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by

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ABSTRACT

Three different dosimeters were used to calibrate Savannah River Plant ^{60}Co sources having intensities greater than 10^7 rads/hr. These dosimeters are (a) ceric sulfate dissolved in 0.4M H_2SO_4, (b) oxalic acid dissolved in water, and (c) a commercially available nylon film containing a radiochromic dye. Response per unit dose to these dosimeters is independent of radiation intensity at 10^4 to 10^{11} rads/hr. The dosimeters were calibrated at 6.0×10^5 rads/hr with a ^{60}Co source whose intensity was determined with the standard Fricke dosimeter. For the sources at 10^7 rads/hr or greater, intensities were calculated from slopes of linear plots of dosimeter response versus irradiation time. Individual dose rates varied from 1.0×10^7 to 4.6×10^7 rads/hr. Each source was calibrated with at least two different dosimeters. Relative standard deviations varied from 2 to 9%. A conservative estimate of the uncertainty in the accuracy of these dosimeters is 10%. Of the three dosimeters, the nylon film is easiest to use and is therefore recommended for future calibrations.

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CALIBRATION OF INTENSE ^{60}Co GAMMA RAY SOURCES AT THE SAVANNAH RIVER PLANT

INTRODUCTION

The Savannah River Plant (SRP) has seven ^{60}Co γ-ray sources with intensities greater than 10^7 rads/hr and one of less than 10^7 rads/hr. These sources are used for investigating the effects of radiation on materials of construction and on chemical reactions. For quantitative evaluation of these effects, the intensity of each source must be known. These intensities are determined by irradiating calibrated chemical dosimeters, which are solutions or materials that possess some property whose change can be correlated with radiation dose. The most widely used dosimeter is the Fricke dosimeter,[1] which is a solution of ferrous sulfate in 0.4M H_2SO_4. In this solution, radiation causes oxidation of ferrous ion to ferric ion. Because the yield of ferric ion also depends on the amount of dissolved oxygen in the solution, this dosimeter cannot be used to calibrate sources with intensities greater than $\sim 3 \times 10^6$ rads/hr.[1]

Sources of high intensities can be measured with dosimeters which can be calibrated at lower intensities with the standard Fricke dosimeter. This calibration is valid only if the dosimeter response per unit dose is independent of the radiation intensity. Three dosimeters that have this property are a ceric sulfate solution,[2] an oxalic acid solution,[3] and a commercially available nylon film containing a radiochromic dye.[4,5]

This report describes the methods for calibrating the high intensity ^{60}Co γ-ray sources at SRP with the three dosimeters. Methods for calibrating the three dosimeters and a low intensity ^{60}Co source with the Fricke dosimeter are also presented. The eight ^{60}Co sources are briefly described.

DOSIMETER CALIBRATION TECHNIQUES

^{60}Co Sources at SRP

SRP has eight ^{60}Co γ-ray sources for studying the effects of radiation on materials of construction and on chemical reactions. The lowest intensity source is a commerically available *Gammacell 220* (Atomic Energy of Canada, Ltd., Ottawa, Canada) containing 7800 Ci of ^{60}Co (as of January 1976) in a lead shield. Samples are irradiated by mechanically moving them into the radiation field. The other seven sources were constructed at SRP and contain an array of ^{60}Co slugs submerged in ∿24 ft of water for shielding purposes. The largest source contains approximately 1×10^6 Ci of ^{60}Co.[6] Samples are irradiated by manually lowering them into the radiation fields of the sources through air-filled tubes or directly through the water.

Figure 1 is a photograph of three sources taken through 24 ft of water. The two smaller sources are positioned around air-filled access tubes, which are curved for shielding purposes. The large source surrounds a sample holder that has been lowered directly through the water.

