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Radiation-induced changes on some physical parameters of gelatine for protection-level dosimetry utilization

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The induced effects of gamma and thermal neutrons of respective absorbed dose ranges 10^{-2}–10^{3} Gy and 10–500 mSv, on the physical changes of the gelatine electrical resistivity, hardness, and luminescence intensity (RPL), have been investigated. The results indicated that the hardness indentation number and RPL luminescence emission intensity increase with increasing gamma and neutron absorbed doses, while the electrical resistivity decreases. The dose sensitivity relation with these induced physical changes is beneficial for dosimetric utilization. The changes are described by semi-empirical regressions formulae.

1. Introduction

The sensitivity of gelatine to gamma and neutron radiation results in physical rather than chemical changes in the behaviour of the material. In analogy with the radiation-induced effects on polymers (as amorphous material), the interaction of radiation with gelatine induced different orders of degradation and cross-linking effects in the material structure [1–10]. This revealed that physical changes induced by the effect of radiation could be manifested as colour centres. Furthermore, these changes in the cloud density of the material colour centres are mainly accompanied by other physical and chemical behaviour changes in gelatine, such as electrical, mechanical and optical changes. Information about radiation-induced effects on these different physical parameters of gelatine is still limited and poorly known. Solid-state dosimetry, in general, is based on these induced physical changes.

The present work investigated (a) gamma and neutron radiation-induced effects on the electrical resistivity, hardness and luminescence behaviour of gelatine, and (b) the beneficial utilization of such induced changes in the field of dosimetry, especially as protection-level dosimeters.

2. Experimental procedure

2.1. Sample preparation

Gelatine was prepared from collagen (extracted from fibrous protein tissue, i.e. meat) in neutral salts solution and by thermal treatment. The gelatine was extracted by dissolving in hot distilled water. The prepared solution was stirred for 2 h and then poured in moulds with 1 mm thick walls, mounted on an accurately levelled table and allowed to dry for 24 h in the open air. The samples were prepared as small pieces of uniform dimensions, 3 cm length, 2 cm width and 1 mm thickness.

2.2. Sample irradiation

The prepared samples of gelatine were irradiated to different gamma doses in the range 0.01–10^{3} Gy and neutron radiation doses in the range 4–500 mSv. The estimated absorbed doses of gamma and neutron radiation at the position of sample irradiation were determined using a secondary standard ionization chamber and a neutron rem meter, respectively, which facilitates Sievert and rem performances.

The effect of radiation on the gelatine samples were investigated using three different physical techniques.

(a) Electrical resistivity of the sample. The samples irradiated by gamma and neutron radiation, as well as the non-irradiated one, were sandwiched between two copper electrodes using a specially designed cell holder. The electrodes were optically polished to optical quality to ensure good electrical contact. A high-impedance digital Keithley Electrometer, model 616C, was used to measure the changes induced in the gelatine resistance by the gamma and thermal neutron radiations. The sample resistance was measured at 308 K, through the potential drop across the sample using an electrical circuit of a known series standard resistance.

(b) Vickers diamond pyramid indentation test. The microhardness indentation numbers of the irradiated

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and non-irradiated samples were measured by using a Vickers diamond pyramid indentation tester at a loaded force of 200 g for a fixed loading time of 15 s. The microhardness force number, HV, was taken as an average value of three different pieces irradiated to each specified gamma dose; for each sample piece, ten different indentations of HV at different selected locations were measured.

(c) Luminescence emission intensity. Measurements of the induced radiophotoluminescence emission intensity (RPL) at 385 and 495 nm band of the gelatine samples irradiated to different gamma and neutron doses, using excitation ultraviolet light of wavelength 265 nm, were carried out using a Shimatzu Spectrofluorophotometer model RL 540.

3. Results and discussion

Fig. 1a and b show the relation between the induced percentage changes of the gelatine electrical resistivity ($\Delta R = R_{\text{irrad.}}/R_{\text{blank}}$) as a function of different absorbed gamma and neutron doses, respectively. The results in Fig. 1 indicate that with increasing the irradiation dose, the resistivity decreases. Analysis of the data presented in Fig. 1 showed a linear relation between the absorbed dose and resistivity changes ($\Delta R\%$) over the range 0.01–10^2 Gy for gamma radiation and from 4–400 mSv dose equivalent, for thermal neutron radiation. The plotted relation in Fig. 1 is found to be expressed by a simple semi-empirical formula

$$\Delta R = 79.265 - 5.57 \ln D_\gamma \quad (1)$$

for gamma absorbed dose in Gy, and

$$\Delta R = 107.89 - 7.611 \ln D_N \quad (2)$$

for neutron absorbed dose equivalent in mSv, with a fit better than $\pm 6\%$.

