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Stability of PVA Fricke gel as a radiochromic indicator for blood irradiation

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HIGHLIGHTS

- PVA-Fricke gel was proposed as a gamma radiation indicator for blood irradiators.
- Color change of the samples was observed from orange to purple proportional to increasing absorbed dose.
- Irradiated samples kept at dark and refrigerator were stable for seven days.
- The proposed gel is also a good quantitative dosimeter for gamma rays.

ABSTRACT

Gamma radiation indicators are appropriate tools for monitoring visually whether or not the irradiation process has been carried out properly. Among chemical radiation indicators available worldwide, a few are suitable for monitoring low dose ranges (especially for blood irradiation, below 50 Gy). Addressing this scope, PVA Fricke gel was proposed in this work. Irradiation of the prepared PVA Fricke gel samples was performed by Co-60 gamma cell unit up to a dose of 80 Gy. Color change of the samples was observed from orange to purple proportional to increasing absorbed dose. Prepared samples were divided into three groups, kept at different environmental conditions, to investigate stability of the gel against temperature and light. Results revealed that the irradiated samples kept at dark and refrigerator were stable for seven days. Optical absorbance measurement of the samples also estimated pre- and post-irradiation color stability. The gel can be easily used to identify processed and unprocessed products in blood irradiation. Although the gel is designed to be a qualitative indicator, it is also a good quantitative dosimeter for gamma rays.

KEYWORDS

Fricke gels
Chemical radiation indicators
Optical absorbance
Gamma exposure
Blood irradiation

HISTORY

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1 Introduction

Gamma radiation is applied for blood irradiation to prevent transfusion associated graft-versus-host disease (ISO/ASTM-51939, 2017), for food irradiation to minimize the microbial load, also to enhance the shelf life (ISO-14470, 2011) and for medical sterilisation (ISO-11140-1, 2021). During irradiation treatments, dosimeters and indicators are used to follow the process and to monitor absorbed doses. Radiation sensitive indicators may be labels, papers, inks or packaging materials which undergo a visual change when exposed to ionizing radiation. The purpose for using such indicators is to determine visually whether or not a product has been irradiated, rather than to measure dose levels (ISO/ASTM-51539, 2003). Chemical gamma indicators are usually self-adhesive labels that undergo a clear and distinct colour change when exposed to radiation. The chemical reaction which causes the color transition is a radiation specific one and is usually irreversible. The processed indicators may be retained as part of the quality control record for validation purposes (Gammatex, 2023). So far, some radiation indicators have been developed and utilized. Particularly useful as radiation dose indicators are radiochromic dyes (Rahim et al., 1985) and those based on monomers (Patel, 1981). Gamma ray polymerization of diacetylenes and the potential use of these compounds as radiation dose indicators were discussed in 1981 (Patel, 1981). A radiochromic film based on n-butyl urethane was prepared for high dose range of 3 to 150 kGy in 2012 (Abdel-Fattah et al., 2012). A monomer of p-toluene sulfonyl urethane was synthesized to use for dosimetry applications in 2018.

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The appropriate dose range of these films was 10 Gy to 15 kGy depending on the monomer content (Soliman et al., 2018). In 2019, preparation of radiochromic indicator labels made from carmoisine dyes with a matrix of paper was reported. The prepared label was tested using a Co-60 source for dose range of 5 to 100 kGy (Widodo et al., 2019). Synthesis and use of GMA-IDA monomer capable of forming colored complexes with ion metals to achieve an optically enhanced dose response with potential applications in polymer gel dosimeters was studied in 2021 (Wolfel et al., 2021). The radiation sensitive material of the radiochromic films developed in 2021 was amine substituted diacetylene compound which exhibited significant radiation induced coloration from white to different shades of blue in the dose range of 0 to 10 Gy (Mittal et al., 2021). A radiation sensitive material, PCDA, was incorporated into polyvinyl butyral (PVB) films to develop dosimeters for blood and food irradiation in 2017 (Abdel-Fattah and Soliman, 2017). The useful dose range was 5 Gy to 4 kGy depending on the PCDA content in the films. Agarose Fricke gel as a radiochromic indicator for blood irradiation, which uses a maximum dose of 50 Gy, was proposed by the author in 2018 (Edalatkhah and Rezaeian, 2018). In this work, we proposed polyvinyl alcohol Fricke gel as a radiochromic indicator for blood irradiation.

