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The Use of Tragacanth as a Gelling Agent in Roselle Flower Extract based Radiochromic Indicators

Goib Wiranto [a,*], Slamet Widodo [a], I Dewa Putu Hermida [a], Yudi Y. Maulana [b], Deni P. Kurniadi [b], Shenny Oktaviana [c], Atiek R. Noviyanti [c], Juliandri [c]

[a] *Research Center for Electronics, National Agency for Research and Innovation (BRIN), Indonesia*
[b] *Research Center for Telecommunication, National Agency for Research and Innovation (BRIN), Indonesia*
[c] *Department of Chemistry, Faculty of Mathematics and Natural Sciences, Padjajaran University, Indonesia*
*Corresponding author: *gwiranto@gmail.com*

***Abstract*—Gamma irradiation techniques have been used in various fields, such as sterilizing medical equipment and food products. In such a process, monitoring the dose of radiation using radio radiochromic dosimeter is important to guarantee the accuracy of the radiation dose absorbed by the irradiated materials. Radio chromic dosimeters usually consisted of radiation-sensitive dyes that could change colors when exposed to radiation. In this research, natural dyes in the form of anthocyanins obtained from roselle flower extract (Hibiscus sabdariffa L.) have been used as radio chromic indicators. The radio chromic indicators have been made in liquid using water solvent and gel using a tragacanth gelling agent. Each radiochromic indicator's pH values varied between 2 and 11 using NaOH and HNO_3 solutions. All radio chromic indicators were then gamma irradiated using a Co-60 radiation source and tested for stability at 8°C under closed storage conditions. The characterization was performed using UV Vis spectrophotometry. The results showed that roselle flower extract was sensitive to gamma radiation as indicated by decolorization, and their sensitivity increased when made in the form of gel at pH 2 and 5. The stability test also showed that the anthocyanin content was stable after being maintained for 28 days in a closed storage at 8°C. With such differences in sensitivity of the roselle flower extract when used in either liquid or gel solution, the radio chromic indicators should find wide application in the future, depending on the radiation doses given to the solutions.**

***Keywords*—Radio chromic indicator; tragacanth gel; roselle flower extract; gamma radiation; natural dyes.**

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I. INTRODUCTION

Gamma irradiation is an ionizing radiation process arising from the decay of radioactive materials such as Co-60 or Ni-60. Due to the many advantages such as high penetrating power to the target materials, non-residual, clean, and cold process [1], gamma irradiation is often used for sterilization of food products [2]–[4] and medical equipment [5]**.** The gamma radiation technique's success depends on the radiation dose received by the target materials. According to Indiarto et al. [3], an inaccurate radiation dose could cause imperfect or damaged products. Monitoring the radiation dose has become crucial to maintaining product quality. In fact, the guidelines for treating food products using gamma radiation have been specified under ISO 14470:2011.

A dosimetry system has been used to detect and monitor the amount of radiation dose using sensitive materials being exposed to radiation, and one of the measurement methods is known as radio chromic in which indicators inside a dosimetry system provide information about radiation dose based on color changes of the sensitive materials. The degree of colorization will vary depending on the radiation dose the materials receive [6]. Such dosimeter devices are popular because the measurement principle is simple instant, and changes in radiation dose can be observed visually [7]. In addition, it can be used to detect gamma radiation doses up to 1000 Gy [8].

A radio chromic dosimeter thus requires dyes that serve as a colorization indicator, and the degree of color changes of the used dyes will be directly proportional to the amount of radiation dose absorbed by the dyes [9], [10]. However, based on the international standard (ISO 51540: 2004), measurement results from a radio chromic dosimeter must be obtained using a spectrophotometry or photometry system. This is because the colorization process in a radio chromic dosimeter is an optical density change phenomenon and must

be analyzed using UV-Vis spectrophotometry. Dye indicators in a radio chromic dosimeter can be used in a liquid, gel, or film form, each of which should have a different responsibility to the same radiation dose. This variation capability has made a radio chromic dosimeter being applied in many fields [8], [11].

