A MODIFIED CERIC SULPHATE DOSIMETRIC SYSTEM USING SODIUM OXALATE FOR HIGH-DOSE DOSIMETRY

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Abstract - The radiolytic reduction of the Ce^{4+} in a 0.8N sulfuric acid was examined, with the objetive of measuring large radiation doses. The ceric sulphate dosimeter was tested in the range from 1 to 20 kGy. The 0.01 M sodium oxalate solution was chosen instead of the arsenious oxide solution (used in the conventional method), in order to obtain a less expensive dosimetric system than that normally used for routine chemical dosimetry. The absorption spectrum of the Ce^{4+} solution presents an absorption peak at 370 nm (2.99×10^{-4}M); the obtained molar extinction coefficients of Ce^{4+} and Ce^{3+} were 5670 and 2.7 $M^{-1}.cm^{-1}$ respectively. The purpose of this work was to establish a simple, reliable and inexpensive routine chemical dosimetric system for high doses.

INTRODUCTION

There are several applications for radiation processing in medical products sterilization, polymer modification and food treatment. An adequate dosimetry is required to assure that the products are appropriately treated.

The ceric sulphate system has been used for about 40 years to measure radiation doses, usually by the technique of spectrophotometry of ceric ion absorption at the 320 nm wavelength. Doses that can be measured with the ceric sulphate solution range from 10 Gy to 200 kGy, that is one of the widest ranges of any dosimeter. It is particularly useful in the 1 to 40 kGy range, which covers the doses normally used for preserving food and sterilizing articles[1]. Its main advantages in routine dosimetry are the excellent stability before and after irradiation, the practically yield $G(Ce^{3+})$ independence of dose rate within the range normally used and the well defined reaction mechanism. The most serious disadvantage of the ceric sulphate dosimeter is its relatively high sensibility to impurities and to light[2].

The ceric sulphate solution was chosen for this study, and the process consists in measuring the absorbed dose by the reduction of ceric to cerous ions in 0.8 N sulfuric acid[3]. The absorbed dose is derived from the difference in the ceric ion concentration measured before and after an irradiation. The accuracy of such systems is better than $\pm$ 2%[4]. The conventional chemical dosimetry method using the ceric sulphate system has been described by some authors for high-dose dosimetry[5,6].

At higher concentrations of ceric ions (> 10 mM) the change can be measured either by titration with ferrous sulphate or potentiometrically[3]. The degree of reduction of ceric ions varies

linearly with the dose at concentrations below 50 mM of Ce^{4+} ions until the ceric ions are completely reduced. The yield $G(Ce^{3+})$ is dependent of the Ce^{4+} ion concentration from 0.1 to 50 mM[4].

In the present work the ceric sulphate system was prepared using the ferroin indicator solution and a 0.01 M sodium oxalate solution, establishing a standard dosimetric system that is less expensive than that normally used for routine chemical dosimetry.

DOSIMETRIC SOLUTIONS

The ceric sulphate dosimeter consists of the ceric ammonium sulphate, $(NH_4)_4\ Ce(SO_4)_4.2H_2O$, in 0.8 N sulfuric acid, using the ferroin indicator solution and a 0.01 M sodium oxalate solution, establishing a standard dosimetric 0.1 N solution. The sodium oxalate solution was chosen instead of the arsenious oxide solution (conventional method).

The dosimeter solutions were prepared with bidestilled water and reagents of analytical grade. The dosimetric solution 0.02 N was prepared by diluting the stock solution (0.1 N) that was used as standard solution to calibrate the spectrophotometer and determine its molar extinction coefficient. Before this solution could be used, however, the ceric ion concentration had to be accurately determined.

In the standardization of the 0.1 N solution, it was employed a 0.1 N ferric ammonium sulphate solution diluted in 0.8 N sulfuric acid and slowly titrated with ceric ammonium sulphate using a 0.025 M ferroin indicator.

This study was realized using a ^{60}Co source (185 TBq) of the Department of Nuclear Techniques Applications in Engineering and Industry at IPEN. The dosimeters were always positioned at 5 cm

from the source, using adequate cells (internal diameter of 13.3 mm). The dosimeters were always prepared by filling 3 mL clean glass with the ceric dosimetry solution. After the irradiations the absorbance of the diluted solutions (both the dosimeter and the blank solutions) was measured using a FEMTO 482 spectrophotometer at 370 nm and matched special glass cells. As reference blank a solution of 0.8 N sulfuric acid was employed.

RESULTS

The modified ceric ammonium sulphate dosimeter was carefully standarized for ceric ion concentration and accurately pipeted (different volumes) into volumetric flasks, where each one was completed to volume with 0.8 N sulfuric acid.