Fricke dosimeters, introduced in 1927 in solution form, are sensitive in the dose range of 40 to 400 Gy (Fricke and Morse, 1927). In order to increase the sensitivity and stability of Fricke dosimeters, gelling agents were added to the solution, resulted in Fricke gel dosimeters (Schreiner, 2004). Since, addition of the gelling agent increases the sensitivity and lowers the range of response due to the chain reactions, Fricke gels can be used in the dose range below 50 Gy which corresponds to blood irrdiation (Farajzadeh and Sina, 2021). It has been shown that the chemical yield of a Fricke gel in comparison to that of a Fricke solution increases by a percentage of 150 (Leong et al., 2007). Polyvinyl alcohol (PVA), is the recently introduced gelling agent with the lowest diffusion coefficient. As well, it has a high gelling strength which gives an elastic gel, suitable for an indicator fabrication (Marrale et al., 2017). Hence, PVA Fricke gel seems to be a good candidate as a radiochromic indicator for blood irradiation. As stability is an important parameter for a radiation indicator to be used, we decided to survey the stability of PVA Fricke gel in this work. After preparation of PVA Fricke gel and exposure of the samples to gamma radiation, color change depending on the dose value was investigated. Then, with optical absorbance measurement of the samples, effects of environmental conditions, pre-irradiation and post-irradiation color stability were surveyed.

2 Material and Methods

The PVA Fricke gel is obtained by incorporating ploy vinyl alcohol (PVA), a gelling agent, into the ferrous sulphate solution. The gel was prepared from a 10% w/v aqueous solution of PVA (Merck), 1% w/v glutaraldehyde (Merck), 25 mM sulphuric acid (Merck), and 0.5 mM ferrous ammonium sulfate hexahydrate (Merck) and 0.165 mM xylenol orange (Merck). The gel was made following the procedure described in (Gallo et al., 2019). PVA solution was prepared by dissolving PVA in ultrapure water (80% of the total water volume). The dissolution was performed under stirring ($\sim$ 500 rpm) and the temperature raising up to 70 °C. Afterwards, Fricke solution was prepared by adding sulfuric acid, ferrous ammonium sulfate and xylenol orange sodium salt into water (10% of the total water volume). Then, gutaraldehyde solution was prepared by adding glutaraldehyde into water (the remaining 10% of the total water volume). Final solution were obtained by slowly incorporating Fricke solution into the PVA solution and subsequently by adding the glutaraldehyde solution. After gentle stirring to achieve homogeneity, the prepared solution was poured into the cuvettes as shown in Fig. 1 and underwent gelation while reaching to room temperature. Cuvettes are plexiglass containers with dimensions of $1 \times 1 \times 4$ cm^3 which are routinely used for spectroscopy. The gel was typically affected by no significant toxicity. After thirty minutes, the cuvettes were irradiated by Co-60 gamma rays using a gamma-cell 220 unit (Nordion, Canada) and the color change was surveyed with dose increase.

a) b)

Figure 1: a) The prepared gel, b) Cuvettes' filling by the prepared gel.

In order to investigate stability of the prepared PVA Fricke gel against temperature and light variations, the developed samples were divided into three groups, each one kept at a specific condition. Group 1 included the cuvettes stored in the dark and refrigerator (Ref), group 2 included the cuvettes stored in the dark and room temperature (RT) and group 3 included the cuvettes stored in light and room temperature (RT). This grouping is represented in Fig. 2. Samples of each group were irradiated up to 50 Gy by Co-60 gamma rays. For each dose value, three cuvettes were irradiated and analyzed to reduce the statistical errors. At time of irradiation, the dose rate was 0.95 Gy.s^{-1}. Thirty minutes after the irradiation, the optical analyses of the samples were carried out using a spectrophotometer (BECKMAN COULTER-DU-800). The measurement of the samples was done at wavelength of 585 nm (the wavelength with the maximum absorbance

value, generally used in the literature for such analyses). For each cuvette, the absorbance was measured at different time intervals after irradiation and the absorbance curve as the function of time over a period of 10 days was plotted for different doses. Before each measurement, we removed the samples from the refrigerator and kept them at room temperature for 30 minutes.

