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Formation and Decay of Colour Centres in a Silicate Glasses Exposed to Gamma Radiation: Application to High-Dose Dosimetry

K. Farah[1,2], A. Mejri[1], F. Hosni[1],
A. H. Hamzaoui[3] and B. Boizot[4]

[1]*National Center for Science and Nuclear Technology, Sidi-Thabet,*
[2]*ISTLS, 12, University of Sousse,*
[3]*National Centre for Research in Materials Science, Borj Cedria, Hammam-Lif,*
[4]*Laboratory of Irradiated Solids, UMR 7642 CEA-CNRS -*
Polytechnic School, Route de Saclay, Palaiseau,
[1,2,3]*Tunisia*
[4]*France*

1. Introduction

The interactions of ionizing radiation with glass matrix produce ionization, excitation, and atomic displacement. The main modification induced during γ, X rays, or electron irradiation on the glass structure is the creation of stable defects and the changes of the valence state of lattice atoms or of the incorporated impurities in glass. Some of the modified electronic configurations or defects cause preferential light absorption. Thus, glass becomes coloured and consequently these defects are called "colour centres". These centres are of many types and depending on the glass composition (Yokota, 1954, 1956) and are associated with optical absorption bands and EPR signals.

The change of optical properties of glasses when subjected to ionizing radiation has been investigated by many authors due to wide applications of this kind of material. The earlier studies were focused on ways to prevent the darkening in glasses used in reactor or hot-cell windows and optical devices (Friebele, 1991). Recently, many studies have been concentrated on the application of the irradiation induced colour to develop recyclable colour glasses which is of great interest in the glass industry from the economical and environmental point of view (Sheng et al., 2002a).

In addition to the applications mentioned above, the ionizing radiation induced colour centres in some glasses have been found wide application in radiation dosimetry (Farah et al., 2010; Fuochi et al., 2008, 2009; Mejri et al., 2008). This material is very interesting for dosimetry and very useful for many applications such as food irradiation, sterilization of medical devices, radiation treatment of industrial and municipal waste-water and radiation processing of materials.

In the present work, we investigated the effect of the irradiation dose and thermal annealing on the formation and the decay of the induced colour centres in gamma irradiated silicate

glass. The activation energies of the two colour centres were calculated from Arrhenius equation. In order to evaluate its potential as radiation-sensitive material in high dose dosimetry, the main dosimetric properties were also studied in details.

2. Materials and methods

2.1 Glass composition

The glass samples were obtained from the same glass sheets purchased from the local market and were cut into pieces of 11 x 30 x 3 mm^3 dimensions for optical measurements. The chemical composition of the glass samples were determined by the Prompt Gamma Activation Analysis technique (Anderson et al., 2004) in the Budapest Neutron Centre (wt%: 68.52 SiO_2, 13.77 Na_2O, 8.19 CaO, 4.34 MgO, 1.003 Al_2O_3, 0.588 K_2O, 0.105 Fe_2O_3 and about 3.5 % of other components). 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. Then the samples were wrapped in aluminium papers and stored in a dust free dark place to avoid any possible light effect.

2.2 Irradiation sources and procedures

Irradiations of the glass samples were done at the Tunisian semi-industrial ^{60}Co gamma irradiation facility at the dose rate of about 2 kGy/h (Farah et al. 2006). Dosimetry was done using Fricke and Ethanol-ChloroBenzene chemical dosimeters and the traceability was established with alanine/EPR dosimetry system in terms of absorbed dose to water traceable to Aérial Secondary Standard Dosimetry Laboratory (SSDL), Strasbourg-France (Aérial, 2011). All the irradiated samples were stored in the dark in a room where the temperature was maintained between 20 and 25°C and humidity 45-60% R.H.

2.3 Optical absorption measurements

When exposed to gamma radiation, glass turns to a brown colour in a quantifiable and reproducible manner. Measurement of the change in absorbance with calibrated spectrophotometers at specified wavelengths provides a method for accurately determining absorbed dose.

Optical absorption spectra were taken with a Perkin Elmer spectrophotometer Lambda 20 in the range 350-800 nm. The optical spectra of non irradiated samples were measured with reference to air. All optical spectra of the samples after irradiation were measured against to non irradiated sample in order to obtain the net induced changes of absorption. Genesis 5 spectrophotometer and Käfer MFT 30 thickness gauge were used to measure the specific absorbance changes produced in glass (i.e. absorbance divided by dosimeter thickness). An electrical furnace and a freezer were used to reach the desired temperatures.

2.4 Temperature control during irradiation

To control and maintain the glass samples at the desired temperature during gamma irradiation, Julabo refrigerating circulator type F25-EC, with ultra purified water for the temperature range 5-90°C or a mixture of water and ethylene glycol for the range –25°C to +50°C as coolant liquid, was used. Samples were placed inside an aluminium cylinder in which the coolant liquid was circulating thus maintaining the desired temperature. Before

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irradiation, the set-up was kept for 20 minutes to reach the equilibrium temperature. Temperature fluctuation inside the aluminium cylinder during irradiation was within ±1°C.

2.5 Method and conditions of calibration

In order to minimize the contribution of influence quantities to the overall uncertainty and to ensure similar irradiation conditions both for calibration and routine dosimetry during the production run, full in-plant calibration of the glass samples was performed by irradiating them at the Tunisian semi-industrial cobalt-60 gamma irradiation facility together with transfer standard dosimeters in the Risø HDRL calibration phantoms (Sharpe&Miller, 1999). Four glass samples were placed inside the phantom, together with four alanine transfer standard dosimeters in their small 3 mm thick polyacetal holders as shown. The phantoms containing the alanine pellets and the glass samples were then taped on cartons of simulated product (dummy product box) placed on the test site facing the cobalt source. Care was taken to minimize self shielding effects between the dosimeters. The distance of the dummy product box from the source pencils was such as to reduce the spatial variations in the radiation field over the surface area of the dosimeter package to negligible small values.

The alanine pellets were sent back after irradiation for evaluation to the Aérial SSDL. The absorbed doses, as measured by the alanine dosimeters, were then used to establish the calibration curves for the glass.

Temperature strips were placed in the dosimeter packages during the calibration irradiations to record maximum temperature. Temperatures were found to vary around a mean value of 26°C with a maximum variation of +3°C.

3. Results and discussion

3.1 Effect of gamma radiation dose

Before gamma irradiation, the glass used in this study was transparent. When irradiated, two induced bands have been observed at 410 and 600 nm leading to the development of the brown colour. The intensities of the overall absorption spectra are observed to increase progressively with increasing doses between 1 and 1200 kGy (Fig.1). It is obvious that the broad band at 600 nm is less sensitive to radiation than that at 410 nm.

The induced optical absorption by gamma irradiation in the visible range of this silicate glass is due to the generation of two Non Bridging Oxygen Hole Centres (NBOHCs) (≡Si-O°): HC1 at 410 nm and HC2 at 600 nm (Griscom, 1984).

Fig.2 shows the two bands separated in the region between 375 and 800 nm of the absorption spectrum of glass irradiated to 10 kGy. The induced absorption spectra were well modeled through a Gaussian shape with correlation coefficients (R^2) better than 0.99.

Table 1 shows the results of the best fit with two Gaussian bands (correlation coefficients (R^2) better than 0.99) of the induced absorption spectra in the region between 380-800 nm. It was found that the gamma radiation dose had no influence on the absorption band position. The band peak positions the FWHM were relatively constant and only the heights and the area under the bands changed, suggesting that only an increase of the number of induced colour centres were affected by the increasing of the irradiation dose.

These results show that this glass is radiation-sensitive material and the induced colour centres may be used for dose determination in large dose range.
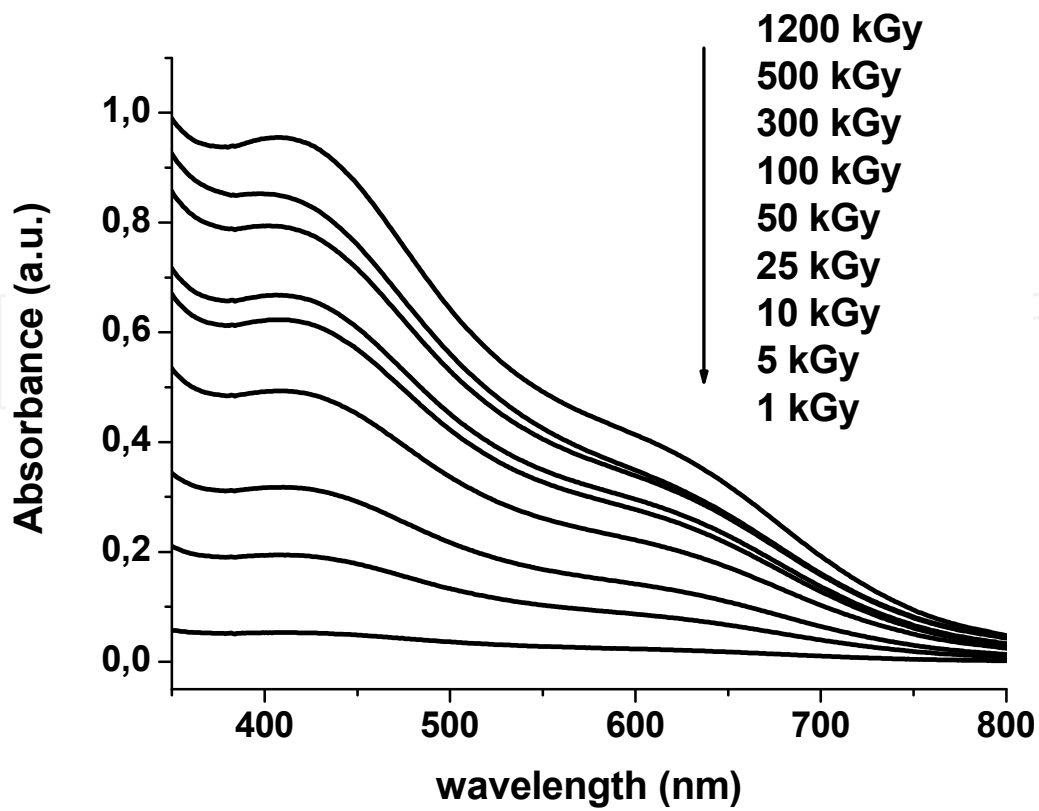
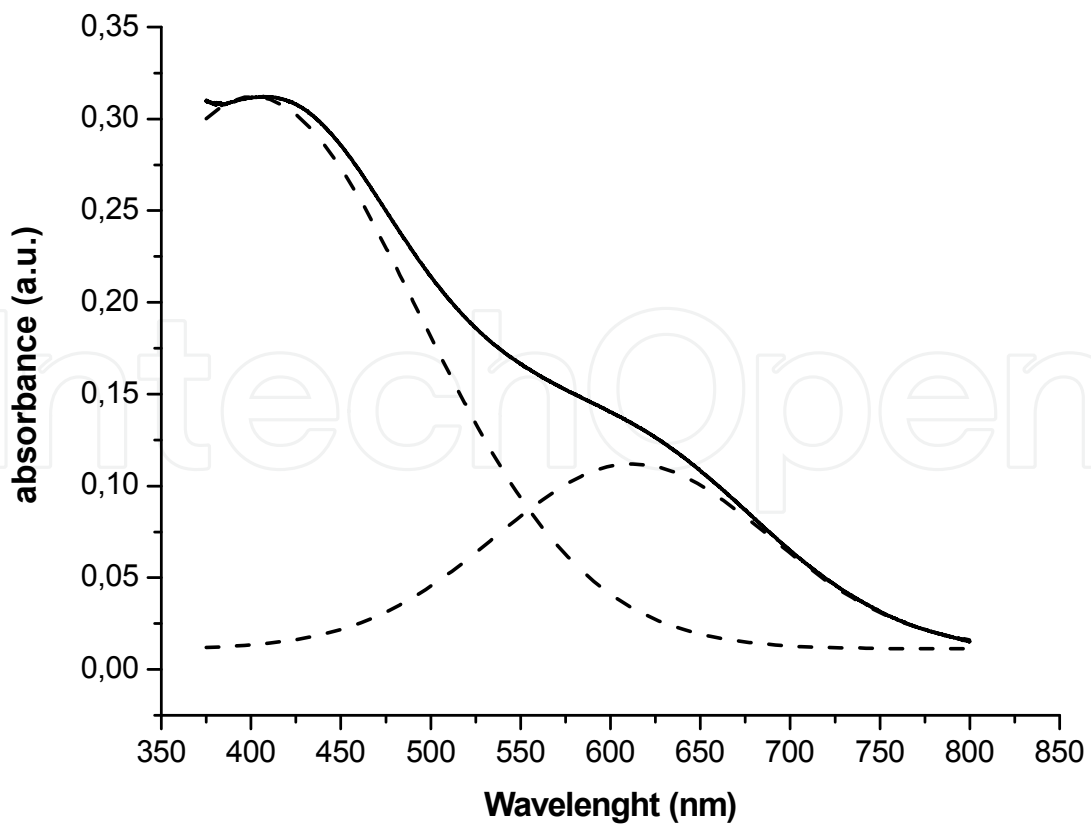Fig. 1. Optical absorption spectra for the gamma irradiated glass samples in the range 1-1200 kGy

