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A simple gamma dosimeter using a film label made of polyvinyl alcohol and bark of *Peltophorum ferrugineum* extract

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Abstract. Gamma radiation is considerably dangerous, but at certain doses, it can be beneficial to humans, for example, in food sterilization applications. Therefore, the development of simple and practical dosimeter devices is essential. Herein, we developed a simple dosimeter label made of *Peltophorum ferrugineum* bark extract separated from its fat content and mixed in a polyvinyl alcohol (PVA) matrix (prepared using an ammonium peroxide crosslinker). The film obtained from this mixture had a yellowish-brown color that changed to dark brown after gamma irradiation. The film label structure was investigated via Fourier transform infrared spectroscopy, and the absorbance properties of the film labels were measured using Ultraviolet-Visible (UV-Vis) spectrophotometer. The intensity of the color change was quantified using a colorimeter. The response of the film label to gamma radiation was measured via irradiation at 0–66 kGy. We found that gamma radiation caused a decrease in the intensity of the R and G color components of the film. Additionally, swelling tests demonstrated that the films exhibited good plastic properties after gamma irradiation. These results suggest that film labels made of PVA-extracted *P. ferrugineum* can be used as a simple dosimeter label for sterilization purposes.

Keywords: Dosimeter, Bark of *Pelthophorum ferruginum* , PVA

1. Introduction

The enormous energy of gamma radiation can be used for sterilization purposes in the food and medical equipment industries. The use of radiation for food sterilization must adhere to international standards [1], and the absorbed dose of radiation is a crucial factor to observe. Dosimeters, which are used to determine the exposure to gamma rays and other types of ionizing radiation, play an important role in providing this information. There are two types of dosimeters: *active dosimeters*, which can read data immediately, and *passive dosimeters*, which need further data processing.

Radiochromic devices are a type of passive dosimeter that is often used to measure the exposure to ionizing radiation. Such dosimeters have been developed in the form of gels, liquids, and films and can be applied in a varying dose range. Gel-type devices irradiated by low-dose (1–15 Gy) ^{60}Co gamma rays have been prepared using ferrous xylenol orange gel for radiotherapy measurements [2]. Radiochromic dosimeters for medium absorption doses (10–1000 Gy) have also been developed using synthetic nitro blue tetrazolium chloride (NBT). Such devices have been used for food radiation, pest control, and other applications, as reported by in a previous study [3]. Dosimeters for high absorption doses (1–125 kGy), obtained from diethyl fumarate (DEF) in a dimethylformamide (DMF) solution, have also been studied for routine dosimetry purposes [4].



Development and innovation in the fabrication of dosimeters are crucial factors for monitoring radiation exposure. Materials for dosimeters comprise a matrix and a coloring agent. In previous studies, plastic film matrixes made of polyvinyl alcohol (PVA) [5], polyvinyl chloride [6], polyvinyl butyral [7], and other polymers have been used for this purpose. PVA has been chosen for this research because it is a water-soluble and non-toxic biodegradable synthetic polymer [8], and these safety parameters are important for radiation on food and medical equipment.

The coloring agents for radiochromic dosimeters used in previous studies were synthesized from synthetic matter. Natural coloring agents that have already been used in previous research include turmeric roots, walnut bark, and henna leaves in gel form [9]. Natural coloring agents of local origin are mostly used for textiles and batik processes. One of them is synthesized from the bark of *Peltophorum ferrugineum*, which is taxonomically classified into the *Fabaceae* family that includes the *Caesalpinioideae* sub-family containing flavonoids and tannins [10,11].

The purpose of this research is to produce radiochromic film labels from a PVA matrix using the natural coloring agent synthesized from the bark of *P. ferrugineum*. This should result in color changes in the film label after irradiation by high-dose gamma rays. The film label can thus function as a simple dosimeter, which can be used to sterilize food and medical equipment at permitted dose levels.

2. Experimental

2.1. Preparation of film labels using *P. ferrugineum* extract

The *P. ferrugineum* bark used as the coloring agent was obtained from local production. The bark was first dried at 40°C for 48 h to remove water and then milled using a dry milling process to obtain a powder. Then, 40 g of this powder was mixed with 150-ml ethanol (J.T Baker; 100%) and stirred at 300 rpm and 55°C for 4 h using a magnetic stirrer. Next, the solution was filtered using Whatman No. 1 paper. The resulting solution of the *P. ferrugineum* extract had a yellowish-brown color.

