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EPR dosimetry using commercial glasses for high gamma doses

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Abstract

Commercial transparent and colored (bronze, brown, and green) glasses were studied as possible dosimeters for high gamma doses using electronic paramagnetic resonance (EPR). All EPR spectra showed the characteristic Fe^{3+} signals, $g = 4.27$ and 2.01. The signal at $g = 2.01$ presented a more useable behavior for the calibration curve. All samples showed their usefulness as high dose dosimeters.

Keywords: Glass; Dosimeter; EPR; High doses

1. Introduction

Several kinds of dosimeters have been proposed, tested, and are presently in use for high dose dosimetry (McLaughlin et al., 1989; Farrar, 1998). The dosimetric properties of colorless window glass manufactured in Brazil have been studied using different evaluation techniques (Quezada and Caldas, 1999; Caldas and Quezada, 2002; Caldas and Teixeira, 2002; Rodrigues Jr. and Caldas, 2002). Special glasses have been studied as possible radiation dosimeters at doses up to 100 kGy (Debnath, 1995; Teixeira et al., 1996; Randhawa and Virk, 2000; Dogan and Tugrul, 2001).

These kinds of glasses showed various advantages in relation to other systems, such as: easy handling, small size, and low cost. However, thermal decay after room temperature irradiation is one disadvantage in relation to other dosimeters. This problem may be avoided by standardizing the time between irradiation and measurement (Quezada and Caldas, 1999; Rodrigues Jr. and Caldas, 2002; Caldas and Teixeira, 2002) or by applying special post-irradiation treatments to the material (Caldas and Quezada, 2002).

In this work, a new high dose dosimetry system utilizing glass samples for dose measurements is worked out for applications in industrial areas of commercial sterilization processes, pasteurization, food preservation, and treatments of several materials. Glass samples were studied for their main dosimetric characteristics, using electronic paramagnetic resonance (EPR), for high dose dosimetry.

2. Materials and methods

Commercial glass samples (transparent, bronze, brown, and green), manufactured (float bath process at 1600 °C) by Cebracê, Brazil, with dimensions of $100 \times 100 \times 3\,mm^3$ were tested as radiation detectors. The main difference between this kind of glass and common glass is the high optical quality provided by the

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Table 1
Results of neutron activation analysis of colored glass samples (the intervals correspond to 1σ)

Element	Concentration ($\mu g/g^{-1}$)			
	Transparent glass	Bronze glass	Brown glass	Green glass
Ca	66000 ± 6000	68000 ± 5000	65000 ± 6000	73000 ± 5000
Na	9.97 ± 0.04	9.75 ± 0.04	9.86 ± 0.04	9.61 ± 0.04
Rb	52 ± 10	24 ± 4	28 ± 6	24 ± 5
Fe	645 ± 69	2409 ± 256	2980 ± 317	3368 ± 358
Co	0.28 ± 0.04	29 ± 3	54 ± 6	0.43 ± 0.05

float manufacturing process. This technology provides uniform thickness of a homogenous mass, free of optical distortions. These samples were cut in the dimensions of $4 \times 7 \times 3\,mm^3$, with an average mass of about 0.210 g. Table 1 shows the results of a neutron activation analysis of the four types of glasses, performed by the Radiochemistry Department of IPEN. The concentrations of Fe and Co in the colorless samples were only 645 and 0.28 μg/g, respectively. Note that the results in Table 1 represent the total amount of iron ions, both Fe^{2+} and Fe^{3+} ions.

In the mixture used for fusion at Cebracê, the bronze color was achieved by adding 24 ppm Se, 0.38% Fe and 34 ppm Co; in the case of the brown samples, 16 ppm Se, 0.44% Fe and 68 ppm Co was sufficient. The green color was obtained by adding only 0.53% Fe.

The irradiations were performed in air (room temperature) in the radiation field of a Gamma-Cell 220 (^{60}Co) system (dose rate of 4.96 kGy/h) under electronic equilibrium conditions, achieved by covering the samples with 6-mm-thick Lucite plates.

Thermal treatments (30 min at 400 °C) were applied to the glass samples intended for repeated utilization (reutilization). The EPR measuments were carried out using a BRUKER EMX spectrometer with a rectangular cavity (ER4102 ST), at room temperature, with microwave frequency of 9.75 GHz (band X), microwave power of 20 mW and with frequency and field modulation amplitude of 100 kHz and 0.1 mT, respectively. The magnetic field was varied between 1000 and 4000 G. This EPR spectrometer belongs to the Multi-users group of the Institute of Physics/USP. A pure quartz rod, flattened in one of the extremities, was necessary to insert the glass samples into the equipment's resonant cavity; the samples were fixed on the quartz rod with silicone paste. Due to the thermal fading of the glass spectra, all measurements in this work were taken exactly one hour after irradiation.