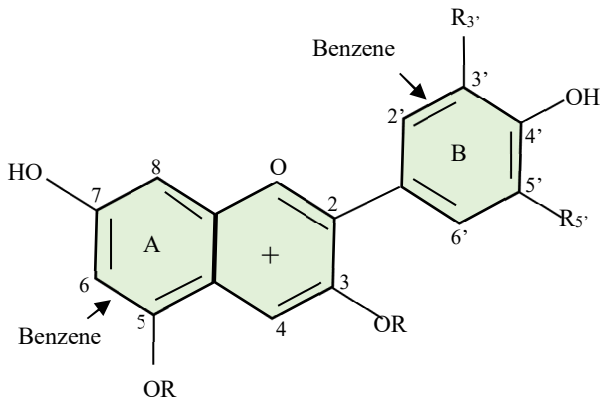
Several radio chromic dosimeters have been available commercially such as FTR-125 by Fujifilm using cellulose triacetate (CTA) [12], FWT-60 by Far West Technology using hexa (hydroxyethyl) pararosaniline cyanide (HPR-CN) [13], B3 by GEX using pararosaniline leucocyanide [14], and Gafchromic using polydiacetylene (PDA) [15]. Several researchers have also used synthetic dyes as radio chromic indicators, such as thymol blue [16], xylenol orange [17], dithizone [10], and methyl red [9]. The use of synthetic dyes has some limitations because of their negative impacts on the environment and being carcinogenic and not easily degradable. Such negative impacts have prompted using natural dyes, such as chlorophyll extracted from dandelion leaves [18], which are more environmentally friendly. The use of anthocyanins as radiochromic dyes has also been demonstrated because of their potential for color degradation upon radiation. These anthocyanins have been extracted from red cabbage [19] and from rosella flower [20], [21].

According to Enaru et al. [22], the stability of anthocyanin structure is highly dependent on environmental conditions, such as the degree of acidity, temperature, radiation exposure, and even the presence of oxygen. Changes in the structure of anthocyanin lead to colorization that can be detected from the shift of the absorbance peaks. As such, this research was intended to develop radio chromic indicators based on natural dyes using anthocyanins extracted from roselle flowers (Hibiscus sabdariffa L.). In addition, a gelling agent made from tragacanth gum was used as a medium, for it was known to be non-toxic and have pH-dependent viscosities.

II. Materials and Method

A. Materials (Anthocyanin)

Anthocyanins are colored water-soluble pigments found in red, orange, blue, purple, and black fruits and vegetables. Examples of anthocyanin pigments can be seen in tomatoes, carrots, and cherry fruits [23], and in flowers such as rosella and rose, red cabbage, and berry fruits [24]. The structure of anthocyanins is polyphenols since they are a subgroup of flavonoid compounds. As shown in Figure 1, the basic chemical structure is flavylium cation (2-phenyl-1-benzopyrilium), which links hydroxyl (-OH) and/or methoxyl (-OCH3) groups and one or more sugars. It has two benzene rings connected by three carbon atoms. In this case, the sugar-free molecule is called anthocyanidins. Depending on the number and position of hydroxyl and methoxyl groups attached to R3' dan R5' positions, various anthocyanidins can be formed, and six are commonly found in vegetables and fruits: pelargonidin, cyanidin, delphinidin, petunidin, peonidin, malvidin.



Type of Anthocyanidin	R_3	R_5
Delphinidin	OH	OH
Petunidin	OH	OCH_3
Malvidin	OCH_3	OCH_3
Cyanidin	OH	H
Peonidin	OCH_3	H
Pelargonidin	H	H

Fig. 1 General structure and various types of anthocyanidins

The sugar group in anthocyanins can be found in glucose, rhamnose, galactose or arabinose [24], [25] and can be detected within the UV range at a wavelength of around 275 nm. The anthocyanin peak lies within the visible range at 475 – 550 nm [23].

The existence of conjugated bonds of the chromophore groups in the anthocyanin structure has allowed light absorption in the visible range through the delocalization of electrons. The length and number of the conjugated bonds determine the depth of the anthocyanin color. Hence, the larger number and longer bonds also mean a stronger color that allows anthocyanin to absorb light within the UV and visible range. In other words, the required energy for the electron transition is smaller in the conjugated bonds, such that light absorption occurs within the longer wavelengths. In this research, the anthocyanins have been prepared from roselle flower extract.

1) Roselle flower (Hibiscus sabdariffa L.)

Roselle is a species of flower plant originating from Africa, but nowadays can be found in most countries in Asia, the Caribbean, Central and South America, and Hawaii. Roselle is rich in anthocyanins, making it highly potential as a natural medicine for cardiovascular, hypertension, cancer, etc. As shown in Figure 2, the anthocyanins within roselle flower are dominated by dark red pigments, making it suitable for making natural dyes. These types of red anthocyanins contain delphinidin-3-O-sambubioside, cyanidin-3-O-sambubioside, delphinidin-3-O-glucoside, and cyanidin-3-O-glucoside [26]. Roselle flowers can be extracted using the maceration method to obtain the main compound within the flower. The maceration extraction method is a separation process done by immersion of samples that have been previously ground using a polar solution such as water or alcohol.