Film labels were then made using this coloring extract and PVA. Fully hydrolyzed PVA, which was used as the matrix, was obtained from Merck, Germany. The PVA powder (2 g) was diluted in 100 ml (w/v) of distilled water, and the resulting solution was stirred at 300 rpm and room temperature until it reached homogeneity. The solution was then mixed with ammonium peroxodisulfate [APS; (NH₄)₂S₂O₈] at 4% of the PVA weight. APS, obtained from Emsure, Germany, served as a crosslinker for PVA. The PVA–APS solution was then stirred at 200 rpm and room temperature for 4 h. Next, 50 ml of the solution was mixed with 10 ml of brown *P. ferrugineum*, after which the mixture was stirred for 15 min. This final solution was poured into an acrylic mold (size: 15 cm × 15 cm) and dried at room temperature for 48 h to synthesize the film labels.

2.2. Characterization of the dosimeter film

A Thermo Genesys 105 UV/Vis spectrophotometer was used to measure the maximum absorbances of a PVA film label and a film label containing *P. ferrugineum* at wavelengths between 300 and 800 nm. The optical spectra were measured before and after irradiation, and the Origin 9.1 software package was used to calculate the maximum absorbances and optical spectra for both film labels.

The RGB intensity of film labels containing *P. ferrugineum* was measured using a colorimeter purchased from Nix Pro, Canada. The transparent film labels were placed on a board with a white background, and the intensity measurements were performed five times to obtain the standard deviation. The total RGB intensity was calculated using the luminance formula (Y) and the color variance recommended by the International Telecommunication Union ITU-R BT.601-7:

$$Y = 0.299(R) + 0.587(G) + 0.114(B). \quad (1)$$

The molecular structures of the bonds in the film labels were determined via Fourier transform infrared (FTIR) spectroscopy (Thermo Scientific, Nicolet iS5). The tested samples included film labels made of PVA, PVA–APS, PVA–APS–*P. ferrugineum* before being irradiated at 0 kGy and PVA–APS–*P. ferrugineum* after being irradiated at 66 kGy. The spectra were recorded at 400–4000 cm⁻¹. Absorbance spectra of all the four film labels were then compared. Origin 9.1 was used to illustrate the film labels' spectra.

Swelling tests were performed on the *P. ferrugineum* extract film labels before and after gamma irradiation. Both film labels were weighed separately using an analytical balance and then washed with distilled water for 30 min. Next, they were kept aside and reweighed. These steps were repeated for washing periods of 60, 90, and 120 min.

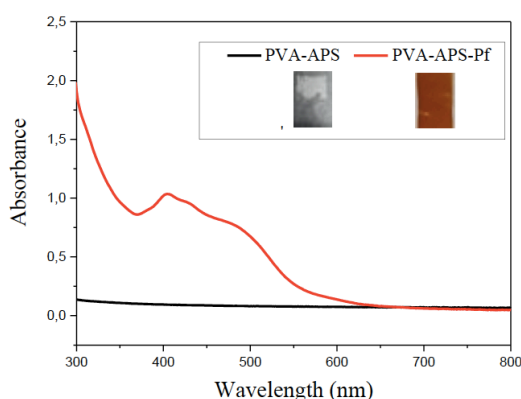


Figure 1. Absorbance spectra of polyvinyl alcohol (PVA) film labels and film labels containing *Peltophorum ferrugineum*.

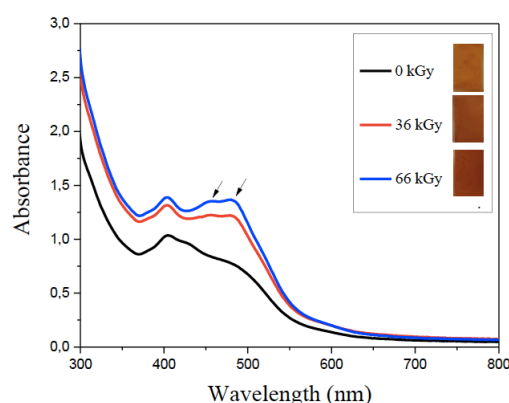


Figure 2. Absorbance spectra of film labels containing *P. ferrugineum* before and after irradiation at varying doses.

The film labels containing *P. ferrugineum* were cut into small pieces (2 cm × 3 cm) and irradiated using a Gamma Cell ^{60}Co device at the Irradiation Facility owned by BATAN, Jakarta. The radiation dose of the gamma cell was set to 6 kGy/h with a measurement range of 36–66 kGy. The irradiated *P. ferrugineum* film labels were then analyzed for color changes using the UV/Vis spectrophotometer, colorimeter, and FTIR equipment.

