

Optical Properties of Polyvinyl Alcohol-Based Polymer Films Containing Methylene Blue and Trichloroacetic Acid for Gamma Radiation Dosimetry Applications

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Abstract

The primary objective of this study is to investigate the optical properties of polyvinyl alcohol (PVA) based polymer films, incorporating methylene blue (MB) dye and trichloroacetic acid (TCA), for their potential application in gamma radiation dosimetry. Specifically, this research aims to explore the effects of gamma radiation on the color change characteristics, optical absorption spectra, activation energy, and optical band gap energy of the PVA-MB-TCA polymer films. Additionally, the study seeks to assess the stability of these polymer films under varying doses of gamma radiation, ranging up to 14 kGy. The PVA-MB-TCA polymer films were prepared using a solvent-casting method. The polymer film samples were then exposed to gamma radiation from a ⁶⁰Co source, with doses up to 14 kGy. The study observed significant color changes in the polymer films, transitioning from blue at 0 kGy to light blue-near transparent at 14 kGy. Spectrophotometric analysis identified three distinct wavelengths of maximum absorption at 360 nm, 440 nm, and 560 nm. As the radiation dose increased, absorption values decreased at 360 nm and 440 nm, while an opposite trend was noted at 560 nm. Furthermore, the activation energy of the polymer films was found to decrease with increasing radiation doses, indicating a reduction in the energy barriers for internal reactions. Similarly, the optical band gap energy also showed a decreasing trend with higher radiation doses across all types of transitions. These results demonstrate that the PVA-MB-TCA polymer films undergo significant optical and structural changes when exposed to gamma radiation, highlighting their potential utility as reliable high-dose radiation dosimeters. The stability of these films under radiation further supports their applicability in various fields requiring precise radiation dose monitoring, such as medical sterilization, food processing, and environmental safety.

Keywords: Optical properties; Polymer films; Gamma irradiation; Radiation dosimetry.

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INTRODUCTION

The use of gamma radiation has significantly increased across multiple sectors, impacting areas such as occupational safety, food processing and preservation, pharmaceuticals, sterilization of medical products, material modification methods, and other applications (Handayani & Permawati, 2017). Alongside this expansion, the safety of workers in the radiation industry has become a critical concern. In many radiation-related fields, radiation control is essential to prevent the adverse effects of radiation and to ensure its use according to requirements, one of which is achieved through dosimetry (as a radiation dose monitoring instrument). Radiation

dosimeters are concerned with measuring the radiation dose absorbed by materials when exposed to ionizing radiation (Doyan et al., 2021; Rabaeh et al., 2021).

In food processing, radiation dosimeters are particularly beneficial when radiation systems are legalized for food. This is intended to control the radiation dose exposure in food. In the food industry, the purpose of radiation varies and is associated with the given radiation dose (Handayani & Permawati, 2017). For example, to delay the ripening process and reduce microbes in food, doses around 0.4 to 2.5 kGy are administered (referred to as the radurization process); to kill all bacterial spores and for food preservation (fruits and vegetables), doses of approximately 1 to 10 kGy are administered (known as the radication process); and to extend the shelf life of food and kill all pathogenic microbes, doses of about 30 to 50 kGy are administered (called the radappertization process) (Eskin & Robinson, 2001). Dosimeters can represent the permissible doses according to food processing requirements, necessitating the development of suitable radiation dosimeters as dose monitoring instruments.

The development of radiation dose monitoring instruments has significantly progressed with the advent of advanced composite materials. Extensive research has been conducted on creating effective and reliable dosimeters using these composites (Gafar et al., 2017). One notable advancement is the use of dyed polymer films for routine dosimetry, which has become increasingly prevalent in measuring absorbed radiation doses (Kattan et al., 2011). These polymer-based dosimeters offer several technical benefits that make them highly suitable for various applications. They are notably portable and lightweight, allowing for easy handling and deployment in different environments (Akhtar et al., 2016). Furthermore, the manufacturing costs of these dosimeters are relatively low, making them an economical choice for widespread use (Akhtar et al., 2013). These attributes not only enhance the practicality of dyed polymer film dosimeters but also ensure their accessibility and efficiency in diverse fields. As the need for accurate and reliable radiation dose measurement grows, the integration of advanced materials in dosimetry continues to drive innovation and improve the effectiveness of radiation safety protocols.

Film dosimeters are extensively used for monitoring low-dose radiation in various consumables, including food, beverages, and other processed products intended for human consumption (El-Kelany & Gafar, 2016). These dosimeters ensure that radiation levels remain within safe limits, protecting public health. Additionally, film dosimeters play a crucial role in high-dose radiation applications, such as the sterilization of medical instruments, where they help verify that the necessary radiation dose has been applied to achieve sterility (Gafar & El-Ahdal, 2014). In the field of radiotherapy, these dosimeters are used to measure and control the precise radiation doses delivered to patients, ensuring effective treatment while minimizing harm to surrounding healthy tissues (Hassani et al., 2014).

In contemporary industrial settings, dye-based dosimeters are increasingly employed as reliable indicators of radiation exposure. These dosimeters are used to assess the radiation doses that materials are subjected to during various processes, providing a visual and quantitative means of monitoring radiation levels (Ali-Omer & Ali-Bashir, 2018; Aydarous et al., 2016; Basfar et al., 2012). The use of

dye-based dosimeters spans a wide range of industries, from food safety and medical sterilization to material processing and environmental monitoring, reflecting their versatility and importance in ensuring radiation safety and efficacy.