Fig. 2. Induced optical absorption with band separation of glass irradiated at 10 kGy.

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Dose (kGy)	λ (nm)	FWHM (nm)	A
1	402	163.93	10.57
	598	174.93	4.50
10	405	162.42	62.31
	599	182.78	29.04
100	403	156.12	140.48
	602	174.82	43.93
1200	403	165.50	200.88
	603	182.07	60.31

Table 1. Results of band separation for the induced absorbance spectra

Where λ is the peak wavelength of band (nm), FWHM is the full width at half-maximum of the band (nm) and A is the area of the band
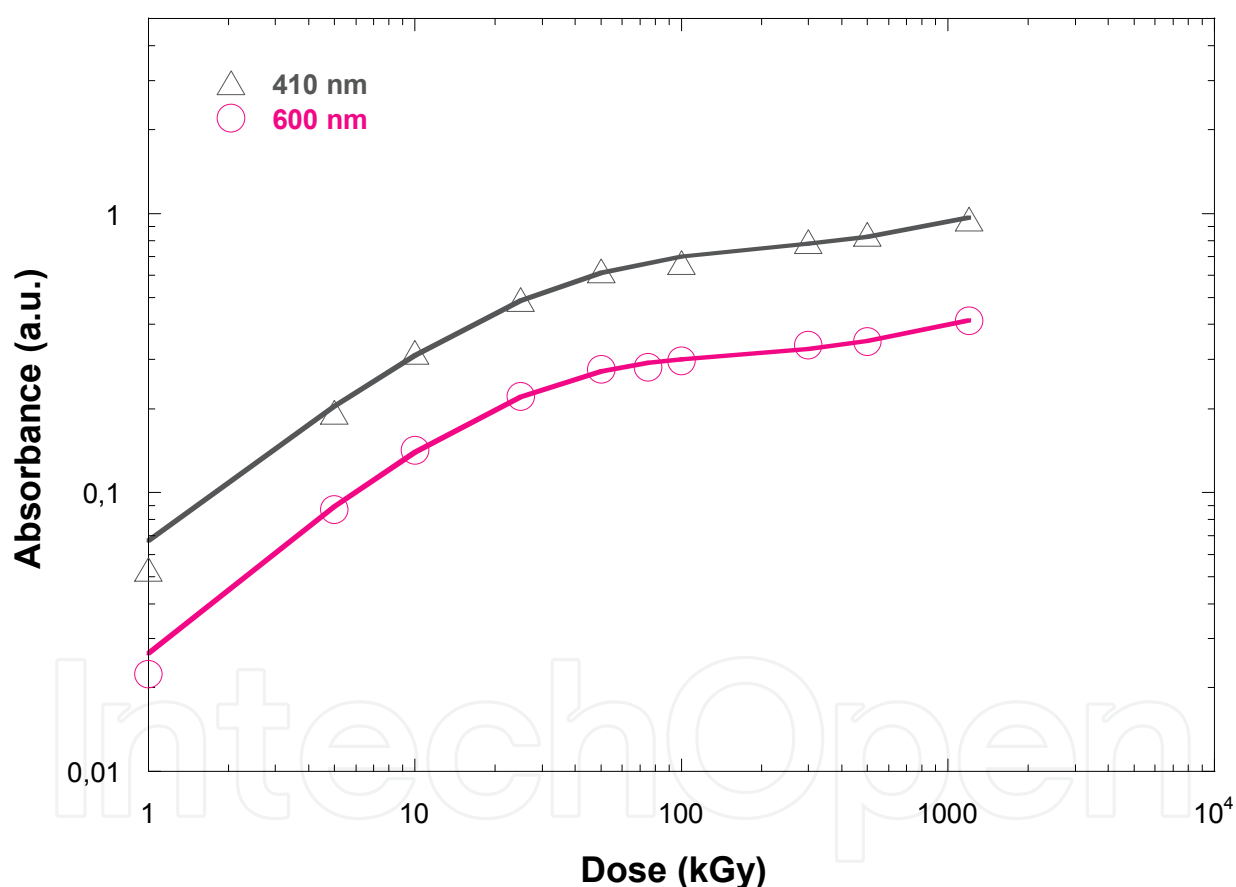
Fig. 3. Dose response curves of gamma irradiated silicate glass

The dose response curves shown in Fig.3 are successfully fitted following Mashkov equation (Eq.1) (Mashkov et al.,). The fitting parameters are given in Table 2.

$$Q(D) = Q_c (kD)^b + Q_a [1- \exp [-(kD)^b]] \tag{1}$$

Where $Q(D)$, $Q_a(D)$ et $Q_c(D)$ represent the measured quantities (Optical Absorption) proportional respectively to the total concentration of the colour centres for an accumulated

dose D, the concentration of the colour centres created by an extrinsic process of activation and the concentration of the colour centres created during an intrinsic process of the rupture, k is a rate constant and b is a number between 0 and 1. The nonlinear dose dependence of the specific absorbance as function of the dose can be interpreted in terms of two different processes involved in the creation of colour centres: the creation of colour centres induced by activation of precursory defects which saturate with the dose because their concentration in glass is limited, and an "unlimited" creation of new colour centres during an intrinsic process of Si-O-Si bond rupture (Boizot, 1997).

Fitting parameters	Wavelengths	
	410 nm	600 nm
R^2	0.995	0.996
k (kGy^{-1})	0.049 ± 0.007	0.052 ± 0.005
b	0.72 ± 0.06	0.86 ± 0.07

Table 2. Fitting parameters for Figure 3

The results of fitting our experimental data by Mashkov Equation showed that the values obtained for the k parameter (defects formation rate or rate constant) are equal to 0.049 and 0.052 kGy^{-1} for the two bands at 410 and 600 nm respectively. The similarity of this parameter for the two bands suggests that the related optical transitions correspond to the same type of NBOHCs in different configurations.

3.2 Activation energy

Figures 4 and 5 show the kinetics, for the absorption band at 410 nm, of temperature annealing performed between (-20 °C) and 150 °C of the glass samples irradiated at 30 kGy. The annealing process can be described by a sum of two first order decay kinetic functions (exp (-t/τ)) where t is the annealing time and τ is an appropriate time constant.

The activation energy characteristic of the annealing process was calculated from the Arrhenius equation. The obtained values for the fast and slow components of the 410 nm and 600 nm bands are presented in Table 3. The similarity of the activation energy values for both bands may suggest that the related optical transitions correspond to the same type of NBOHC's in different configurations (Griscom, 1984). The HC1 centre is a hole trapped in the 2p orbital of one non-bridging oxygen (NBO), analogue to NBOHC in silica glass, to which is correlated the absorption band at 410 nm. The HC2 centre is a hole trapped on two or three NBO's bonded to the same silicon to which is correlated the absorption band at 600 nm (Suszynska& Macalik, 2001).

