BONE EQUIVALENT CALCIUM-CONTAINING PHOSPHATE GLASS FOR RADIATION DOSIMETRY USING OPTICAL ABSORPTION

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الخلاصة :

يُعنى هذا البحث بدراسة (مقياس) الجرعات الإشعاعية المصنوع من نوع مطوّر حديثاً من زجاج (Ca–Al–Na phosphate) وذلك لشدة حساسيته الضوئية لتأيين الفوتونات photons ضمن مدى طاقة من (او . إلى ١٠ Mev) . وعند تعريض هذا المقياس المصنوع من الزجاج غير الملون إلى أشعة جاما يتكون لون يميل إلى الإحمرار وتزداد درجة إحمراره بزيادة الجرعة الإشعاعية الساقطة عليه .

كما تم ملاحظة ثلاث مناطق للامتصاص في القياسات الصيفية الضوئية لهذا الزجاج : الأولى ضمن منطقة الإشعة الفوبنفسجية (180–300 nm) ، والمنطقتان الأخريتان في المنطقة الضوئية (450–600 nm) وقد وُجد أنَّ الامتصاص الضوئي عند طول موجة 496 nm يكون خطيّاً لغاية جرعةٍ إشعاعيةٍ مقدارها (٢) كيلوجراي ، ثم يميل إلى التشبع عند (٢ - ٨) كيلوجراي .

يمتاز هذا المقياس الزجاجي الجديد بامتصاصه لإشعة جاما وذلك في مدى واسع من الطاقة (او . إلى ١٠ مليون ألكترون فولت) وقد وُجد أن معامل امتصاصه للطاقة لايعتمد كثيراً على تغير الطاقة الساقطة لذا يمكن استخدامه لقياس الجرعات الإشعاعية في الأغذية . وقد تم دراسة ثبات مراكز اللون المتكونه نتيجة الشتعيع في ظروف التخزين العادية في المعمل ، حيث وُجد أن نسبة العودة العفوية من حالة الإثارة تقارب ٣٧٪ خلال ٢٥ يوماً .

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ABSTRACT

A newly developed Ca–Al–Na–phosphate glass dosimeter was studied for its spectral sensitivity to ionizing photons in the range 0.1 MeV to 10 MeV. The dosimeter is colorless before exposure to ionizing radiation and then takes on an reddish color upon increasing irradiation with ^{60}Co gamma rays. Three radiation-induced bands are observed in the spectra of irradiated glasses. The first is in the UV region (180–300 mm) and the other two bands are in the visible region (450–600 nm). The change in absorbance is linear up to 2 kGy and then tends to saturate from 2 to 8 kGy, when measured at its 496 nm absorption band maximum.

The new glass dosimeter simulates compact bone in terms of gamma-ray absorption properties over broad radiation spectra. It has a relatively small energy dependence and can also be used for food irradiation dosimetry.

The stability of color centers formed upon irradiation was studied at normal laboratory storage conditions and the spontaneous deexcitation of the glass is about 37% in 25 days.

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INTRODUCTION

Irradiation can change the optical properties of a glass, and this effect has been used as a means of measuring irradiation dosages. Several commercial glasses and certain crystalline materials develop color centers under irradiation. Although this induced coloration is generally unstable, the measurement of absorbed doses from 10^2 to 10^6 Gy is possible using spectrophotometry for analysis [1 – 11].

A source of error in glass and solid-state dosimetry is the difference in radiation absorption properties (spectral energy dependence) of these relatively high atomic-number systems as compared with irradiated materials such as foodstuffs. These results in errors as great as 40 or 50% with degraded gamma-ray spectra, and even greater for lower energy X-rays.

In radiation processing applications in the sterilization of medical devices, food preservation, *etc.*, and in medical and radiation protection applications, the main interest is in measuring the absorbed dose in plastics and biological tissues.

In the present work, a Ca-containing phosphate glass dosimeter is designed to match as closely as possible compact bone, and to examine the radiation response characteristics of the dosimeter.