Fig. 2a and b show the relation between the resultant changes of radiation-induced microhardness indentation number, HV, with the variation of the gamma and neutron radiations doses, respectively, absorbed by the gelatine samples. The data show a distinct gradual increase of microhardness number as the irradiation absorbed dose increases in the linear range 10^{-2}–10^3 Gy for gamma absorbed dose and 4–400 mSv for neutron dose equivalent. The experimentally deduced results, HV–D_γ and HV–D_N of Fig. 2, may be expressed by Equations 3 and 4 with an accuracy of fit ranging over 4%–6%

$$V_{HF} = 662.85 + 113.5 \ln D_\gamma \quad (3)$$

for gamma absorbed dose in Gy, and

$$V_{HF} = 27.07 + 6.596 \ln D_N \quad (4)$$

for neutron absorbed dose equivalent mSv.

Fig. 3 shows the luminescence emission spectra of non-irradiated as well as gamma- and neutron-irradiated gelatine samples excited by 265 nm ultraviolet light. The emission spectra curves of the 385 and 495 nm bands did not show any significant changes induced by the effect of neutron and gamma radiation. The depicted experimental relations of Fig. 4a and b between the intensity of luminescence emission and the radiation absorbed dose are linear, with a range extending over 10^{-2}–10^2 Gy for gamma absorbed dose and 4–400 mSv neutron equivalent absorbed dose. The mathematically predicted regressions fit to the data of Fig. 4 are found to be expressed by

$$(\text{RPL})_{385} = 8.2 \ln D_\gamma + 60.84 \quad (5a)$$

$$(\text{RPL})_{495} = 8.7 \ln D_\gamma + 66.21 \quad (5b)$$

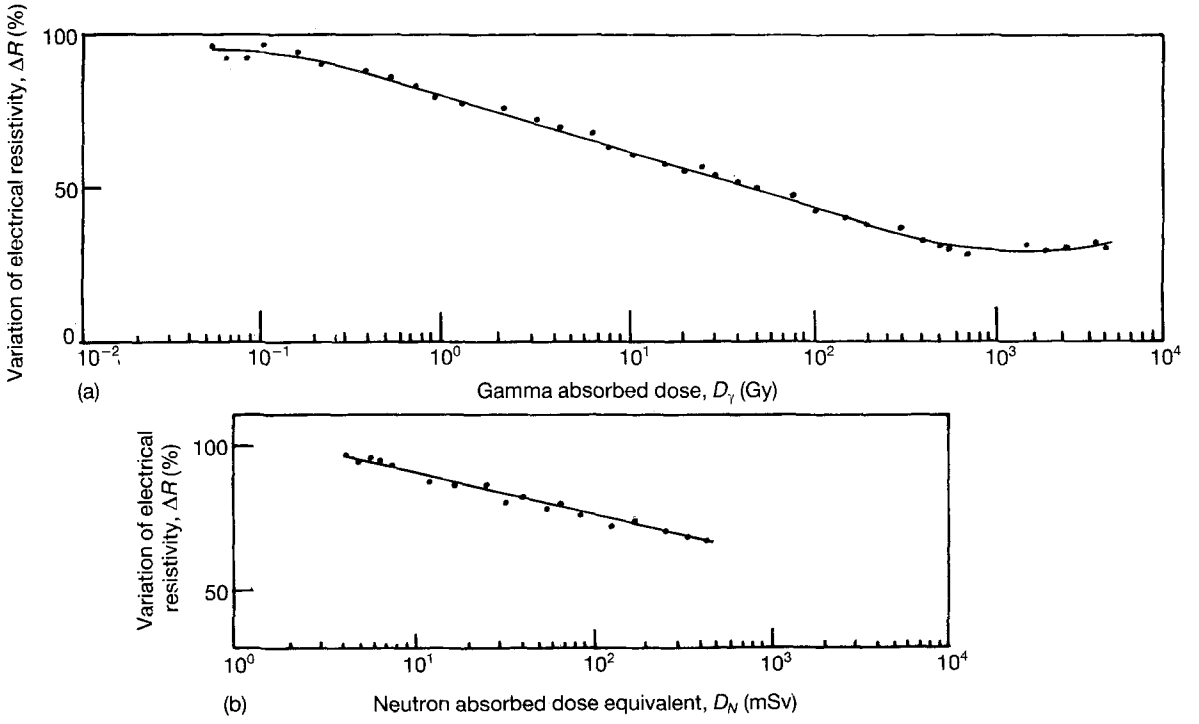

Figure 1 Variation of gelatine resistivity, $R_{\text{irrad.}}/R_{\text{blank}}$ (%), as a function of (a) gamma absorbed dose, and (b) neutron absorbed dose.

