China Instrument and Control Society
Instrumentation
INSTRUMENTATION, Vol. 11, No. 3, September 2024 https://doi.org/10.15878/j.instr.202400168

Article

Enhanced Sensitivity and Stability in Gelatin-Based Fricke gel Dosimeters: Impact of Benzoic Acid on Low-Dose Radiation Measurements

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Abstract: Fricke gel dosimetry is a critical technique for accurate radiation dose measurement, leveraging the radiation-induced oxidation of ferrous to ferric ions within a hydrogel matrix. This study aimed to advance the field by preparing and evaluating seventy-one Fricke gel dosimeter (FGD) samples with varying chemical compositions, focusing on the incorporation of benzoic acid. Two gelatin-based samples demonstrated superior sensitivity, particularly for low-dose measurements. Our findings indicated that the sample without benzoic acid exhibited a 1.75 times higher sensitivity compared to the sample containing benzoic acid, with lower limits of detection at 0.04 Gy and 0.1 Gy, respectively. Stability tests revealed temperature-dependent responses, with better performance observed under refrigerated conditions. Reproducibility was confirmed through consistent calibration curves across multiple trials. Additionally, the dosimeters' responses varied with different radiation types, underscoring the need for specific calibrations. The study concludes that while benzoic acid slightly reduces sensitivity, it provides consistent responses across various radiation energies, suggesting its potential as a beneficial additive in diverse clinical scenarios. This research contributes to the ongoing optimization of FGDs for precise radiation dose assessment.

Keywords: chemical dosimeter; freak dosimeter; benzoic acid

Citation: Saeedi-Sini, Seyed Amir, Sedigheh Sina, Mohammad Hossein Sadeghi, and Ebrahim Farajzadeh. "Enhanced Sensitivity and Stability in Gelatin-Based Fricke Gel Dosimeters: Impact of Benzoic Acid on Low-Dose Radiation Measurements." Instrumentation 11, no. 3 (September 2024). https://doi.org/10.15878/j.instr.202400168.

1 Introduction

Fricke gel dosimetry has been a cornerstone in the accurate measurement and quantification of ionizing radiation doses, utilizing the principle that radiation-induced chemical changes within the dosimetric material can be directly correlated with absorbed dose levels. This technique, which includes Fricke gel (FG) and polymer gel dosimetry (PGD), is renowned for its tissue-equivalent properties and its ability to provide precise dosimetric analysis[1]. FG dosimetry, in particular, is based on the oxidation of ferrous to ferric ions within a hydrogel matrix, typically composed of materials such as polyvinyl alcohol-gelatin-tributylphosphate (PVA-GTA) or gelatin, which allows for the homogeneous distribution of ferrous ammonium sulfate[2]. This method has been extensively studied for its simplicity and effectiveness in dose measurement[3].

In recent years, the incorporation of Xylenol Orange (XO) into FG dosimetry has significantly advanced the field by enabling the dosimetric signal to be determined via optical absorbance. XO chelates ferric ions, forming

highly colored complexes, which enhances the visibility and accuracy of dosimetric readings. Despite these advancements, challenges such as ion diffusion and self-oxidation phenomena have been identified, which can affect the consistency and reliability of FG dosimeters. To address these issues, recent research has explored various modifications, including the addition of organic additives like antioxidants, saccharides, and nanocomposites, as well as the use of alternative chelating agents and gel matrices[4-32].

Building on this foundation, our previous work validated gel dosimeters by comparing traditional spectrophotometric methods with a novel approach using a camera and MATLAB software. This new method demonstrated comparable sensitivity, offering a viable alternative for dosimetric measurements[33].

In the current study, we aim to further advance FG dosimetry by preparing and evaluating seventy-one Fricke gel dosimeter samples, each with distinct chemical compositions. This comprehensive evaluation seeks to identify formulations with superior sensitivity, particularly for low-dose measurements. Preliminary results have highlighted the promise of two gelatin-based samples due to their enhanced sensitivity. To deepen our understanding of factors influencing dosimetric performance, this study specifically examines the effect of benzoic acid.