3. Results and discussion

Fig. 1 shows the optical absorption spectra of glass samples before irradiation, just to verify the absorption bands of the iron ions. In all cases of these glasses the main absorption band can be observed in the infra-red range, with a maximum around 1100 nm that is known to be related to Fe^{2+} ions. Two bands concerned with Fe^{3+} ions were observed at 380 and 420 nm and their intensities were lower than the Fe^{2+} band intensity. The spectra of bronze and brown glasses presented additional bands due to Co^{2+} ions at 520, 590 and 650 nm (Debnath, 1998).

Fig. 2 shows EPR spectra of glass samples (transparent, bronze, brown and green) irradiated with 5 kGy. These spectra present two signals, at $g = 4.27$ and $g = 2.01$. The Fe^{3+} ions can exist in silicate glass either in the substitutional sites or in the interstitial positions. The signal at $g = 4.27$ is referred to in the literature (Loveridge and Parke, 1971; Griscom, 1980; Wenbiao et al., 1985; Berger et al., 1995) as due to the tetrahedral Fe^{3+} ions in the substitutional silicon site under a rhombic distortion due to the presence of compensating cations (alkali metals) in its neighborhood. The pairs of exchange coupled Fe^{3+} ions can arise from interstitial Fe^{3+} ions after heat treatment and irradiation (Griscom, 1980; Griscom et al., 1999 and Debnath, 2001), which causes a resonance with g-factor around 2.0. The two signals can be independent of each other, because the amount of interstitial Fe^{3+} ions can vary from sample to sample, depending on the manufacturing process.

Another interpretation about the origin of the signal around $g = 2.0$ is related to the oxygen hole centers (Griscom, 1980; Ikeya, 1993), that should arise with electron centers. In this study, it would be expected that the Fe^{3+} ions are such electron centers, consequently the signal intensity at 4.27 should decrease with the absorbed dose. It can be observed in Fig. 3, however, that this intensity increases with the absorbed dose. Therefore, this fact corroborates that the signal at $g = 2.01$ is probably due to pairs of exchange coupled Fe^{3+} ions.

The details of the signal at $g = 2.01$ can be seen in Fig. 4; this signal was chosen for dose evaluation because the signal intensity at $g = 4.27$ does not show an uniform behavior, as can be seen in Fig. 3.

Fig. 5 shows EPR spectra of transparent glass samples irradiated with different absorbed doses of 1, 5, and

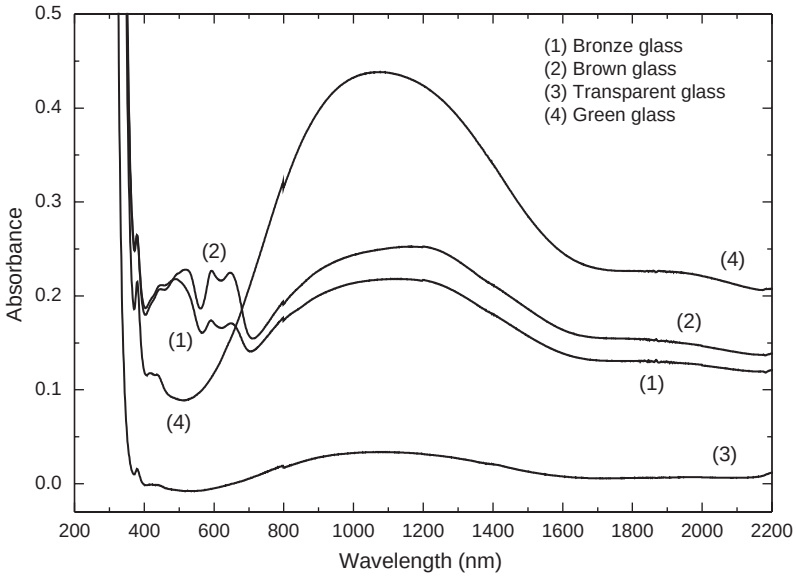

Fig. 1. Optical absorption spectra of the different glass samples before irradiation.

