

Dosimetric properties of gamma- and electron-irradiated commercial window glasses

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Abstract. Two different types of window glasses have been investigated as possible routine dosimeters in the range of 1–50 kGy. Glass samples were irradiated with 3.4 MeV and 8.4 MeV electron beams and the results compared with those obtained by ^{60}Co γ irradiation. Due to the strong optical absorption at wavelengths < 310 nm, even at low doses, optical measurements were limited to the range 320–700 nm. In both types of glasses, irradiation induced two absorption bands around 410 and 600 nm the intensities of which are proportional to dose; the 600 nm band being less sensitive to radiation. These bands have been attributed to non bridging oxygen hole centres existing in two different configurations. Because of the rapid fading of the optical absorbance observed at room temperature immediately after irradiation ($> 15\%$ in two days), the samples underwent a post-irradiation thermal treatment (150°C for 20 min) to improve the stability of absorbance. The fading characteristics of the irradiated and thermally treated glasses, kept in the dark at room temperature, were studied for several weeks. A fast decay, whose intensity depends on the type and energy of the ionizing radiation, was observed for few days after irradiation and thermal treatment, followed by a much slower decay. The results show that this kind of material could be used as routine dosimeters within a certain dose range, as long as proper calibrations are made for each batch of glass and the appropriate precautions are taken when doing calibration and performing routine dosimetry.

Key words: silicate glass • gamma rays • electron beam • optical absorption • dosimeter • fading behaviour

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Introduction

The interactions of photons (γ and X rays) and high energy electrons produce with glass mainly ionization and excitation, but also cause a certain amount of atomic displacement depending on the energy of the radiation. The displacement of lattice atoms and/or electron defects and production of electron-hole pairs, which may be individually trapped at various defect sites in the glass structure, imply changes in the valence state of lattice atoms or inclusion of impurity atoms in glass. Some of the modified electronic configurations or defects cause preferential light absorption; the glass thus becomes coloured, and hence these defects are called “colour centres”. These centres are of many types and depend on the glass components and have associated characteristic optical absorption bands and ESR spectra. The radiation-induced colour centres in some commercial glasses have been found suitable for radiation dosimetry [2, 9, 13]. The use of glass samples as radiation dosimeters presents some advantages that make them attractive for the scope: they are recyclable (a thermal treatment at 300°C for time > 20 min is enough for

re-use of the sample [13]), chemically inert, fast to measure, with little or no dependence on humidity [15]. Beside, window glass is a common material that is easily available, has the advantage to be very inexpensive and does not require special preparation. The main disadvantage presented by all types of glass dosimeters is the significant spontaneous post-irradiation fading [3, 13, 14] even at low temperature (-20°C) [4]. To overcome this problem, it is advisable to submit the glass samples to post-irradiation thermal treatment at 150°C for 20 min [4] before measuring the optical density at different time intervals. In this way, calibration curves can be obtained for different time intervals after irradiation and subsequent thermal treatment. If the behaviour of these glasses under γ and electron irradiation is fully studied and well characterized they may become a suitable material for radiation processing dosimetry.

The aim of this work is:

- 1) to verify the possibility of the application of commercial window soda silicate glass for high dose dosimetry by irradiation with 3.4 MeV and 8.4 MeV electron beams and compare the results with those obtained with ^{60}Co γ rays [4, 11],
- 2) to compare the response to irradiation with 8.4 MeV electrons of two commercial soda silicate glasses having different chemical composition, and

- 3) to follow the fading behaviour of the irradiated samples after post-irradiation thermal treatment.

Materials and methods

Two transparent commercial soda silicate glass sheets (≈ 3 mm thick), type A and B, purchased from a Tunisian market were cut into pieces of 11×30 mm for irradiation and optical measurements. Their chemical compositions were determined by the prompt gamma activation analysis (PGAA) technique at the Budapest Neutron Centre and the results are reported in Table 1. To avoid grease contamination on glass surface, which may affect the absorbance measurements, the samples were carefully cleaned with ethyl alcohol. A thermal treatment at 300°C for 1 h was used to eliminate any spurious optical signal. The optical absorbance spectra of non-irradiated samples were measured against air and used as reference for the optical spectra of the corresponding irradiated samples in order to obtain the radiation-induced changes in absorption.

Irradiations of the glass samples were done with electron accelerators and a ^{60}Co γ source having the characteristics reported in Table 2. Routine dosimetry was done using Fricke and ethanol-chlorobenzene chemical dosimeters. Traceability was established with alanine/EPR dosimetry system in terms of absorbed dose by water traceable in the UK National Physical Laboratory for gamma irradiation [4] and the combined uncertainty in absorbed dose administered by the glass samples was 2.7% at $k = 1$ [11].