2 Materials and Methods

2.1 Fricke Gel Preparation Method

In this study, seventy-one Fricke gel dosimeter samples were prepared, each with a distinct chemical composition, to comprehensively evaluate their performance. Among these, two gelatin-based samples demonstrated superior sensitivity, particularly for low- dose measurements. To better understand the factors influencing dosimetry performance, we specifically examined the effect of benzoic acid.

The dosimeter preparation process is detailed below, with visual guides in Figs.1 and 2, which depict the step-by-step construction of the gelatin-based samples. The composition of the materials used is provided in Table 1.

1. Preparation of materials: Four beakers were prepared with the following contents:

• Beaker 1: Gelling agent (gelatin) mixed with water.
• Beaker 2: Xylenol orange, water, and sulfuric acid.
• Beaker 3: Benzoic acid and water.
• Beaker 4: Ferrous ammonium sulfate (FAS) and water.

2. Heating of gel-water mixture:

• For the gelatin-based formulation, Beaker 1 was heated on a heater-stirrer until the mixture reached 45 °C.
• For the second gelatin-based sample, another beaker similar to Beaker 1 was heated on a heater-stirrer until the mixture reached 45 °C.

3. Cooling process: Once the mixtures reached the desired temperature, Beaker 1 and the second beaker were removed from heat. The gelatin mixtures were then cooled to approximately 35 °C in a water bath.

4. Combining solutions: The contents of Beaker 2 were added to Beaker 1, now at ambient temperature, to form a transparent, homogeneous solution with a light orange hue.

5. Addition of FAS: The solution from Beaker 4 was then added to Beaker 1, ensuring thorough mixing.

6. Incorporation of benzoic acid: The solution from Beaker 3 was added last to Beaker 1, followed by comprehensive mixing to complete the preparation.

7. Cuvette transfer: The final solution was transferred into cuvettes (1 × 1 × 4.5 cm^3) and stored in a refrigerator overnight in preparation for subsequent irradiation.