For polymer-based materials to be effective as radiation dosimeters, they must undergo extensive research, development, and rigorous testing across a spectrum of radiation doses, encompassing both high and low levels (Abdel-Fattah et al., 2014; Soliman et al., 2018; Ticoş et al., 2019). The accuracy and precision in calculating the exposed radiation dose are critical, necessitating meticulous calibration and validation to ensure reliable measurements (Hosni et al., 2013; Raouafi et al., 2018). To date, the development of film dosimeters has primarily focused on using polymer bases combined with various dye indicators. These polymer films, serving as the foundational material for dosimeters, include a range of substances such as polyvinyl butyral (Abdel-Fattah et al., 2014), polyvinyl chloride (Kattan & Daher, 2016), and polycarbonate (Galante & Campos, 2012). Among these, polyvinyl alcohol (PVA) has emerged as the most extensively utilized polymer due to its favorable properties, such as excellent film-forming capabilities, flexibility, and chemical stability (El-Kelany & Gafar, 2016; Raouafi et al., 2018). The combination of PVA with specific dye indicators enhances the dosimeter's sensitivity and accuracy, making it a robust choice for various radiation monitoring applications. This ongoing research and development in polymer-based dosimetry are essential for advancing the field and ensuring the availability of reliable and efficient tools for radiation dose measurement.

Polyvinyl alcohol (PVA)-based polymer films offer numerous advantages over other materials, making them particularly suitable for various applications. These advantages include excellent mechanical properties, such as strength and durability, which contribute to the film's robustness. PVA films are also water-soluble, allowing for easy processing and manipulation in aqueous environments. Furthermore, they exhibit remarkable flexibility and elasticity, enabling them to conform to different shapes and surfaces without breaking. One of the most crucial benefits of PVA films is their non-toxicity, which makes them safe for use in applications involving direct contact with food, medical products, and other sensitive materials (Ang et al., 2020; Chaturvedi et al., 2015; Gadhawe et al., 2019; Wong et al., 2020). Various dyes are used as indicators in these polymer films to enhance their functionality for specific applications. These dyes include cresol red (CR) (Ebraheem et al., 2002), methylene red (Akhtar et al., 2013), ethyl violet and bromophenol blue (Ebraheem & El-Kelany, 2013), methyl thymol blue (MTB) (Rabaeh et al., 2021), thymolphthalein (TP) (El-Kelany & Gafar, 2016), tetrabromo phenolphthalein ethyl ester (TBPE) and acid yellow (AY) (Gafar et al., 2017), methyl viologen (Lavalle et al., 2007), and tetrazolium violet (Emi-Reynolds et al., 2007). These dyes provide specific optical and colorimetric properties that can be exploited for various dosimetry and sensing applications.

In addition to these dyes, some studies have explored the incorporation of chlorine-containing compounds into polymer blends (Abdel-Fattah et al., 1996, 1997; Doyan et al., 2021; Gafar et al., 2017). The inclusion of chlorine is believed to improve dye solubility within the polymer matrix, enhancing the uniformity and effectiveness of the film (Abdel-Fattah et al., 1996, 1997). Furthermore, the low pH environment created by chlorine compounds can increase the sensitivity of dye

components in the polymer film mixtures, leading to more pronounced and detectable changes in response to external stimuli, such as radiation exposure (Doyan et al., 2021; Gafar et al., 2017). This combination of advantageous properties and enhanced functionality underscores the versatility and potential of PVA-based polymer films in a wide range of industrial, medical, and environmental applications.

The color in mixed polymer films acts as an indicator in the dosimetry system, showing the amount of ionizing radiation exposure the material has received. Different radiation doses cause color changes in polymer films (Doyan et al., 2021). In application, thin polymer films are placed (embedded) on the material to be irradiated, and when the material is exposed to radiation, the color change in the polymer film indicates the radiation dose on the material. In gamma radiation dosimetry studies, the useful radiation dose range is a primary factor in its application. Above a certain dose threshold, polymer film materials do not indicate color changes and are thus considered unusable. Previous studies have shown that PVA polymer films with MTB dye have a useful gamma radiation dose range from 2.5 to 20 kGy (Rabaeh et al., 2021). PVA polymer films with TP dye are utilized as new gamma radiation detector systems in the dose range of 1 to 6.5 kGy (El-Kelany & Gafar, 2016). The addition of TBPE and AY dye indicators in PVA has shown promising results as new dosimeters in the gamma radiation dose range of 0.1 to 5 kGy (Gafar et al., 2017). Recent studies have demonstrated the useful dose range and stability of PVA-CR polymer films with added chlorine, which is in the range of 1 to 14 kGy (Doyan et al., 2021).