By the mean of real-space multigrid electronic structure calculations, Jin and Chang proposed a diffusion mechanism of interstitial oxygen ions generated from O_2 under the UV irradiation with activation energies of 0.27 eV for O^- and 0.11 eV for O^{2-} (Jin & Chang, 2001). The value of 0.27 eV corresponding to the O^- diffusion activation energy in the glass network under UV irradiation is identical, within experimental error, to our values of activation energies calculated from Arrhenius plots of the slow component τ corresponding to the long-time isothermal annealing. Approximately the same value of activation energy was found by Tsai and al. for the long-time slow thermal annealing of radiolytic atomic hydrogen in OH containing amorphous silica (Tsai et al., 1989).
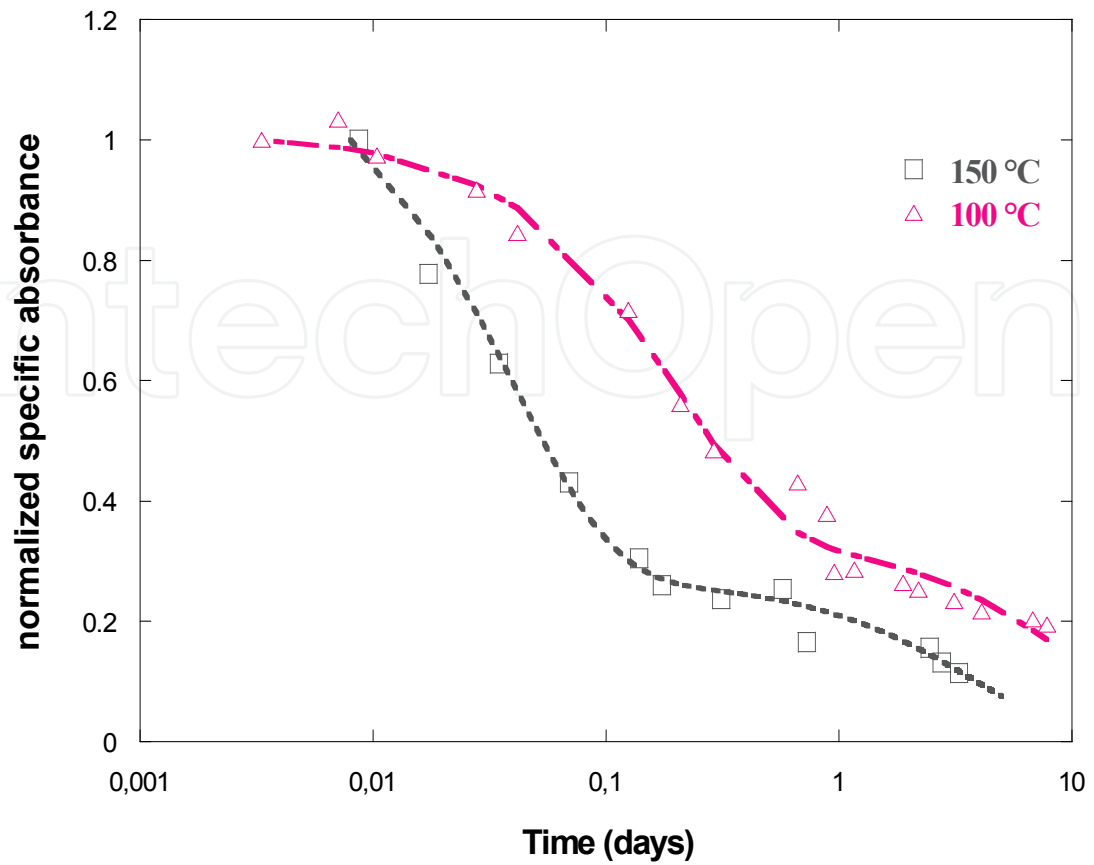
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Fig. 4. Isothermal annealing of glass samples at high temperatures.
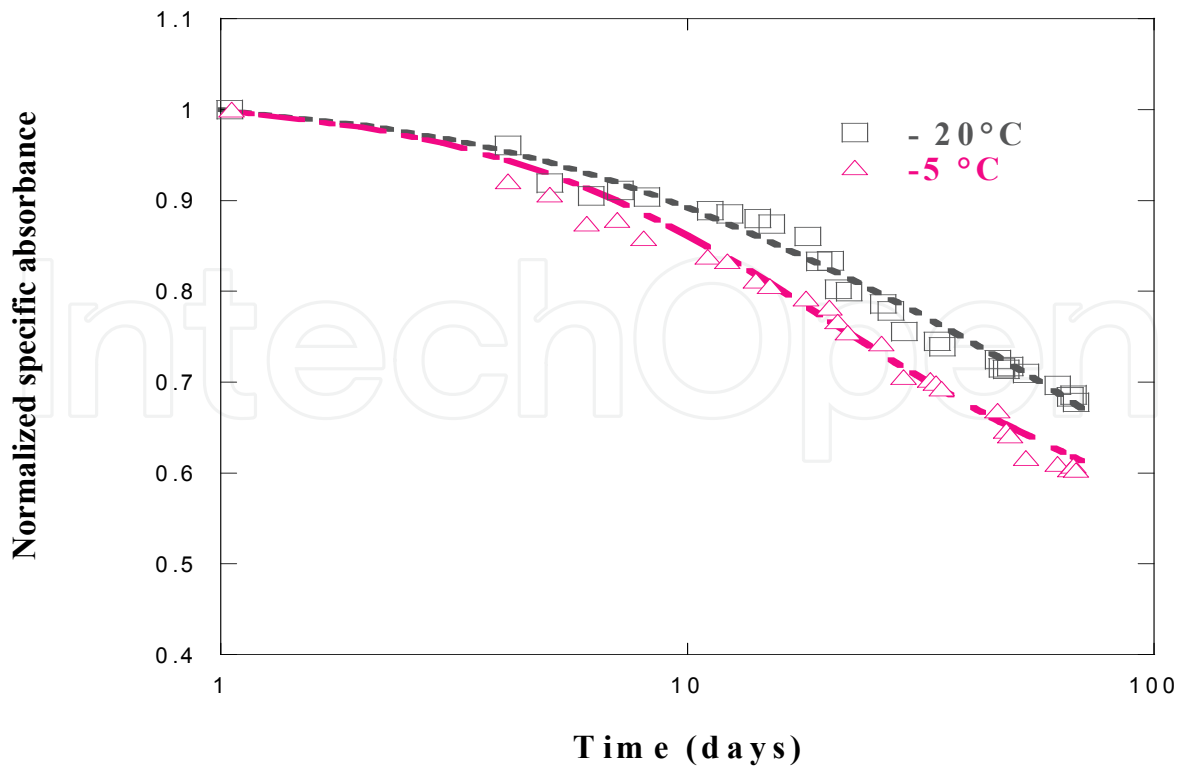
Fig. 5. Isothermal annealing of glass samples at low temperatures.

	Activation energy (eV)	
Wavelength(nm)	Fast component (τ_1)	Slow component (τ_2)
410	0.251± 0.022	0.310 ± 0.015
600	0.261± 0.022	0.353 ± 0.030

Table 3. Activation energy for the fast and slow components of both bands.

3.3 Application to high dose dosimetry
3.3.1 Room temperature fading behaviour

Five replicate glass dosimeters were irradiated with ^{60}Co gamma rays to 30 kGy. The changes of absorbance were followed up to 535 days. After each measurement glass samples were stored in the dark at room temperature. Fig. 6 presents the fading behaviour of the 410 nm absorption band at room temperature respectively for the long-term and the short-term period. A strong fading can be observed in the first 9 days followed by a slow fading up to 535 days. The 600 nm absorption band showed similar behaviour.

The data in Fig. 6 were fitted using first-order kinetics based on the data after the first 10 days. The coefficient of correlation (R^2) was 0.99.
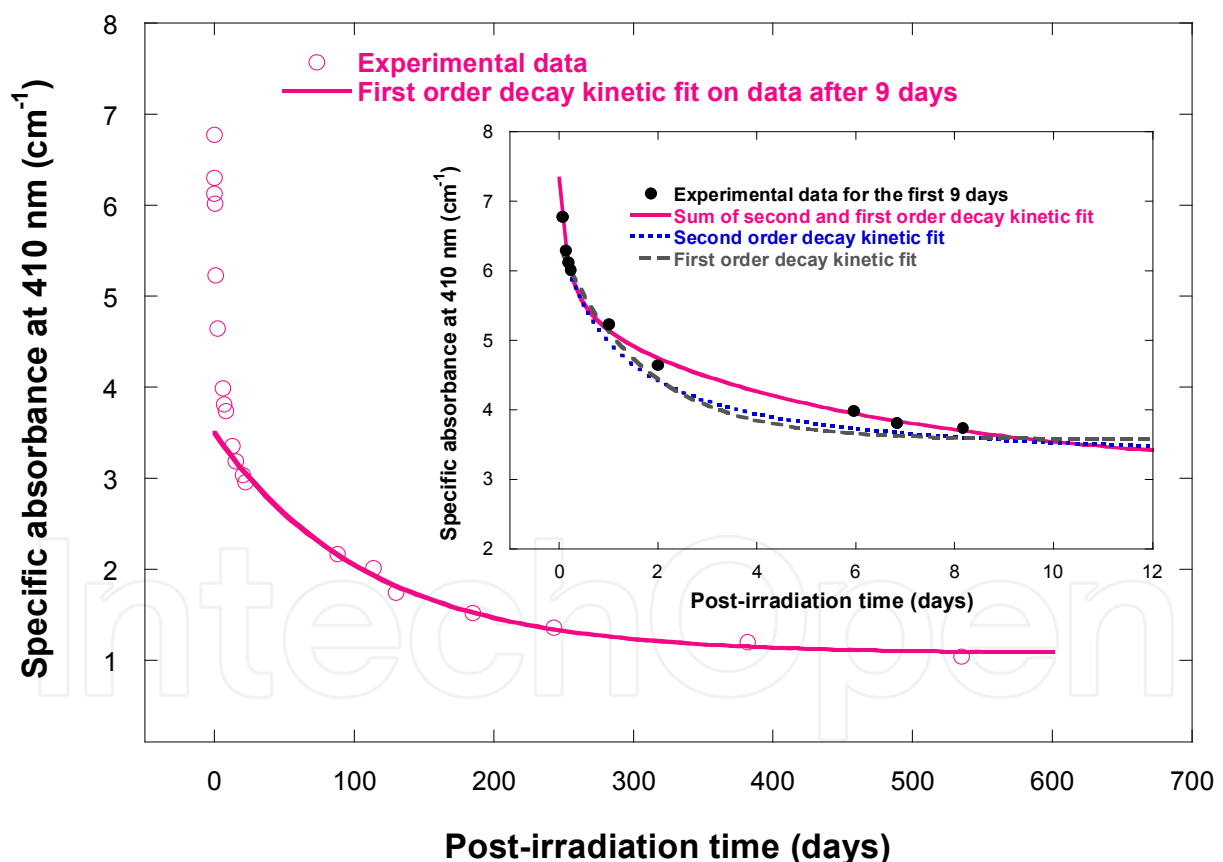

Fig. 6. Post-irradiation stability at room temperature of response of glass samples irradiated at 30 kGy. Inset: kinetics of the fading behaviour at room temperature for the short-term period.

The stability of radiation induced colour centres at room temperature was mainly controlled by the initial strong fading process. This initial fading process seems to follow neither a

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simple first-order nor second-order kinetics, but it can be well described by a sum of a first order decay kinetics function (exp (-t/τ)) and second order decay kinetics function
[1+ k (t/κ)]⁻¹, where t is the annealing time, τ and κ are appropriate time constants and k is a fitting parameter.