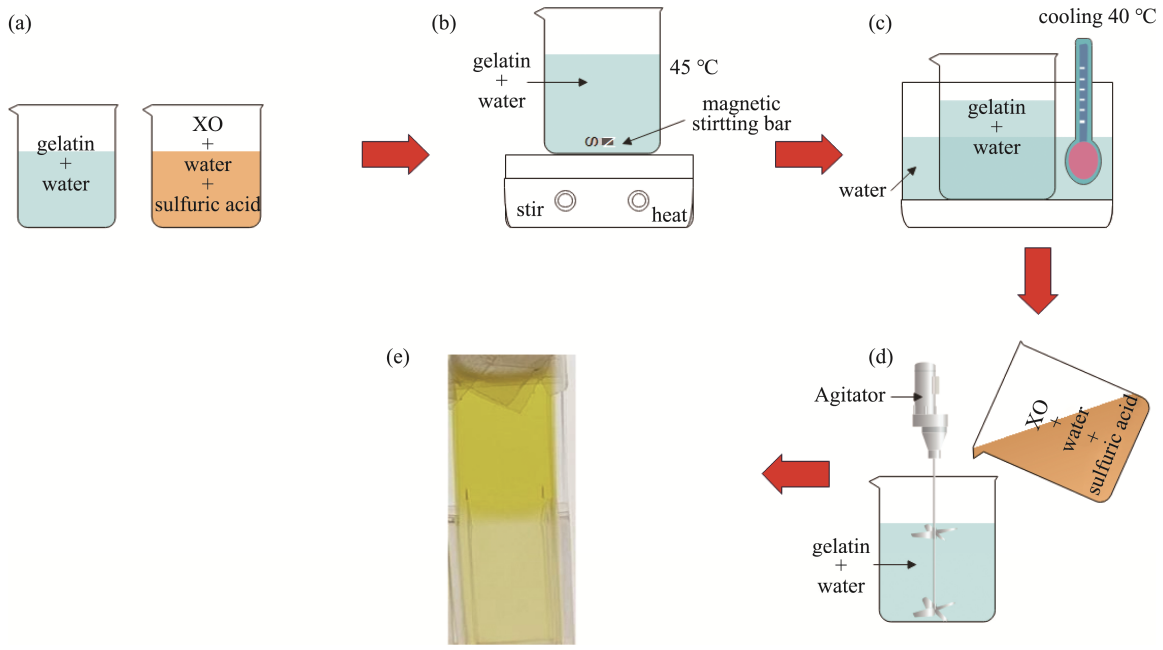

Fig.1 Fricke gel preparation procedure, sample 1.

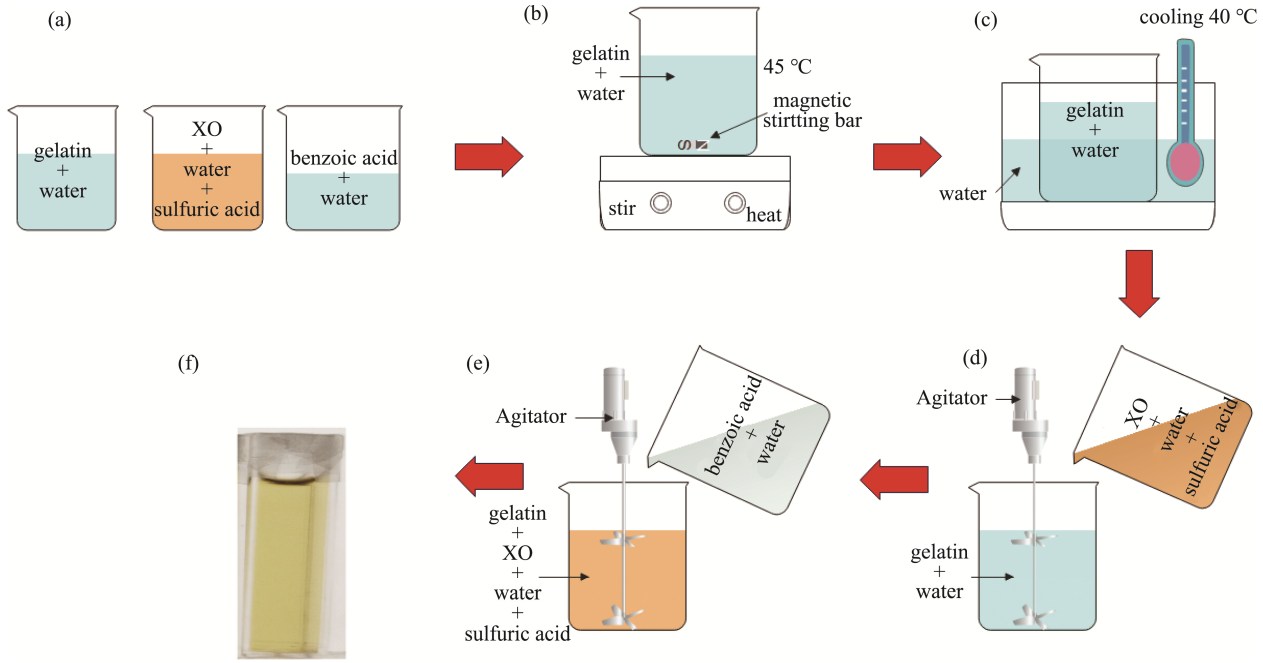

Fig.2 Fricke gel preparation procedure, sample 2.