Fig. 2 Petals of dried roselle flower [27]

Closed storage at cold temperature is required for more than 24 hours with less frequent stirring. The maceration method is an effective technique when the compound to be extracted has the characteristic of being thermolabile or easily degradable at high temperatures. This technique is also time-consuming and requires a large volume of solvent solution. Tazoho et al. [20] have demonstrated this maceration technique to extract roselle flowers using ethanol solution.

2) Tragacanth (Astragalus gummifer)

Tragacanth is a natural gum obtained from the dried sap of the tragacanth plant, which is part of the Middle Eastern legumes of the genus Astragalus. Tragacanth plants can be found in Asia, Turkey, Afghanistan, and Iran. Commercially, Tragacanth is packed in small pieces of 0.5 – 2.5 mm thick or in powder shape. It is transparent, smell-free, tasteless, viscous, and color in white to yellowish. The application of Tragacanth in industrial, food, and pharmacy fields is mostly as an emulsifier, stabilizer, and thickener. Its superior water-absorbing qualities make it an excellent agent in increasing solution viscosity.

As shown in Figure 3, tragacanth gum contains mixtures of L-fucose, D-xylose, and D-galactose and small amounts of cellulose, starch, and protein. The viscosity of gel formed by tragacanth increases as the temperature and concentration increase. In contrast, the viscosity of tragacanth gel decreases with increasing pH values.

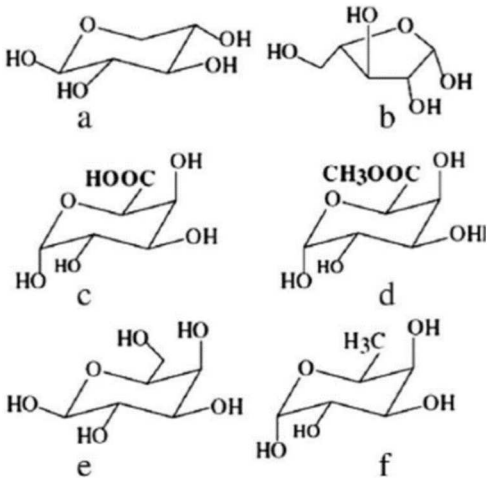


Fig. 3 Chemical structure of the tragacanth gum, a) β -D-xylose, b) L-arabinose, c) α -D-galacturonic acid, d) α -D- galacturonic acid methyl ester, e) β -D-galactose, f) α -L-fucose [28]

B. Method

Figure 4 shows the general steps in this research, which involved extraction of roselle flower, synthesizing radio chromic indicators, gamma irradiation of the radio chromic indicators, stability testing, and characterization using UV-Vis spectrophotometer.

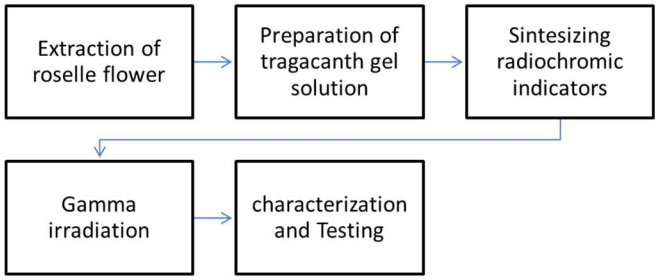


Fig. 4 Steps involved in the development of radio chromic indicators using Roselle Flower extract and Tragacanth as a gelling agent

3) Material preparation

The main materials used in this research were dried roselle flowers and tragacanth gum. Other chemicals used were 70% ethanol, 65% HNO3, NaOH, and aqua bides. Laboratory tools needed to develop the radio chromic indicators include various glassware such as reagent bottles, glass stirrers, petri dish, glass funnel, beaker glass, measuring cup, Whatman filter paper, disposable cuvette, mortar and pestle, drop pipette, thermometer, 2 ml-microtubes, rotary evaporator, and aluminum foil. In addition, for further material processing and characterization, the required equipment includes hot-plate with magnetic stirrer, digital scales (AND, EK 610), pH meter (LAQUA 1100), micropipette with volume of 100–1000 µL (WATSON), tissue rupture (QIAGEN), Spectrophotometer UV Vis (GENESYS 10S), and Gamma cell 220 irradiator.

4) Extraction of roselle flower

The roselle flower has been extracted using the maceration method to obtain natural dyes. Initially, dried roselle flower was ground manually using mortar and pestle. Then, 50 g of the resulting roselle powder was mixed with 1000 ml of 70% ethanol solvent inside a Reagan bottle. The solution was then stored in cold storage at 5 – 8 °C with aluminum foil covering the bottle. After 24 h, the mixture was filtered using Whatman paper, and the resulting filtrate was then concentrated using a rotary evaporator. The concentrated roselle extract was mixed with aqua bides to prepare the radio chromic indicators to get a 2.5% roselle solution. Finally, NaOH was added to the roselle extract solution to obtain variation in the pH values from 2 – 11. All the resulting anthocyanin solution indicators were then characterized using UV-Vis Spectrophotometer.