The thermal decay of colour centres after irradiation is mainly controlled by the diffusion of species in glass and it is generally attributed to diffusion - limited processes (Agnello & Boizot, 2003; Sheng et al., 2002c). Agnello and Boizot speculate that reaction of mobile species induced by irradiation with the induced colour centres is in the origin of their thermal decay (Agnello& Boizot, 2003). These mobile species are for example oxygen ions or anions and cations impurities released by broken bonds. In fact, a large number of studies have demonstrated that alkalis ions are movable and diffusible under irradiation. The association with NBO confines the alkali ion to local motion, whereas the absence of a co-ordinating NBO allows the alkali ion to explore more easily its environment (Ojovan & Lee, 2004).

By the combination of (MAS NMR) and (XPS) spectroscopy investigations, (Boizot and al., 2000) provided additional evidence in favour of a long range migration of sodium in glass.

Sheng et al., 2002c), gave evidence that the long-term fading process of colour centres induced by X-rays in soda-lime silicate glass was dominated by a first order kinetics, while, both the first and the second order kinetics played role in the short-term fading process (Sheng et al., 2002). Indeed, each NBOHC induced by gamma irradiation is surrounded by electrons and other NBOHCs. They assumed that the recombination of NBOHCs is controlled by the diffusion of electrons in the glass network (reaction 4) and/or by reaction of NBOHC with neighboring NBOHCs (reactions 2 and 3):

$$\equiv Si - O° + e^- \rightarrow \equiv Si - O^- \qquad (2)$$

$$2(\equiv Si - O°) \rightarrow \equiv Si - O - O - Si \equiv \qquad (3)$$
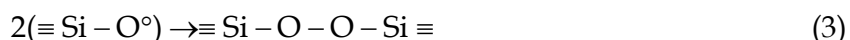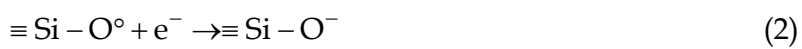
It is obvious that the reaction (2) dominate the recombination process at short-term range because of the small initial distance between NBOHCs (Waite, 1957). Sheng and al. demonstrated that in addition to reaction (2), reaction (3) played also role in the short-term recombination process, while the long-term recombination process was dominated only by reaction (2).

3.3.2 Effect of post irradiation heat treatments on the room temperature fading

The effect of post irradiation heat treatments on the glass response fading was studied in the temperature range of 60-150 °C using sets of three glass samples. After gamma irradiation with 30 kGy absorbed dose, dosimeter sets were immediately submitted to the different heat treatments for 20 minutes, which was found to be the best treatment time, and stored after irradiation in the dark at room temperature. Optical absorbance measurements were carried out up to two months. The specific absorbance values were normalized to the first measurements taken 5 minutes after the heat treatments. The results are presented in Fig. 7 and Table 4. The best results have been obtained with heat treatments at 150 °C (20 min). This procedure is very effective for the removal of unstable entities responsible for the initial strong fading. The standard deviation of glass dosimeters response measurements is about 0.5 % (1σ) within the first two hours after irradiation. The response decay of irradiated glass dosimeters is about 8 % between the first 24 h and 20 days. This means that glass dosimeters can be evaluated either within the first two hours or just after one day after irradiation and heating.
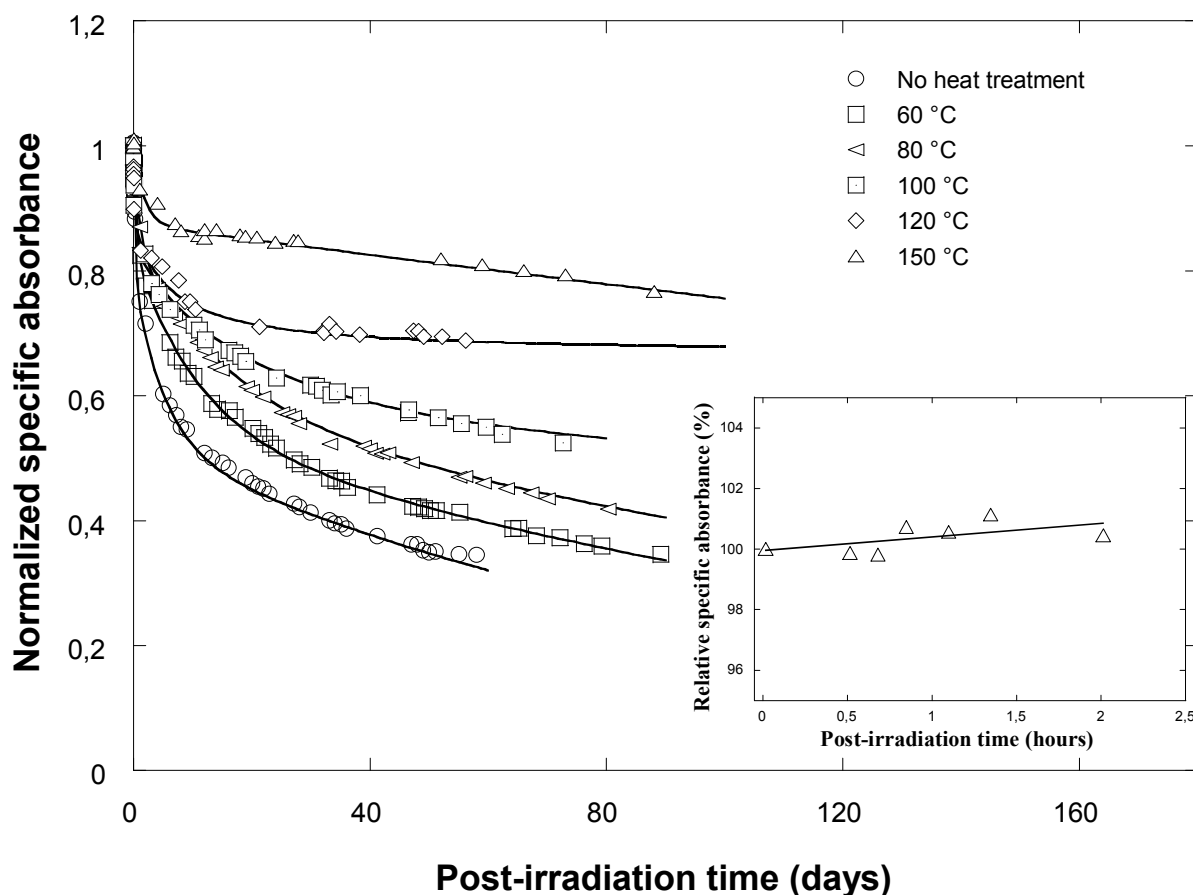
Fig. 7. Post-irradiation stability at room temperature of glass samples after 30 kGy irradiation and different heat treatments for 20 minutes. Inset: behaviour of glass samples, heated at 150 °C for 20 min, within the first two hours after the heat treatment.

Storage time	Reduction of relative specific absorbance (%)							
	Room temperature	-5 °C	-20 °C	60 °C	80 °C	100 °C	120 °C	150 °C
24 h	25	5	5	17	16	15	12	7
20 days	54	40	39	49	45	35	29	15
60 days	65	66	65	60	54	45	31	19

Table 4. Effect of thermal treatments on fading of glass samples after an irradiation of 30 kGy with gamma rays

3.3.3 Dose response curves

In order to find out the useful dose range for this silicate glass, the response curve (specific absorbance versus dose) was measured in the dose range 0.5 kGy– 87 kGy. Irradiations were carried out at the 60Co Gamma cell 220 excel of the Egyptian Radiation Technology Centre with the dose rate of 4.78 kGy/h at controlled temperature of 34°C. All data for the dose response curve were taken 24 h after irradiation. The reported specific absorbance was measured at 410 nm (Figure 8). The specific absorbance shows a rapid growth up to 40 kGy.

At higher doses the specific absorbance continued to grow slowly up to 87 kGy which was the upper dose level of the present experiments. The glass response had not yet reached saturation at this dose level. The dose response curve taken after irradiation and heat treatment at 150°C for 20 min for the dose range of 0.5–87 kGy is shown as inset of the Fig.8. The dose response curves shown in Fig.8 are successfully fitted following Mashkov equation.

T °C	First-order kinetics τ (h)	Second order kinetics κ (h)
Room temperature	189.6	30.9
-5	1128	60
-20	2184	117
60	169.5	7.5
80	168	0.43
100	110.4	0.024
130	6.25	0.0006
150	3.15	0

Table 5. Fading time constants for temperature range from -20 to 150 °C
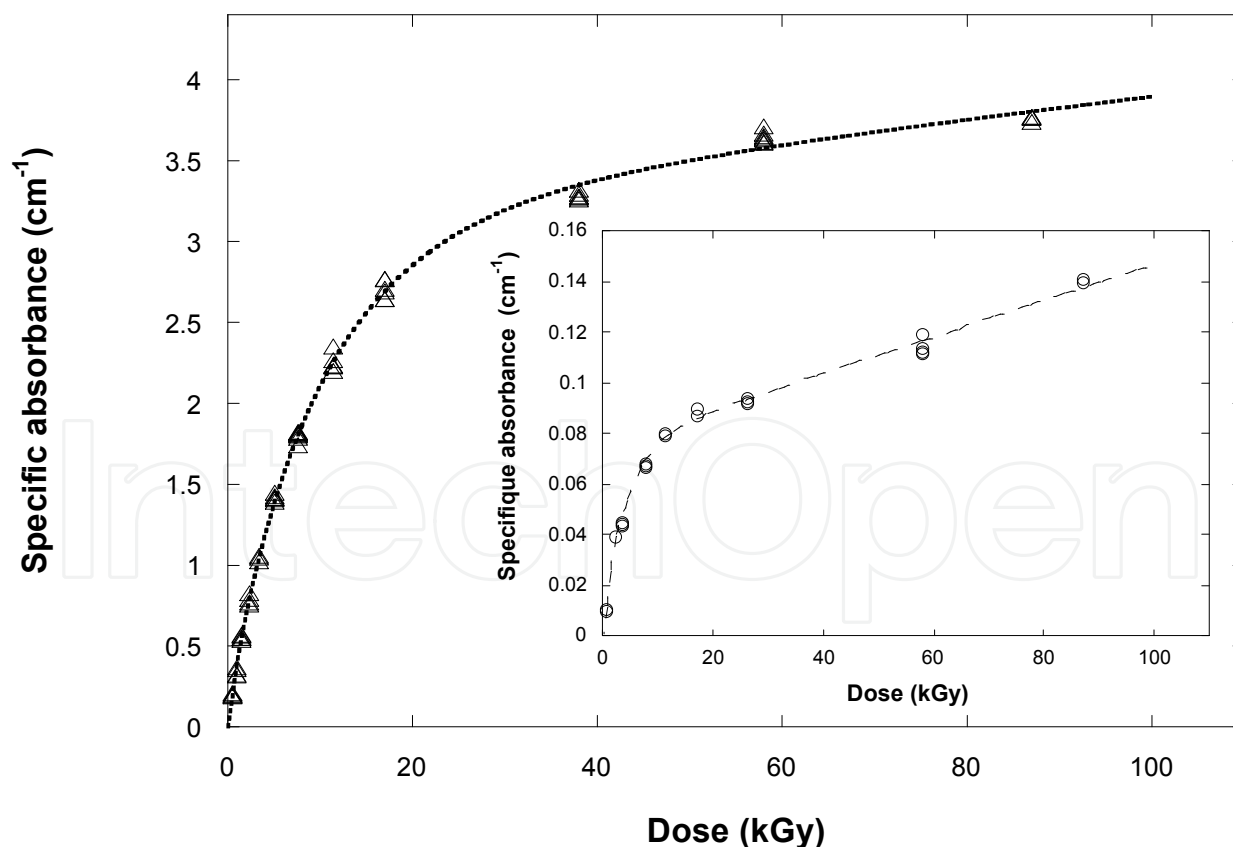
Fig. 8. Response curve for gamma irradiated glass in the dose range of 0.5-87 kGy (dose to water), absorbance at λ = 410 nm, dose rate: 4.78 kGy/h. Inset: response curve after heat treatment at 150°C for 20 min.