Table 1 Materials utilized in the construction of the gel dosimeter samples, detailing their specific roles and quantities

Component	Role in dosimetry process	Chemical formula
Gelatin	Gelling agent	$(C_{17}H_{32}N_5O_6)_X$
Poly(vinyl alcohol)	Gelling agent	[CH2CH(OH)]n
Benzoic acid	Activator-Catalyzer	$C_7H_6O_2$
Xylenol orange	$C_{31}H_{28}N_2O_{13}$	Chemical dye
Ferrous ammonium sulfate	Activator-Catalyzer	$Fe(NH_4)_2(SO_4)_2{\cdot}6H_2O$
Sulfuric acid	Solvent	H_2SO_4
di-ionized water	Solvent	H_2O

2.2 Irradiation of Gel Dosimeters

Prior to irradiation, the gel dosimeter samples were conditioned at 5 °C for approximately 12h. The irradiation was conducted using a Cs-137 source, calibrated following the standards of a secondary standard dosimetry laboratory. The samples were placed 60 cm from the source, exposed to a dose rate of 2 Gy/h within a 10×10 cm² field.

Fig.3 depicts the radiation-induced color changes in dosimeter samples 1 and 2, highlighting their dose-dependent responses.

2.3 Reading the Dosimeters

The method for reading dosimeter responses was adapted from Farajzadeh et al.[33]. A negatoscope was used to illuminate the samples, and images were captured with a camera positioned 15 cm away. Image analysis was conducted using a custom MATLAB GUI tool, which focused on changes in optical absorption at 589 nm, the peak absorption wavelength between the green and red spectrums. The blue channel was discarded, and the red and green pixel values were averaged to quantify the optical absorption changes of the dosimeters, denoted as A in units of 1/cm.

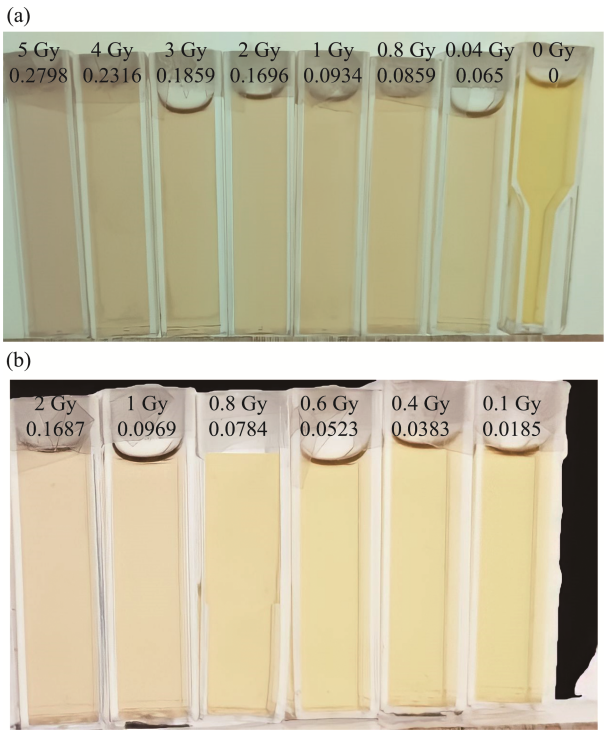

Fig.3 Color change of dosimeter due to radiation; (a) sample 1 and (b) sample 2.

2.4 Calibration Curve and Linearity

Calibration curves were generated by plotting dosimeter readings against absorbed doses. The linearity of these curves was evaluated by comparing the coefficient of determination (R^2) from linear fits. The lower limit of detection (LLD) for each dosimeter was determined by comparing readings from irradiated samples with those from control (unirradiated) samples.

2.5 Sensitivity of the Dosimeter

Sensitivity, defined by the slope (S) of the calibration curve as shown in Equation (1):

$$\mathrm{L}\left(\frac{1}{\mathrm{cm}}\right) = S \times D + b \qquad (1)$$

where S represents the sensitivity of the dosimeter, D is the absorbed dose, and b is the intercept of the calibration curve. Higher sensitivity indicates a greater dosimeter reading at a specific dose, which is crucial for low-dose measurements. This parameter was essential in comparing the two dosimeters, with higher sensitivity indicating greater utility in low-dose applications.