5) Preparation of tragacanth gel solution

Tragacanth gel solution was prepared by mixing tragacanth powder with aqua bides in 1:40 (2.5% w/v) composition. Initially, 1 g of tragacanth powder was added by 40 ml aquabides that had been previously heated at 70°C using a hot-plate. After 10 min, the mixture was stirred using tissue rupture at 15000 rpm for 15 min. A uniform, soft, and clear gel solution was resulted after the process. Then, the tragacanth gel solution was mixed with aqua bides containing 0.1 M HNO3 to obtain a gel solution with pH value of 2. Likewise, the gel solution was mixed with aquabides

containing 0.1 M NaOH to obtain a solution with pH value of 8. This technique prepared tragacanth gel solution at pH values of 2, 5, and 8.

6) Radio chromic indicators with roselle extract and tragacanth gel solution

The radio chromic indicators have been prepared by mixing the roselle extract and tragacanth gel solution with a ratio of 1:1 for each pH value of the tragacanth gel, thus, a total of 21 samples resulted. Constant stirring for 10 min was required to get a homogenous mixture. Each sample was finally prepared in a 2-ml glass tube and ready for gamma irradiation.

7) Gamma irradiation and characterization

The gamma irradiation process has been performed in the Center for Application of Isotope and Radiation, National Agency for Nuclear Energy (BATAN), using Gamma cell 220 with a radiation source of Co-60. With a radiation speed of 3.6 kGy/h, all radio chromic indicator samples were irradiated with 1, 2, 4, 6, 10, 15, and 20 kGy doses. The optical properties of each sample were characterized before and after the gamma radiation using UV-Vis spectrophotometer. The colorization of the radio chromic indicators was observed within the visible range between 400 – 800 nm spectrums.

Fig. 5 Color change of the roselle extract solutions due to variation in pH values

8) Stability test of the radio chromic indicators

The final observation of the radiochromic indicators was performed to study the effect of environmental conditions on the anthocyanins. In this case, samples with roselle extract solutions were stored in closed storage at a temperature of 8°C for 28 days. The absorbance spectrum of each sample was then measured at the end of day 28.

III. Results and Discussion

A. Identification of Anthocyanin in Roselle Extract

The roselle extract mixed with aquabides resulted in a dark red color solution with an average pH value of 2.51. At this low pH value, the roselle extract solution was in an acidic condition. The addition of 0.1 M NaOH was able to increase their pH value up to 11. At every increase in pH value, the color of the solution changed from dark red to more and lighter red. This proved the existence of anthocyanins in the roselle extract solutions, as shown by their sensitivity to pH change. Figure 5 shows the colorization of the roselle extract samples.

At pH values of 2 – 4, the color of the roselle extract solutions was red, indicating that the anthocyanin structures were in flavylium cation form. At pH 5, the color slightly changed to lighter red, possibly due to the presence of colorless carbinol pseudo-base structures. As the pH increased to 6 and 7, the solution color turned purplish red, which means that the anthocyanins have formed quinoidal base structures known to have blue color. From pH 8 to 11, the roselle extract solutions entered basic (alkaline) conditions, where the anthocyanins were forced to degrade by forming a chalcone structure, as indicated by their yellow color. The work of Rakića et al also confirmed these phenomena.[29].

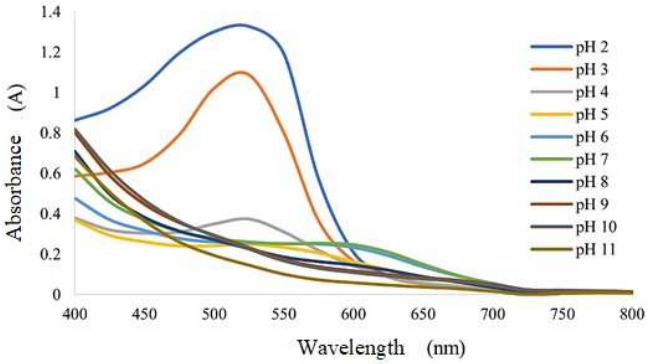


Fig. 6 UV-Vis spectra of the roselle extract solutions at pH values of 2 - 11

Figure 6 shows the UV-Vis spectra of the roselle extract solutions at various pH values. The peak of the absorbance occurs at a wavelength of 525 nm. According to Koulani et al. [30] and Indah et al. [21], the peaks corresponding to the anthocyanins in the roselle extract solution should be found between 475 – 550 nm. With the change in pH value of the solution, the absorbance peaks have shifted in intensity but still stayed between the same wavelength regions. Based on these results, the existence of anthocyanins in the roselle extract were identified.