3 Results and discussion

Fig. 3 represents samples of the irradiated PVA Fricke gel. Three cuvettes were irradiated at each dose from 0 to 80 Gy (0, 10, 20, 30, 40, 50, 60, 70, 80 Gy). The delivered dose increases from left to right as shown by the labels on top of each cuvette.

The first three cuvettes on the left observed in orange color correspond to unirradiated samples and the last three cuvettes on the right observed in purple color correspond to the samples irradiated up to 80 Gy. Color change of the samples started immediately after the irradiation and completed in thirty minutes. As it can be seen in Fig. 3, color of the gel changes from orange to purple proportional to the absorbed dose. As mentioned in methods, we used xylenol orange (XO) in the composition of the gel as a radiochromic dye which is orange in acidic medium. Changes in color and opacification of the gel occurred upon irradiation when the gel was irradiated, the ferrous ions (Fe^{2+}) oxidized to ferric ions (Fe^{3+}), combined with XO to form XO-Fe^{3+} complex. Actually, the color change is due to complex formation of XO-Fe^{3+} which shifts the absorption peak to the visible region. There is a good degree of contrast from unirradiated to irradiated in given dose region, which is an important parameter in selecting the material of an indicator.

The colorimetric response of the samples to radiation was also measured in terms of optical density. Absorbance of the gel samples versus time in different environmental conditions are reported in Figs. 4 to 9 for different doses. Each point of each figure is the mean of the measurements on three samples. Error bars, reported in Table 1, are smaller than the plot symbols. When the sample was not irradiated, the absorbance was measured at 435 nm related to ferrous ions since there was no radiation induced ferric ions in the sample. When the sample was irradiated, the absorbance was measured at 585 nm related to ferric ions produced due to radiation induced oxidation of ferrous ions.

Figure 2: Classification of the cuvettes into three groups (1) stored in the dark and refrigerator (2) stored in the dark and room temperature (3) stored in light and room temperature.

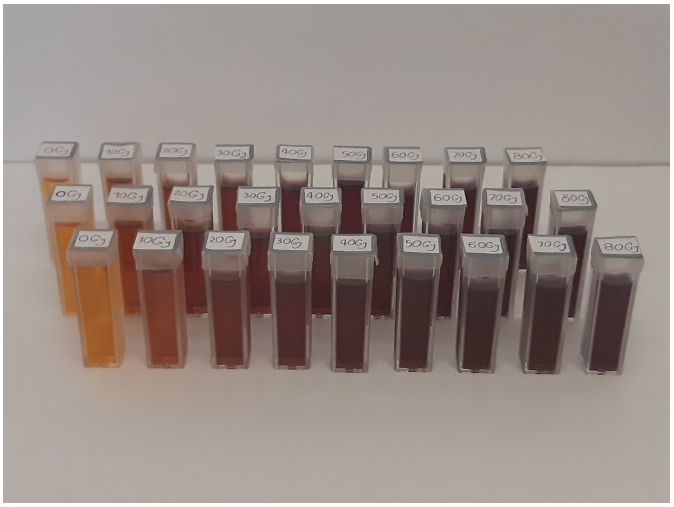

Figure 3: Color change of the prepared gel 30 min after gamma irradiation.

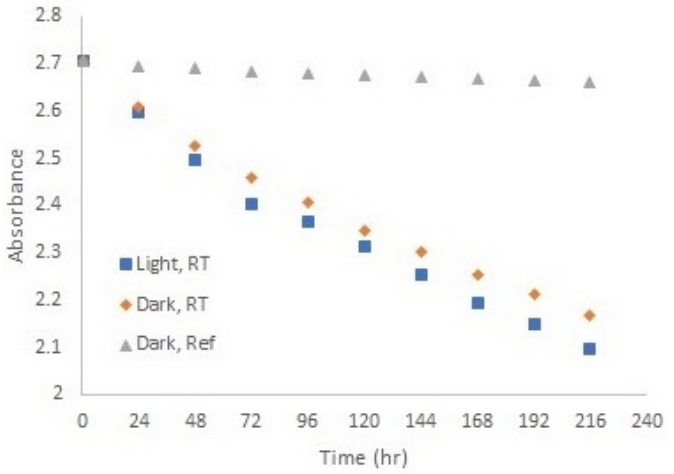

Figure 4: Stability of the pre-irradiation samples under different conditions.