2.6 Stability and Reproducibility

The stability of the dosimeters was assessed over different time frames and storage conditions to ensure consistent post-irradiation responses. Additionally, the reproducibility of the dosimetry process was verified through repeated construction and calibration of the dosimeters, and by comparing the repeatability of responses with calibration curves from similar studies.

3 Results

3.1 Composition of Dosimeters for Low-Dose Measurement

We compared two FXG dosimeters selected from seventy-one prepared samples for their ability to detect very low minimum detectable doses. The key distinguishing factor was the presence of benzoic acid, as detailed in Table 2.

Table 2 Material composition of selected samples for low-dose measurement

Material	Sample 1	Sample 2
Gelatin	131 mM	120 mM
XO	0.0446 mM	0.0505 mM
Sulfuric acid	80.9 mM	84.53 mM
Benzoic Acid	---	3.357 mM
FAS	1.025 mM	0.9843 mM
Water	100 ml	100 ml

3.2 Linearity and Lower Limit of Detection (LLD)

The dosimeters demonstrated a linear response to radiation doses, as evidenced by the calibration curves with R^2 values close to 1 (0.9921 for Sample 2 and 0.9978 for Sample 1). The lower limit of detection (LLD) was determined to be approximately 0.04 Gy for one gelatin-based dosimeter (Sample 1) and 0.1 Gy for the other gelatin-based dosimeter (Sample 2), as illustrated in Fig.4.

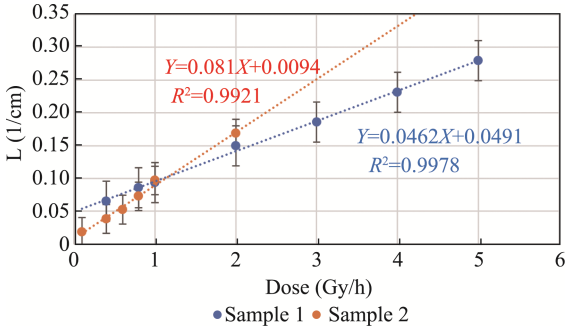

Fig.4 Calibration curves of the two gelatin-based dosimeters.

3.3 Sensitivity of the Dosimeters

The sensitivity, defined by the slope of the calibration curve, differed between the two dosimeters. The gelatin-based dosimeter (Sample 1) exhibited a sensitivity 1.75 times higher than that of the other gelatin-based dosimeter (Sample 2), highlighting its superior efficacy in low-dose measurements.

3.4 Stability Variation with Storage Temperature

Stability testing indicated temperature-dependent responses for both gelatin-based dosimeters, with improved stability observed when stored in a refrigerator. Notably, Sample 1 exhibited greater stability at room temperature compared to Sample 2, as shown in Fig.5.

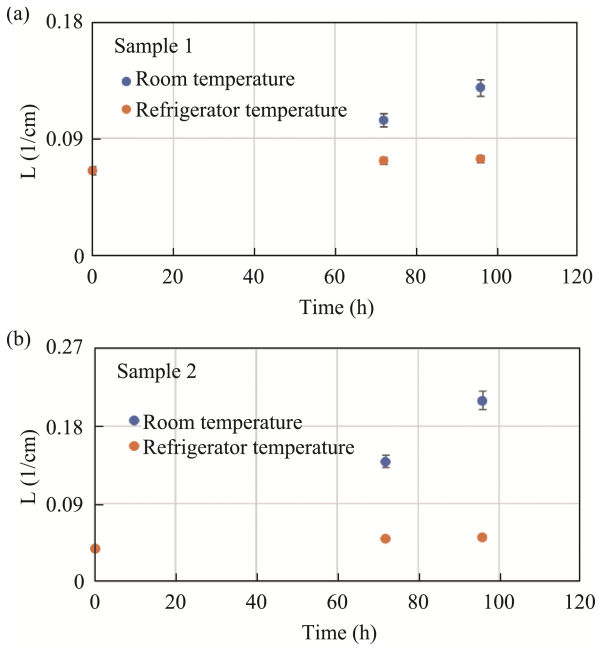

Fig.5 Stability variation with storage temperature for (a) sample 1 and (b) sample 2.