EXPERIMENTAL PROCEDURES

Glasses in the system $P_2O_5 - CaO - Na_2O - Al_2O_3$ were prepared in 100 gram batches by mixing metal oxide or metal carbonate powder with ammonium dihydrogen phosphate. Each batch was mixed and transferred to a Pt – 2% Rh crucible, then calcined at 300°C and melted at about 800°C. The melt was cast onto a preheated plate (to about 200°C) and samples were transferred into an annealing furnace which was then switched off to cool to room temperature. Glass specimens were then ground and polished for optical measurements (thickness = 2.97 mm).

Irradiation of glasses was carried out in the ^{60}Co gamma chamber 4000A (product of India). The absorbed dose rate in water in the irradiation facility was determined at the central position by Fricke dosimetry [$G(Fe^{3+}) = 1.62$ μmol/J = 15.6×10^{-2} eV^{-1}]. The dose rate was found to be 166.6 rad/s for the ^{60}Co source at the time of the experiments. Irradiations were carried out with samples held between layers of dosimeter material, in order to maintain conditions of approximate electron equilibrium [12]. Optical absorption measurements were carried out within 10 minutes after irradiation, using UVIKON 860 spectrophotometer, and were taken at room temperature (28 ± 2°C).

RESULTS AND DISCUSSION

Radiation Induced Color Centers

The glass was colorless before irradiation, and attained a reddish coloration upon increasing doses of gamma radiations. Figure 1 shows the optical absorption spectra of the irradiated and unirradiated glass samples. Curve 1 indicates the absence of any absorption bands in the visible region, nevertheless an absorption band in the UV region can be inferred in the range 180 – 300 nm. As the gamma dose increases, the intensity of the UV-band seems to increase as indicated by the shift of the UV-cut off towards larger wavelengths.

The spectrum recorded after receiving the highest dose (curve 10, Figure) shows a broad absorption band in the visible region with a peak at 492 nm. The spectra of the glass samples subjected to 1.0 and 1.5 kGy (curves 4 and 5 respectively) indicate that this band is an overlapping of two bands. Assuming that the shape of an absorption is Gaussian in shape, two absorption bands can be predicted at 450 – 580 nm and 530 – 570 nm.

Similar radiation-induced absorptions have been studied by different authors [13 – 15] and were attributed to formation of positive hole color centers. The longer wavelength-band was assigned to positive holes in the structures, containing P and O ions only. The shorter wavelength-band was assigned to holes in structures involving the metal ions in addition to the P and O ions [16]. Gaussian resolution of the induced spectra of different phosphate glasses [13 – 17] indicated the presence of two absorption bands, in the visible region, at about 427 nm and 539 nm. In the present phosphate glass the position of these two bands is confirmed by recording the 4th derivative (abs/nm^4) [18] of the radiation-induced spectrum in the range from 400 to 600 nm. Figure 2 shows that at