A polymer film material can be applied as a radiation dosimeter if there is a color change in the polymer due to radiation exposure (sensitivity to color change due to radiation exposure). The color change in polymer films along with the rate or change in radiation dose is a crucial factor in dosimetry studies (Gafar & El-Ahdal, 2014). The optical response characteristics of materials to radiation doses are critical parameters evaluated in radiation dosimetry studies. Each polymer film material typically shows different optical responses to radiation doses, related to optical absorption and energy gap (Saion et al., 2005; Susilawati et al., 2021). The threshold of optical absorption against radiation doses is studied to evaluate the maximum allowable radiation dose, also confirming data quantitatively on the color change of polymer films due to gamma radiation exposure (Susilawati, 2009). In dosimetry systems, the linearity of the energy gap (bandgap energy) is observed at each radiation dose, which can be linear, supralinear, saturation response, and even polymer film damage with increasing radiation doses (Horowitz, 2001, 2014). Finally, the stability of polymer films is related to storage time and testing whether the material undergoes changes in optical characteristics over time or not.

In our research, we employed PVA as the base material, incorporating methylene blue (MB) dye and trichloroacetic acid (TCA) to enhance the film's properties. The inclusion of TCA is particularly significant as it increases the dye's sensitivity within the polymer matrix, thereby improving the film's overall responsiveness to gamma radiation (Susilawati, 2009). Additionally, TCA acts as an electrocatalyst, contributing to the film's stability and functional performance (Dhara et al., 2016). To determine the feasibility of using PVA-MB-TCA polymer films as gamma radiation dosimeters, it is essential to thoroughly investigate their optical

properties. In this experiment, we conducted a comprehensive analysis of the PVA-MB-TCA polymer films subjected to gamma irradiation. We specifically examined the color change characteristics, optical absorption spectra, activation energy, and optical band gap energy of the films when exposed to gamma radiation doses up to 14 kGy. These parameters are crucial for understanding how the polymer films react to radiation and for assessing their potential use in dosimetry. Furthermore, we evaluated the stability of the polymer films both with and without gamma radiation treatment.

Objective of Study

The primary objective of this study is to investigate the optical properties of polyvinyl alcohol (PVA) based polymer films, incorporating methylene blue (MB) dye and trichloroacetic acid (TCA), for their potential application in gamma radiation dosimetry. Specifically, this research aims to explore the effects of gamma radiation on the color change characteristics, optical absorption spectra, activation energy, and optical band gap energy of the PVA-MB-TCA polymer films. Additionally, the study seeks to assess the stability of these polymer films under varying doses of gamma radiation, ranging up to 14 kGy. By thoroughly examining these parameters, the study aims to determine the feasibility and effectiveness of PVA-MB-TCA polymer films as reliable dosimetric materials for use in medical, industrial, and environmental applications where accurate radiation dose measurement is critical.

METHODS

The PVA-MB-TCA polymer films were created using polyvinyl alcohol (PVA), methylene blue (MB) dye, trichloroacetic acid (TCA), and color diluents (ethanol and NaOH) as the primary components. The solvent-casting technique was employed for the preparation of these samples. Initially, a dye solution was prepared by dissolving 0.08 g of MB crystals ($W = 25$ g granules/crystals, Merck, Germany) in 50 ml of ethanol (96% technical, Merck), along with 10% NaOH (0.1M, Merck). This mixture was stirred for 15 minutes at room temperature to ensure uniformity and then stored in a sealed container at room temperature.

To prepare the polymer solution, 3.5 g of PVA ($M_w = 72,000$ g/mol, Sigma-Aldrich, USA) was dissolved in 100 ml of distilled water in a beaker. This solution was heated to 100°C while being stirred with a magnetic stirrer at 200 rpm for 5 hours until the volume was reduced to 60 ml. Subsequently, 2 g of TCA ($M_w = 163.4$ g/mol, Sigma-Aldrich) was added to the PVA solution, and the mixture was stirred for an additional 30 minutes. The heating temperature was then lowered to 25°C, and the previously prepared MB dye solution was added and stirred for 20 minutes until a homogeneous mixture was obtained. This final solution (PVA-MB-TCA) was evenly spread onto a glass plate and left to dry for 120 hours at 25°C, resulting in solid polymer films. These films were then cut into 3 x 3 cm pieces and placed in special plastic envelopes to protect them from dirt, dust, and sunlight, maintaining a constant room temperature. The thickness of the polymer films was approximately 0.19×10^{-3} m.

The PVA-MB-TCA polymer films were subjected to gamma irradiation using the IRPASENA Irradiator with a Cobalt-60 source (type C-188, BARC, India), which has an activity of 80 kCi and an average energy of 1.25 MeV. The irradiation was

performed at the Research and Development Center for Isotope and Radiation Technology, National Nuclear Energy Agency, Indonesia. Fourteen polymer film samples were irradiated sequentially with radiation doses ranging from 1 to 14 kGy at room temperature. A control sample, which was not irradiated (0 kGy), was also included. Initial tests indicated that radiation doses below 1 kGy did not alter the color of the polymer samples, maintaining the same appearance as the non-irradiated sample. For applications in food dosimetry, a minimum dose of approximately 1 kGy is required.