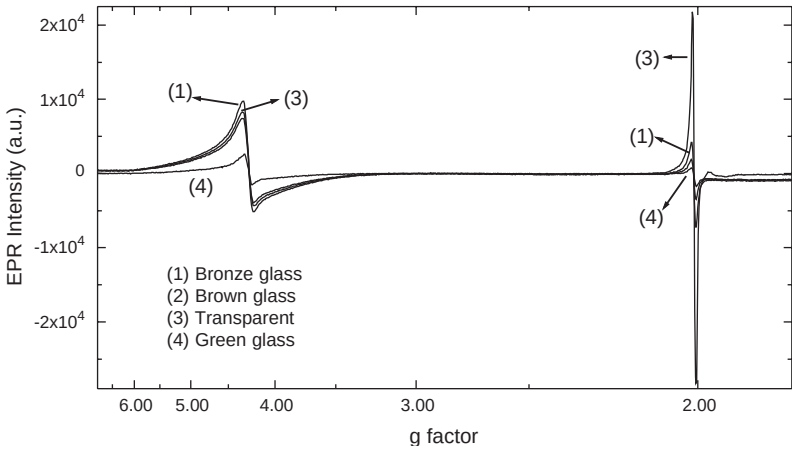

Fig. 2. EPR spectra of the different glass samples irradiated (^{60}Co) with 5 kGy.

10 kGy. The bronze, brown and green glass samples presented similar EPR spectra to that of transparent glass around $g = 2.0$.

Reproducibility of the EPR signal was verified utilizing groups of five samples of each type of glass. The groups were submitted five times to the same procedure of thermal treatment at 300 °C for 30 min (defined for reutilization) followed by irradiation with 5 kGy. The maximum standard deviations of 2.5%, 3.3%, 2.7%, and 3.8% were obtained for transparent, bronze, brown, and green glass samples respectively.

The EPR spectra were observed at room temperature up to 20 days after an irradiation of 5 kGy of each group of glass samples. After the first 24 h post-irradiation time, the response presents a reduction of about 14%, 21%, 17%, and 19% in the cases of transparent, bronze, brown, and green glass sample, respectively. Afterwards, the response decrease slowed down tending to a constant value (after about 14 days).

The glass samples were irradiated to various doses in the range between 100 Gy and 75 kGy. Fig. 6 presents the calibration curves for all four types of samples. Maximum uncertainties of 1.9%, 2.9%, 1.8%, and 2.8% were obtained for the transparent, bronze, brown, and green glass samples, respectively. The transparent glass sample presented the most intense signal of the glass group. Linearity can be observed in all curves of Fig. 6