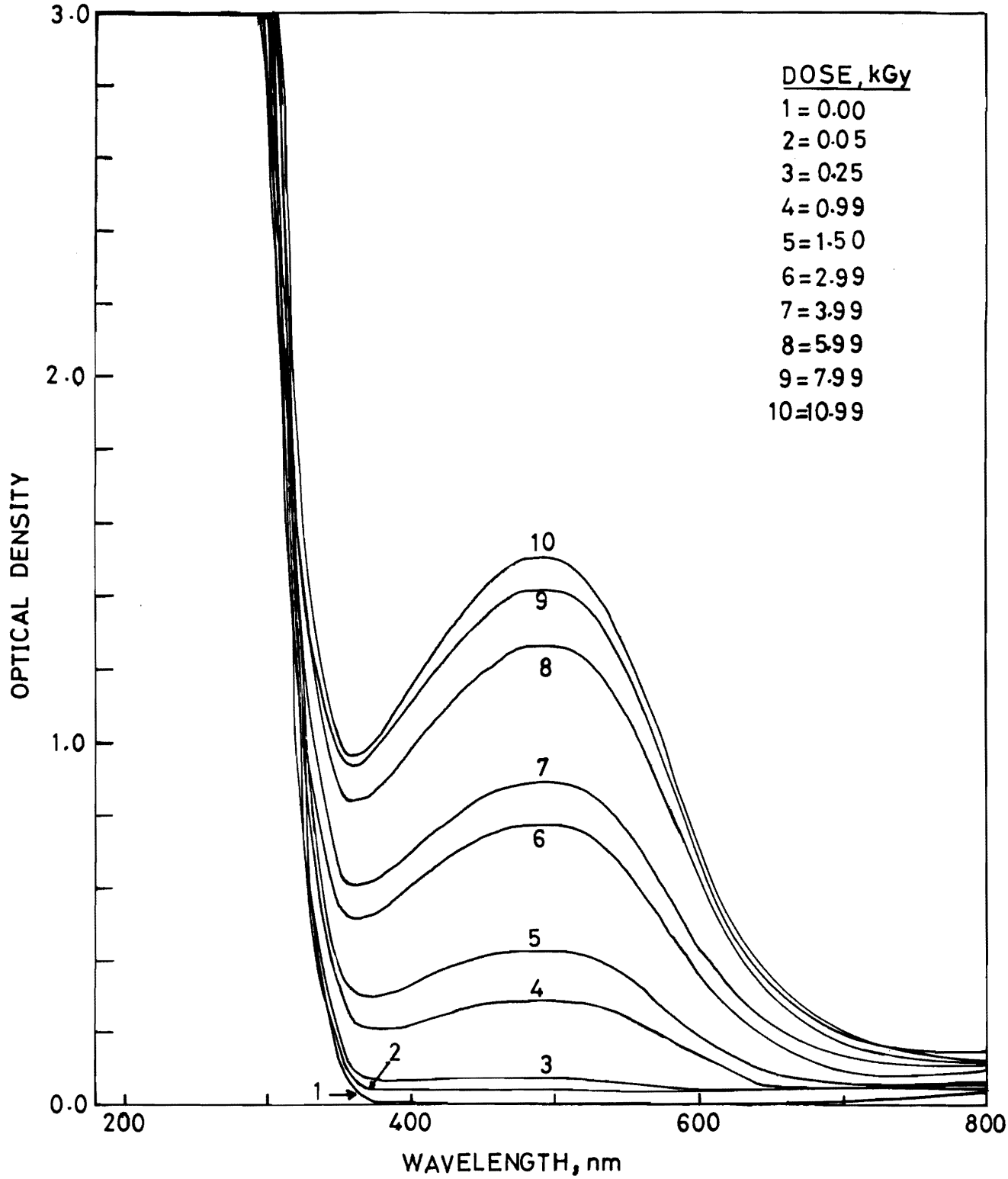

Figure 1. Absorption Spectra of Unirradiated and Irradiated Calcium Phosphate Glass.