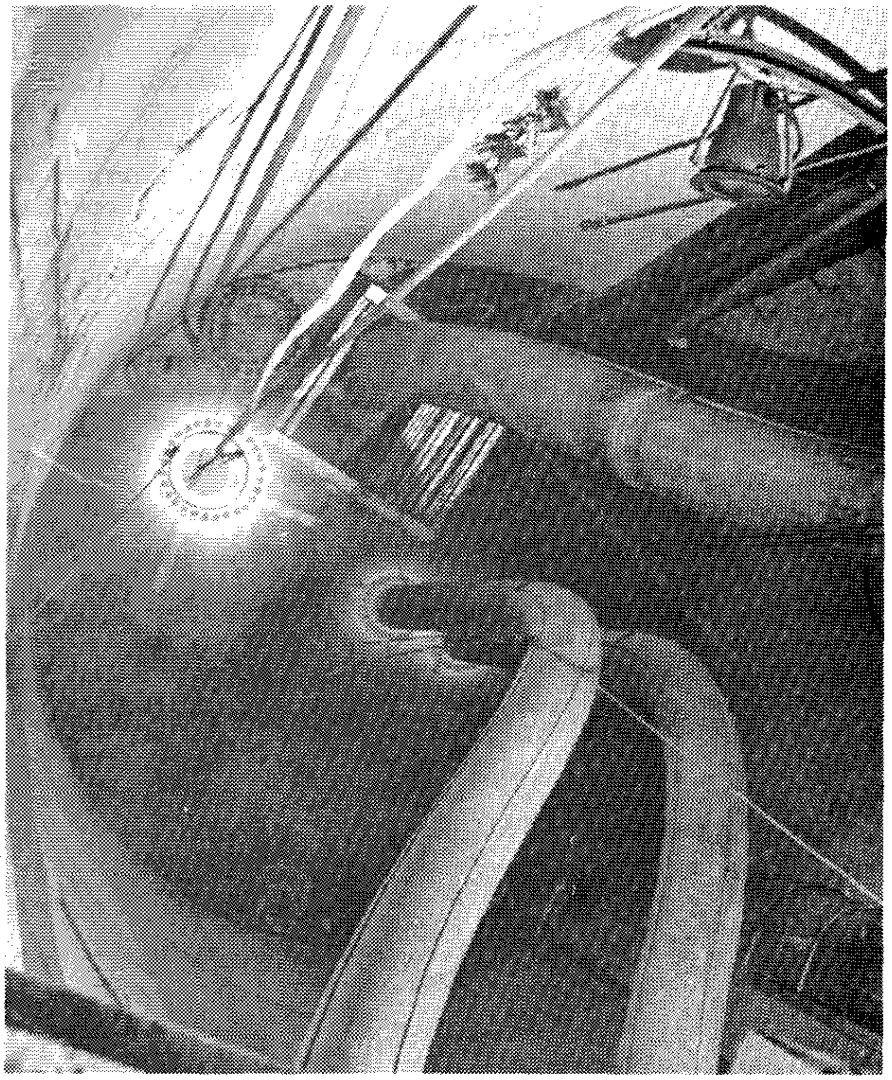

FIGURE 1. Three Intense ^{60}Co Sources at SRP

Properties of the Dosimeters

All dosimeter solutions were prepared with reagent-grade chemicals and triply distilled water. Composition, pertinent properties, and analytical techniques for these dosimeters are listed in Table 1.

TABLE 1

Dosimeter Solutions for Calibrating SRP ^{60}Co Sources

Composition	*Response Measured*	*Yield/100 eV*[a]	*Analytical Technique*
10^{-3}M $FeSO_4$, 10^{-3}M NaCl, 0.4M H_2SO_4[b]	Appearance of Fe^{3+}	15.6	Spectrophotometric at 304 nm
4×10^{-4}M $Ce(SO_4)_2$, 0.4M H_2SO_4	Disappearance of Ce^{4+}	2.3 - 2.6[c]	Spectrophotometric at 320 nm
10^{-2}M $H_2C_2O_4$	Disappearance of $H_2C_2O_4$	4.9[c]	Titration of H^+ with standard NaOH

---

[a] Ions or molecules formed or destroyed per 100 eV of radiation absorbed.