For electron irradiations care was taken to place both the reference dosimeters and the glass samples at the same depth in a plastic phantom and to insert a thermocouple in the phantom to measure the temperature reached by the samples during irradiation. The maximum temperature registered was 52°C for the 50 kGy dose. Traceability was established with alanine/EPR dosimetry system in terms of absorbed dose by water traceable in the Risø High Dose Reference Laboratory. The absorbed dose to the glass samples had a combined uncertainty of 2.6% at $k = 2$. All the samples, after irradiation, were stored in the dark in a room where the temperature was maintained between 20 and 25°C and humidity 45–60% RH.

Spectrophotometers and a thickness gauge were used to measure the specific absorbance changes (absorbance/dosimeter thickness = mm^{-1}) produced in the glass. Since at $\lambda < 310$ nm the absorbance of the

Table 1. Chemical composition of the two types of glass: determination by PGAA technique

Elements	Type A (%)	Type B (%)
H	0.037	0.04
F	3.161	1.917
Na	13.770	14.11
Mg	4.340	4.298
Al	1.003	1.185
Si	68.520	69.08
S	0.201	0.249
Cl	0.013	0.013
K	0.588	0.487
Ca	8.190	8.433
Ti	0.051	0.017
Mn	0.027	0.028
Fe	0.105	0.124
Sm	4×10^{-5}	2×10^{-5}
Gd	6×10^{-5}	2×10^{-5}
Co	–	8×10^{-4}
Ni	–	0.021

Table 2. Characteristics of the facilities used for irradiations

	Nominal beam energy (MeV)	Energy* (MeV)	Beam characteristics and dose rate
Institute of Isotopes, Budapest	4	3.4	LINAC 2.6 μs p.l. – 50 p.p.s. 2.17 Gy/pulse
ISOF-CNR, Bologna	10	8.4	LINAC 2 μs p.l. – 50 p.p.s. 6.95 Gy/pulse
Tunisian semi-industrial ^{60}Co γ source		1.25	1 kGy/h, 6 kGy/h

* In the case of the electron accelerators this refers to the most probable beam energy.

irradiated glasses was too high thus loosing linearity of the signal we limited the analysis of the spectra to the range 320–700 nm.

Results and discussion

Effect of gamma and electron irradiation

Type A glass

Samples of type A glass were irradiated to different doses in the range of 1–100 kGy. The damage introduced in the glass structure together with the trapping of electrons and holes in defect states give rise to colouration of the glass samples. The absorption spectra of the irradiated glasses show two large optical absorption bands at around 410 and 600 nm; the band at 600 nm is less sensitive to radiation than that at 410 nm (Fig. 1). The position of the absorbance bands were the same for both types of radiation (Fig. 1), only the intensity and the area of the bands were different due to the different number of colour centres depending on the type of radiation and dose. The bands at 410 and 600 nm in irradiated soda

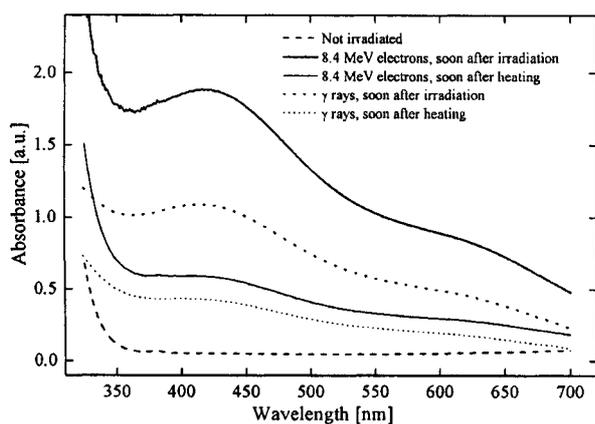


Fig. 1. Absorption spectra of type A electron- and gamma-irradiated glasses at 25 kGy (dose to water) taken soon after irradiation and 30 min after post-irradiation thermal treatment (150°C for 20 min).