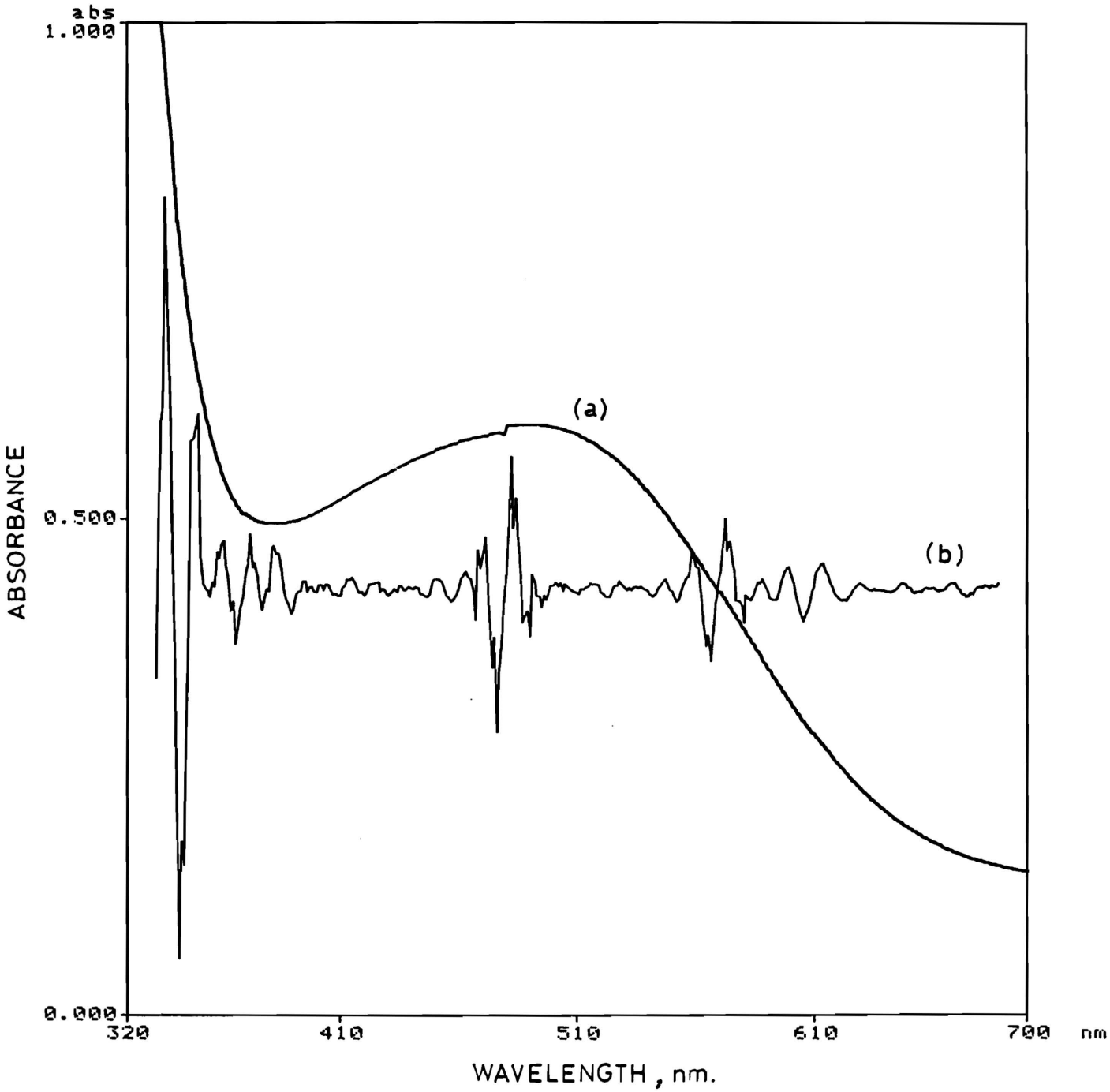

Figure 2. (a) Absorption Spectra of Irradiated Samples of Calcium Phosphate Glass. (b) Fourth Derivative of Spectral Intensity of Absorbance with Respect to Wavelength.

least two induced bands are present at about 479 nm and 570 nm.

Figure 3 shows the relation between absorbed dose and the change in absorbance per thickness at 496 nm. The response is linear up to 2 kGy and then tends to saturate from 2 to 8 kGy.

Design of a Bone-Equivalent Glass Dosimeter

The main purpose of using this dosimeter system is to simulate the ionizing photon properties of bone, so that a meaningful absorbed dose in bone can be measured. In other words, errors due to the presence of dosimeter probe materials placed in biological tissues and irradiated with broad gamma-ray spectra are thus diminished.

A formulation has been devised previously as a solid medium simulating soft muscle and plastic in terms of radiation absorption properties [19]. This system consisted of 66.8% SiO_2, 31.2% Li_2O, and 2% K_2O and showed a linear response in the range 0.1–4.5 kGy. To simulate other biological tissues, specially bone, several combinations of Ca – Al – phosphate oxides were theoretically tested and compared with bone in terms of their radiation absorption properties. The most successful combination of oxides in the glass was found to be 64.67% P_2O_5, 11.34% CaO, 18.83% Na_2O, and 5.16% Al_2O_3.