The optical absorption measurements for the PVA-MB-TCA polymer films across all radiation doses were precisely conducted using a UV-Vis spectrophotometer (Genesys 180-type, USA), which operates with a double beam configuration and covers a wide wavelength range from 190 to 1100 nm, ensuring high accuracy with a wavelength precision of ± 0.5 nm. The choice of this instrument was driven by its capability to provide detailed spectral data, which is crucial for analyzing the subtle changes in optical properties induced by gamma radiation. The measurements focused on the specific wavelength range of 300 to 600 nm, where significant absorption peaks were anticipated based on the composition of the polymer films and the known absorption characteristics of the dye and polymer matrix. By systematically recording the absorbance across this range, we generated comprehensive absorption spectra for each sample. These spectra were then carefully plotted to illustrate the relationship between wavelength and optical absorption, allowing for a clear visual representation of how the absorption properties of the PVA-MB-TCA films change with varying radiation doses.

In the subsequent analysis, several key parameters were evaluated, including the critical dose required to induce color changes in the polymer films, the response of optical absorption to varying radiation doses, the absorption edge (AE), activation energy (ΔE), and the energy gap (E_g). To determine the absorption edge and activation energy, the Urbach edges method as described by Skuja et al. (2004) was employed. This method involves examining the exponential tail of the absorption edge to extract precise information on the activation energy, which provides insights into the disorder and electronic transitions within the material. Meanwhile, the optical energy gap, an essential parameter for understanding the electronic structure and behavior of the polymer films, was determined using the model proposed by Mott and Davis (2012).

RESULTS AND DISCUSSION

Color Change in Polymer Films Due to Gamma Radiation

The color of the PVA-MB-TCA polymer film samples exhibited significant changes before and after gamma radiation. Figure 1 illustrates various stages of the polymer samples. Increasing the radiation dose physically altered the color of the polymer film samples, changing from blue (0 kGy) to a yellowish-blue (8 kGy), and at the highest dose (14 kGy) becoming light blue (nearly transparent). In this study, we did not observe the morphological structure of the polymer film samples due to radiation, but the color changes due to radiation were very evident as a result of gamma-ray energy exposure at different doses.

The color change in the polymer films is considered to be due to a decrease in pH (increase in acidity) of the polymer film samples, resulting from the interaction of TCA with gamma rays. This is consistent with previous studies, which reported

that dry polymer films made of PVA mixed with chlorine-containing compounds (such as trichlorethylene) undergo pH degradation when irradiated with gamma rays, leading to color changes in the polymer film, where without radiation (0 kGy) the pH was found to be > 8.8 and after radiation (12 kGy) the pH ranged from 2.8 to 7.2 (Doyan et al., 2021). In our experiments, PVA polymer films with MB dye (without TCA) showed no color change even when irradiated with gamma rays up to 14 kGy. This serves as empirical evidence that only the TCA molecules in the polymer film samples are affected by gamma-ray irradiation within the radiation dose range established in this study.

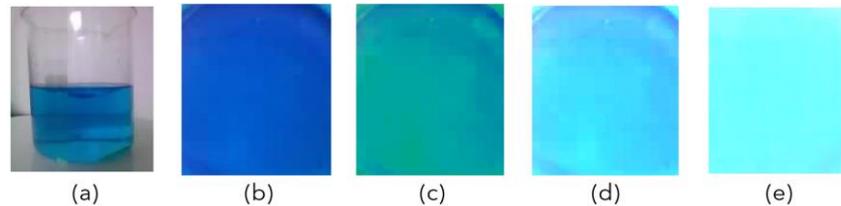


Figure 1. Samples of PVA-MB-TCA polymer films before and after gamma radiation: (a) liquid polymer sample before drying, (b) dried polymer film without radiation (0 kGy), (c) dried polymer film with 8 kGy radiation dose, (d) dried polymer film with 12 kGy radiation dose, and (e) dried polymer film with 14 kGy radiation dose.

Previous research has demonstrated that the blue color intensity in mixed polymer films (PVA-methyl thymol blue) progressively diminishes as the gamma radiation dose increases. This fading of color is linked to the generation of numerous free radicals as a result of radiation exposure, which in turn accelerates the degradation of the blue color within the polymer (Rabaeh et al., 2021). Gamma rays interact with the polymer, generating hydrated electrons and free radicals that damage the dye molecules and erase the chromophores, which are the parts of the molecules responsible for their color (Aldweri et al., 2017; Rabaeh & Basfar, 2020). This interaction results in a visible bleaching effect, where the blue color gradually fades as the radiation dose increases, indicating a loss of the dye's structural integrity and its chromophoric properties.

Additionally, the TCA (trichloroacetic acid) bonds in the mixed polymer films undergo a process known as dehydrochlorination due to gamma radiation. This process increases the concentration of chlorine ions within the polymer film (Susilawati, 2009). The presence of these chlorine ions, resulting from the breakage of TCA bonds, further contributes to the chemical changes in the polymer matrix under radiation. The combined effects of free radical formation and dehydrochlorination not only lead to the degradation of the dye molecules but also alter the overall chemical environment of the polymer film, enhancing the rate of color bleaching. This phenomenon underscores the significant impact of gamma radiation on both the physical and chemical properties of polymer-dye composites, making them effective for applications where radiation-induced changes need to be monitored.

Optical Absorption Spectrum of the Polymer Films

The optical absorption spectrum of the PVA-MB-TCA polymer film was analyzed using spectrophotometric techniques. Measurements were taken for the polymer film samples exposed to various radiation doses, from 0 to 14 kGy. The

results showed that the highest absorption was observed at three specific wavelength peaks: 360 nm, 440 nm, and 560 nm (Figure 2).