The absorbance of this chemical dosimeter was measured varying the Ce^{3+} concentration up to 8.4×10^{-4}M, using an initial Ce^{4+} solution of 1×10^{-4}M. In Fig. 1 a linear relationship can be observed. In this low concentration range of Ce^{4+} and Ce^{3+} ions the molar extinction coefficients were determined as 5670 and 2.7 $M^{-1}.cm^{-1}$ respectively, using the data of Fig. 1.

In order to test the chemical dosimeter in gamma radiation in the range 0.3 to 20 kGy, it was necessary to divide the stock solution in two different parts. For the lower dose range, an initial Ce^{4+} ion concentration of 2.99×10^{-4}M was prepared. The dosimeters were irradiated between 0.3 and 1.0 kGy. A linear response was obtained as can be seen in Fig.2.

In the case of the higher dose range, the initial Ce^{4+} concentration was 1.72×10^{-2}M. The dosimeters were exposed to the ^{60}Co source up to 20 kGy. In Fig.3 the linearity can be observed

until 5 kGy. After this dose the dosimeter presents a saturation at 10 kGy.

The data presented in Fig.2 and 3 cannot be directly compared with those of Yefsah et al[6] because the experimental procedures of the stock solution preparation were different.

CONCLUSION

A modified ceric sulphate dosimeter was developed for routine dosimetry using sodium oxalate instead of the conventional arsenious oxide solution. It showed to be suitable and reliable. The methodology for use of this dosimetry system was established in the range of 0.3 to 10 kGy.

