ASSESSMENT OF DOSE RATES IN INDUSTRIAL GAMMA IRRADIATION FACILITIES USING FRICKE AND THERMOLUMINESCENT DOSIMETRIC SYSTEMS

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ABSTRACT

The assessment of gamma dose rates in irradiation facilities allows an easy previous estimation of absorbed dose of a product during the irradiation process. The liability of dose measurements is assign to the metrological procedures adopted including the uncertainty evaluation. Fricke and thermoluminescent (TLD-800) dosimetric systems were used to measure several high-dose points inside the irradiation room of a gamma irradiation facility. The results have shown the applicability of such dosimetric systems in quality assurance programs, assessment of dose rates, dose uniformity and routine dose measurements in a range between 40 Gy and 1 kGy.

1. INTRODUCTION

The ionizing radiation is widely used in gamma irradiation facilities to induce physical, chemical or biological benefits on the irradiated products. Typical examples are food treatment for parasite and pathogen control and shelf-life extension, sterilization of medical devices, cosmetics and phytoterapics, enhancement of polymers, modification of gemstones colors, stuff disinfections, conservation of art objects and other various applications [1, 2]. For this irradiation technology, dosimetric tests must be accomplished to evaluate the dose distribution throughout the sample being irradiated. By the assessment of dose rates inside the irradiation room it is possible to have a previous estimation of the absorbed dose of a product located at a specific distance from the irradiation source during a definite time of exposure.

The gamma irradiation facility used in this practice consists on a multipurpose panoramic irradiator category II (dry storage), model IR-214, fabricated by MDS Nordion, with a Cobalt-60 source. Cobalt-60 emits photons with approximately 1.17 and 1.33 MeV in equal proportions, and its half-life is approximately 5.27 years [3, 4].

The dosimetric systems used were the Fricke and thermoluminescent systems.

The Fricke dosimeter is a liquid chemical dosimeter composed by ferrous ammonium sulfate (1.0 mol.L\(^{-1}\)) in sulfuric acid solution (0.4 mol.L\(^{-1}\)). Its response to ionizing radiation is based on a process of oxidation of ferrous ions to ferric ions and the absorbed dose is quantified by
a change in the absorbance at a specified wavelength (303 nm). It is considered a reference-standard dosimetry system in the absorbed dose range of 40 to 400 Gy [5].

Thermoluminescent dosimeters (TLDs) are widely used for the measurement of absorbed dose. They have many useful features, such as small size, wide dynamic range and dose-rate independence, but also a number of disadvantages, including a complicated anneal cycle to erase the past radiation history, nonlinear response for high absorbed doses and loss of sensitivity with accumulated dose [6]. The thermoluminescent dosimeter used in this practice was TLD-800 (Li₂B₄O₇: Mn), which works in the absorbed range of µ Gy to 10⁵ Gy.

2. EXPERIMENTAL PROCEDURE

2.1. Preparation of Fricke Dosimetric Solution

The Fricke dosimetric solution was prepared dissolving 0.392 g of ferrous ammonium sulfate, (NH₄)₂Fe(SO₄)₂·6H₂O, and 0.058 g of sodium chloride, NaCl, in 12.5 mL of 0.4 mol.L⁻¹ sulfuric acid, H₂SO₄ and then diluting it to 1000 mL in a volumetric flask with 0.4 mol.L⁻¹ sulfuric acid [7].

An alternative method that has also been used consists on preparing concentrated stock solutions of 0.5 mol.L⁻¹ ferrous ammonium sulfate and 0.5 mol.L⁻¹ sodium chloride, dissolving separately 9.804 g of ferrous ammonium sulfate and 1.462 g of sodium chloride in about 30 mL of 0.4 mol.L⁻¹ sulfuric acid and then diluting to 50 mL in volumetric flasks with the same acid solution. These solutions are stored in the dark and the dosimetric solution is then prepared diluting 1 mL of each concentrated stock solution to 500 mL in a volumetric flask with 0.4 mol.L⁻¹ sulfuric acid.

The 0.4 mol.L⁻¹ sulfuric acid solution is prepared diluting 22.5 mL of concentrated sulfuric acid solution (98%) in about 500 mL of triply-distilled water in a volumetric flask. It is then diluted with triply-distilled water to give 1000 mL of solution.