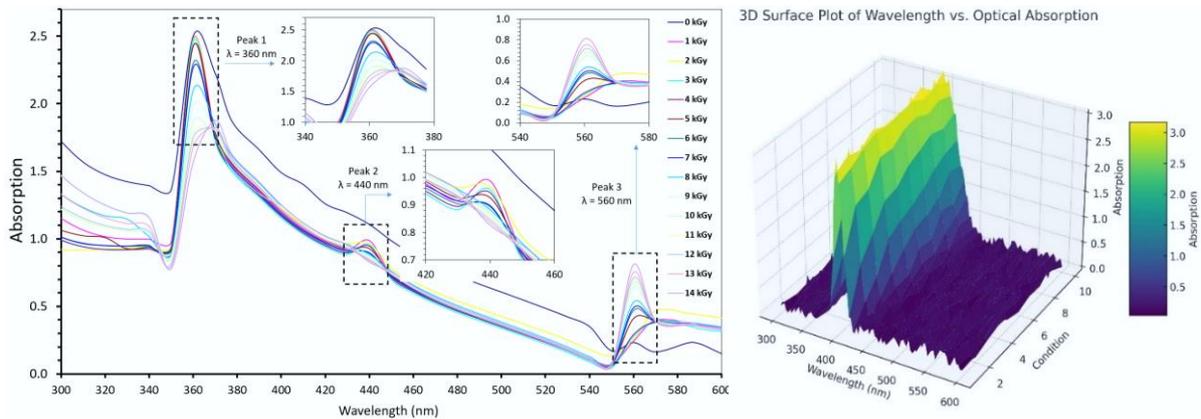


Figure 2. Relationship between wavelength (λ) and optical absorption (A)

The first peak is located in the ultraviolet (UV) region, while the subsequent two peaks are situated in the visible region of the spectrum (see Figure 2). The absorption trends at these three peaks (360 nm, 440 nm, and 560 nm) exhibit distinct behaviors as a result of gamma radiation, as illustrated in Figure 3. The UV peak at 360 nm is particularly sensitive to changes in radiation dose, often associated with electronic transitions within the polymer matrix. As the radiation dose increases, the absorption at this UV peak may either increase or decrease, indicating alterations in the polymer's electronic structure. In the visible region, the peaks at 440 nm and 560 nm correspond to the characteristic absorptions of the dye molecules within the polymer film. The absorption at 440 nm typically reflects the primary color change response of the dye to radiation, while the peak at 560 nm may indicate secondary interactions or changes in the dye's molecular environment. The variation in absorption at these visible peaks with increasing gamma radiation doses suggests a complex interaction between the dye molecules and the polymer matrix, influenced by the formation of free radicals and other radiation-induced chemical changes.

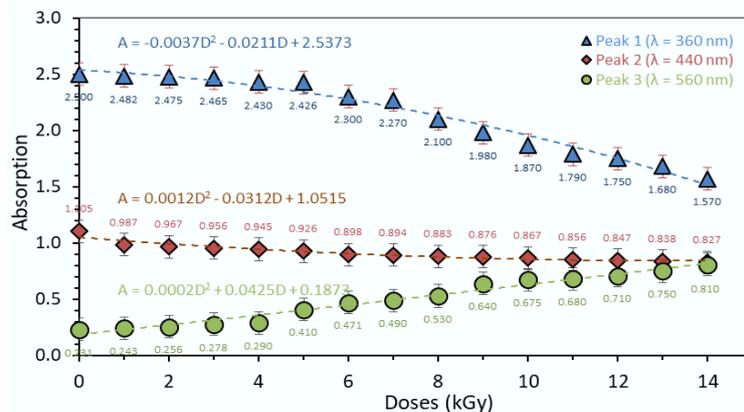


Figure 3. Relationship between radiation dose and optical absorption

The highest absorption peak occurs in the UV region at a wavelength of 360 nm, with a maximum absorption of 2.500 a.u. observed at 0 kGy. As the radiation

dose increases, this absorption value decreases, dropping to a minimum of 1.570 a.u. at 14 kGy. The relationship between absorption (A) and radiation dose (D) is characterized by the quadratic equation: $A = -0.0037D^2 - 0.0211D + 2.5373$. This trend indicates that gamma radiation significantly impacts the UV absorption characteristics of the PVA-MB-TCA polymer film, potentially due to the formation and interaction of free radicals within the polymer matrix, leading to changes in electronic transitions that are sensitive to UV light.

The second peak, found in the visible region ($\lambda = 440$ nm), also shows a decreasing trend in absorption with increasing radiation dose, although this decrease is less pronounced compared to the first peak. The maximum absorption at this peak is 1.105 a.u. at 0 kGy, which diminishes to 0.827 a.u. at 14 kGy. The relationship for this peak is given by the equation $A = 0.0012D^2 - 0.0312D + 1.0515$. This less significant decrease suggests that the dye molecules in the polymer matrix, which absorb light in the visible range, are less susceptible to radiation-induced damage compared to those absorbing in the UV range, possibly due to the different energy levels and stability of the involved chromophores.