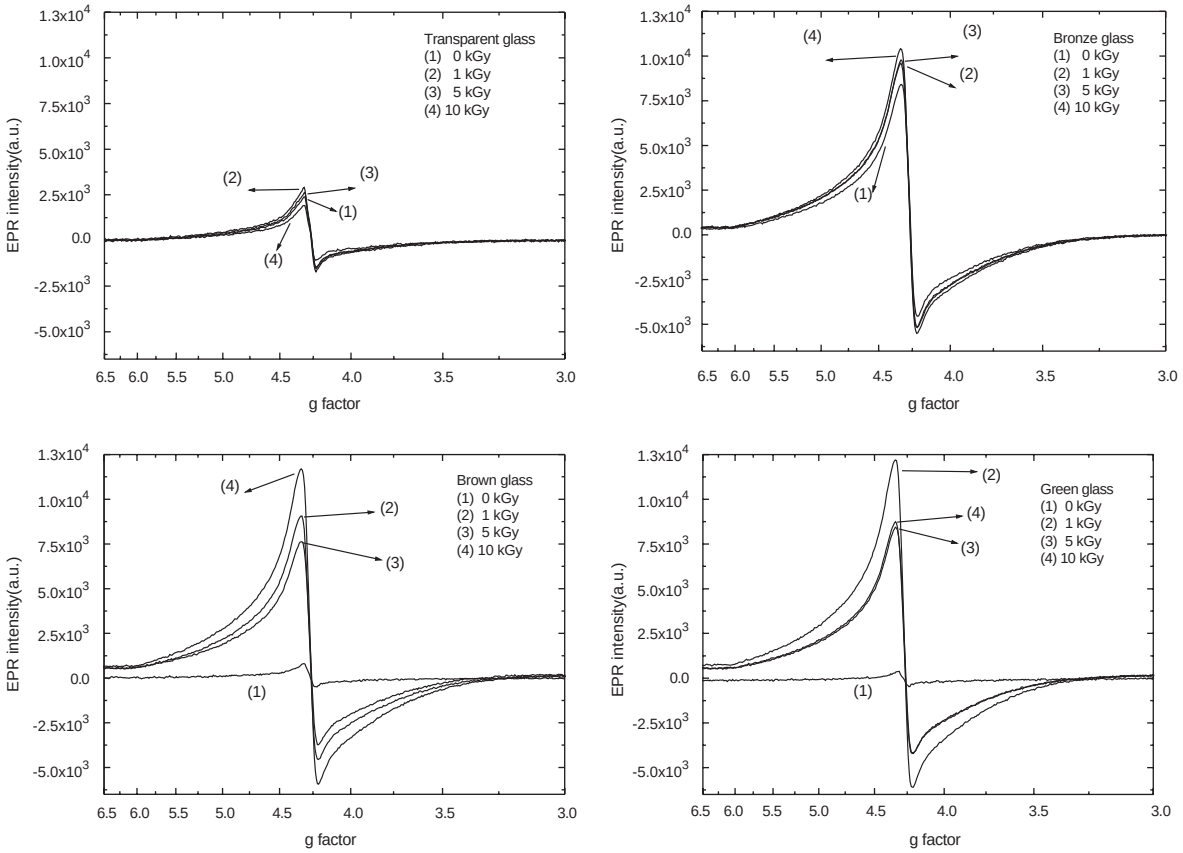

Fig. 3. EPR spectra of the different glass samples around $g = 4.3$. The glasses were irradiated to different absorbed doses (^{60}Co).

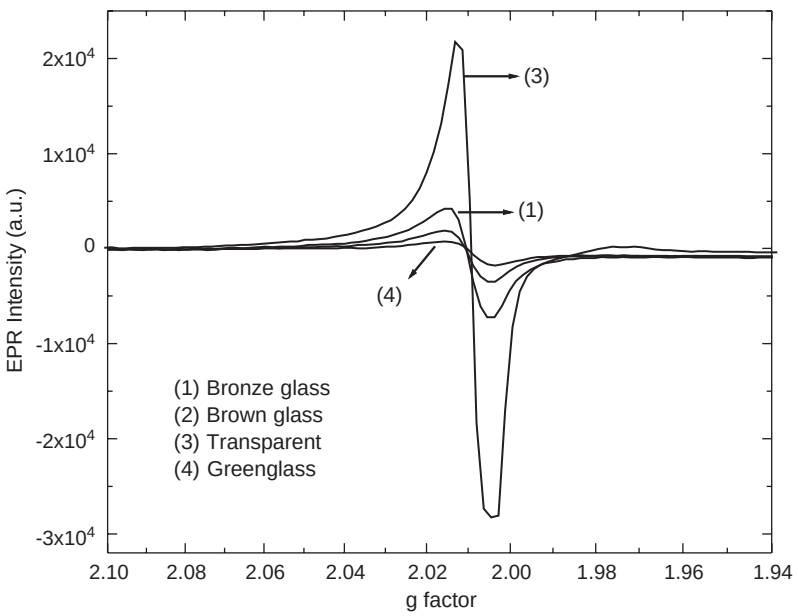

Fig. 4. EPR spectra of the different glass samples around $g = 2.0$. The glasses were irradiated with 5 kGy (^{60}Co).