Table 1 lists the computed photon mass energy-absorption coefficients of glass dosimeter *versus* photon energy, as well as for the compact bone [200]. From these data, it is evident that, for the broad photon spectral region the glass dosimeter is similar to compact bone. The comparison is illustrated graphically by comparing absorption coefficients and their ratios *versus* radiation spectral energy (see Figure 4). Figure 4 shows plots of mass-absorption coefficients (left ordinate) *versus* photon energy, which predict a flat energy dependence of response of the glass dosimeter relative to compact bone. The ratio of absorption coefficients (right ordinate) is very close to unity over the entire photon range energy (see Table 1).

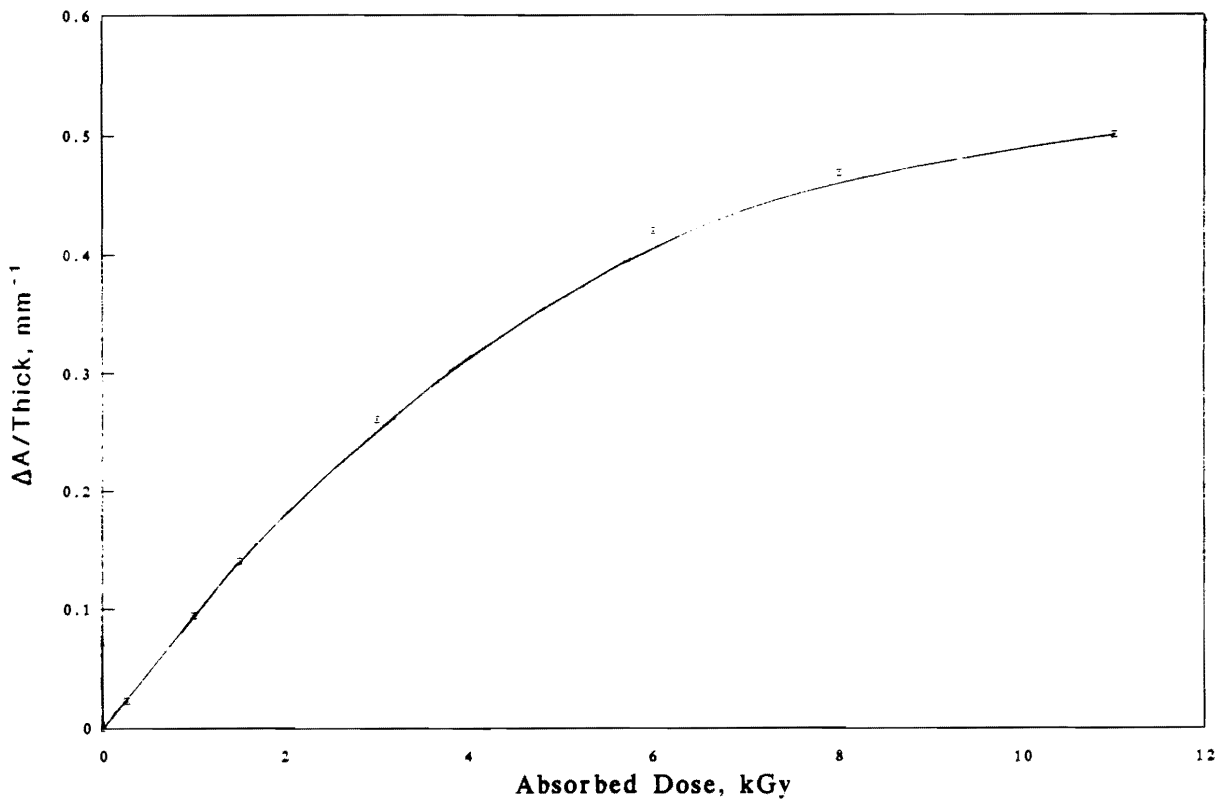

Figure 3. Response of Calcium Phosphate Glass in Terms of Change in Optical Absorbance per Unit Thickness.

Table 1. Mass Energy Absorption Coefficients μ_{en}/ρ (m^2/kg) of Ca-Containing Phosphate Glass.