3.3.4 Effect of temperature during irradiation

The response of the majority of dosimeters is affected by the temperature during irradiation (Abdel-Fattah and Miller, 1996, Farah et al., 2004). In large gamma ray irradiators, the dosimeter temperature can reach 60°C for high doses. This effect should be carefully investigated especially if the dosimeters are going to be used for dose measurement at temperatures different than that for which they were calibrated.

In order to study the effect of temperature during irradiation on response of the glass dosimeters, samples were irradiated to an absorbed dose of 5 kGy at temperatures in the range from –3°C to +80°C, temperature range used during the usual irradiation processing. After irradiation, dosimeters were stored for 24 h in dark at room conditions (25 ± 3°C, 40-60% R.H.) then specific absorbance was measured at 410 nm. Figure 9 shows the specific absorbance, normalized at -3°C, plotted as a function of temperature during irradiation. The response of glass dosimeter decreases from –3°C to +80° C and the temperature coefficient of optical absorption was negative corresponding to a mean value of (-0.53 ± 0.02) %°C-1 and (-0.45 ± 0.04) % °C-1 respectively for 410 nm and 600 nm. These results extend the published data of (Zheng et al., 1988) and (Zheng, 1996). These authors observed, for glass dosimeters irradiated with 3 kGy, a negligible variation of absorbance at the wavelength of 500 nm between 0 and 50 °C followed by a fast decrease between 50 and 80 °C.

These differences can be explained by the fact that the colour centre studied by (Zheng et al., 1988) is probably due to free electrons trapped at some imperfection in the glass structure. While the induced absorption bands observed in our glass can be attributed to a trapped holes (NBOHCs colour centres).
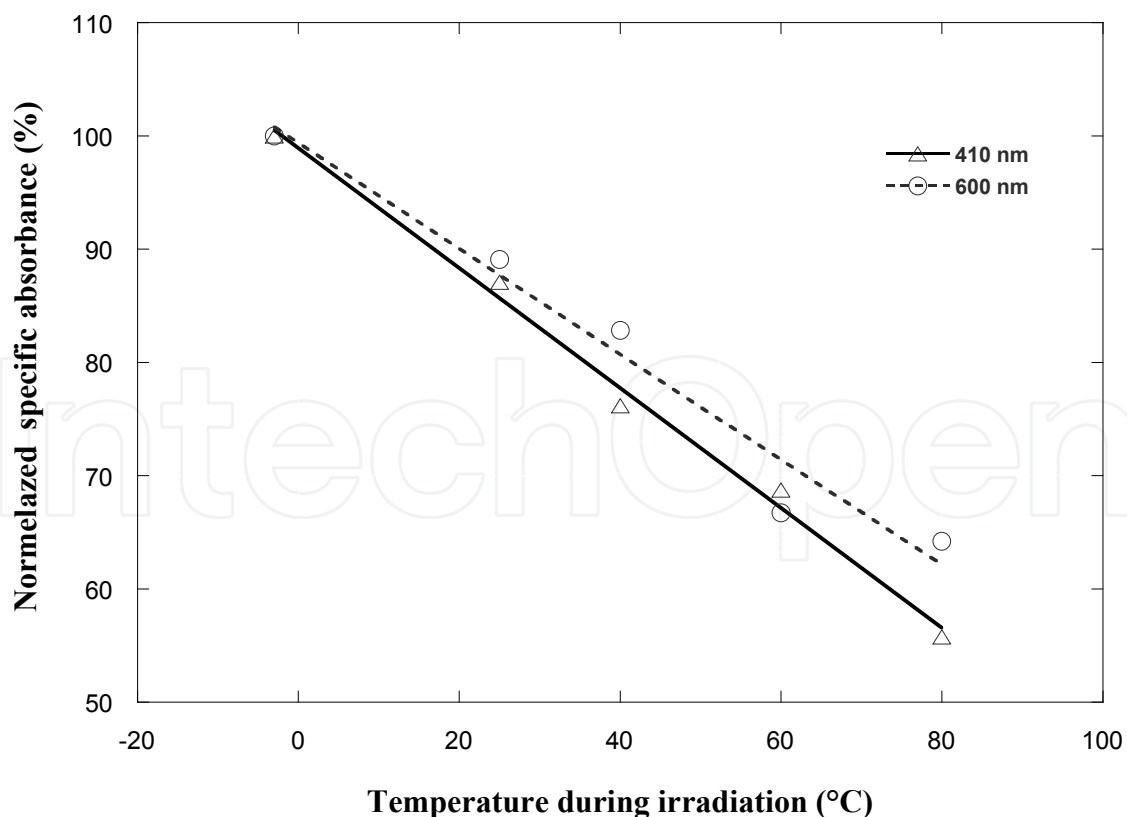

Fig. 9. Effect of temperature during irradiation on the response of glass samples irradiated at 5kGy and measured at 410 and 600 nm absorption bands

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3.3.5 Relative humidity effect during post-irradiation storage

In order to investigate the effect of Relative Humidity (R.H.) during post-irradiation storage on the response of glass dosimeter, sets of three replicate glass samples were irradiated using 60Co gamma rays at a dose of 7 kGy. Following irradiation, glass samples were stored in the dark under different extreme R.H. conditions, dried condition (about 0%), moist condition (in water) and room condition (between 40-60%). The specific absorbance was measured at 410 and 600 nm up to 22 days. As seen in Figs. 10 and 11, small differences of specific absorbance have been observed between the glass dosimeters stored in the two extreme R.H. conditions (0 and 100%). Relative differences of specific absorbance compared to the ambient conditions of glass dosimeters stored in extreme R.H. conditions (0 and 100%) are significant for the two wavelengths 410 and 600 nm.
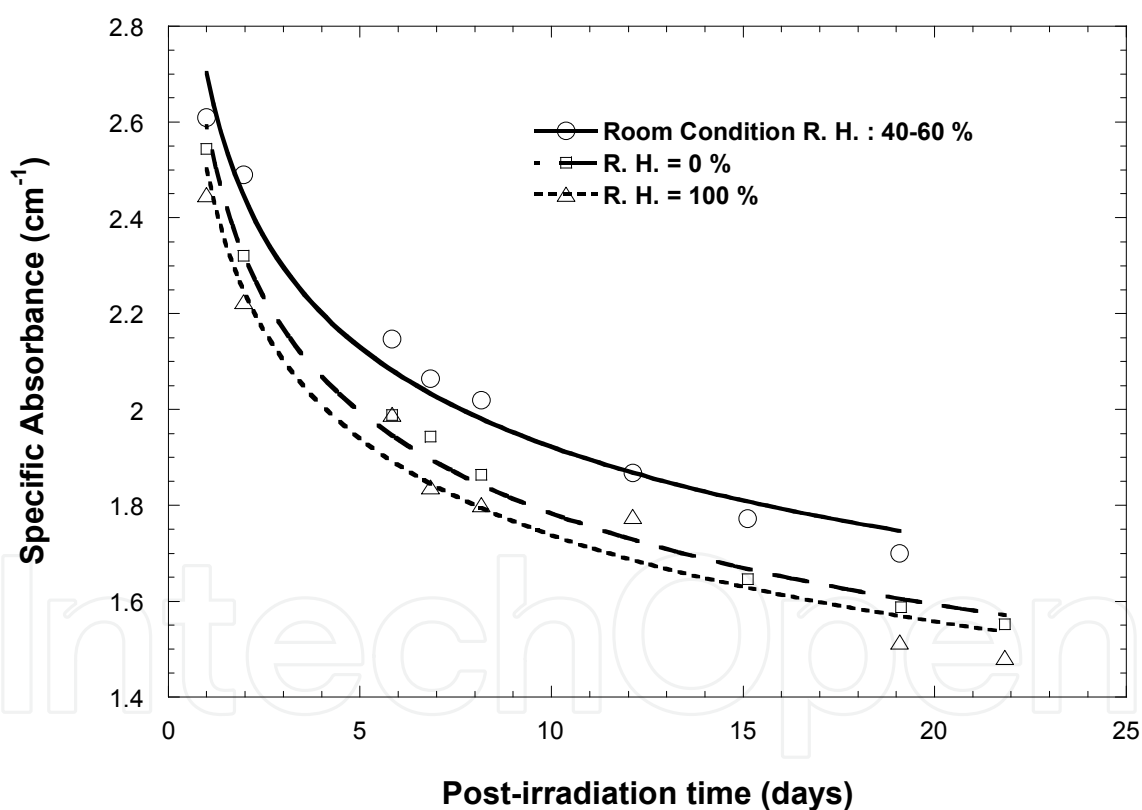
Fig. 10. Post-irradiation stability of irradiated glass to 7 kGy and stored at a different humidity conditions (410 nm band).
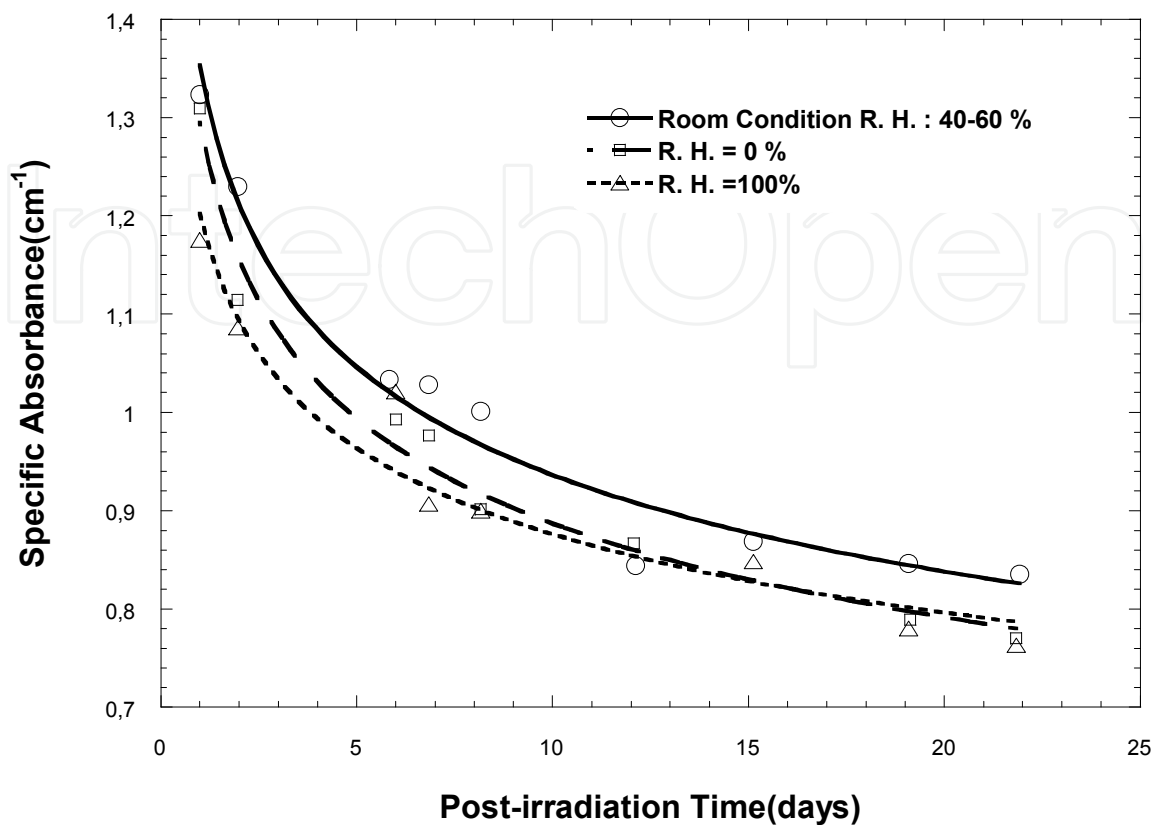
Fig. 11. Post-irradiation stability of irradiated glass to 7 kGy and stored at a different humidity conditions (600 nm band).