B. Radio chromic Properties of the Roselle Flower Extract

After the gamma irradiation process to the roselle extract solutions, visual observation was performed to see the effect of radiation on the anthocyanin. A radiochromic property is the ability of a compound to change color in response to radiation exposure. Visually, these phenomena can be seen from the color degradation of the roselle extract solutions. As shown in Figure 7, at radiation doses of 1 – 2 kGy, the color of the solution was dark red but lighter than the color before radiation. As the dose of radiation increases to 4 – 6 kGy, the lighter color change is observed and becomes significantly colorless as the dose of radiation approaches 20 kGy).

Fig. 7 Color change of the roselle extract solutions due to gamma radiation

The results were confirmed by the absorption spectra obtained using UV-Vis spectrophotometer. As can be seen in Figure 8, the roselle extract solutions have experienced a gradual downshift in their absorbance peaks because of radiation exposure. The decrease in the absorbance peaks can

be considered as a percentage of decolorization that can further be calculated using equation [31]

$$\% \text{ decolorization} = \left[\frac{A_0 - A_i}{A_0}\right] \times 100 \quad (1)$$

where A_0 and A_i are the absorbance values of the materials before and after radiation exposures, respectively. Using the above equation, the roselle extract solutions started to experience a significant decolorization at a radiation dose of 4 kGy, with a drop in absorbance reaching 52% compared to its original state. The decolorization percentage increases with increasing gamma radiation dose and reaches 86% at a radiation dose of 20 kGy.

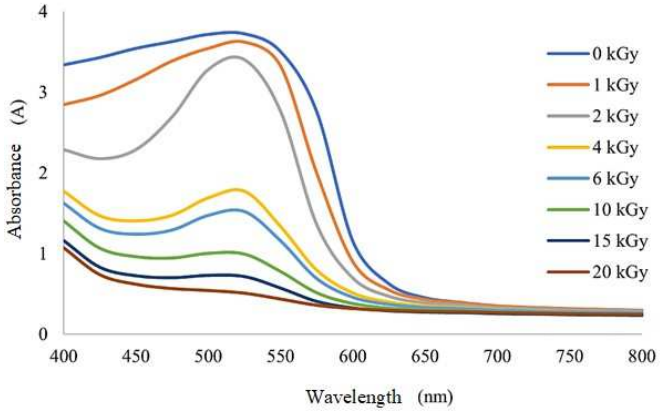


Fig. 8 UV Vis spectra of the roselle extract after gamma radiation at various doses

The response curves from the UV Vis spectra described how individual roselle extract solutions responded to radiation. Since the absorbance peak of the irradiated solution occurred at 525 nm, measuring the response of each solution is equivalent to calculating the difference between the absorbance peaks before and after radiation. The resulting net absorbance can then be plotted as shown in Figure 9, in which a significant increase in net absorbance was started at a radiation dose of 4 kGy. The net absorbances were still increasing at higher radiation doses, but less significantly.

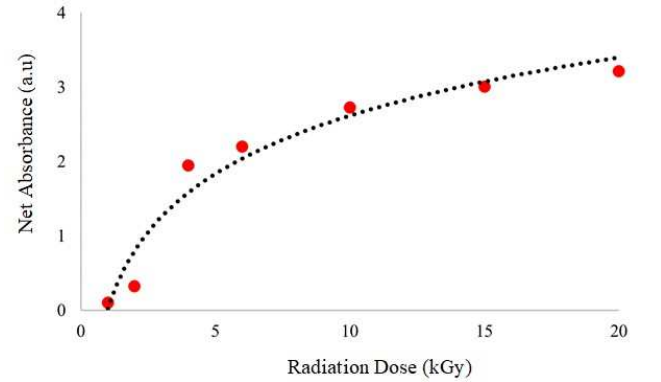


Fig. 9 Net absorbance curve of the roselle extract as a response to gamma radiation.

The sensitivity values of the roselle extract to gamma radiation can be determined from the slope of the net absorbance curve. However, due to the non-linear shape of the net absorbance curve, the linear slope can be obtained by dividing the curve into three regions. The first region represents a small radiation dose of less than 1 kGy, in which the sensitivity of the roselle solution was calculated as 0.099/kGy. The second region represents a medium radiation dose between 1 to 4 kGy, where the sensitivity of the solution was found to be 0.645/kGy. The third region has a high radiation dose above 4 kGy. The sensitivity of the solution in this region was 0.079/kGy. Since the highest sensitivity occurs at the medium dose region, it can be said that the anthocyanin content of the roselle extract was highly degraded after 4 kGy radiation dose.