3. Results and discussion

3.1. Absorbance spectra of the film labels

The absorbance spectra of the PVA film labels and the film labels containing *P. ferrugineum* prior to gamma irradiation are shown in figure 1. The absorbance spectra of the transparent PVA film labels show a straight line without any peaks in the wavenumber range 300–800 nm. On the contrary, the film labels containing *P. ferrugineum* show an absorbance peak at 403 nm. This wavelength at 403 nm shows a complimentary color of yellowish green. These results show that the color of the PVA film labels, which are identified as the matrix, does not affect the film labels containing *P. ferrugineum*, which exhibit a yellowish-brown color. The colors of the film labels are dominated by the dark color of the bark extract.

The absorbance spectra of film labels containing *P. ferrugineum*, irradiated at 36 and 66 kGy, can be shown in figure 2. These film labels undergo a visually perceptible color change: their yellowish-brown color changes to solid brown after irradiation. This color change phenomenon in films containing synthetic colors, such as methyl viologen [12], bromothymol blue (BTB) [13], and methyl orange [14], when irradiated with a high dose of gamma rays has been reported before. The film label spectra reported herein show an increase in absorbance parallel to an increase in the gamma-ray dose. Gamma radiation ionizes the film labels and creates hydrogen radicals from PVA [14]. Additionally, the optical spectra of the irradiated film labels show a different maximum absorbance in comparison with the spectra taken before irradiation, and this spectral difference is marked by the appearance of new peaks at 455 and 482 nm, which show a color change from yellowish brown to solid brown. However, the FTIR results for all four-film labels do not show different peaks after irradiation.

3.2. RGB Intensity of film labels containing *P. ferrugineum*

The film labels containing *P. ferrugineum* were evaluated to determine the relative RGB intensities before and after irradiation, as illustrated in figure 3. After irradiation, the R, G, and total RGB intensities of the film labels decreased depending on the given radiation dose, while the intensity of B remained considerably low. It can be observed that the irradiation dose is inversely proportional to the intensity of R and G, with the relative intensity of G undergoing a more significant decrease compared to the other values. This result is consistent with the color changes in the film labels (from yellowish

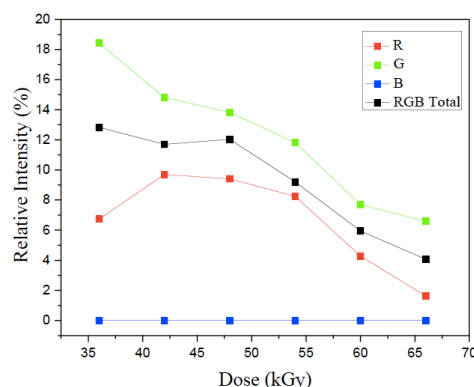


Figure 3. Δ Relative intensities of film labels containing *P. ferrugineum* in the dose range 36–66 kGy.

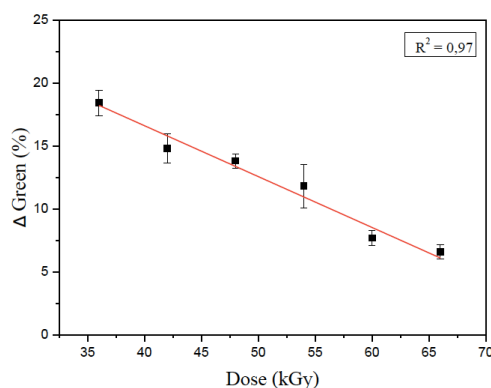


Figure 4. Linear fit of the green intensity (with standard deviation) for film labels containing *P. ferrugineum*.

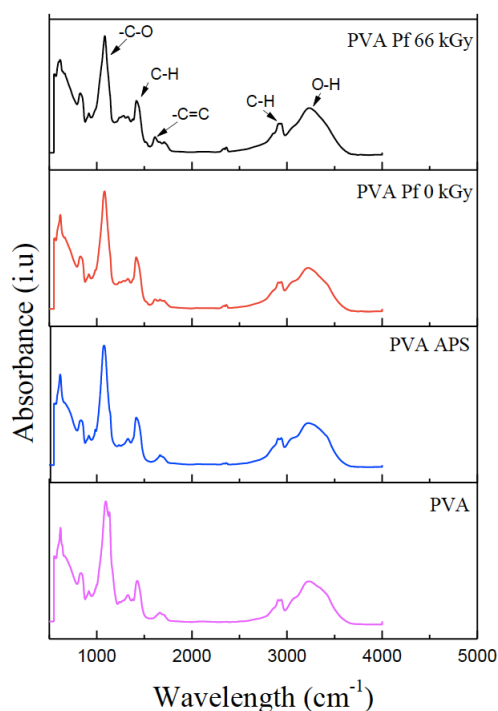


Figure 5. FTIR spectra of the PVA, PVA–ammonium peroxodisulfate (APS), and PVA–APS–*P. ferrugineum* samples irradiated at 0 kGy and that of a PVA–APS–*P. ferrugineum* sample irradiated at 66 kGy.