3.3.6 Dose rate effect

In the semi-industrial applications that depending on plant design, products may be treated in different positions with different dose rates, whereas, dosimeters were calibrated at a fixed dose rate. In order to investigate the effect of the dose rate on the response of glass dosimeter, several groups of glass samples were irradiated to different doses in the range of 0.5-50 kGy at the following dose rates: 1 and 6 kGy/h. Glass samples were submitted to a thermal treatment at 150°C for 20 minutes immediately after irradiation. Specific absorbance measurements were carried out after 24 hours. As seen in Fig. 12, the dose rate effects did not appear to be significant in the dose range of 0.5-20 kGy. Only a weak change of response within 1-2% was observed. This result is in good agreement with published data (Engin et al., 2006; Zheng et al., 1988). Above 20 kGy, glass samples exhibit significant dose rate dependence within 8-11%. In fact, the absorbance increases within 8–11% when the dose rate decreases. This observation is in agreement with the data published by (Ezz-Eldin et al., 2008). For low dose rate the rate of electrons production is low, thus the ejected electrons has enough time to annihilate a glass defect or forming non-bridging oxygen centers which causes an increase in the glass absorbance.

For the high dose rate, the rate of electrons production become high, which gives a better chance for their fast recombination rather than for annihilation resulting in less absorbance.
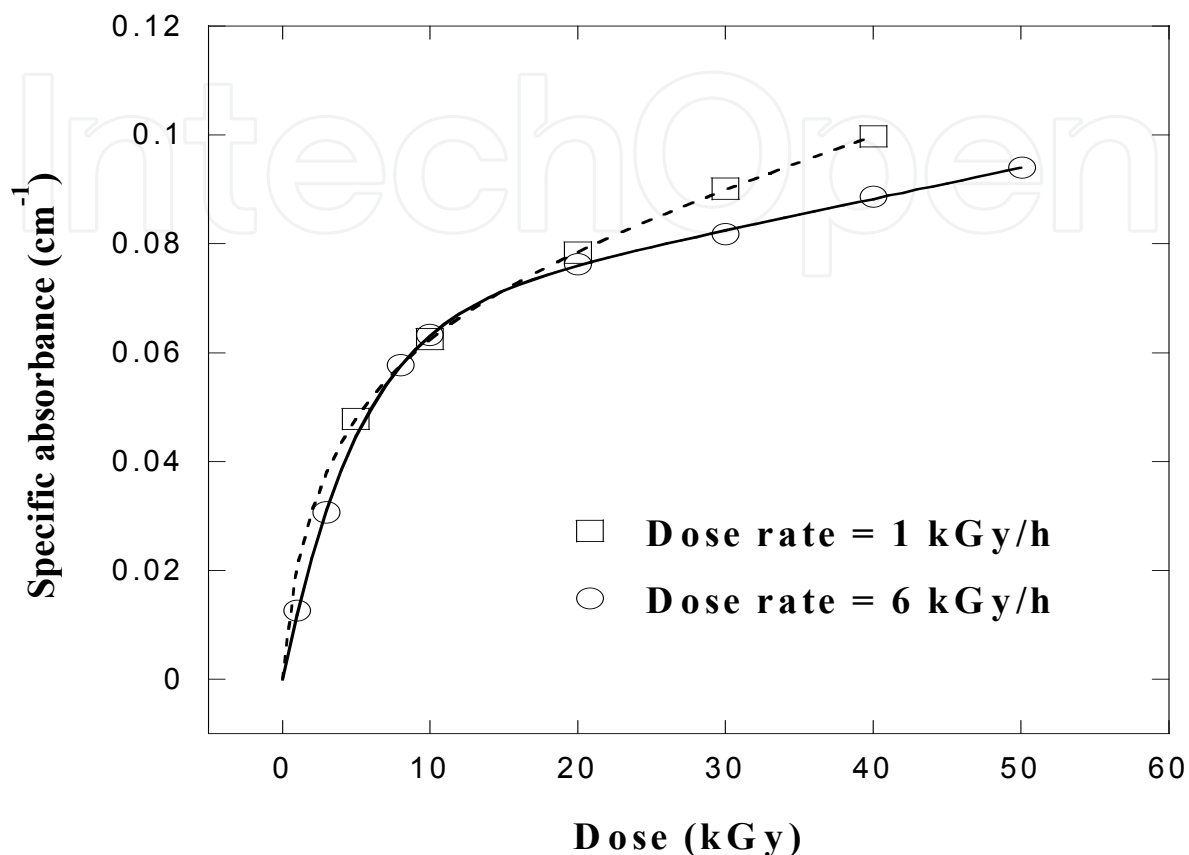
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Fig. 12. Dose rate effect on the response of glass samples irradiated at 1 and 6 kGy/h and measured at 410 nm absorption band.

3.3.7 Reuse and reproducibility of glass dosimeter

Few investigations were found in the literature (Sheng, 2002b) about the reuse of glass dosimeters, by thermal bleaching of the radiation induced coloration.

In order to study the possibility of re-using glass dosimeter, three sets of glass samples, each containing three, were irradiated, at room temperature, with 60Co gamma rays at three different doses 1, 5 and 20 kGy.

Measurement of the specific absorbance was taken 30 min after irradiation. Then the irradiated samples were submitted to a heat treatment of 300°C for 30 min, sufficient to remove the radiation induced colour centres, before performing a new irradiation. This procedure was repeated six times giving good reproducibility as it can be seen from the results shown in Fig. 13. The standard deviation of the measurements was found to be lower than 4% (1σ).
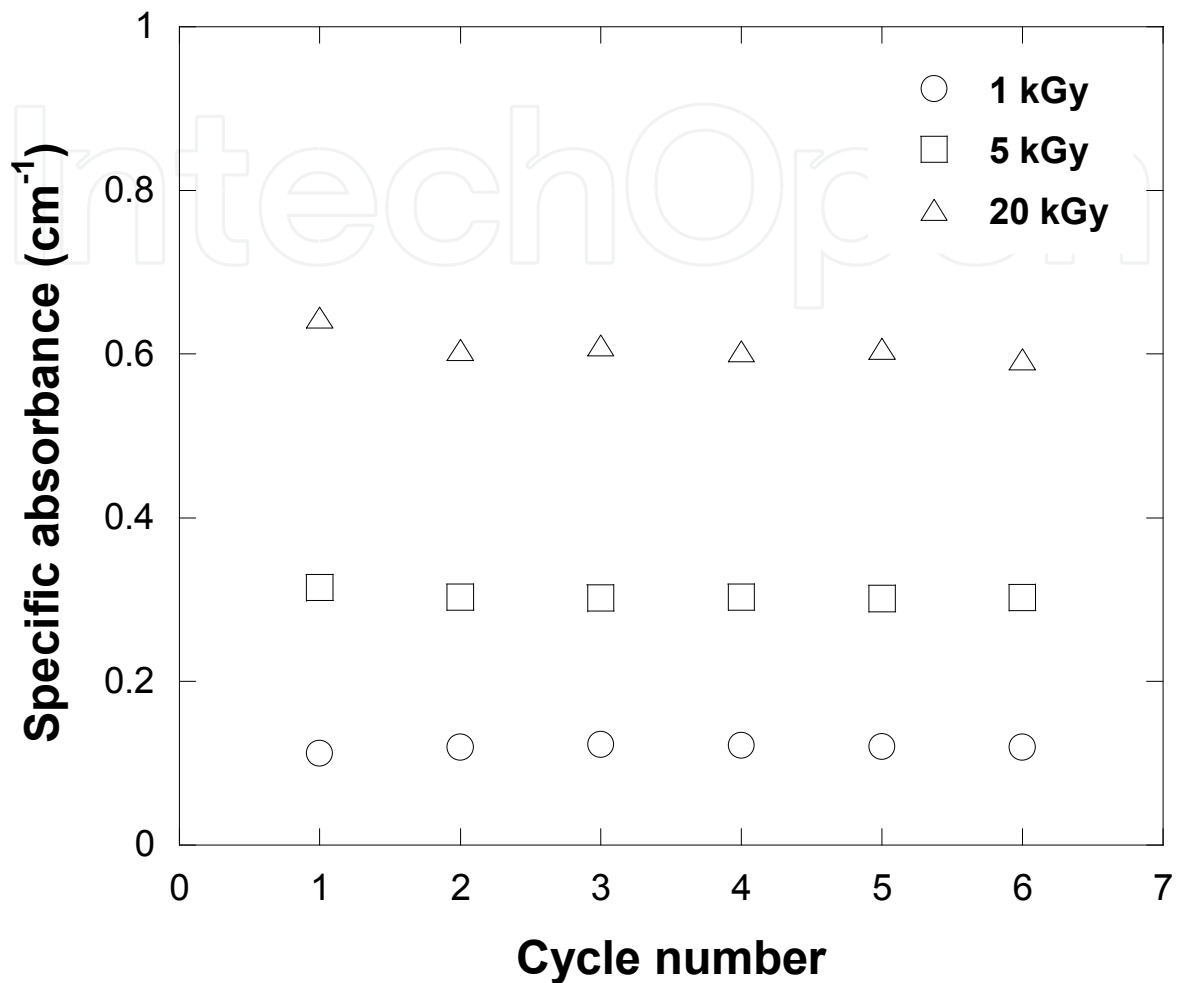
Fig. 13. Reproducibility of the results by reuse of glass dosimeter.