Photon Energy keV	Ca-Phosphate Glass[1]	Compact Bone[2]	Ratio Glass/Bone
30	0.08617	0.0729	1.18
60	0.01117	0.00999	1.12
100	0.00391	0.00384	1.02
150	0.00291	0.00303	0.96
200	0.00281	0.00299	0.94
600	0.00291	0.00314	0.93
1000	0.00274	0.00296	0.93
1500	0.00250	0.00270	0.93
4000	0.00190	0.00200	0.95
10 000	0.00161	0.00160	1.00

[1] Calculated Ca-phosphate glass mass energy absorption coefficient

[2] Compact bone mass energy absorption coefficient (quoted from Hubbel [19])

Post-Irradiation Stability

Post-irradiation stability of the glass dosimeter was checked for a storage period of 73 days. The irradiated sample (11 kGy) was stored under normal laboratory conditions in the dark. The absorption at 496 nm was read at different intervals of time, and the results are presented in Figure 5. From these results, it can be seen that the radiation-induced optical absorbance of the dosimeter decreased by 16% of its initial value during the first 4 days after irradiation. Afterwards, a long slow rate of decrease in absorbance is noticed up to 73 days (48%).

Figure 6 shows the stability of the radiation induced optical absorption of the glass within 6 days at 100°C. It is clear that the intensity of the main absorption bands in the visible region of the spectrum decreased markedly. In other words, heating of glasses after irradiation eliminates a large portion of the unstable radiation-induced color centers. These results indicate that the intensity of the bands decreases significantly by 70% of its initial value within 24 hours after irradiation when kept at 100°C. This is followed by a long term slow decrease within 6 days. It was also observed that irradiated glasses become pale after heating.

