PAPER • **OPEN ACCESS**

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To cite this article: P Widodo *et al* 2019 *IOP Conf. Ser.: Mater. Sci. Eng.* **496** 012006

IOP Conf. Series: Materials Science and Engineering **496** (2019) 012006 doi:10.1088/1757-899X/496/1/012006

Radiochromic film label made from chitosan and *Taraxacum officinale* leaf extract

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Abstract. Monitoring the dose of gamma-ray radiation is important in the healthcare field. A simple indicator for dose monitoring is provided by radiochromic labels. This paper reports the preparation and characterization of radiochromic labels prepared from the natural dye of *Taraxacum officinale* (TO) leaf extract mixed with a chitosan matrix. The radiochromic label was fabricated in the form of a colored plastic biodegradable film. The film structure was characterized by Fourier transform infrared spectroscopy. The color of the film label was quantitatively measured with a UV-Vis spectrophotometer at 433 and 630 nm. The ability of the labels to detect gamma-ray radiation was evaluated for a dose range from 0 to 30 kGy. Gamma-ray radiation caused changes in the color intensity of the film labels. Spectral analysis revealed that the absorbance increased when the film label was exposed to gamma-ray radiation. The results of these experiments show that the radiochromic film label made from TO leaf extract in a chitosan matrix can be used as a radiochromic indicator to determine the dose of gamma radiation and is a good candidate for use in the sterilization of health equipment.

Keywords: Radiochromic film label, Taraxacum officinale, chitosan

1. Introduction

In the industrial field, ionizing radiation is used to improve the quality of a product because it can change the physical, chemical, and biological properties of irradiated objects. Ionizing radiation is used to sterilize health products [1], inhibit budding, promote disinfestation of insects and pathogens, prolong shelf life, and sterilize foodstuffs and agricultural products [2] and modify materials by inducing polymerization [3].

The absorption dose plays an important role in the irradiation process; thus, its monitoring is essential [4]. The "dose" refers to the energy absorbed per unit mass [5]. Various dosing tools have been developed, including radiochromic films, chemical solutions, calorimeters, fluorescent systems, organic crystals, inorganic crystals, semiconductors and diamonds [6].

A radiochromic film label comprises two parts: a dye and a matrix. Several studies have reported the use of dyes as a radiation indicator; examples include leuco brilliant blue cyanide [7], calcein [8], leucocrystal violet [9], and leucomalachite green [10]. The matrix is the material used to immobilize the dyes. Materials used as a matrix include polyvinyl alcohol, films of PVA [8], polyvinyl butyryl, films of PVB [9], solutions, ethanol as a solvent [11], and gels, agarose as gelling agent [12].

In the present study, we report the preparation of a radiochromic film label made from *Taraxacum officinale* (TO) leaf extract immobilized with a chitosan matrix. The TO extract is a biodegradable and nontoxic natural dye. Chitosan is a copolymer derived from the diacetylene chitin process; it is also biodegradable and nontoxic and can form a film [13].

2. Materials and methods

2.1. Materials
The materials used in these experiments were TO leaves extract, ethanol (J.T. Baker), chitosan in powder form (Pusat Penelitian Bioteknologi dan Bioindustri (PPBB), Bogor, Indonesia; degree of deacytelation >80%), and glacial acetic acid (Merck).

2.2. TO leaf extraction
TO leaves were washed with tap water and then dried in an oven at 40 °C for 72 h. The dried leaves then milled using dry milling until they became powder. TO leaf extraction was performed by mixing 40 g of leaf powder with 200 mL of 100% ethanol, and then stirred using a magnetic stirrer at 300 rpm for 24 h at room temperature. The solution was filtered using Whatman No. 1 filter paper. The pH of the resultant extract was 6.2.

2.3. Film label preparation
The chitosan matrix was prepared by dissolving 4 g of chitosan powder into 200 mL of 1% (w/v) acetic acid and stirring the resultant mixture at 300 rpm for 3 h at 55 °C using a magnetic stirrer. The chitosan–TO film label was prepared by mixing 10 mL of TO extract solution with 50 mL of chitosan solution and stirring the resultant mixture for 15 min at room temperature using a magnetic stirrer. The chitosan–TO solution was poured onto a 15 × 15 cm^2 acrylic board and left for 48 h at room temperature to form the film.