3.5 Stability Over Time

Both gelatin-based dosimeters exhibited a few degrees of degradation over time, even under refrigerated conditions. The stability variation remained below 0.3% for both samples up to 120 hours post-irradiation, with an overall change of 0.284% observed (Fig.6).

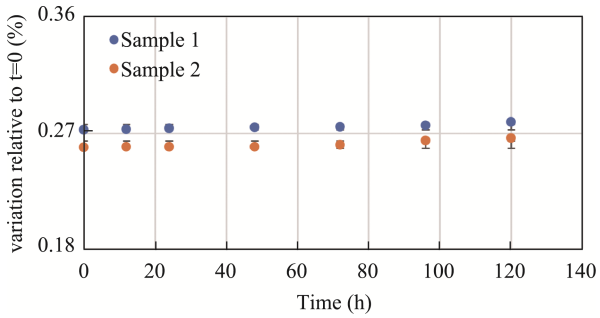

Fig.6 Stability variation over time for sample 1 and sample 2.

3.6 Reproducibility of Response

The reproducibility of the dosimeters was verified through repeated construction, irradiation, and calibration. The results demonstrated consistent calibration curves across multiple trials, as illustrated in Fig.7.

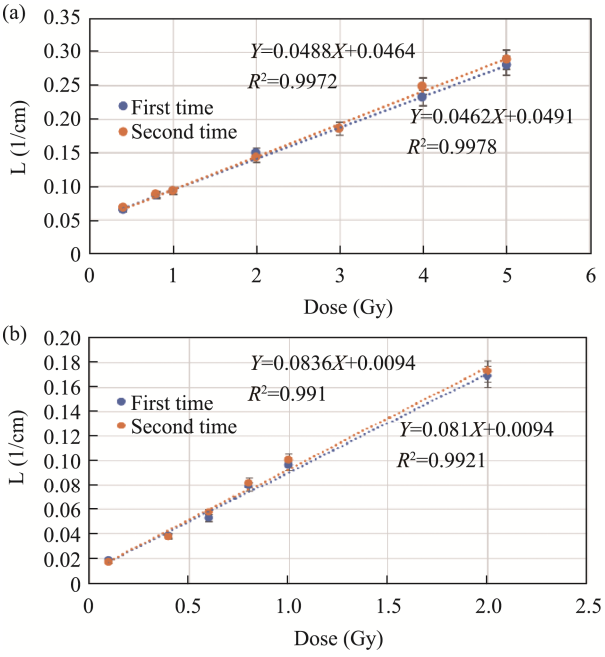

Fig.7 Calibration curves illustrating the reproducibility of response for (a) sample 1 and (b) sample 2 across repeated trials.

3.7 Beam Type and Energy Dependence

Experiments were conducted with both Cs-137 and X-rays at 120 keV to assess the dosimeter's response consistency across different radiation types. The findings indicate that the response of the dosimeters shows notable variations depending on the radiation beam's energy. For both radiation types, the measured absorption demonstrated discernible changes, highlighting the dosimeter's performance sensitivity to the energy of the radiation beam. This energy dependence suggests the need for calibration when using these dosimeters for diverse radiation measurement applications.

For dosimeter sample 1, the percentage error in absorption was 0.1503 for X-ray and 0.1496 for Cs-137, with an overall percentage error of 0.468%. For dosimeter sample 2, the percentage error in absorption was 0.1683 for X-ray and 0.1687 for Cs-137, with an overall percentage error of 0.237%. These results indicate that while dosimeter sample 2 exhibited more consistent absorption measurement across different radiation sources, sample 1 showed slightly higher variability.