3.4 In-plant calibration of glass dosimeter for gamma irradiation
3.4.1 Calibration conditions
The in-plant calibration of glass dosimeters was done in the Tunisian semi-industrial cobalt-60 gamma irradiation facility rigorously following the procedure described in section 2.5 Method and condition of calibration. The phantoms, containing the alanine transfer standard dosimeters and the glass samples, were fixed on dummy product boxes and irradiated to nominal doses from 0.5 to 17 kGy at the dose rate of 1 kGy/h.

All dosimeters were read 24 hours after irradiation. Figs. 14 and 15 represent the response functions where the specific absorbance of the irradiated glasses vs dose to water, as measured by the alanine dosimeters, is reported. Here the mean values of the specific absorbance for each group of glass samples are plotted.

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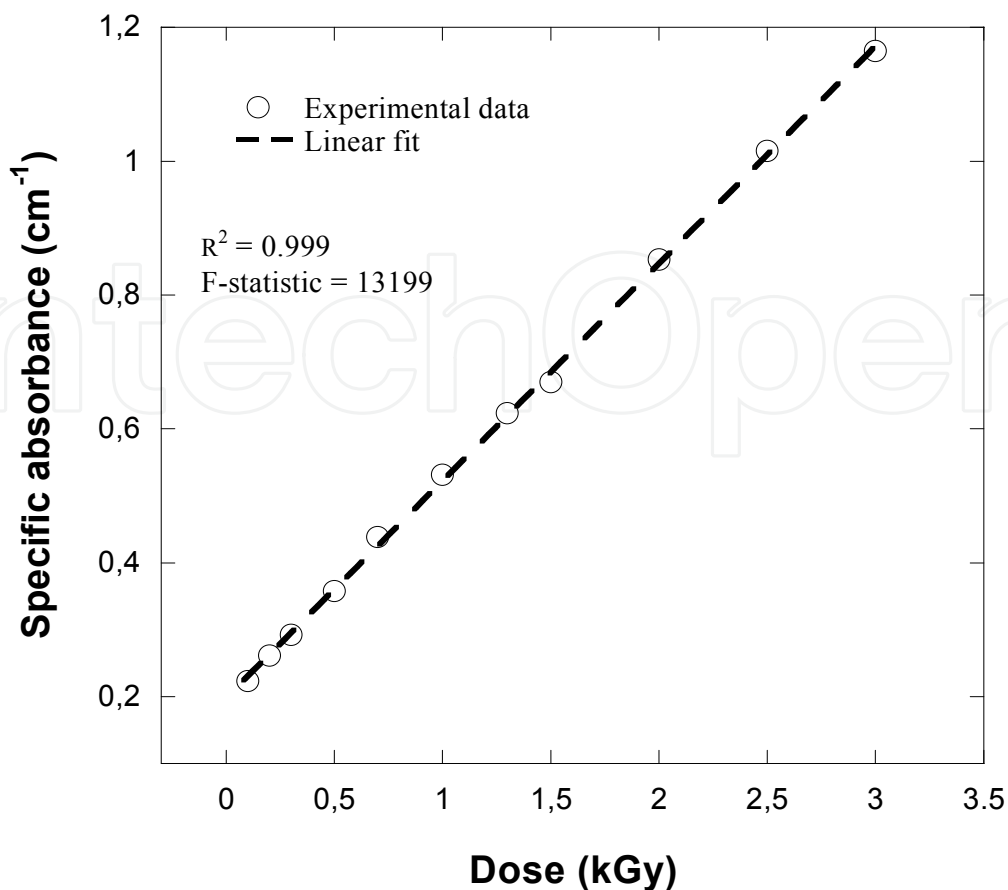
Fig. 14. Calibration curve of glass dosimeter in the dose range 0.1-3 kGy (dose to water).
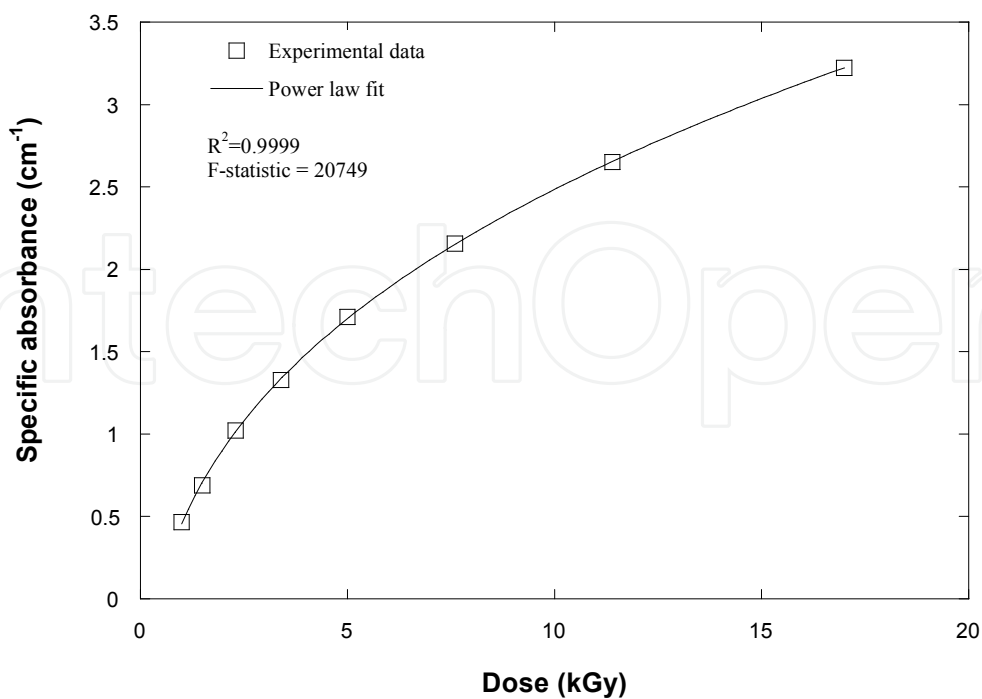Y = 0.1982 +0.3245.X equation is used for linear fit.

Fig. 15. Calibration curve of glass dosimeter in the dose range of 1-17 kGy (dose to water).
Y = - 1.262 + 1.717.X 0.34 equation is used for power law fit.

3.5 Estimation of uncertainty

The procedure outlined in ISO/ASTM Standard 51707 (ISO/ASTM, 2002) was followed to estimate the overall uncertainty in the measurement of absorbed dose from the glass dosimeter. Three components of uncertainty were taken into account. These included, (1) uncertainty in absorbed doses reported by the Aérial, (2) uncertainty in the spectrophotométric analysis of dosimeter response and measurement of dosimeter thickness (Eq. 4), and (Eq. 5) goodness of fit of the power law function to the calibration data.

$$CV_{overall}(\%) = \sqrt{\frac{\sum\limits_{i}(n_i - 1)S_{i-1}^2/\overline{k}^2}{\sum_i(n_i - 1)}} \times 100\% \qquad (4)$$

Where:
C.V. (%) = overall coefficient of variation
S_{i-1} = sample standard deviation for ith set of measurement,
(n_i-1) degrees of freedom for ith set of data,
$\overline{k}$ = average value of specific absorbance for ith set of measurement,
n_i = number of replicate measurements for ith set of data.

$$u_f = \sqrt{\frac{\sum\limits_{i=1}^{i=n}(y_i - f(x_i))^2}{n - m}} \qquad (5)$$

Where:
u_f = fit standard uncertainty
y_i = y data value
$f(x_i)$ = predicted y value using the power law function
n = number of data points
m = number of coefficient fitted

Results of this analysis are given in Table 6. The components of uncertainty combined in quadrature gave an estimate of overall uncertainty at a 95% confidence level of ± 7.58% (2σ) which is in basic agreement with the expected uncertainty for the routine use of these glass dosimeters and acceptable for the intended applications.

Uncertainty	Type A (%)	Type B (%)
Reference Dose (Aérial)	2.50	
Uncertainty in calibration curve	2.43	.48
Overall uncertainty (%)	3.79	
Expanded overall uncertainty (%) (k=2)	7.58	

Table 6. Components of uncertainty.

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3.6 Field trial irradiations

In order to check the applicability and the performance of glass for routine application, field trial irradiations were carried out with the same irradiation arrangement for used the in-plant calibration. Five sets of three glass dosimeters and five sets of three Gammchrome dosimeters were irradiated, together in the polystyrene phantom in close contact, without shielding each other, to nominal doses of 0.2, 1, 1.5, 2.5, and 3 kGy. Four sets of three glass dosimeters and four sets of three Amber Perspex dosimeters were irradiated, together in the polystyrene phantom in close contact, without shielding each other, for the nominal doses: 1, 3, 7, and 15 kGy.

Dose determination for the irradiated glass was done using the established in-plant calibration curves of Figs.14 and 15. The evaluation of the absorbed dose with Harwell Perpsex dosimeters was done using our own calibration function traceable to the Aérial SSDL.