Unlike the first two peaks, the third peak, located in the visible region at a wavelength of 560 nm, shows an increasing absorption trend with higher radiation doses. The absorption at this peak starts at a minimum value of 0.231 a.u. at 0 kGy and rises to a maximum of 0.810 a.u. at 14 kGy. The relationship for this peak is described by the equation $A = 0.0002D^2 + 0.0425D + 0.1873$. This increasing trend may be attributed to the formation of new absorbing species or structural changes within the polymer matrix that enhance the absorption at this wavelength. Notably, at the highest radiation dose of 14 kGy, the maximum absorption at the third peak nearly equals the minimum absorption at the second peak, indicating a balance between radical formation and consumption in target reactions (Rabaeh et al., 2021).

The absorption spectra for the first and second peaks exhibit a decreasing trend with increasing radiation doses, while the third peak shows an increase as the radiation dose becomes higher. These findings contrast with previous studies, which identified the absorption peaks of chlorine-containing organic compounds in PVA-trichloroethylene (PVA-TCE) polymer films within the visible spectrum at wavelengths of 438 nm and 575 nm (Doyan et al., 2021). In those studies, the absorption at 438 nm was observed to rise with radiation doses ranging from 7 to 12 kGy, whereas the absorption at 575 nm declined with radiation doses from 0 to 6 kGy at a 35% TCE concentration in the polymer film. The key similarity in our current study is the optical absorption response to gamma radiation doses, highlighting the varying effects of radiation on different polymer-dye compositions.

The colorimetric properties related to the gamma radiation dose response at these optical absorption peaks are crucial factors in radiation dosimetry studies (Gafar et al., 2018). These properties provide a visual and quantifiable means of assessing the amount of radiation absorbed by the material. The shift in color and corresponding changes in optical absorption at specific wavelengths allow for precise monitoring of radiation doses, which is essential for ensuring safety and efficacy in various applications. Previous studies have emphasized that these colorimetric changes are vital for determining a material's suitability as a radiation dosimeter, as they offer a straightforward and reliable method for dose

measurement (El-Kelany & Gafar, 2016; Gafar et al., 2017; Gafar & El-Ahdal, 2014). Materials that exhibit clear and consistent color changes in response to different radiation doses can be effectively used in diverse fields, including medical, industrial, and environmental monitoring, where accurate radiation dose assessment is critical.

For dosimetry applications in food processing, our study identifies that the PVA-MB-TCA polymer film irradiated within the dose range of 1 to 14 kGy is highly suitable. This range is particularly relevant to food processing, where different radiation doses are applied to achieve various objectives such as delaying ripening, reducing microbial load, and ensuring food safety (Eskin & Robinson, 2001). The observed colorimetric response within this dose range indicates that the PVA-MB-TCA polymer film can provide reliable and precise measurements, making it a valuable tool for controlling and monitoring radiation exposure in food products. Additionally, our findings reveal that at radiation doses above 14 kGy, the polymer film becomes nearly transparent and indistinguishable, rendering it ineffective for further use as a dosimeter. This transparency at higher doses highlights the limitations of the material and underscores the importance of defining an optimal dose range for its application. Thus, the PVA-MB-TCA polymer film not only meets the requirements for effective dosimetry in the specified dose range but also provides insights into its operational limits, ensuring its practical applicability in food processing and other relevant fields.

Activation Energy and Gap Energy of Polymer Films

Activation energy in a chemical reaction refers to the minimum energy needed to start the reaction. This energy is necessary to form a transition state during the collision of reactant molecules (Otero & Martinez, 2011). For the mixed PVA-MB-TCA polymer film, we established the relationship between $h\nu$ (eV) and $\ln(\alpha)$, as shown in Figure 4. This relationship is critical for understanding how the polymer's electronic properties change with radiation exposure. Furthermore, we examined the correlation between activation energy (ΔE) and radiation dose, as shown in Figure 5. This analysis provides insights into how gamma radiation influences the energy barriers that need to be overcome for reactions within the polymer matrix to occur. Additionally, we explored the energy gap (E_g) for each radiation dose, as illustrated in Figure 6. The energy gap is a crucial parameter that indicates the electronic properties of the polymer film and how these properties are modified by radiation.