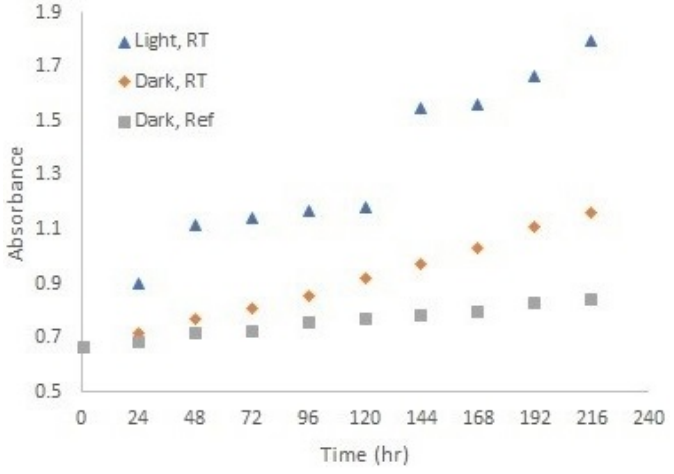

Figure 5: Absorbance over time after irradiation of 10 Gy under different conditions.

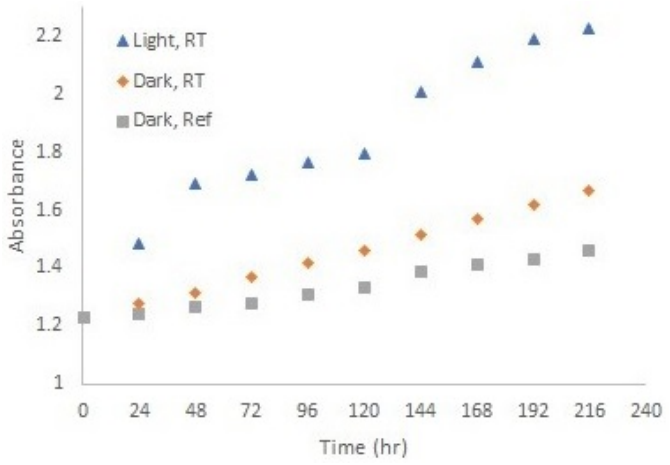

Figure 6: Absorbance over time after irradiation of 20 Gy under different conditions.

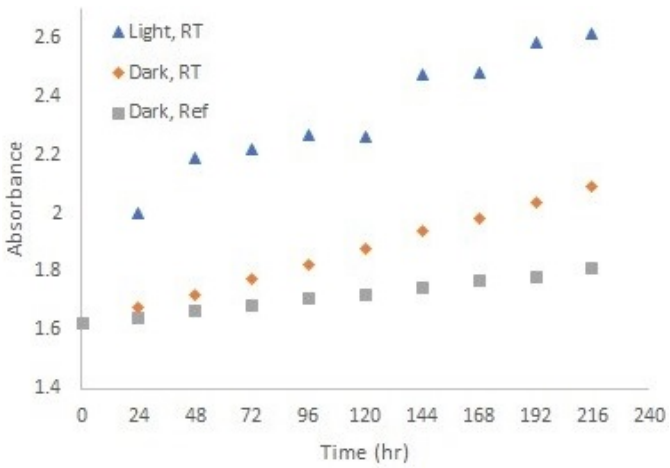

Figure 7: Absorbance over time after irradiation of 30 Gy under different conditions.

Table 1: Absorbance variation percentages.