2.4. Collection of UV-Vis absorption spectra
Absorption spectra were recorded for the TO extracts, chitosan film label, nonirradiated chitosan–TO film, and the chitosan–TO film label, which were tested for exposure to gamma-ray radiation over the dose range 3–30 kGy. The measurements were performed using a UV-Vis spectrophotometer (Thermo Genesys 10S) in the wavelength range 200–800 nm.

2.5. Film-label irradiation tests
The chitosan–TO film-label response to exposure to gamma-ray radiation was examined using a Gammacell Irradiator 220 (Nordon, Canada), which had a dose rate of 5.7 kGy/h. The dose rate of the Co-60 source in the Gammacell Irradiator 220 was calibrated using a Fricke dosimeter. The chitosan–TO test label, which was 2 × 3 cm^2 in area, was placed on a special holder. The film label was then exposed to gamma-ray radiation with doses of 3, 6, 12, 15, 21, 27, and 30 kGy at chamber temperature.

2.6. FTIR absorption spectroscopy
Fourier transform infrared (FTIR) absorption spectra were collected for the TO extract solution, chitosan–TO film label, and the chitosan–TO film label irradiated at a dose of 30 kGy. Before the spectra were recorded, the TO extract solution was evaporated until it exhibited an opaque color; subsequently, the solution was dripped onto the sample holder. Neither the nonirradiated chitosan–TO film label nor the chitosan–TO film label irradiated at a dose of 30 kGy were subjected to a special treatment prior to measurement. Measurements were conducted using a double-beam spectrophotometer (Thermo Nicolet iS5) in the attenuated total reflectance (ATR) Fourier transform mode. The FTIR absorption spectra were recorded for 500–4000 cm^{-1}.

3. Results and discussion

3.1. Absorption spectra of TO extracts and chitosan film labels
The TO extract solution had a pH of 6.2 and was dark green. Figure 1 shows the absorbance spectra of the TO extract solution, chitosan film label, and the chitosan–TO film label. The absorbance spectrum

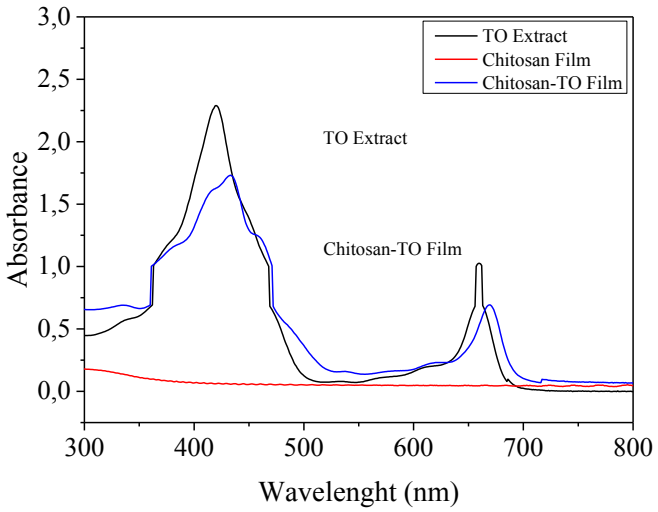

Figure 1. Absorbance spectra of the TO extract, chitosan film, and the chitosan–TO film.

Figure 2. The color changes of chitosan–TO film labels after irradiation.

of the TO extract solution exhibited maximum absorption peaks at 420 and 660 nm. The peak at about 420 nm corresponds to the absorption of purple light and the reflection of yellow-green light, whereas the peak around 660 nm corresponds to the absorption of red light and the reflection of green-blue light. The combination of green-yellow and green-blue colors reflection produces a dark-green color, as shown in figure 1. The chitosan film label has a flat absorption spectrum because of its transparent and colorless nature. The absorbance spectrum of the chitosan–TO label exhibited two absorption peaks at wavelengths of 433 and 670 nm. The absorption peak of the chitosan–TO film label shifted compared with the absorbance peak of the TO extract. The peak at about 433 nm corresponds to blue absorption and yellow reflection, whereas the peak around 670 nm corresponds to red absorption and green-blue reflection. The combination of yellow and green-blue colors reflection produces a lighter green color of the chitosan–TO film label compared with that of the TO extract.