Linear regressions were applied to compare the absorbed dose results obtained in the gamma plant, measured by glass dosimeters and by the Gammchrome and Amber Perspex dosimeters (Figs. 16 and 17). The values of slope lines and correlation coefficients calculated from the data of Figs. 16 and 17 indicate the good agreement.
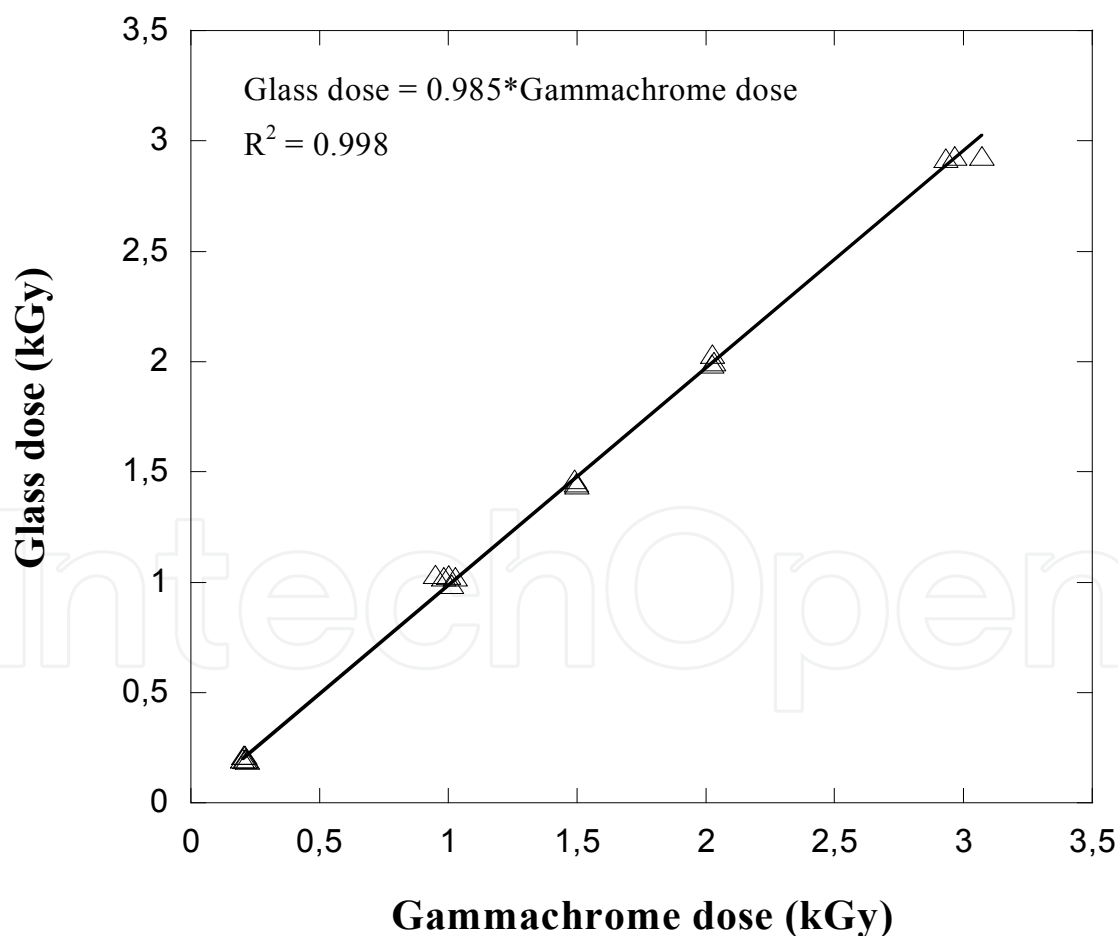

Fig. 16. Comparison of doses measured by Gammchrome dosimeters and glasses irradiated together during production runs at the gamma plant.
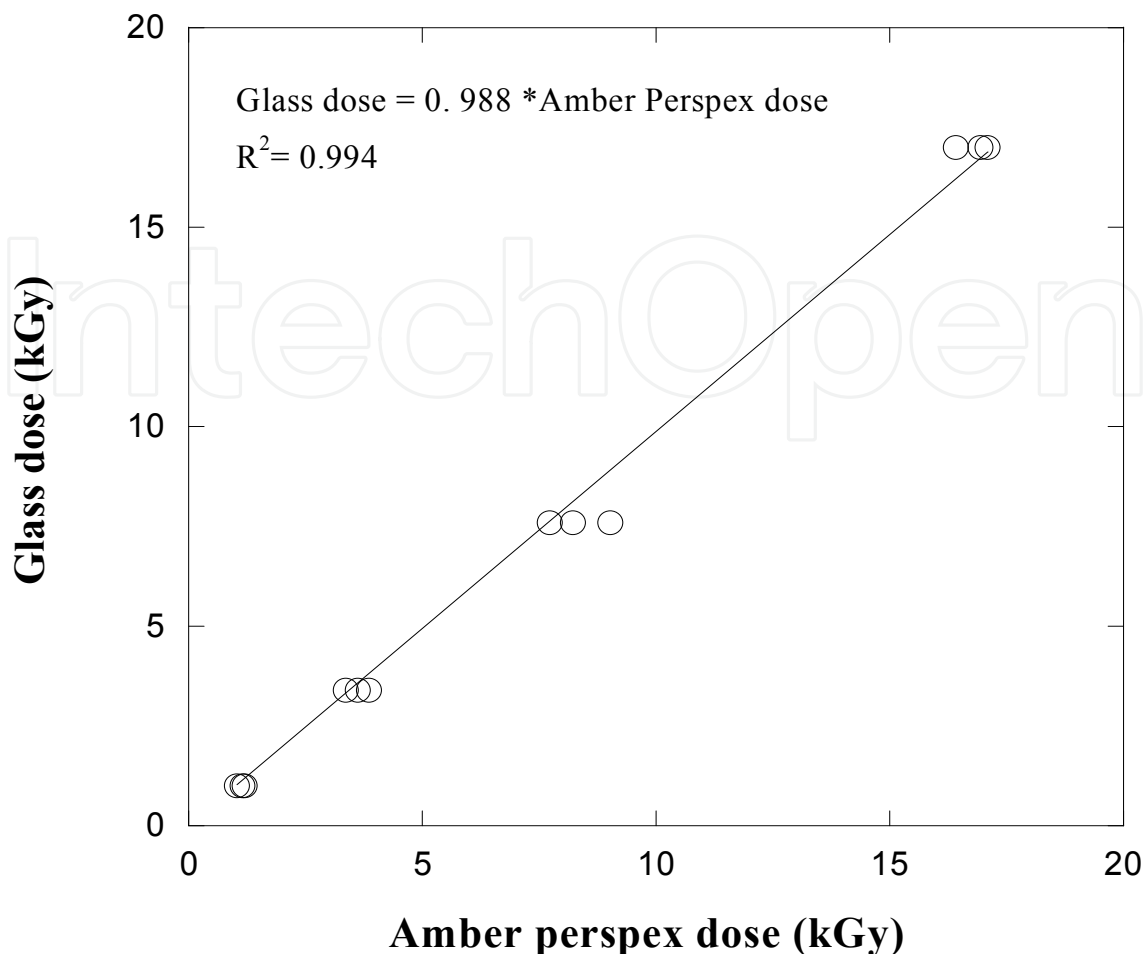
Fig. 17. Comparison of doses measured by Amber Perspex dosimeters and glasses irradiated together during production runs at the gamma plant.

4. Conclusion

The induced optical absorption by gamma irradiation in the visible range of this silicate glass is due to the generation of two Non Bridging Oxygen Hole Centres: HC_1 at 410 nm and HC_2 at 600 nm. The similarity of the activation energy values of the annealing process for both bands suggests that the related optical transitions correspond to the same type of NBOHCs in different configurations. The increase of gamma irradiation dose had no influence on the band peak positions and the FWHM of the induced colour centres and only the heights and the areas under the bands changed.

The nonlinear growth of the absorbance with increasing of dose can be explained by the competition of two different colour centres creation processes. The first is an intrinsic process corresponding to the rupture of the bond $\equiv$ Si-O-Si $\equiv$ in which the concentration of colour centres increases linearly with the dose. The second one is an extrinsic process corresponding to the activation of a fixed number of pre-existing precursors which must therefore saturate.

These colour centres were unstable at room temperature or at heating conditions. Most induced colour disappeared after being treated at 300 °C for 20 minutes and completely disappeared at 500 °C. The initial strong post-irradiation decay can be explained by reaction

of NBOHC with neighboring NBOHCs because of the small initial distance between them. At long-term stage the slow decay is only controlled by the diffusion of impurities and electrons in the glass network.

The present study reveals that the optical absorbance response of this type of glass to γ can be quantified reasonably well in the 0.1-50 kGy range. The tests conducted on this type of glass show its good performances in the production irradiators and if the glass samples are calibrated in the plant they are reliable dosimeter material that can be used as a routine dosimeter for measuring doses between 0.1-17 kGy ensuring that the environment conditions are carefully controlled during the use of this dosimeter. The effect of influence quantities on the glass response could be minimized by performing the in-plant calibration. Even if the fading effect 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.

These results indicate that this material is very interesting for dose evaluation in many radiation processing applications.

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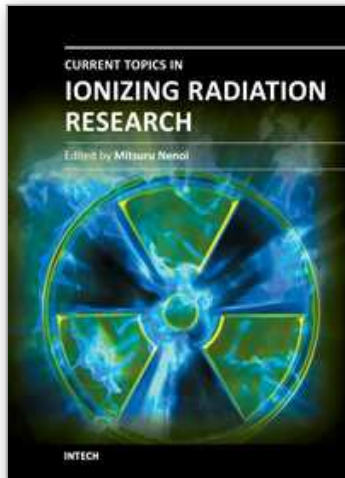
Current Topics in Ionizing Radiation Research

Edited by Dr. Mitsuru Nenoi

Since the discovery of X rays by Roentgen in 1895, the ionizing radiation has been extensively utilized in a variety of medical and industrial applications. However people have shortly recognized its harmful aspects through inadvertent uses. Subsequently people experienced nuclear power plant accidents in Chernobyl and Fukushima, which taught us that the risk of ionizing radiation is closely and seriously involved in the modern society. In this circumstance, it becomes increasingly important that more scientists, engineers and students get familiar with ionizing radiation research regardless of the research field they are working. Based on this idea, the book "Current Topics in Ionizing Radiation Research" was designed to overview the recent achievements in ionizing radiation research including biological effects, medical uses and principles of radiation measurement.

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