[b] Fricke dosimeter.

[c] Values reported in the literature vary widely.

The Fricke dosimeter has been extensively studied, and the values determined by several workers for the radiolytic yield of ferric ion are in excellent agreement.[1] This dosimeter is not significantly affected by impurities and is currently accepted as a primary standard for calibrating low-intensity ^{60}Co sources. It cannot be used to calibrate intense sources as it is only accurate up to about 4×10^4 rads because of its dependence upon dissolved oxygen. A dose of 4×10^4 rads corresponds to only 12 seconds irradiation in a source with an intensity of 10^7 rads/hr.

The ceric sulfate dosimeter is based upon the radiolytic reduction of ceric ions. It is only slightly dependent on dissolved oxygen and thus can be exposed to megarad doses.[2] Because ceric ion is easily reduced, the response of this dosimeter is sensitive to trace impurities which accounts in part for the range of yields reported in the literature.[2] For this reason, this dosimeter can be used only as a secondary standard, i. e., solutions must be calibrated against the Fricke dosimeter before they can be used to calibrate ^{60}Co sources.

The oxalic acid ($H_2C_2O_4$) dosimeter is based on radiolytic degradation of the oxalic acid. Products are carbon dioxide and small amounts of glyoxal and glyoxalic acid.[3] This dosimeter can also be irradiated to megarad doses, but suffers the dual disadvantage of responding non-linearly with dose and being affected by impurities. This dosimeter is also a secondary standard and must be calibrated against the Fricke dosimeter before use.

The nylon film dosimeter is impregnated with an aminotriphenylmethane dye derivative. The dye turns blue (λ_{max} = 600 nm) when irradiated. The dosimeter is available in batches of 1-cm x 1-cm squares approximately 0.06 mm thick.[7] Because of variations in thickness from batch to batch, this dosimeter is also only a secondary standard and must be calibrated against the Fricke dosimeter. Precautions to be followed when using this film are: (1) it should be protected from fluorescent light or bright daylight, (2) the absorbance should be measured 24 hr after the dosimeters are irradiated to eliminate any possible time dependence of color development, and (3) the films should be kept dry and handled with tweezers. Once developed, the color is stable for several days.

Calibration of the Dosimeters

The intensity of the *Gammacell 220* source for calibrating the dosimeters was measured with the Fricke dosimeter. Aliquots (5 ml) of the dosimeter solution were irradiated for various times at 23°C, and absorbance of the solutions was measured in a 1-cm cell at 23°C with a *Beckman DU* Spectrophotometer. The ferric ion concentration was calculated from the equation

$$[M] = (Abs)/\varepsilon \qquad (1)$$

where (Abs) is the absorbance (corrected for the absorbance of the solution before irradiation), and ε is the molar absorptivity of ferric ion ($2167 M^{-1} cm^{-1}$ at 23°C). Typical results are shown in Figure 2. The dose received by a sample is calculated from the equation

$$D = It + D_o \qquad (2)$$

where D is the dose in Mrads, t is the irradiation time, I is the intensity of the source, and D_o is the dose to the sample while it was traveling into and out of the radiation field. The intensity is calculated from the equation

$$I(\text{rads/hr}) = \frac{(S_2)(6.02 \times 10^{23}\ \text{ions/mole})(60\ \text{min/hr})}{(G)(6.24 \times 10^{13}\ \text{eV/g-rad})(\rho)(10)} \qquad (3)$$

where S_2 is the slope of Figure 2 in moles/ℓ-min, G is the 100-eV yield of ferric ions (15.6 ions/100 eV), and ρ is the density of 0.4M H_2SO_4 (1.04 g/mℓ at 23°C).[8] Average values for I and D_o

determined from least squares analysis of data from three determinations were 0.601 (±0.004) Mrad/hr and 7.05 (±0.04) Mrad, respectively.