3.2. Absorption spectra of chitosan–TO film labels at various radiation doses

The chitosan–TO film labels respond to exposure to gamma-ray radiation. The color of the chitosan–TO film labels darkened in proportion to the increased absorbed radiation dose. The color changes of chitosan label colors irradiated with doses of 3–30 kGy are shown in figure 2. The film labels irradiated with a dose of 30 kGy exhibit a darker green color compared with other labels irradiated with lower doses.

The chitosan–TO film label response is also shown in the irradiated-film-label absorption spectra. The UV-Vis absorbance spectra of the chitosan–TO film irradiated at doses from 3 to 30 kGy show absorption peaks at 433 nm and 670 nm (figure 3). No absorption peak shift is observed in the absorption spectra. The absorption peak increases inversely proportional to increasing radiation dose. The higher the absorption peak, the darker the color appears. Soliman *et al.* [14] reported similar results wherein the film color intensity darkened in proportion to increasing radiation dose.

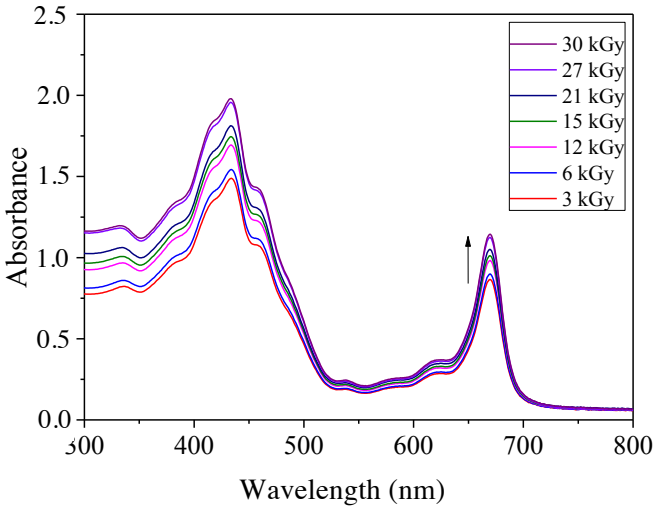

Figure 3. UV-Vis absorption spectra of irradiated chitosan–TO-based film labels.

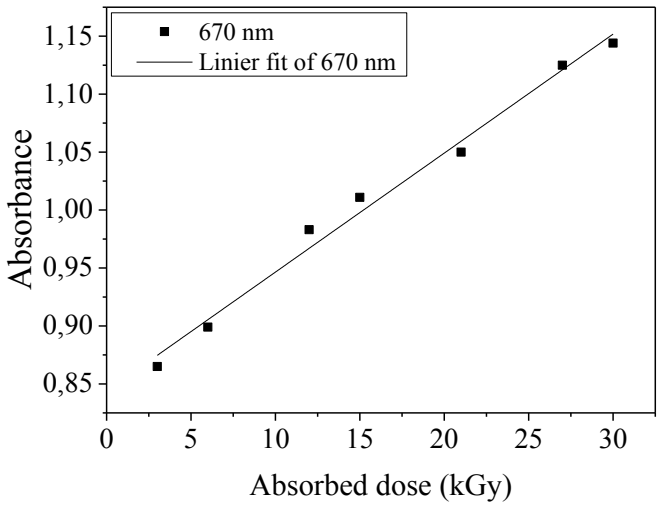

Figure 4. Absorbance of chitosan–TO film labels at a wavelength 670 nm after the film labels were subjected to radiation doses from 3 to 30 kGy.

