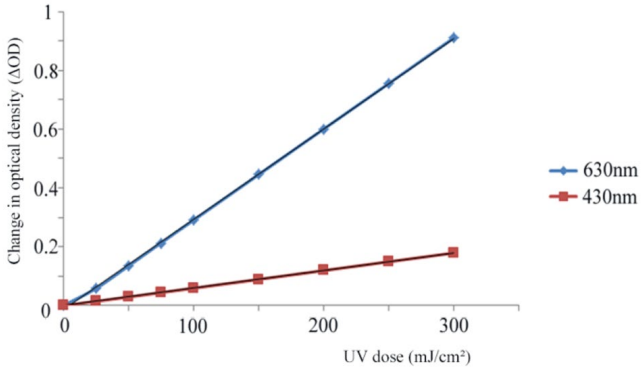


Figure 5. UVC dose–response curves at 430 nm and 630 nm.

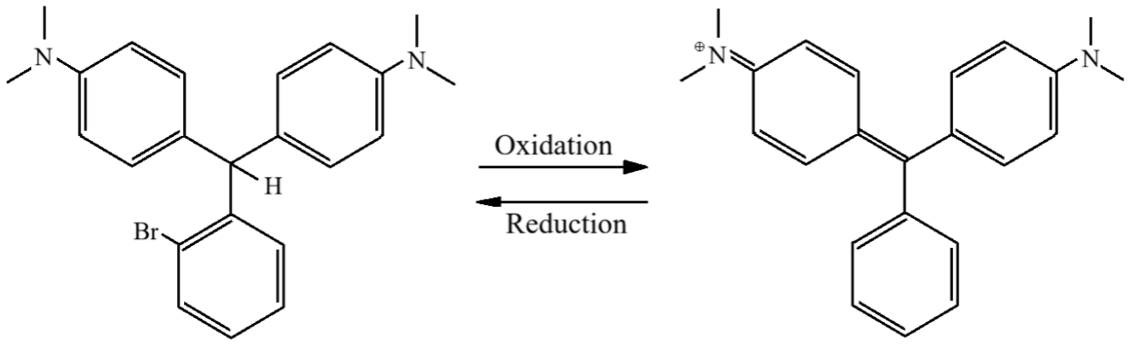


Figure 4. UVC-induced redox reaction mechanism of Br-LMG.

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range, demonstrating that the film follows a simple first-order-like photoconversion kinetics under the experimental conditions. The sensitivity (slope) was 0.006 $(mJ/cm^2)^{-1}$ at 430 nm and 0.0031 $(mJ/cm^2)^{-1}$ at 630 nm, meaning that ΔOD at 430 nm is approximately twice as sensitive to dose. However, the absolute ΔOD values at 630 nm are much larger, making this wavelength more practical for both instrumental readout and visual inspection.

The linear dose–response relationship without saturation up to 300 mJ/cm² is particularly advantageous for UVC disinfection applications, where doses typically range from tens to hundreds of mJ/cm². Notably, even at the lowest dose tested (25 mJ/cm²), a discernible colour change was observable by the naked eye, indicating that the film can serve as a qualitative pass/fail indicator for low-dose UVC exposure. The dose dependence of the change in optical density (ΔOD) shows high linearity over 0–300 mJ/cm². While Seyfi Cankal *et al.* [8] reported saturation above 60 mJ/cm² for their radiochromic film, the present film maintains linearity up to 300 mJ/cm², extending the measurable dose range by a factor of five.

3.4. Post-irradiation Stability and Storage Stability

For practical applications, a dosimeter must maintain its response after irradiation until readout. Short-term stability tests revealed a slight but notable post-irradiation effect at high doses. Specifically, for the 300 mJ/cm² group, the optical density increased from 0.95 immediately after irradiation to 1.03 after approximately 5 min, after which it remained stable. This 8.4% increase is attributed to a delayed equilibrium of the photo-oxidation reaction within the solid polymer matrix, where trapped radicals or residual reactive species continue to react with LMG for a short period after UVC exposure ceases. For doses ≤200 mJ/cm², no significant post-irradiation change was observed (variation <1%), suggesting that the reaction is essentially complete immediately upon irradiation at lower doses. From a practical standpoint, these results imply that for high-dose measurements (≥200 mJ/cm²), a stabilisation time of at least 5 min is recommended before accurate readout. For routine UVC disinfection monitoring (typically ≤100 mJ/cm²), immediate readout is acceptable.

Long-term storage stability is equally critical. After 30 days of dark storage at room temperature, the optical density at 630 nm increased by only 5.2–9.2% relative to the immediate post-irradiation value (Figure 6). This modest increase is within the acceptable range for most dosimetric applications and is likely due to slow continued oxidation by residual oxygen or trapped radicals. Importantly, no fading (decrease in optical density) was observed, which is advantageous compared with some commercial radiochromic films that exhibit significant signal loss over time.

For each dose, the increase in optical density after 30 days relative to the immediate post-irradiation value is in the range 5.2–9.2%, indicating stable performance. Compared to Seyfi Cankal *et al.* [8] who reported stability for only 15 days, the present film remains stable for 30 days with a minimal increase of 5.2-9.2% and no fading, which is superior to some commercial films showing dose-dependent signal loss [6].