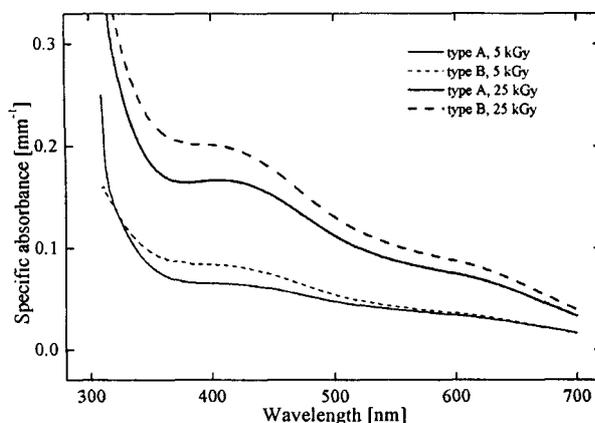


Fig. 2. Absorption spectra of type A and type B glasses irradiated with 8.4 electron beams at 5 and 25 kGy (dose to water) taken 30 min after post-irradiation thermal treatment (150°C for 20 min).

silicate glasses have been attributed to “non-bridging oxygen hole centre” (NBOHC: $\equiv \text{Si}-\text{O}^{\bullet}$) [1, 6, 10], a hole trapped on the unbounded $2p$ orbital of a non-bridging oxygen atom, which most probably overlaps with the band of peroxy radical (POR: $\equiv \text{Si}-\text{O}-\text{O}^{\bullet}$), a hole trapped on the unbounded $2p$ orbital of the singly bonded oxygen atom in a dangling peroxy radical, at 600–630 nm [6]. The notation ‘ \equiv ’ represents three bonds with other oxygens in the glass network and ‘ \bullet ’ denotes an unpaired electron.

Type B glass

Samples of type B glass were irradiated with 8.4 MeV electron beams in the same range of dose (1–100 kGy) and the results obtained were similar to those of type A glasses. The absorption spectrum shows the same absorption bands (Fig. 2); however, for the same dose absorption is about 20% larger than that for type A. This difference might be due to the different concentrations of various components and the presence of other impurities such as Co and Ni.

Fading characteristics

The radiation-induced colour centres are unstable at room temperature and showed a rapid fading of the optical absorbance (Fig. 3). To improve their stability the irradiated samples were submitted to post-irradiation thermal treatment at 150°C for 20 min, which was proven to be the optimal treatment as far as time and temperature are concerned [4]. Optical measurements were taken at 410 and 600 nm about 30 min after irradiation and heat treatment, and then repeated over several weeks. After each measurement, the glass samples were stored in the dark at room temperature. The results, with the first measurements taken 30 min after irradiation and heat treatment, are shown in Figs. 3 and 4. As it can be seen, the glass initially has a fast fading rate; the decrease of the optical absorbance is from 5% to 10% after 24 h and is dose and energy dependent, reaching a 20% up to a 30% decrease, respectively for the 3.4 and 8.4 MeV electron irradiation after 20 days. After that, the

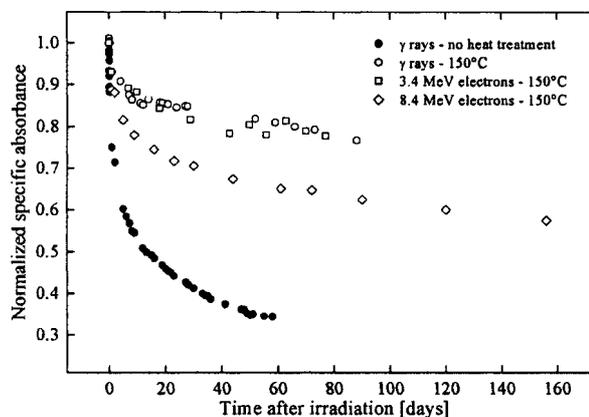


Fig. 3. Post-irradiation fading at room temperature of absorbance at 410 nm of type A glass samples after 25 kGy irradiation (dose to water), with and without thermal treatment. The gamma irradiation data are from Ref. [11].

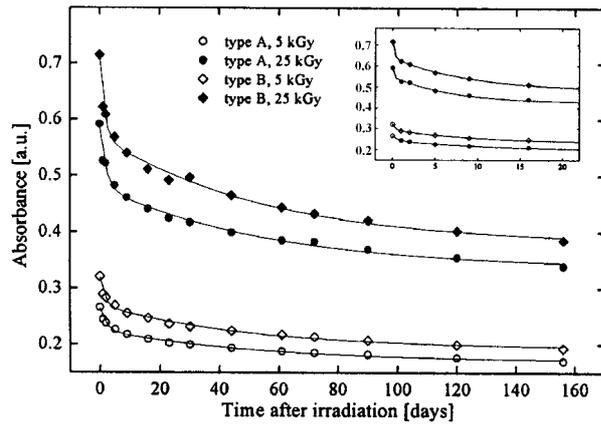


Fig. 4. Post-irradiation fading at room temperature of type A and type B glasses irradiated with 8.4 MeV electrons at 5 and 25 kGy (dose to water) and heat treated at 150°C for 20 min (absorbance measured at $\lambda = 410$ nm) (fitting with double exponential function). Inset: behaviour of glass samples within the first 20 days after post-irradiation heat treatment.

fading rate slows down significantly and the intensity tends to approach a constant value with time.