3.3. Response of the chitosan–TO film label to radiation

The absorption peak representing the response of the chitosan–TO film label to the radiation dose absorbed by the film label were obtained at a wavelength of 670 nm. Figure 4 shows the absorbance response of the chitosan–TO film label to the radiation dose absorbed by the film label in the 3–30 kGy range. The absorbance point value on the graph increases nonlinearly in proportion to the increased radiation dose absorbed by this film label. The points on the graph can be approximated using the linear function described in equation (1):

$$Absorbance = 0.843 + 0.010(Absorbed\ dose(kGy)) \quad (1)$$

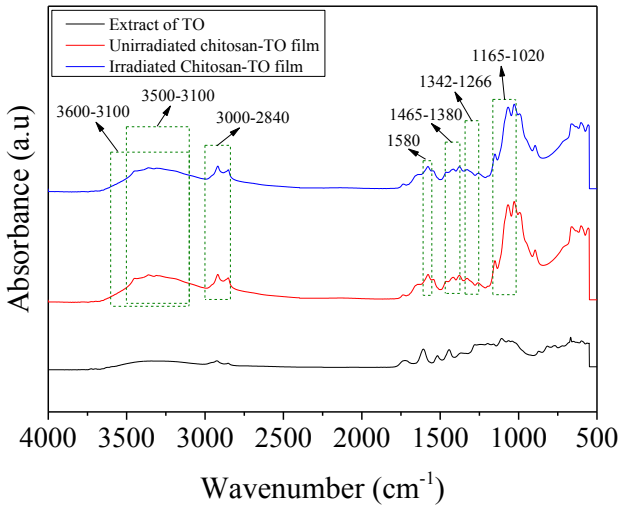

Figure 5. FTIR spectra of the TO extract, a nonirradiated chitosan–TO film label, and a chitosan–TO film label irradiated at 30 kGy.

Equation (1) has a coefficient of determination value of 0.986, which shows that the absorbance value is 0.986 dependent on the absorbed dose value.

3.4. FTIR spectra of the chitosan–TO film label

The chemical structure of the chitosan–TO film label before and after irradiation was characterized via FTIR spectroscopy. Figure 5 shows the FTIR spectrum of the TO extract, the nonirradiated chitosan–TO film label and the chitosan–TO film label irradiated at a dose of 30 kGy. There is no TO extract peak observed in the FTIR spectrum of the chitosan–TO film labels because the volume of TO extract in the chitosan–TO film labels is much smaller than the chitosan matrix volume. In the spectra of the nonirradiated chitosan–TO film labels and the chitosan–TO film labels irradiated at 30 kGy, the O–H stretching vibration is observed in the wavenumber range 3600–3100 cm^{-1}, which overlaps with an N–H stretching vibration at 3500–3100 cm^{-1} [15]. In the range of 3000–2840 cm^{-1}, the stretching vibration of C–H is experienced [16,17]. The N–H bending absorption band also appears at 1580 cm^{-1} [15]. The C–H bending, C–N stretching, and C–O stretching bands appear in the wavenumber ranges of 1465–1380 cm^{-1}, 1342–1266 cm^{-1}, and 1165– 1020 cm^{-1}, respectively. No substantial change was observed in the spectrum of the chitosan–TO film label before and after irradiation at a dose of 30 kGy.

4. Conclusions

Radiochromic film labels prepared from TO leaf extract on a chitosan matrix were used as radiochromic indicator to determine the dose of gamma radiation in the dose range from 3 to 30 kGy. The color of the chitosan–TO film label developed was dark green. The intensity of the color of the chitosan–TO film labels darkened in proportion with the radiation doses. Changes in the color intensity of chitosan–TO film labels were confirmed by increases in the intensity of the absorption peaks at 433 nm and 670 nm in proportion to the radiation doses. The absorbance peaks can be approximated using equation (1) with a coefficient of determination of 0.986 for chitosan-TO film labels.

Acknowledgements

This research was completed with a funding by the PITTA (Publikasi Internasional Terideks untuk Tugas Akhir) Grant in 2017 from Universitas Indonesia. The authors would like to thank Einago (www.einago.com) for the English language review.

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