Comparison with previous studies showed that a reduction in anthocyanin content after gamma radiation also occurred when sumac fruit extract was used as a radiochromic indicator [32]. According to Tena et al. [33] and Choong et al. [34], the chromophore groups in the anthocyanin acted as coloring agents and antidotes to free radicals. The process of opposing radical compounds is done by the conjugated bonds of the chromophore through electron delocalization. In addition, the hydroxyl (OH) groups at the benzene rings also oppose the radical compounds by donating hydrogen atoms (H+) and transferring electrons to the radical compounds. Because of the interaction between chromophore and hydroxyl groups with radical compounds, anthocyanin becomes unstable as a result of electron deficiency. This unstable form of anthocyanin can cause conjugated bonds to break so that the roselle extract solution will experience decolorization. The higher the radiation dose, the freer radicals created during the process of water radiolysis. Consequently, the number of anthocyanins reacting with the free radicals also increases and therefore the roselle extract solution color will be more degraded.

C. Radiochromic Properties of the Roselle Extract in Tragacanth Gel

The observation aimed to study the effect of Tragacanth as a gelling agent for roselle extract solutions. The initial result showed that roselle flower extract mixed with tragacanth gel has an absorbance peak at 526 nm, as shown in Figure 10. The slight increase indicated that the tragacanth gel did not affect the anthocyanin in the roselle extract.

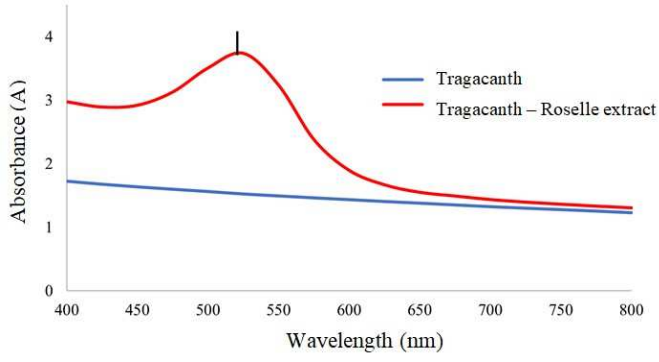


Fig. 10 UV Vis spectra of Tragacanth and Tragacanth – roselle flower extract

After gamma irradiation, the Tragacanth – roselle extract gel solution experienced color degradation as can be seen in Figure 11. Visually, the Tragacanth – roselle extract gel at pH 2 and pH 5 showed significant color degradation. At pH gel solution of 8, the color of the Tragacanth – roselle extract turned from red to brown.

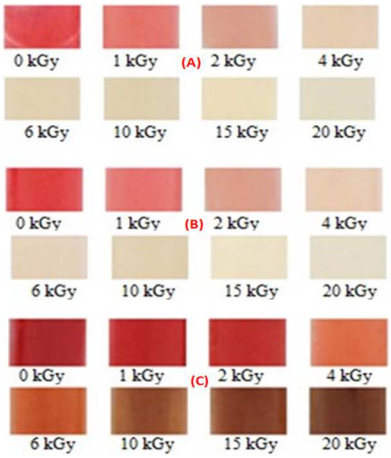


Fig. 11 Color change of the Tragacanth – roselle flower extract due to gamma radiation, (a) at pH 2, (b) at pH 5, (c) at pH 8

Figure 12 shows the UV-vis spectra of the radiated Tragacanth – roselle flower extract solutions. At a radiation dose of 1 kGy, the Tragacanth – roselle flower extract solution with pH 2 and 5 showed an absorbance decrease of 43%, which decreased to 90% at 20 kGy radiation dose. In the case where the solution with pH value of 8, however, a significant decrease of 53% in absorbance occurred after 4 kGy radiation dose, followed by color changes and a shift in the absorbance peaks as the radiation doses reached 20 kGy. The insignificant response of the Tragacanth–roselle extract gel at pH 8 was due to the low viscosity of the tragacanth gel in the basic (alkaline) phase. As mentioned, tragacanth gel would have low viscosity in the basic phase and was less effective than in the acidic phase in responding to radiation doses.