All Fricke solutions must be stored away from natural and artificial light sources to avoid oxidation. Since the Fricke dosimetric solution response is extremely sensitive to impurities, particularly organic ones, organic materials shall not be used, as well as metal, and all reagents must be of sufficient high purity. Thermal oxidation shall be avoided and temperatures during the irradiation and spectrophotometric analysis must be determined to correct the result of absorbed dose.

2.2. Assessment of Dose Rates

The assessment of gamma dose rates in the irradiation facility was carried out by the determination of dose rates in multiple locations. The absorbed dose was measured using Fricke and thermoluminescent dosimeters in the positions shown in fig. 1.
The samples of the dosimeters were located near the source mid-height and irradiated at electronic equilibrium conditions in appropriate acrylic boxes (Fricke dosimeters were irradiated in acrylic boxes filled with water, as it is shown in Fig.2).
The TLD-800 dosimeters, previously calibrated in the dose range of 25 to 1000 Gy, were irradiated in February 20, 2009. About 6 to 8 samples were irradiated at each location. The Fricke dosimeters were irradiated in March 20, 2009, and 3 samples were used in each location, starting from the distance of 0.05 m to the source. The solution absorbance was read with an UV spectrophotometer (SHIMADZU UV 2401 PC) and the absorbed dose was calculated using the expression

\[
Dose = 2.77 \times 10^2 \frac{\Delta A}{[1 + 0.007(t - 25)] \times [1 + 0.0015(t' - 25)]}
\]

where \(\Delta A\) is the increase in absorbance, \(t\) is the temperature during the spectrophotometric analysis and \(t'\) is the temperature during the irradiation [8].

3. RESULTS AND DISCUSSION

Based on the results obtained with the dosimetric analysis, the estimated dose rate at each location was calculated. The result is given in Fig. 3, and, as it can be seen, there was conformity between the two dosimetric systems (Fricke results were revised and adjusted to the day of TLD dosimetry to make this comparison possible).

![Figure 3. Comparison of dose rate values obtained with TLD-800 and Fricke dosimeters in 02-20-09](image)

The error bars presented in figure 3 represents graphically the uncertainty of each dosimetric system, estimated based on the guide to the expression of uncertainty in measurement –
GUM [9]. The top statistical uncertainty of the dose rate values obtained with the Fricke dosimeter was 5% and, with the TLD-800, 15%, at the 95% confidence level. This high uncertainty value for the TLD is due to the fact that a new batch of dosimeters was used.

Based on this data it was possible to express the dose rate as a function of the distance to the source. The curves that best fit the obtained data are shown in Fig. 4.

![Figure 4: Obtained curves of dose rate versus distance to the source in 02-20-09](image)

Equation 2 represents the red curve (from 0 to 0.20 m to the source) and equation 3, the blue one (over 0.20 m to the source).

\[
\text{Dose rate} = 17353e^{-7.2642d} \tag{2}
\]

\[
\text{Dose rate} = 380.88d^{1.5316} \tag{3}
\]

where \(d\) is the distance to the source.

These equations allow the calculus of the dose rate at any distance to the source in the irradiation facility. Revising and adjusting the dose rate based on Cobalt-60 half-life (5.27 years) it is possible to obtain the dose rate for any date, which is extremely important to determine the position and time of exposure of the products and to have an estimation of the absorbed dose.
4. CONCLUSIONS

The obtained results have shown applicability of Fricke and thermoluminescent dosimetric systems in the assessment of dose rates and in quality assurance programs. Despite being more practical and easy to work with, the TLD 800 system has presented greater uncertainty than the Fricke system which is considered a reference-standard for dose values from 40 to 400 Gy. Therefore, the main advantage of TLD 800 is its wider measurement spectrum of doses from µGy to $10^5$ Gy.

In order to guarantee the quality of the services provided in a gamma irradiation facility it is important to maintain routine dose measurements to control the product absorbed dose for each irradiation process. It is also of great importance to determine the dose distribution throughout the products irradiated to certify the all parts have received the recommended dose, without exceeding the limits.

The Fricke and TLD 800 dosimetric systems were shown practical and effective on the absorbed dose evaluations, allowing an implementation of quality assurance program in the gamma irradiation process. The uncertainty of the dose rate values obtained with the Fricke dosimeter was 5% and, with the TLD-800, 15%, at the 95% confidence level.

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REFERENCES