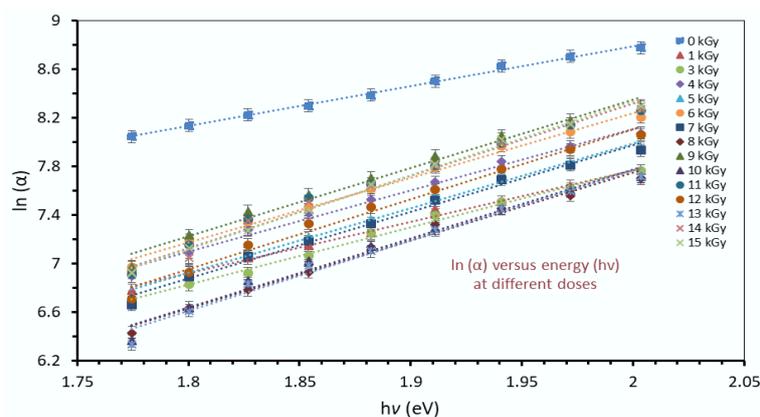


Figure 4. Relationship between $h\nu$ (eV) and $\ln(\alpha)$

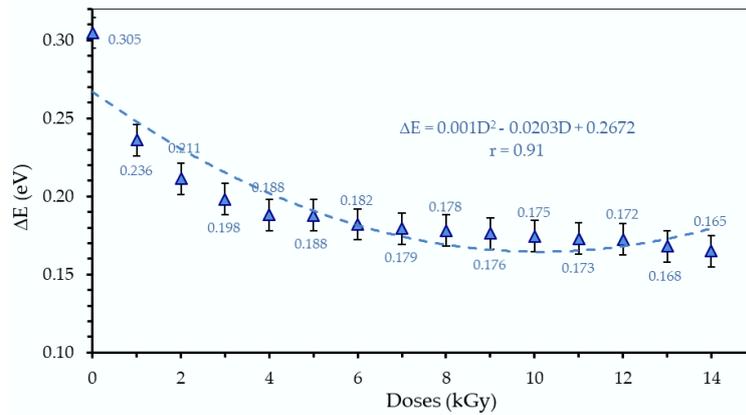


Figure 5. Relationship between activation energy (ΔE) and radiation dose (kGy)

Activation energy (ΔE) was determined following the Urbach edges method (Skuja et al., 2004) based on UV spectra. The optical activation energy was derived by drawing a straight line (exponential function) from the slope of the graph plotting $\ln(\alpha)$ against photon energy ($h\nu$) (Figure 4). The intersection points of this line (as an exponential function) at each $h\nu$ represent the ΔE values for each radiation dose (Figure 5). The results indicate that the activation energy of the mixed PVA-MB-TCA polymer film before irradiation was 0.305 eV, and it showed a decreasing trend with increasing radiation doses. The relationship between ΔE and radiation dose (D) is described by the quadratic equation $\Delta E = 0.001D^2 - 0.0203D + 0.2672$, with a correlation coefficient (r) of 0.91. This trend reflects the decreasing energy barriers for the polymer's internal reactions as the radiation dose increases, culminating in a minimum activation energy of 0.165 eV at the highest dose of 14 kGy.

These findings align with previous studies, which also observed a decrease in activation energy with increasing gamma radiation doses (Susilawati, 2009). This reduction in activation energy is attributed to the chain-scission of polymeric molecules within the polymer samples (Singh & Neerja, 2007). As the radiation dose increases, the molecular chains in the polymer break down, leading to lower energy requirements for further reactions. This phenomenon highlights the significant impact of gamma radiation on the structural and electronic properties of the polymer film. Understanding this behavior is crucial for developing the PVA-MB-TCA polymer film as an effective dosimetric material.

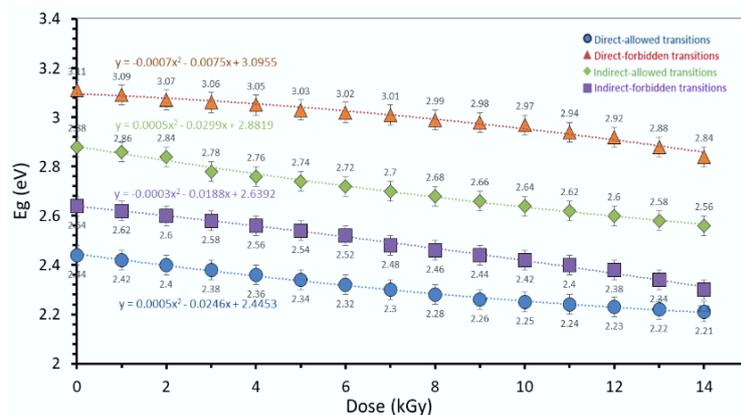


Figure 6. Energy gap (E_g) per radiation dose (kGy)

Band gap energy (E_g) refers to the energy range in a material where no electron states exist; it lies between the valence band and the conduction band (Isac, 2014). For an electron to transition from the valence band to the conduction band, it must acquire sufficient energy to overcome this gap (Aziz et al., 2020). Analyzing the optical absorption spectrum can provide insights into the optical band gap (E_g) between these bands, accounting for both direct and indirect transitions (Costa et al., 2016). This analysis is crucial for understanding the electronic properties of materials and their potential applications in various fields, such as optoelectronics and radiation dosimetry.

The optical band gap (E_g) was determined following the Mott and Davis model (Mott & Davis, 2012). This approach involves extrapolating $(\alpha hv)^m$ versus hv to obtain the band gap values for different radiation doses. Specifically, four types of optical transitions were considered: $(\alpha hv)^{1/2}$ ($m^{-1/2} \text{ eV}^{1/2}$) for allowed direct transitions; $(\alpha hv)^2$ ($m^{-2} \text{ eV}^2$) for allowed indirect transitions; $(\alpha hv)^{3/2}$ ($m^{-3/2} \text{ eV}^{3/2}$) for forbidden direct transitions; and $(\alpha hv)^{1/3}$ ($m^{-1/3} \text{ eV}^{1/3}$) for forbidden indirect transitions (Escobedo-Morales et al., 2019). These various transitions provide a comprehensive understanding of how the polymer's electronic structure responds to gamma radiation. The study's results, illustrated in Figure 6, show that the band gap energy decreases with increasing radiation doses for all types of transitions.