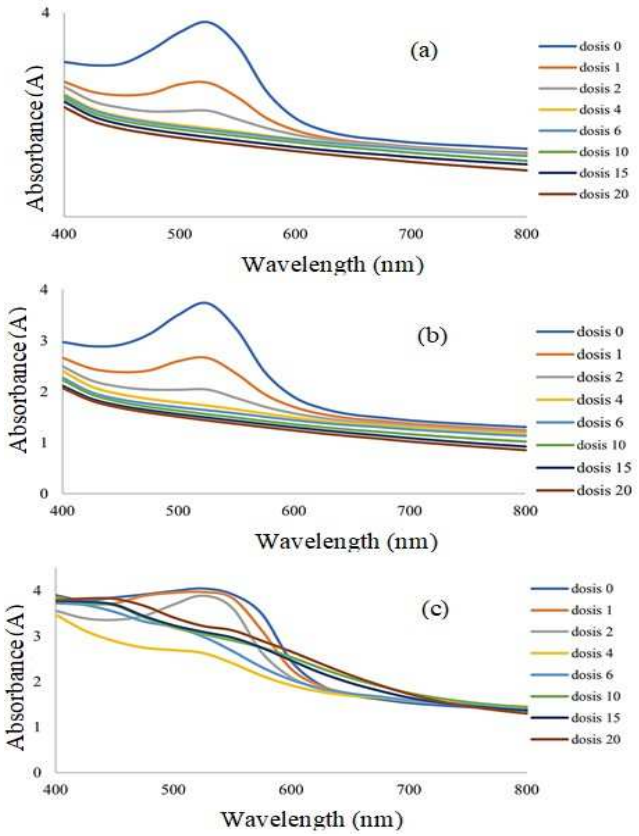


Fig. 12 UV Vis spectra of the Tragacanth – roselle flower extract after gamma radiation, (a) at pH 2, (b) at pH 5, (c) at pH 8

The color changes from red to brown occurred because of the low stability of the flavylium cation structure in the alkaline solution phase. In other words, the structure of anthocyanin pigments was stronger when the solution was in acidic condition [24]. The Tragacanth–roselle extract gel solution with pH 8 experienced a peak absorbance shift with these color changes. Consequently, the net absorbance curve could not be produced since the maximum absorbance for each radiation dose lies at different wavelengths.

Thus, the net absorbance curves for the Tragacanth – roselle flower extract for pH 2 and pH 5 have been calculated from the absorbance peak at 526 nm. As shown in Figure 13, a significant increase in net absorbance occurred at a low radiation dose of 0 – 1 kGy. Although subsequent exposures did not show a more significant increase in the net absorbances, the Tragacanth – roselle extract gel's response was higher than the roselle extract's response in this low radiation dose.

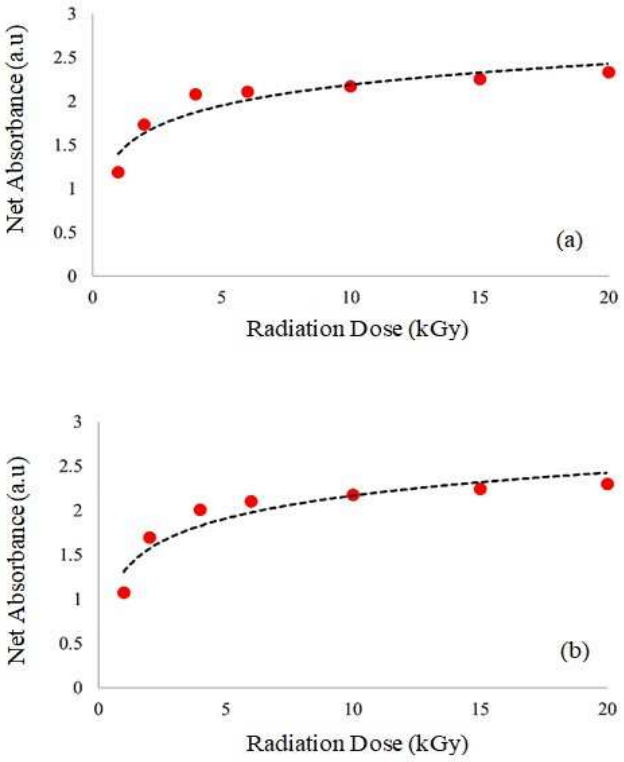


Fig. 13 Net absorbance curves of the Tragacanth – roselle flower extract as a respond to gamma radiation at (a) pH 2, (b) pH 5

For pH 2 and pH 5, the sensitivities of the Tragacanth – roselle extract gel solutions were 1.1/kGy at low radiation doses of less than 1 kGy. The sensitivities decreased to 0.28/kGy for radiation doses of 1 – 4 kGy, and yet smaller (0.02/kGy) for higher radiation doses of 4 – 20 kGy. The gel solution did not experience further significant decolorization at these high doses. To further study the effect of radiation dose on the sensitivity of the solutions, the linearization curves have been plotted for both gel solutions. Figure 14(a) shows the linear regression lines for the solutions with pH 2 and pH 5 at the medium region of radiation dose (1 – 4 kGy). It can be seen that the slopes of these lines precisely represent the sensitivities of the solutions. In some cases, R2 is called the coefficient of determination, which measures how strong

the influence of radiation dose has on the absorbance of the roselle extract in tragacanth gel. A value of close to 1 is considered to have a very strong influence.