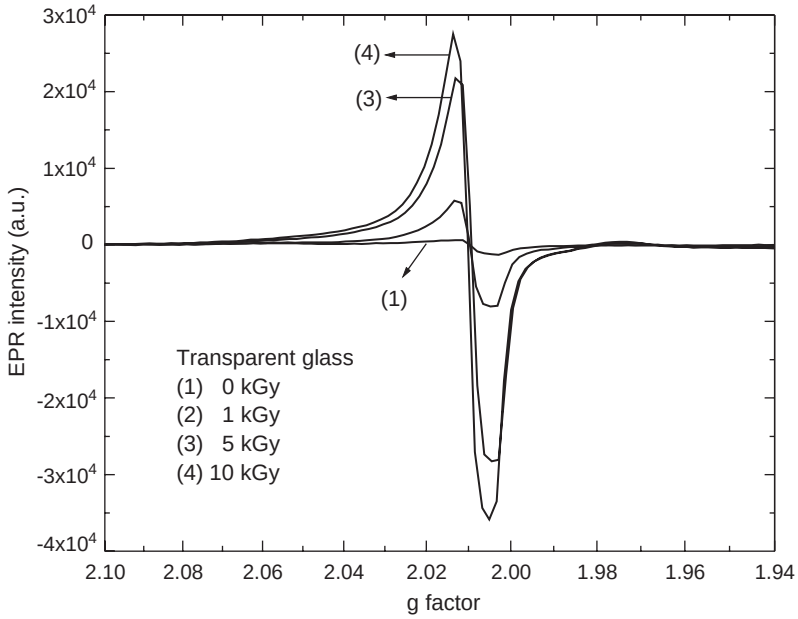

Fig. 5. EPR spectra of transparent glass samples around $g = 2.0$. The glasses were irradiated to different absorbed doses (^{60}Co).

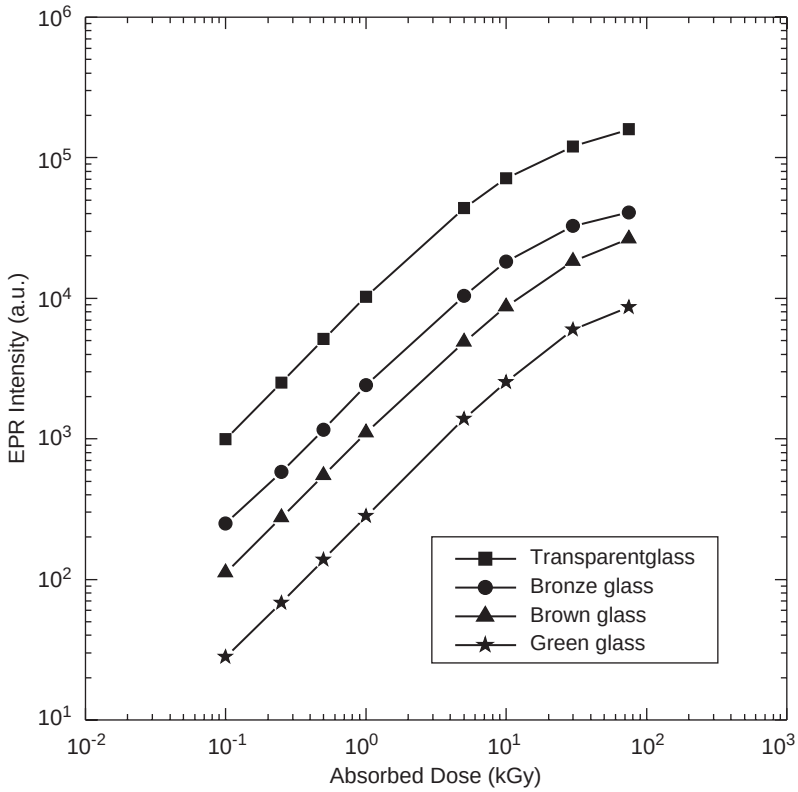

Fig. 6. Calibration curves of EPR signal at $g = 2.01$ for different glass samples for ^{60}Co radiation.

up to 5 kGy; a tendency to saturation effects is clearly seen after 10 kGy.

Due to the thermal fading, which occurs even during irradiations, and is different for sources with different dose rates, the results in this section are directly applicable only to the specific irradiation facility. Each user has to construct his/her own calibration curve, as for any dosimetric system.

4. Conclusions

The basic advantages of commercial glasses are their chemical inertness, insolubility, rigidity, small size, and very low cost. This kind of material is interesting for radiation dosimetry. The results obtained in this work showed that the Fe ions present in the glassy matrix are largely in the form of Fe^{2+} ions. The irradiation induces the increase of Fe^{3+} ions. The EPR spectra observed at $g = 4.27$ and $g = 2.01$ probably are due to tetrahedral Fe^{3+} ions in the substitutional silicon site under a rhombic distortion due to the presence of a compensating cation and to pairs of exchange coupled Fe^{3+} ions, respectively.

The transparent, bronze, brown, and green glass samples can be used as high dose dosimeters using the EPR technique. The dosimetric properties of these glasses were adequate for doses up to 5 kGy, provided that their thermal fading is taken into account. The transparent glass was the most sensitive to irradiation compared to the other glass samples.

The glass samples investigated in this work may be used for dosimetry in the main radiation processes of disinfection, water purification, pasteurization and food sterilization.

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