Storage conditions	Dose (Gy)	Absorbance variation (%)	Standard deviation
Light, RT	0	22	0.0020
Dark, RT	0	19	0.0000
Dark, Ref	0	1	0.0000
Light, RT	10	80	0.0024
Dark, RT	10	46	0.0010
Dark, Ref	10	5	0.0000
Light, RT	20	63	0.0030
Dark, RT	20	23	0.0024
Dark, Ref	20	5	0.0025
Light, RT	30	50	0.0021
Dark, RT	30	19	0.0026
Dark, Ref	30	5	0.0031
Light, RT	40	34	0.0026
Dark, RT	40	15	0.0000
Dark, Ref	40	3	0.0014
Light, RT	50	19	0.0038
Dark, RT	50	12	0.0036
Dark, Ref	50	1	0.0000

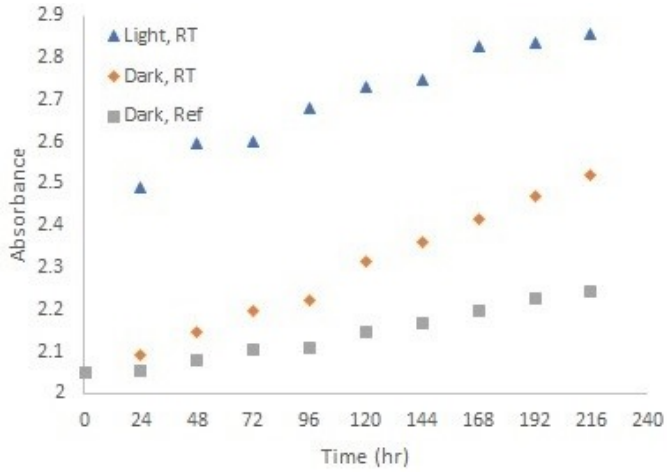

Figure 8: Absorbance over time after irradiation of 40 Gy under different conditions.

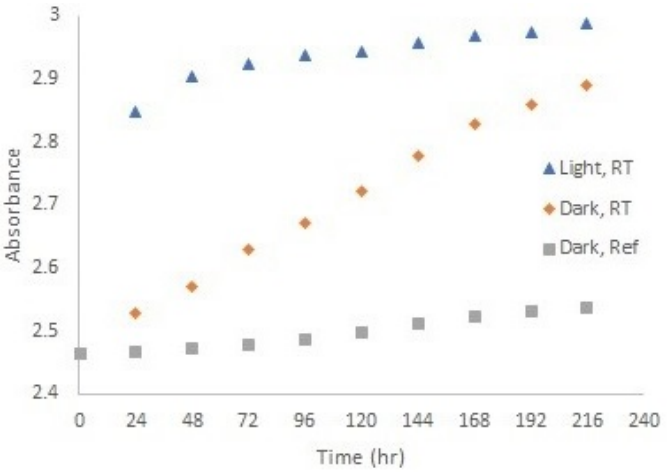

Figure 9: Absorbance over time after irradiation of 50 Gy under different conditions.

As illustrated in Fig. 4, absorbance of the samples stored in the dark and room temperature and the ones stored in light and room was reduced by 19% and 22% after ten days respectively (reprted in Table 1). Smooth trend of the data was placed the criteria for expressing the stability. As it can be seen in Fig. 4, there is a nearly plateau up to ten days for the samples stored in the dark and refrigerator and the absorbance of the samples was reduced by 1%. Therefore, the samples stored in the dark and refrigerator were stable for ten days. So, keeping the gel in the dark and refrigerator is the best condition of samples storage after preparation before use. Indeed, keeping the samples in the dark and refrigerator can minimize possible auto-oxidation of ferrous ions. Passing time, auto-oxidation of ferrous ions to ferric ions occurred, resulted in ferrous ions depletion and absorbance decrease over time.

Figures 5 to 9 show stability of the irradiated samples by dose of 10 to 50 Gy under different conditions. As illustrated in Fig. 5, which is related to irradiation of the samples by 10 Gy, absorbance of the samples stored in the dark and refrigerator, the ones stored in the dark and room