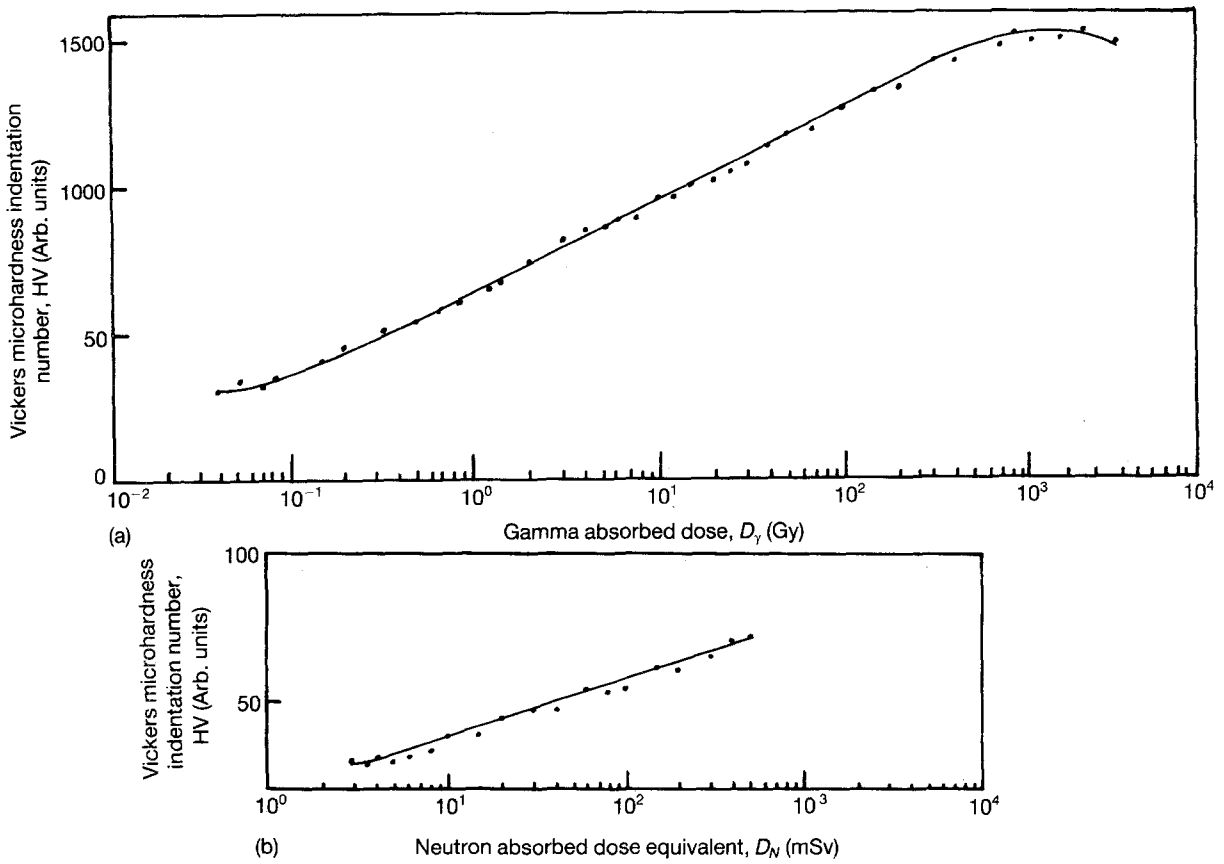

Figure 2 Variation of gelatine Vickers microhardness indentation with (a) gamma absorbed dose, and (b) neutron absorbed dose.

for gamma absorbed dose in Gy, and

$$(\mathrm{RPL})_{385} = 0.06113D_{\mathrm{N}} + 26.83 \quad (6a)$$

$$(\mathrm{RPL})_{495} = 0.0842D_{\mathrm{N}} + 31.84 \quad (6b)$$

for neutron absorbed dose equivalent in mSv, with a fit better than $\pm$ 5%–8%, and where $(\mathrm{RPL})_{385}$ and $(\mathrm{RPL})_{495}$ are, respectively, the luminescence emission intensities at 385 and 495 nm bands.

In analogy between the amorphous structure and gelatine, the gamma radiation-induced effects produce ionization of the gelatine electronic and atomic network bulky structure into cations and anions [11–14]. These charge carriers induced by the effect of radiation, are trapped through the network structure of gelatine by pre-existing flows to form defect centres, i.e. either recombine with the positive-charge holes and/or produce a secondary electron cascade by knock-on collision with bound electrons. These secondary electrons continue to migrate through the matrix structure of the gelatine, producing additional ionization of the bound electrons by Coulomb interaction, and finally they remain at a trap centre at the defect lattices and/or recombine with the positive hole in the network. Furthermore, according to the present predicted results, the damage effect induced by gamma radiation revealed an increase in the Vickers microhardness indentation, because the indentated diagonal diameter decreases with increasing irradiation absorbed dose. Moreover, HV is dependent on the Vickers load, F, and the diagonal diameter, as shown by the relation $HF = 1.8(F/d^2)$. Thus, as the irradiation dose was increased gradually, the elasticity of the gelatine matrix structure was gradually transferred into a more solidified, condensed and compact material. The diamond of the Vickers indentation cannot easily produce diagonal scratches in the network of gelatine. Furthermore, the rate of decrease of the electrical resistivity with increasing gelatine gamma absorbed dose induces a greater bound Coulomb force between the different molecules in the defect side of the gelatine network. In addition, there is a consequent decrease of the charge carriers mobility to the conduction side under the thermal heat induced by the effect of the electric field applied to the irradiated samples. Excitation of the colour centre by ultraviolet light at 265 nm produced increased luminescence emission at wavelength bands 385 and 495 nm with increasing gamma and thermal neutron irradiation doses. This may be ascribed to the increased cloud density of the trapped charge carriers at traps sited in the defect side of the lattice.