ACKNOWLEDGMENTS

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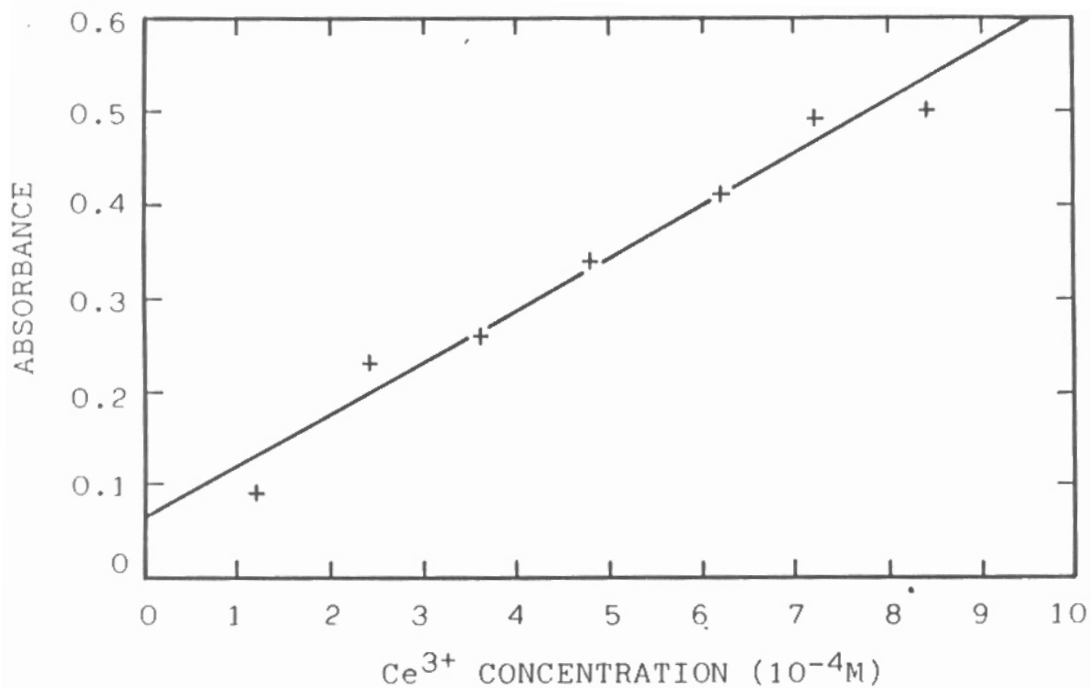
Modified ceric sulphate dosimeter response in function of Ce^{3+} concentration. Initial Ce^{4+} concentration: 1×10^{-4} M.
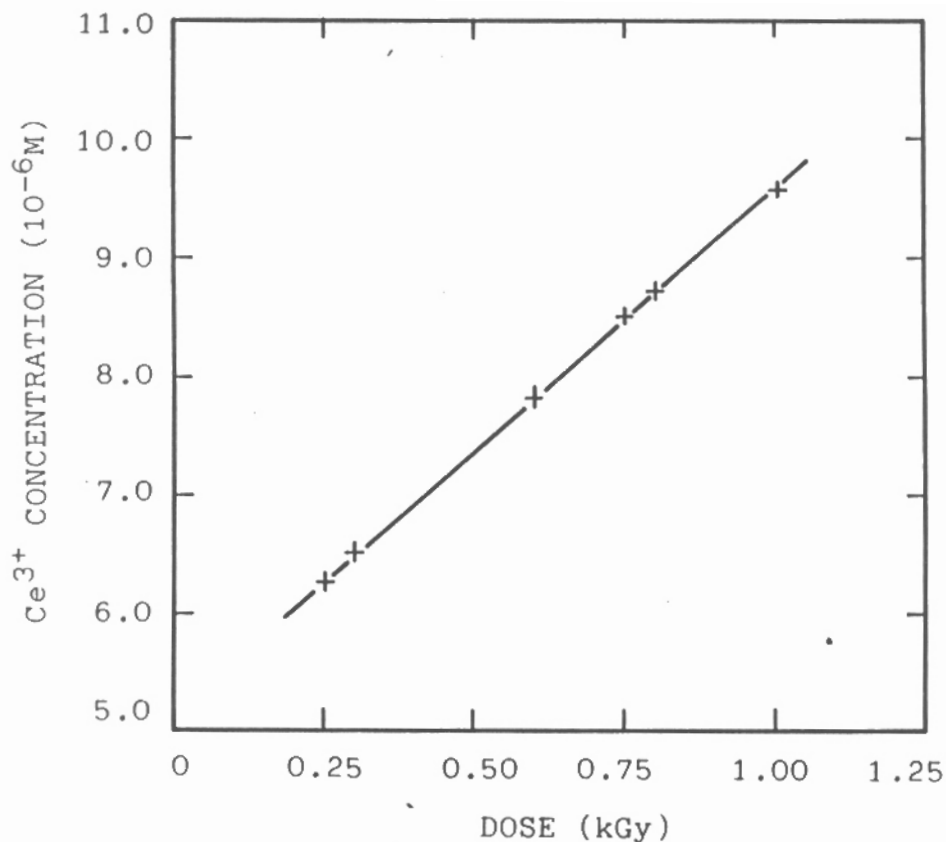
Dose response curve of the modified ceric sulphate dosimeter irradiated with ^{60}Co. Initial Ce^{4+} concentration: $2.99 \times 10^{-4}M$.
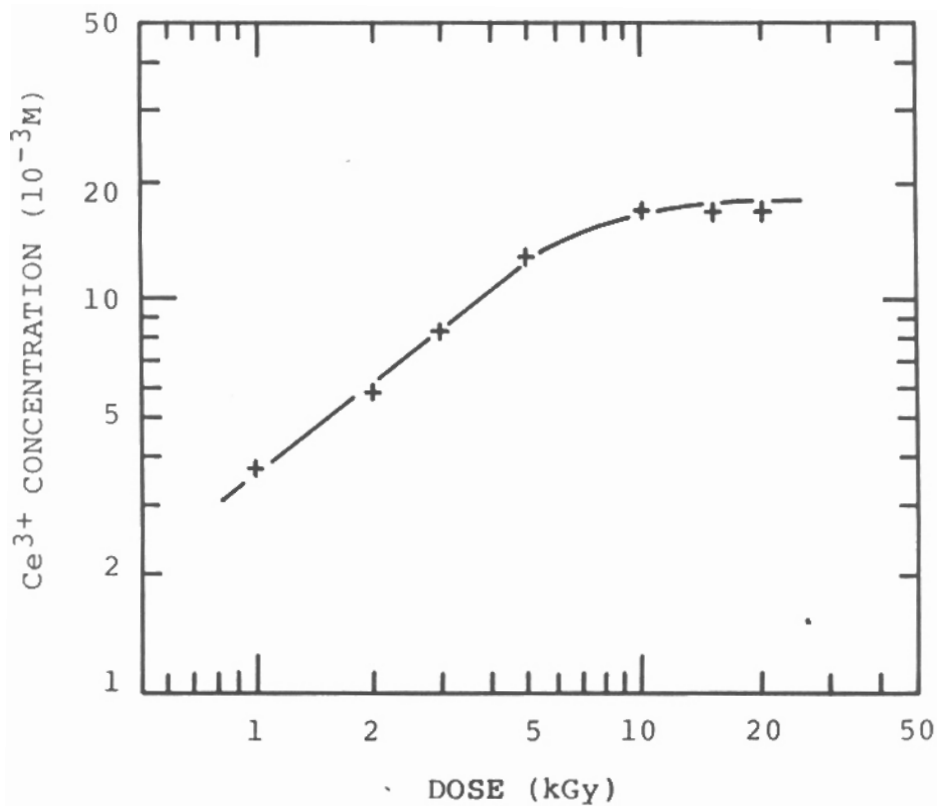
Dose response curve of the modified ceric sulphate dosimeter irradiated with ^{60}Co. Initial Ce^{4+} concentration: $1.72 \times 10^{-2}M$.