temperature and the ones stored in light and room temperature was reduced by 5%, 46% and 80% after ten days, respectively (reported in Table 1). Also, Fig. 9 which is related to irradiation of the samples by 50 Gy, shows that absorbance of the samples stored in the dark and refrigerator, the ones stored in the dark and room temperature and the ones stored in light and room temperature was increased by 1%, 12% and 19% after ten days, respectively (reported in Table 1). Although the gel suffers from auto-oxidation of ferric ions after irradiation, XO as a chelating agent reduces this effect. Actually, auto-oxidation portion was added to induced oxidation portion, resulted in absorbance increase over time. Figure 7 which is related to irradiation of the samples by 30 Gy, represents that absorbance of the samples stored in the dark and refrigerator was reduced by 19% after ten days, whereas it was nearly constant for seven days. In fact, the gel is relatively stable no more than seven days after irradiation. Absorbance variation percentages and the standard deviation of Figs. 4 to 9 are reported in Table 1. As it can be seen, absorbance variation percentages related to the samples stored in the dark and refrigerator are smaller in comparison with absorbance variation percentages related to the samples stored in other conditions. This verifies keeping the gel in the dark and refrigerator is the best condition of samples storage after irradiation.

Figure 10-a shows color stability of the gel at tenth day after preparation. It is related to unirradiated samples: (1) samples kept at dark and refrigerator, (2) samples kept at dark and room temperature and (3) samples kept at light and room temperature. As it can be seen, samples 1 retained the orange color of the first day, whereas color of samples 2 and 3 was changed. This is in line with the data on Fig. 4.

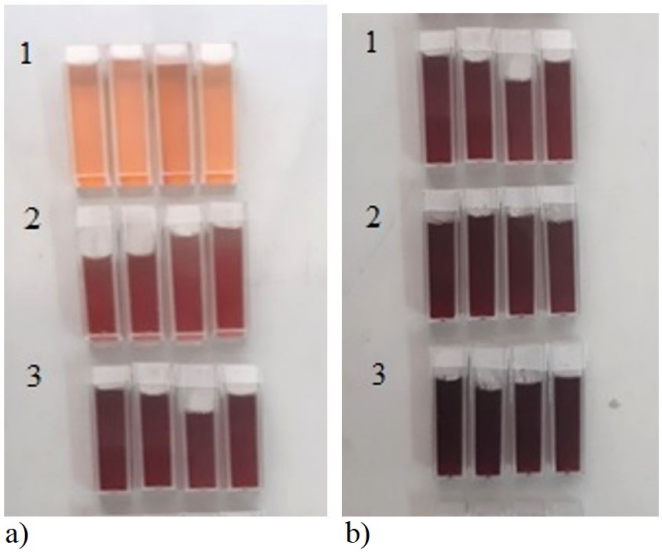

Figure 10: a) Color stability of pre-irradiation samples at tenth day 1) samples kept at dark and refrigerator, 2) samples kept at dark and room temperature, 3) samples kept at light and room temperature. b) Color stability of post-irradiation samples at seventh day 1) samples kept at dark and refrigerator, 2) samples kept at dark and room temperature, 3) samples kept at light and room temperature.

Also, Fig. 10-b shows samples irradiated by 30 Gy at seventh day after irradiation. Samples 1 kept at dark and refrigerator, samples 2 kept at dark and room temperature and samples 3 kept at light and room temperature. As it can be seen, color of samples 1 had nearly no change in comparison with the first day, whereas color of samples 2 and 3 became more intensified. Passing more days, the induced color due to irradiation turned to purple without dependence on absorbed dose due to auto-oxidation of ferrous ions. This verifies that the gel is relatively stable no more than seven days, as mentioned before.

4 Conclusions

After irradiation of the prepared samples of PVA Fricke gel, a visible color change was shown as a function of absorbed dose. There is a good degree of contrast from unirradiated to irradiated samples. Keeping the gel in the dark and refrigerator is the best condition of samples storage post- and pre-irradiation. Good performance of the prepared gel in the dose range below 50 Gy, verify its application as a radiochromic indicator for blood irradiators. The results reveal that the gel, as well its use as a qualitative indication for visual verification of irradiation, is able to provide facility operators with a quantitative dose delivered to products. Due to tissue equivalence composition of the gel, it could provide phantoms for 3D dose validation by molding into the human body organs. Performance improvement of the prepared gel such as extension of its shelf life using new additives is our next goal.

Conflict of Interest

The authors declare no potential conflict of interest regarding the publication of this work.

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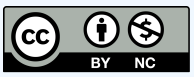

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