brown to brown), as indicated by the peak observed at 482 nm in the optical spectra of the samples. The spectrum intensity (RGB) of the colorimeter depicts that the intensity of G on the film labels reflects the sensitivity to detect gamma-ray detection; the linear compatibility that is observed to be $R^2 = 0.97$ for the relative intensity of G at different doses also supports this analysis (figure 4). The standard deviation appears different for each dose, which is due to two factors: homogeneity differences in the film labels and the stability of the colorimeter.

3.3. FTIR characterization of film labels containing *P. ferrugineum* extract

The FTIR spectra of the PVA, PVA–APS, and PVA–APS–*P. ferrugineum* film labels taken before and

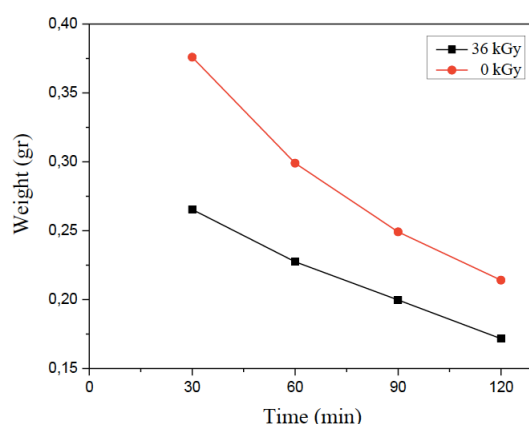


Figure 6. Swelling tests on film labels containing *P. ferrugineum* before (red) and after irradiation (black).

after gamma irradiation at 66 kGy are shown in figure 5. The absorbance spectra show the same pattern for all the film labels at 1089, 1410, 1611, 2918, and 3226 cm^{-1} , and they are identical to the spectra obtained by Sonker *et al.* [8] who studied the effects of the crosslinker on PVA. In both cases, the PVA pattern could be observed for all four-film labels. The signals observed at 1089, 1611, 2918, and 3226 cm^{-1} can be assigned to the stretching vibrations of the $-\text{C}-\text{O}$, $-\text{C}=\text{C}$, $\text{C}-\text{H}$, and $\text{O}-\text{H}$ bonds, respectively. There also exists a band at 1410 cm^{-1} , which corresponds to the bending of the $-\text{CH}$ bond. The wide absorbance band observed at 3226 cm^{-1} is due to $\text{O}-\text{H}$ stretching from the hydroxyl group, which illustrates the hydrophilic properties of PVA [15]. This is also supported by the results of the swelling test. The FTIR characterization results observed from the film labels in this research are depicted in figure 5. No change is observed in the spectral form both before and after irradiation. Similarly, film labels containing PVA extract and *P. ferrugineum* exhibit the dominant PVA spectrum. This is strongly influenced by the composition of label making that is observed between PVA and *P. ferrugineum*.

3.4. Swelling tests

Results of the swelling tests for the film labels containing *P. ferrugineum* before and after gamma irradiation are depicted in figure 6. The weight of the film labels that is observed before and after irradiation is observed to decrease with the increasing test time. This was because some of the PVA molecules that was not well-crosslinked by APS dissolved in water. The weight loss for the film labels before irradiation was observed to be sharper than the weight loss observed in case of the film labels after irradiation. During the irradiation process, the film labels became hot so that the water molecules that were absorbed by the film would evaporate.

4. Conclusions

The plastic film labels made of PVA-APS and natural dye from *P. ferrugineum* extract has been successfully prepared as a simple dosimeter. The color of the film labels changed from yellowish brown to solid brown after gamma irradiation at a dose of 36–66 kGy, followed by a linear decrease in the relative intensity of the green color component ($R^2 = 0.97$). All the characterizations performed in this study showed positive results so that the PVA-APS-*P. ferrugineum* extract film labels could be used as a simple dosimeter label.

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