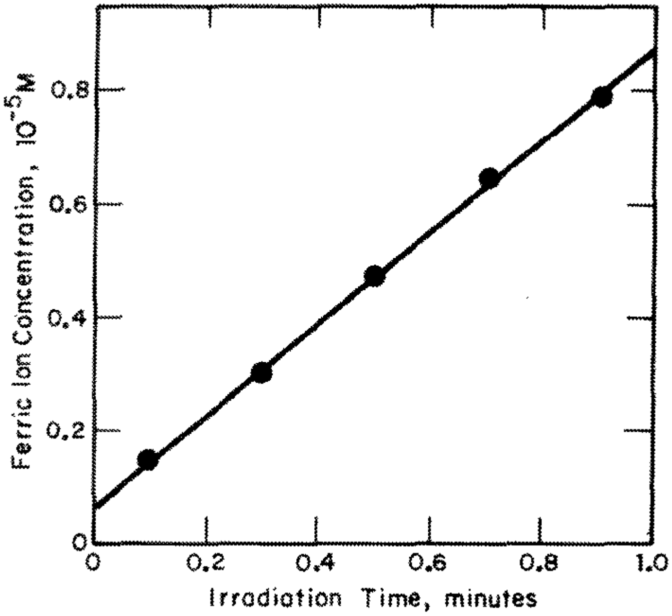

FIGURE 2. Calibration of *Gammacell 220* with Fricke Dosimeter

Ceric Sulfate

Aliquots (5 ml) of a 0.0037M $Ce(SO_4)_2$-0.4M H_2SO_4 solution were irradiated in the *Gammacell* in the same configuration as were the Fricke solutions. For the ceric sulfate dosimeter, irradiation times were ∿100 times longer than with the Fricke solutions. Because this dosimeter is slightly affected by temperature, samples were preheated to 35°C, the ambient temperature in the *Gammacell* during irradiation. Preliminary experiments established that 35°C was approximately the temperature that the samples would reach in the intense ^{60}Co sources. The absorbances of the irradiated aliquots and original solution were measured; the concentration of ceric ion was calculated from Equation 1 with $\varepsilon = 5610\ M^{-1}cm^{-1}$. Corrections for temperature differences of the samples between irradiation and analysis were negligible. Results for a typical calibration are shown in Figure 3. The 100-eV yield for reduction of $Ce^{4+}[G(Ce^{3+})]$ at 35°C was calculated from

$$G(Ce^{+}) = \frac{(S_2)(6.02 \times 10^{23}\ \text{ions/mole})(100)}{(6.24 \times 10^{19}\ \text{eV/g}\cdot\text{Mrad})(\rho)(10^3\ \text{ml/l})} \qquad (4)$$

where S_3 is the least squares slope (M/Mrad) of the line in Figure 3 and ρ is the density of 0.4M H_2SO_4 at 35°C (1.02 g/ml).[8] The result, 2.33 ions/100 eV, was in agreement with published values.[9] The solution was stored in the dark and then used as soon as possible to calibrate the intense sources.