The observed decrease in band gap energy with higher radiation doses is attributed to the increased formation of polarons and free ions within the polymer samples due to gamma radiation exposure (Aziz et al., 2020; Meftah et al., 2014). These radiation-induced species disrupt the electronic structure of the polymer, reducing the energy required for electronic transitions and thus lowering the band gap. This reduction in band gap energy has significant implications for the material's optical and electronic properties, affecting its potential applications in areas where precise control of electronic behavior is necessary. Understanding these changes is essential for optimizing the use of PVA-MB-TCA polymer films in radiation dosimetry, as it highlights the material's sensitivity to radiation and its capacity to undergo significant electronic alterations under exposure.

This findings indicate a clear dependency of the energy gap in irradiated polymer materials on the doses of γ -ray irradiation. Prior research has demonstrated a linear decrease in the energy gaps of CR dyed PVA-TCE films subjected to γ -ray irradiation within the 0 to 12 kGy dose range (Doyan, 2021). Similarly, a linear reduction in the energy gap was observed for KCl-Mn and KCl-Ce phosphorus polymer materials irradiated with γ -rays at doses between 0.08 and 0.75 kGy, resulting in energy gaps of 4.16 eV and 4.34 eV, respectively (Talebi, 2019). This suggests a consistent pattern in which the optical properties of these materials are affected by γ -ray exposure, underlining the material's potential adaptability for various applications, particularly in radiation dosimetry.

Understanding the behavior of optical properties in materials subjected to radiation is crucial for identifying their suitability in radiation dosimetry systems. These materials typically exhibit diverse responses to radiation doses, including linear, supralinear, and saturated responses, as well as potential defects at higher doses (Horowitz, 2014). In the present study, both direct and indirect transitions of the energy gap showed a linear decrease with increasing radiation dose. This linearity suggests that the optical properties of the materials remain stable under

irradiation, reinforcing their potential as reliable components in dosimetry applications (Doyan, 2021; Talebi, 2019). The stability and predictability of the optical response to γ -ray doses are essential attributes for materials used in precise radiation measurement systems.

This results affirm that the energy gap in irradiated polymer materials is significantly influenced by gamma irradiation doses, with previous studies and the current work consistently showing a linear response in the decrease of energy gaps. This linearity is crucial as it indicates stable optical properties, essential for the materials' application in radiation dosimetry systems. By maintaining a predictable response to varying radiation doses, these materials can be effectively utilized in dosimetry, offering precise and reliable measurements. The potential applications of such materials in radiation detection and measurement technologies are vast, given their demonstrated stability and linear response to irradiation.

CONCLUSION

The PVA-MB-TCA polymer film has been introduced as a potential material for gamma radiation dosimetry applications. This study explored the optical properties of the polymer film before and after exposure to gamma radiation up to a dose of 14 kGy. The results demonstrated that increasing the radiation dose physically alters the color of the polymer film samples, changing from blue (0 kGy) to nearly transparent light blue (14 kGy). Spectrophotometric analysis revealed that the maximum absorption occurred at three distinct wavelength peaks: 360 nm, 440 nm, and 560 nm. The absorption values decreased with increasing radiation doses at 360 nm and 440 nm, while the absorption at 560 nm showed an opposite trend, increasing with higher radiation doses. These findings indicate a complex interaction between the dye molecules and the polymer matrix under gamma radiation, affecting the film's optical properties significantly.

Furthermore, the study found that the activation energy of the polymer film decreased as the radiation dose increased. This trend suggests that gamma radiation lowers the energy barriers for internal reactions within the polymer matrix, likely due to the chain scission of polymeric molecules. Additionally, the band gap energy of the polymer film also decreased with increasing radiation doses for all types of transitions. This reduction in band gap energy is attributed to the formation of polarons and free ions within the polymer samples caused by gamma radiation exposure, which disrupts the electronic structure of the polymer. These changes in activation energy and band gap energy highlight the sensitivity of the polymer film to gamma radiation and its potential effectiveness as a dosimetric material.

The implications of this study on the optical characteristics of PVA-MB-TCA polymer films for gamma radiation dosimetry are significant. The demonstrated sensitivity of the polymer films to gamma radiation, evidenced by the distinct color changes and alterations in optical absorption, activation energy, and band gap energy, highlights their potential as effective dosimetric materials. These properties enable precise monitoring and control of radiation doses, which is crucial for applications in various fields, including food processing, medical sterilization, and radiation safety. The ability of the polymer film to provide reliable dose measurements within a specific range (1 to 14 kGy) makes it a valuable tool for ensuring the safety and efficacy of radiation processes. Furthermore, understanding the film's limitations, such as the loss of effectiveness at doses above 14 kGy, allows

for the optimization of its use in practical applications, enhancing its utility in industries that rely on accurate radiation dosimetry.

RECOMMENDATION

Based on the findings of this study, it is recommended that further research be conducted to optimize the composition and processing conditions of PVA-MB-TCA polymer films to enhance their sensitivity and stability as gamma radiation dosimeters. Specifically, exploring the effects of varying the concentrations of methylene blue dye and trichloroacetic acid, as well as testing additional dye combinations, could provide deeper insights into improving the dosimetric performance of these films. Additionally, long-term stability tests and real-world application trials in diverse environments, such as food processing and medical sterilization, should be carried out to validate the practical effectiveness and reliability of these polymer films. These steps will help to refine the material's properties and expand its potential applications, ensuring it meets the stringent requirements of radiation dosimetry in various industries.

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