The decay of absorption can be fitted by the sum of two first order decay kinetic expressions of the type:

$$(1) \quad A(t) = A_0 + A_1 e^{-t/\tau_1} + A_2 e^{-t/\tau_2}$$

where $A(t)$ is the absorbance at time t and τ corresponds to the appropriate time constants.

It is the depth of the traps in the material that determines the decay characteristics [6]. The initial intense fading of the optical absorption clearly indicates that during post-irradiation storage in the dark and at room temperature some energy is available which is sufficient to remove shallow traps or trapping centres which are thermally unstable. The reduced fading of the optical absorption after thermal treatment at 150°C can be interpreted as being partially due to annealing of the radiation-induced shallow traps.

Dose response

Four glass samples of type A were irradiated each at a dose with 3.4 and 8.4 MeV electrons, with doses in the range of 1–100 kGy. At doses ≥ 40 –50 kGy, the specific absorbance at 410 and 600 nm tends to saturation, thus in Fig. 5 only the specific absorbances taken 24 h after irradiation and heat treatment for the dose range 1–50 kGy are shown as dose response curves. In the same figure the data for gamma irradiated samples [11] are shown for comparison with the data obtained with the electron irradiated samples. Similar behaviour was observed for the specific absorbance at 600 nm.

Similar results were obtained with the type B glass (Fig. 5), but with the absorption values much larger than those obtained with type A glass.

The rapid rise in the signal over the initial 10 kGy dose is followed by a more gentle increase over the following 40 kGy, reaching after that a quasi-plateau. According to [5], the nonlinear dose dependence can

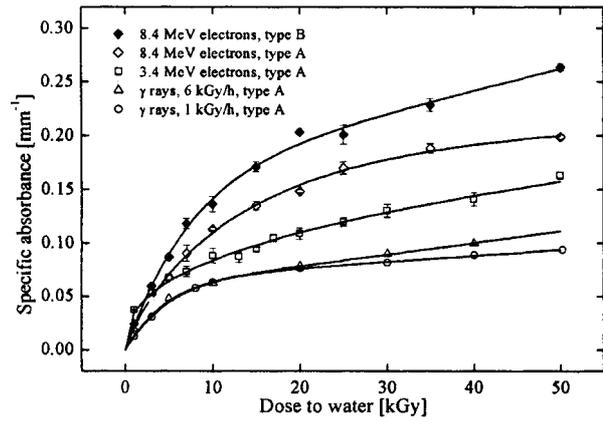


Fig. 5. Response curves for irradiated and heat treated glasses (absorbance measured at $\lambda = 410$ nm). The gamma irradiation data are from Ref. [11].

be interpreted in terms of two different processes: first, the activation of pre-existing defects (precursors) forming colour centres which saturate with dose because of their limited concentration in glass, and then bond breaking or displacements of atoms that creates new defects in the glass structure where electrons or holes could be transferred. The generation mechanism may be complicated by the competition between stabilization and destruction of defects by the reaction with diffusing atoms or molecules arising from bond rupture during irradiation [12].

Response of the glasses to irradiation is rather radiation-energy dependent than dose-rate dependent, at least at the dose rates used in this study. This can be explained by considering that the minimum energy that an electron must have to displace a silicon atom in a silicate glass is approximately 250 keV [8]. The recoil Compton electrons produced by ^{60}Co γ rays by interaction with the material have at most 1 MeV energy (recoiling electrons rapidly lose energy and are quickly degraded below the minimum energy required to produce a displacement. Furthermore, even if the recoiling electron has sufficient energy it may take a “glancing” collision which will not impart the necessary energy to the lattice atom [8]) whereas the electron irradiations were done with 3.4 and 8.4 MeV electrons. From that, it is clear that for the same dose the higher is the energy of the radiation the greater is the number of colour centres formed.

Variability and uncertainty

The coefficient of variation of a set of dosimeters is of great importance for a specific dosimetry system and application because it determines the useful dose range for the dosimetry system and the metrological quality of the dosimeter. Thus, the pooled average of the CV over the dose range of the electron irradiated and heat treated glasses was determined to be within 4 and 5%.