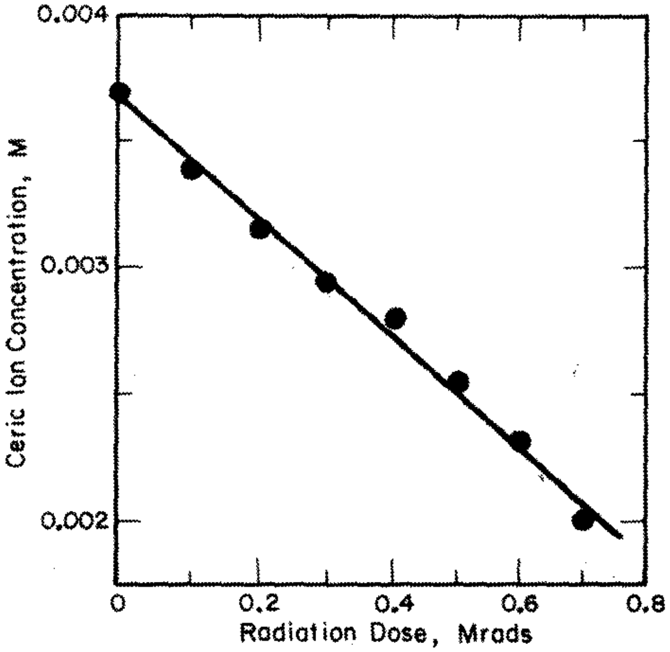

FIGURE 3. Calibration of Ceric Sulfate Dosimeter

Oxalic Acid

Aliquots of 0.02M $H_2C_2O_4$ in water were irradiated in the *Gammacell* by the same procedure used for the other dosimeters. No temperature corrections were made because the response of this dosimeter is not significantly affected by variations between 20°C and 60°C.[10] The amount of oxalic acid remaining after each irradiation time was determined by titrations with sodium hydroxide solutions after complete removal of radiolytically formed carbon dioxide.[3] Because the disappearance of oxalic acid is nonlinear with dose, the results were analyzed with the equation

$$D = k\ C_o \log C_o/C \tag{5}$$

where D is the dose in Mrads, k is a proportionality factor, and C_0 and C are concentrations in moles/ℓ before and after irradiation. Once k is known, the solution can then be used to measure intensities of other sources, since D = It. A typical plot of results for a *Gammacell* irradiation is shown in Figure 4. Least squares analysis of the slope gave a value of 4.15 Mrad M^{-1} for k, which is within the range of published values.[3]

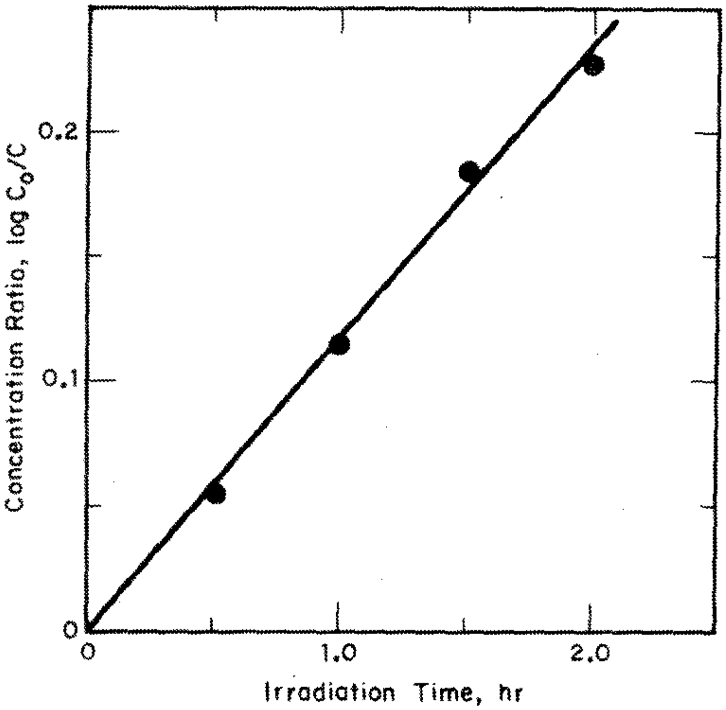

FIGURE 4. Calibration of Oxalic Acid Dosimeter

Film Dosimeter

The nylon dosimeter films were placed between two nylon pieces (1 in. x 0.75 in. x 0.38 in.) and irradiated for various times in the *Gammacell* at 35°C. The change in absorbance was determined at 600 nm with a digital spectrophotometer and plotted as a function of dose (Figure 5). The following calibration equation, where Abs is the absorbance, was derived by numerical regression analysis of the data:

$$D = 0.613 \times 10^{-4} + 1.054\ (\text{Abs}) + 0.0960\ (\text{Abs})^2 - 0.0210\ (\text{Abs})^3 \qquad (6)$$