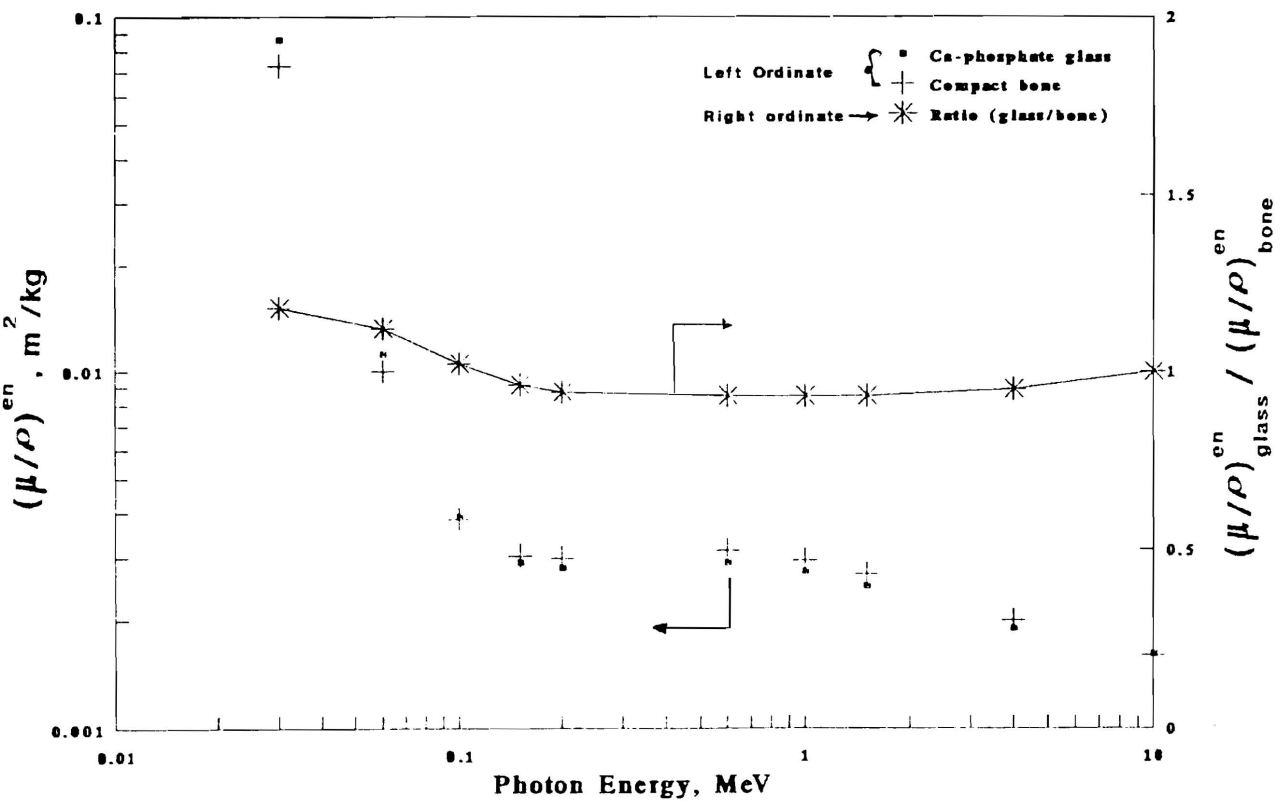

Figure 4. Calculated Mass Energy Absorption Coefficients of Phosphate Glass and Bone versus *Photon Energy; Ratio of Mass Energy Absorption Coefficients* versus *Photon Energy.*

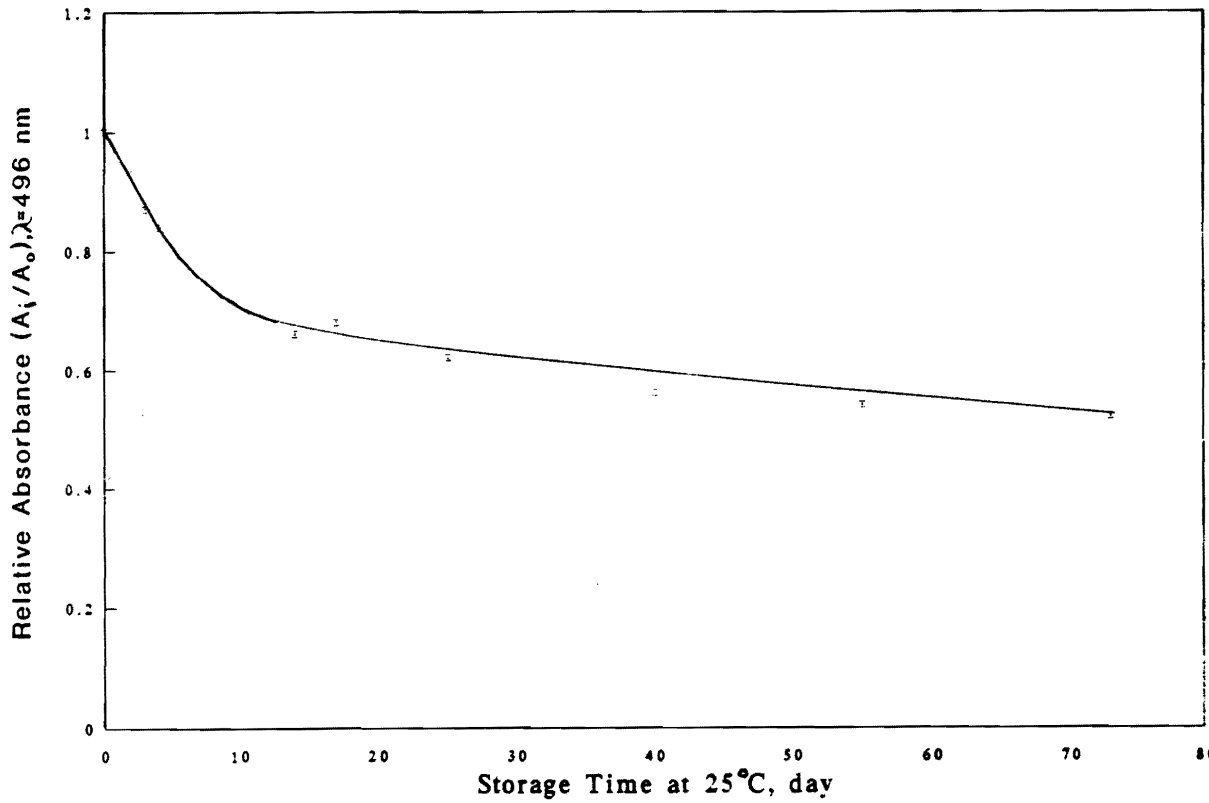

Figure 5. Change in Optical Absorbance Relative to that of Unirradiated Sample as a Function of Storage Time after Irradiation (11 kGy) at Room Temperature.