4 Discussion

The current study aimed to enhance the sensitivity of FGD by exploring various chemical compositions, particularly focusing on the inclusion of benzoic acid. The findings reveal significant insights into the performance, stability, and reproducibility of gelatin-based FGDs in low-dose radiation measurement. Notably, the dose range for Sample 1 is from 0.04 Gy to 5 Gy, while for Sample 2, it is from 0.1 Gy to 2 Gy. The results demonstrated that Sample 1, which did not contain benzoic acid, exhibited a higher sensitivity compared to Sample 2, which included benzoic acid. Specifically, Sample 1 showed a sensitivity 1.75 times greater than that of Sample 2, highlighting its superior efficacy in detecting low doses of radiation. This finding aligns with previous research indicating that the chemical environment within the gel matrix critically affects dosimetric sensitivity[9-11,25,33].

The linearity of the response was high for both samples, with R^2 values of 0.9978 for Sample 1 and 0.9921 for Sample 2, indicating reliable performance across the tested dose ranges. The LLD was found to be 0.04 Gy for Sample 1 and 0.1 Gy for Sample 2, suggesting that Sample 1 is more suitable for very low-dose measurements.

The stability of the dosimeters was evaluated under different storage conditions, revealing temperature-dependent variations. Both samples showed improved stability when stored in a refrigerator, but Sample 1 maintained better stability at room temperature compared to Sample 2. This observation suggests that the absence of benzoic acid in Sample 1 contributes to its enhanced thermal stability, which is consistent with the known effects of benzoic acid on the chemical stability of dosimetric materials.

Reproducibility tests confirmed that the FGDs could consistently reproduce calibration curves across multiple trials. This consistency is crucial for clinical applications where reliable dose measurements are essential. The study also investigated the dosimeters' responses to different types of radiation (Cs-137 and X-rays), revealing notable variations in absorption based on the radiation energy. Sample 2 exhibited more consistent measurements across different radiation types, suggesting

a broader applicability in various clinical settings.

The incorporation of benzoic acid, while slightly reducing sensitivity, did not significantly compromise the overall performance of the dosimeters. Instead, it offered benefits in terms of consistent response to different radiation types, which can be advantageous in diverse clinical scenarios. Future research should explore optimizing the concentration of benzoic acid and other additives to balance sensitivity, stability, and reproducibility. Additionally, the study's methodology, including the novel use of a camera and MATLAB software for dosimeter reading, proved effective and comparable to traditional spectrophotometric methods. This approach offers a practical alternative for dose measurement in resource-limited settings.

5 Conclusion

The study successfully identified a gelatin-based FGD formulation with superior sensitivity and stability, particularly suitable for low-dose radiation measurements. The findings underscore the potential of benzoic acid as a modifying agent to enhance dosimetric properties. These advancements contribute to the ongoing efforts to refine and optimize FGDs for precise and reliable radiation dose assessment in medical and research applications.

Author Contributions:

The research paper was a collaborative effort among Seyed Amir Saeedi-Sini, Sedigheh Sina, Mohammad Hossein Sadeghi, and Ebrahim Farajzadeh, with each author playing vital roles across various stages of the project. Saeedi-Sini and Sina were instrumental in the foundational aspects such as conceptualization, data curation, formal analysis, investigation, and methodology, with Sina also securing funding, administering the project, and supervising the research process. Both contributed significantly to writing the original draft, with Sina also involved in reviewing and editing the manuscript. Sadeghi focused on validating the findings and enhancing their presentation through visualization, also contributing to both the original writing and its revision. Farajzadeh's contributions were centered around methodology and validation, ensuring the research design was robust and results were accurate, and he also aided in visualizing the findings.

Funding Information:

This research received no external funding.

Data Availability:

The authors declare that the main data supporting the findings of this study are available within the paper and its Supplementary Information files.

Conflict of Interest:

The authors declare no competing interests.

Dates:

Received 03 April 2024; Accepted 7 July 2024; Published online 30 September 2024

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