This equation described the data very well below absorbances of 1.0. The standard deviation of the relative error between calculated and observed doses for 14 data points below an absorbance of 1.0 was 4.5%. With large doses, the response of the nylon dosimeters was nonlinear because dosimeter sensitivity decreases.[4,5]

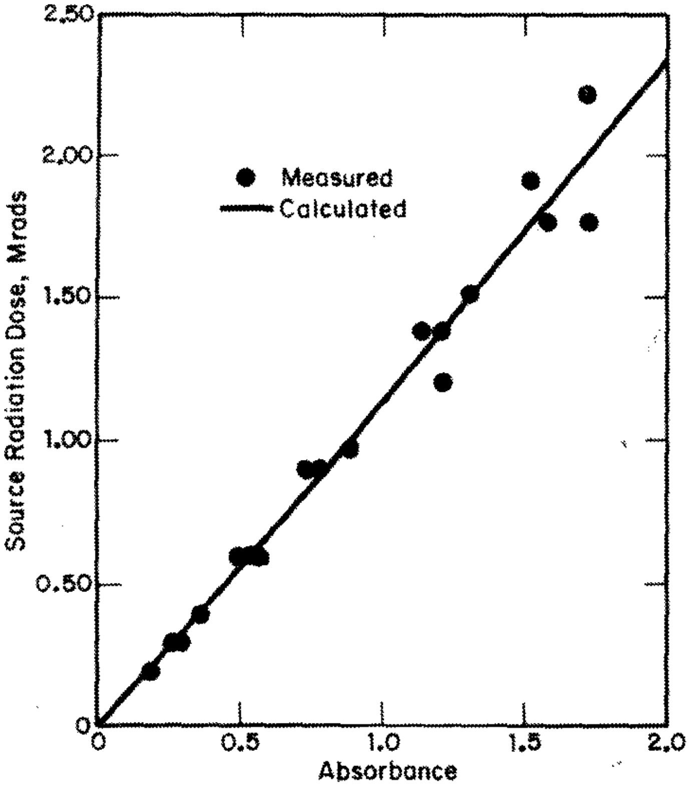

FIGURE 5. Calibration of Nylon Film Dosimeters

CALIBRATION OF THE INTENSE ^{60}Co SOURCES

After the three dosimeters were calibrated against the Fricke dosimeter in the *Gammacell*, they were used to measure the intensities of other ^{60}Co sources.

Results for irradiation of calibrated ceric sulfate solutions in one of the intense sources are shown in Figure 6. Temperature during irradiation was 35°C. Intensity was calculated from Equation (3) with S the slope of Figure 6 in moles/ℓ-hr; G, 2.33 ions/100 eV; and ρ, 1.02 g/ml.

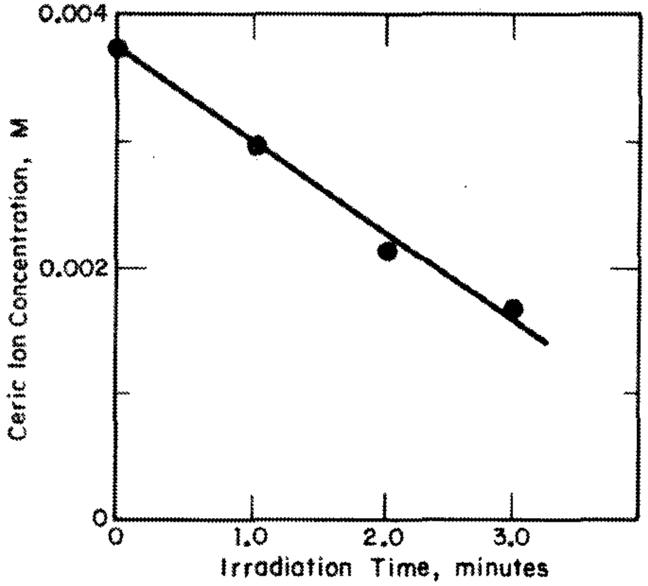

FIGURE 6. Calibration of an Intense ^{60}Co Source with Ceric Sulfate Solution

Figure 7 shows a plot of results for an intensity determination with a calibrated oxalic acid solution. Rearranging Equation (5) with D = It gives