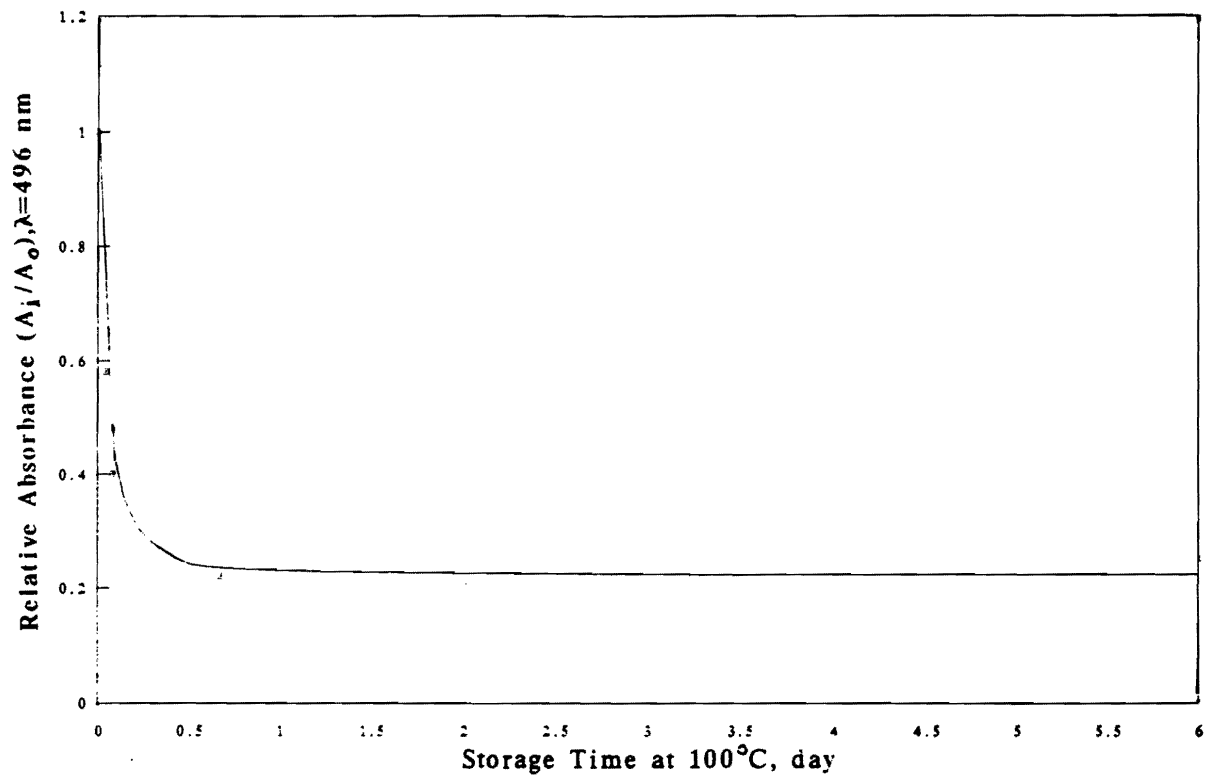

Figure 6. Change in Optical Absorbance Relative to that of Unirradiated Sample as a Function of Storage Time after Irradiation (11 kGy) at 100°C.

CONCLUSION

A new glass dosimeter has been produced for simulating bone in terms of its ionizing photon absorption properties. The effective dose range of this dosimeter is from 0.1 to 10 kGy. The spontaneous deexcitation of this glass is about 37% in 25 days. Therefore, when using this type of glass for dosimetry, the radiation-induced absorbance must be corrected for post-irradiation decay.

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