3.5. Reflectance Spectra and Colour Difference (ΔE) Analysis

For field applications where a spectrophotometer is unavailable, visual colour discrimination is the simplest readout method. To quantify visual perception, the CIE L^*, a^*, b^* colour coordinates and total colour difference (ΔE) were derived from reflectance spectra (Figure 7, Table 1). As the UVC dose increased, L^* (brightness) decreased from 97 to 77, a^* became more negative (from –1 to –44), and b^* also shifted

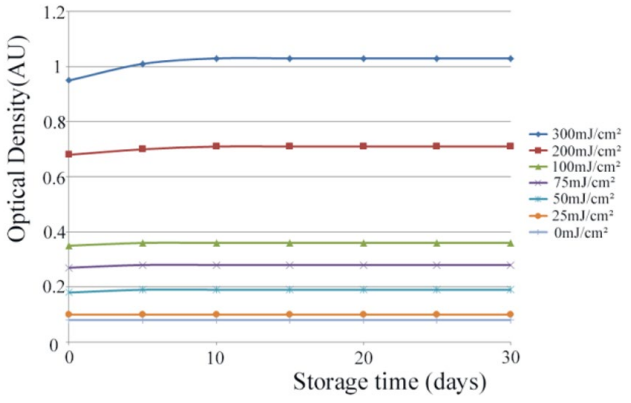


Figure 6. Change in optical density (630 nm) as a function of storage time after irradiation (30 days).

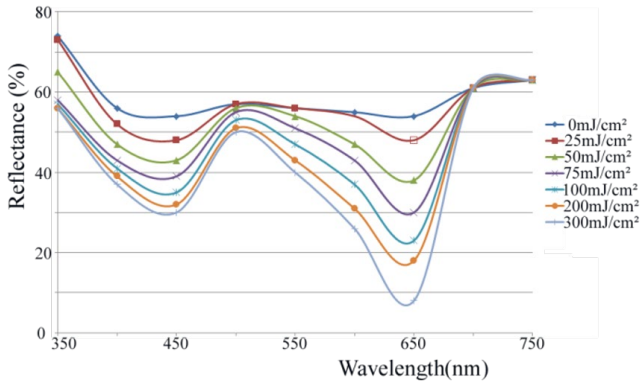


Figure 7. Reflectance spectra of UVC-irradiated Br-LMG/PVB films.

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negatively (from −1 to −14). These changes correspond to a darker, more intensely green-blue colour. The ΔE values increased monotonically with dose: 0 (0 mJ/cm²), 10.05 (25 mJ/cm²), 17.97 (50 mJ/cm²), and up to 50.02 (300 mJ/cm²).

According to the standard interpretation of colour difference, a ΔE value greater than approximately 5–10 is considered distinguishable by an untrained observer, and values above 15–20 are clearly noticeable [15]. In this study, ΔE exceeded 10 at 25 mJ/cm² (threshold of perceptibility) and reached nearly 18 at 50 mJ/cm², where the colour difference becomes conspicuous. Thus, the Br-LMG/PVB film allows not only quantitative instrumental readout but also semi-quantitative visual assessment: doses below 25 mJ/cm² are difficult to discern visually, whereas doses of 50 mJ/cm² and above can be reliably distinguished by the naked eye. This feature is particularly valuable for rapid field screening of UVC disinfection efficacy.

Reflectance (400–700 nm) of specimens irradiated with 0, 25, 50, 75, 100, 200 and 300 mJ/cm². Whilst Seyfi Cankal *et al.* [8] achieved higher sensitivity (2.5 mJ/cm²) using CNN-based image processing requiring computational resources, this film's ΔE threshold of 18 at 50 mJ/cm² enables immediate naked-eye discrimination without any equipment for most UVC disinfection applications (typical target doses: 40-100 mJ/cm²). Mean values for each dose (n = 3) ΔE are calculated according to Equation (2).

4. Conclusion

This study successfully fabricated a Br-LMG/PVB film without an acid generator and evaluated its dosimetric characteristics for 253.7 nm UVC dose monitoring. The film exhibited a maximum absorption peak at 630 nm with spectral sensitivity matching the 254 nm UVC lamp. The dose–response curve showed excellent linearity ($R^2 \approx 1$) over 0–300 mJ/cm² with a sensitivity of 0.0031 $(mJ/cm^2)^{-1}$ at 630 nm. Post-irradiation stability evaluation revealed a 5.2–9.2% increase in optical density after 30 days without fading, and a 5 min stabilization time is recommended for high doses (≥200 mJ/cm²). Colour difference analysis demonstrated that ΔE reached 18 at 50 mJ/cm², enabling naked-eye discrimination. Therefore, the Br-LMG/PVB film serves as a simple, low-cost, field-deployable UVC dosimeter requiring no additional equipment for chemical and environmental engineering applications.

CRedit Authors Statement

Author Contributions: Guk Chol Kim: Conceptualization, Methodology, experiment, Validation; Kye Ryong Ri: experiment, Formal Analysis, Data Curation, Writting Draft Preparation; Song Taek Kim: Material Analysis; Jong Hyok Kim: Review and Editing. All authors have read and agreed to the published version of the manuscript.

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Table 1. CIE *L**, *a**, *b** values and colour difference (ΔE) of Br-LMG/PVB films as a function of UVC dose.

UVC fluence / (mJ/cm²)	*L**	*a**	*b**	ΔE
0	97	-1	-1	0
25	97	-10	-3	10.05
50	93	-18	-5	17.97
75	90	-26	-9	27.75
100	87	-30	-10	32.65
200	81	-40	-13	45.01
300	77	-44	-14	50.02

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