$$\log(C_o/C) = (I/k\ C_o)\ t \qquad (7)$$

Thus, source intensity I can be calculated from the slope of Figure 7 with k = 415 Mrad M^{-1}.

Results from irradiation of six film dosimeters in an intense ^{60}Co source are shown in Figure 8. For this source, the access tube was filled with water for shielding and temperature control. Dosimeters were irradiated in waterproof secondary containers. The positive intercept corresponds to the dose the samples received while they were manually lowered into and pulled out of the radiation field. For the data in Figure 8, the positive intercept corresponds to an irradiation time of 4 seconds. With this type of access tube, results were fitted to Equation (2) by least squares analysis,and source intensity I was calculated from the slope. With sources containing air-filled access tubes where samples could be introduced and removed faster, lines defined by the data passed through the origin. Also, no significant difference in source intensity I resulted whether the data were fitted to Equation (2) or to the equation D = It.

Table 2 compares results for intensity determinations for selected positions of 4 of the 7 intense ^{60}Co sources using at least 2 different dosimeters. All data are corrected for radioactive decay to January 1, 1976. All relative standard deviations are less than 10%. Even though results with the ceric sulfate dosimeter are consistently slightly higher than those with the other dosimeters, the agreement is acceptable. The higher results may be due to the effect of possible impurities in the specific ceric sulfate solutions used to determine the results in Table 2. Since the dosimeters are completely independent of each other and are not affected by dose rate, the above precision can be considered a measure of dosimeter accuracy.