The overall estimated uncertainties for the electron irradiated glasses, calculated according to the procedure outlined in ISO/ASTM 51707 [7], associated with all these measurements are 6% at 1σ , while for the gamma irradiated glass samples a value of 4.7% (1σ) was found [11].

Conclusions

The results reported in the preceding sections have shown that the optical absorbance response of both types of glasses to γ and electron irradiation can be quantified reasonably well in the 1–50 kGy range. The observed properties suggest the possibility of using commercial window glasses, a common and very inexpensive material, as high-dose dosimeters in radiation processing where the required performances of a dosimeter are not so stringent as they are for processes that concern public health and safety. In any case calibrations are required for each batch of dosimeters and appropriate precautions must be taken when doing calibration and performing routine dosimetry. Even if the fading effect is undesirable, because it can give rise to significant errors during the routine use of these dosimeters, it is not of great importance if calibration curves and routine dose evaluation are done at the same time interval. In any case to reduce the rapid fading of the optical absorption, it is advisable to subject the irradiated glass dosimeters to the reported thermal treatment after irradiation and take measurements at the same time interval after irradiation and heat treatment or by applying a correction factor that accounts for the fading for any other post-irradiation time period.

To minimize the contribution of influence quantities (environmental conditions) to the overall uncertainty and to ensure similar irradiation conditions both for calibration and routine dosimetry during the production run, in plant calibration of the glass dosimeters should be performed by irradiating them together with reference or transfer standard dosimeters.

Glass dosimeters may be reused by thermally bleaching the radiation induced colour centres at 300°C for at least 30 min. Besides, as the glass changes colour when irradiated, they might be used as Yes/No irradiation indicators.

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References

1. Bartoll J, Nofz M, Stösser R (2000) Complementarity of ESR and optical absorption of gamma-irradiated silicate glasses. *Phys Chem Glasses* 41:140–149
2. Caldas LVE, de Souza CN (1991) High dose dosimetry using glass detectors in electron beams. In: *Proc of the Int Conf High Dose Dosimetry for Radiation Processing*. IAEA, Vienna, pp 93–99
3. Engin B, Aydaş C, Demirtaş H (2006) ESR dosimetric properties of window glass. *Nucl Instrum Meth Phys Res B* 243:149–155
4. Farah K, Kovács A, Mejri A, Ben Ouada H (2007) Effect of post-irradiation thermal treatments on the stability of gamma-irradiated glass dosimeter. *Radiat Phys Chem* 76:1523–1526
5. Galeener FL, Kerwin DB, Miller AJ, Mikkelsen Jr JC (1993) X-Ray creation and activation of electron-spin resonance in vitreous silica. *Phys Rev B* 47:7760–7779
6. Griscom DL, Mizuguchi M (1998) Determination of the visible range optical absorption spectrum of peroxy radicals in gamma-irradiated fused silica. *J Non-Cryst Solids* 239:66–77
7. ISO/ASTM 51707 (2007) Standard guide for estimating uncertainties in dosimetry for radiation processing. *Annual Book of ASTM Standards* 12.02, Philadelphia, PA, pp 1140–1163
8. Levy PW (1960) The kinetics of gamma-ray induced coloring of glass. *J Am Ceram Soc* 43:389–395
9. McLaughlin WL (1974) Solid-phase chemical dosimeters. In: Gaughran ERL, Goudie AJ (eds) *Sterilization by ionizing radiation*. Multiscience, Montreal, vol. 1, pp 219–252
10. Marshall CD, Speth JA, Payne SA (1997) Induced optical absorption in gamma, neutron, and ultraviolet irradiated fused quartz and silica. *J Non-Cryst Solids* 212:59–73
11. Mejri A, Farah K, Eleuch H, Ben Ouada H (2008) Application of commercial glass in gamma radiation processing. *Radiat Meas*, doi:10.1016/j.radmeas.2008.05.010
12. Pacchioni G, Skuja L, Griscom DL (eds) (2000) *Defects in SiO₂ and related dielectrics: science and technology*. Kluwer Academic Publishers, Dordrecht, The Netherlands
13. Quezada VAC, Caldas LVE (1999) Glass detectors for dose determination in a flower irradiation process. *Radiat Prot Dosim* 85:473–475
14. Sheng J, Kadono K, Yazawa T (2002) Fading behavior of X-ray induced color centers in soda-lime silicate glass. *Appl Radiat Isot* 57:813–817
15. Zheng Z, Jiansheng C, Jiating Z *et al.* (1997) Study on the possibility of reading two kinds of data from one glass detector. *Radiat Phys Chem* 50:303–305