Figure 14(b) shows the linear regression lines for the solutions with pH 2 and pH 5 at high region of radiation dose (4 – 20 kGy). The same analysis can be performed from the curves to confirm that the solutions' radiation dose and absorbance ability were highly interdependent.

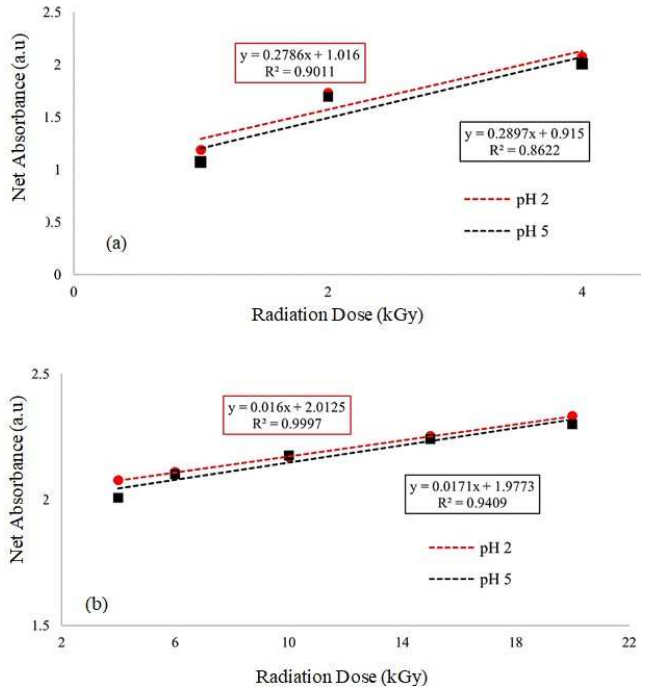


Fig. 14 Linearization curves of the Tragacanth – roselle flower extract as a response to gamma radiation at (a) 1–4 kGy, and (b) 4–20 kGy

Based on the color change, the use of tragacanth gel increased the sensitivity of the roselle extract to gamma radiation, especially with pH 2 and 5. The increase in sensitivity was due to the existence of a three-dimensional network in the gel, which is absent in the case of liquid solution [11]. Thus, the viscosity of the gel solution plays an important role in receiving the gamma radiation dose. Low-viscosity gel solution will be less sensitive to gamma radiation and vice versa.

D. Stability of Radio chromic Indicators

The stability of all radiochromic indicators was tested to study the integrity of the anthocyanin pigments of the roselle extract. After being stored for 28 days at 8°C in closed storage, it was found that no color change was observed, and the anthocyanin compounds remained stable. The absorbance peaks were still the same at 525 nm, as re-measured using a UV-vis spectrophotometer. The same condition was found in the case of Tragacanth – roselle extract gel solutions with pH 2, 5, and 8. This means that the roselle extract solutions have met the requirement for radio chromic indicators as outlined in ISO 51540: 2004.

IV. Conclusion

The radio chromic indicators based on roselle flower extract have been successfully developed in this research. It can be concluded that the radio chromic indicators made from roselle extract solution had responded to gamma radiation through decolorization, reaching 86% at a radiation dose of 20 kGy. The highest sensitivity was 0.645/kGy at medium radiation doses of 1 – 4 kg. The radio chromic indicators made from Tragacanth - roselle extract gel solution with pH 2 and 5 showed more sensitivity to gamma radiation through 90% decolorization at a radiation dose of 20 kGy. The highest sensitivity was 1.1/kGy at a low radiation dose of less than 1 kGy. The tragacanth-roselle extract gel solution with pH 8 responded to gamma radiation by color change from red to brown.

In acidic conditions, the tragacanth gel solution-maintained color stability and increased the roselle extract's sensitivity to gamma radiation. In basic (alkaline) conditions, the tragacanth gel decreased the sensitivity of the roselle extract to gamma radiation and responded by color change from red to brown. The stability of roselle extract in liquid and gel solution could be maintained for 28 days at 8°C in a closed storage.

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