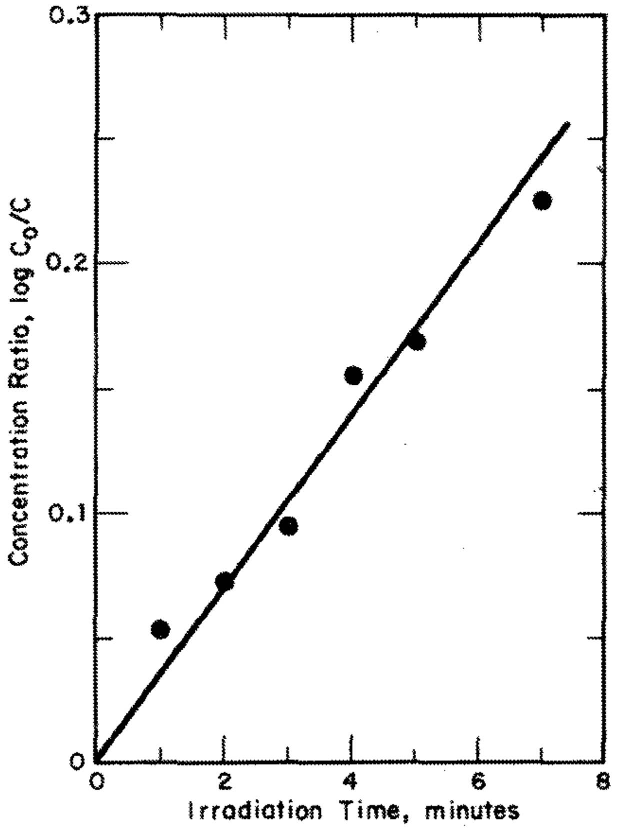

FIGURE 7. Calibration of an Intense ^{60}Co Source with Oxalic Acid Solution

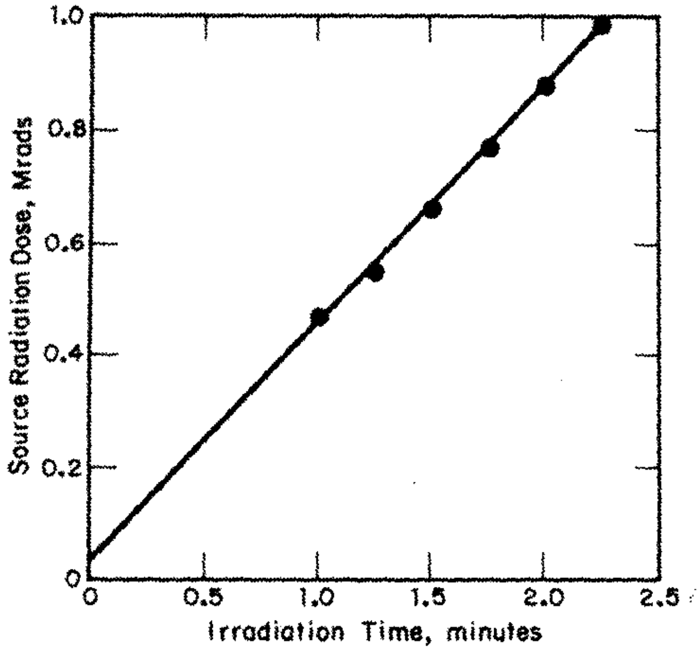

FIGURE 8. Calibration of an Intense ^{60}Co Source with Nylon Film Dosimeters

TABLE 2

Calibration of Intense ^{60}Co Sources

Source[a]	*Dosimeters*	*Intensity, Mrads/hr*	*Average Intensity, Mrads/hr*	*Relative Standard Deviation, %*
Large Source,				
Outer Position	Nylon film	44.8		
	Ceric sulfate	46.1	45.5	2.7
Middle Position	Nylon film	41.6		
	Ceric sulfate	44.6	43.1	6.3
Inner Position	Nylon film	36.0		
	Ceric sulfate	39.6	37.8	8.6
West Source	Nylon film	16.0		
	Ceric sulfate	16.8	16.4	4.3
East Source	Nylon film	12.9		
	Ceric sulfate	14.8		
	Oxalic acid	12.6	13.5	8.0
	Nylon film	13.6		
Source in SRP Reactor Bldg.	Nylon film	10.2		
	Oxalic acid	10.4	10.3	0.2

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[a] Radiation dose corrected for radioactive decay to January 1, 1976.

CALCULATION OF ABSORBED DOSES IN SYSTEMS OTHER THAN 0.4M H_2SO_4

With the above secondary standards, the intensities were measured in rads/hr in 0.4M H_2SO_4, the solvent for the primary standard Fricke dosimeter. Dose rates in systems other than 0.4M H_2SO_4 can be calculated by applying appropriate mass attenuation coefficients.[11] For ^{60}Co gamma radiation, these are proportional to the electrons per unit mass of the material.[11] This function is Z/A where Z is the atomic number and A is the atomic weight. For materials other than the elements, $Z/A = \Sigma_i W_i \, (Z/A)_i$, where W_i is the weight fraction of the ith element. For 0.4M H_2SO_4, Z/A is 0.553; and for water, it is 0.556. Thus, the intensity in water is given by

$$I_{H_2O} = I_{0.4M\ H_2SO_4} \ (0.553/0.556) \qquad (8)$$

CONCLUSIONS

To calibrate ^{60}Co sources with intensities greater than 10 Mrad/hr, the three dosimeters (ceric sulfate solution, oxalic acid solution, and nylon film impregnated with an aminotriphenylmethane dye derivative) are acceptable. The nylon film dosimeter has the added advantages of ruggedness and ease of handling and is thus recommended for future calibrations. As with the others, the nylon film dosimeter requires prior calibration before use.

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