In the same flow, interaction of the thermal neutron radiation with the gelatine network, induced thermal spikes, destroyed bonds and deficient regions [15]. As a result, the internal stresses and configuration changes in the network phase produce an increased rate of microhardness indentation and hence a decrease of the electrical resistivity of the material. Therefore, it could be stated that the ionic and electronic structure of the network will generally produce a deep potential well for the newly formed excess carriers which will consequently occupy bound states, Because they are unable to move without altering the positions of the surrounding atoms, forming polarons. Furthermore, the potential well resulting from the

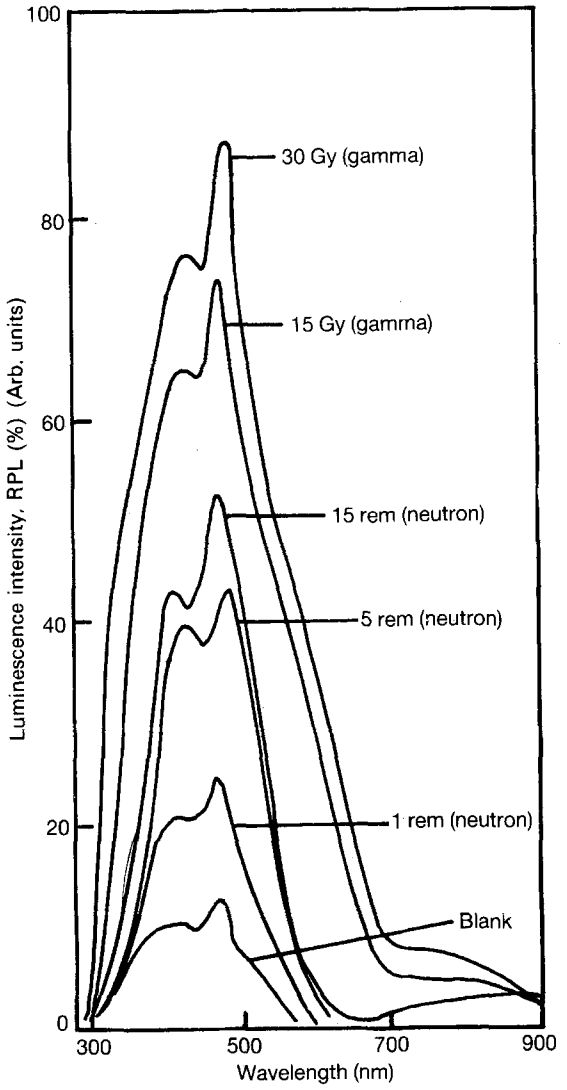

Figure 3 Gamma- and neutron-induced radiophotoluminescence emission spectra, RPL.

carrier-induced displacement, acts as a trap for the carrier itself. Therefore, a pronounced decrease of electrical resistivity and an increase of Vickers microhardness indentation as well as the luminescence emission intensity (RPL) with increasing gelatine equivalent absorbed dose of thermal neutron is expected. Gelatine is found to be more sensitive to gamma radiation. This was attributed to the lower neutron-induced sensitivity with the gelatine when compared to that induced by an equivalent absorbed gamma dose. Table I gives the different experimental values of neutron- and gamma-detected sensitivity applied to the three different measuring techniques. The significant changes may be attributed to the changing techniques of gamma and neutron interaction with the bulky network structure. However, the biologically

TABLE I Relative induced sensitivity of radiation effects on gelatine

Measuring procedure	Equivalent absorbed dose[a] (Gy)	Relative induced sensitivity, S_N/S_γ [b]
Electrical resistivity	0.1	0.95
	0.3	0.97
Microhardness indentation	0.1	0.83
	0.3	0.81
RPL	0.1	0.94
	0.3	0.93

[a] 0.1 Gy γ absorbed dose ≈ 100 mSv equivalent neutron absorbed dose.

[b] Relative induced sensitivity of neutron radiation to equivalent gamma radiation.

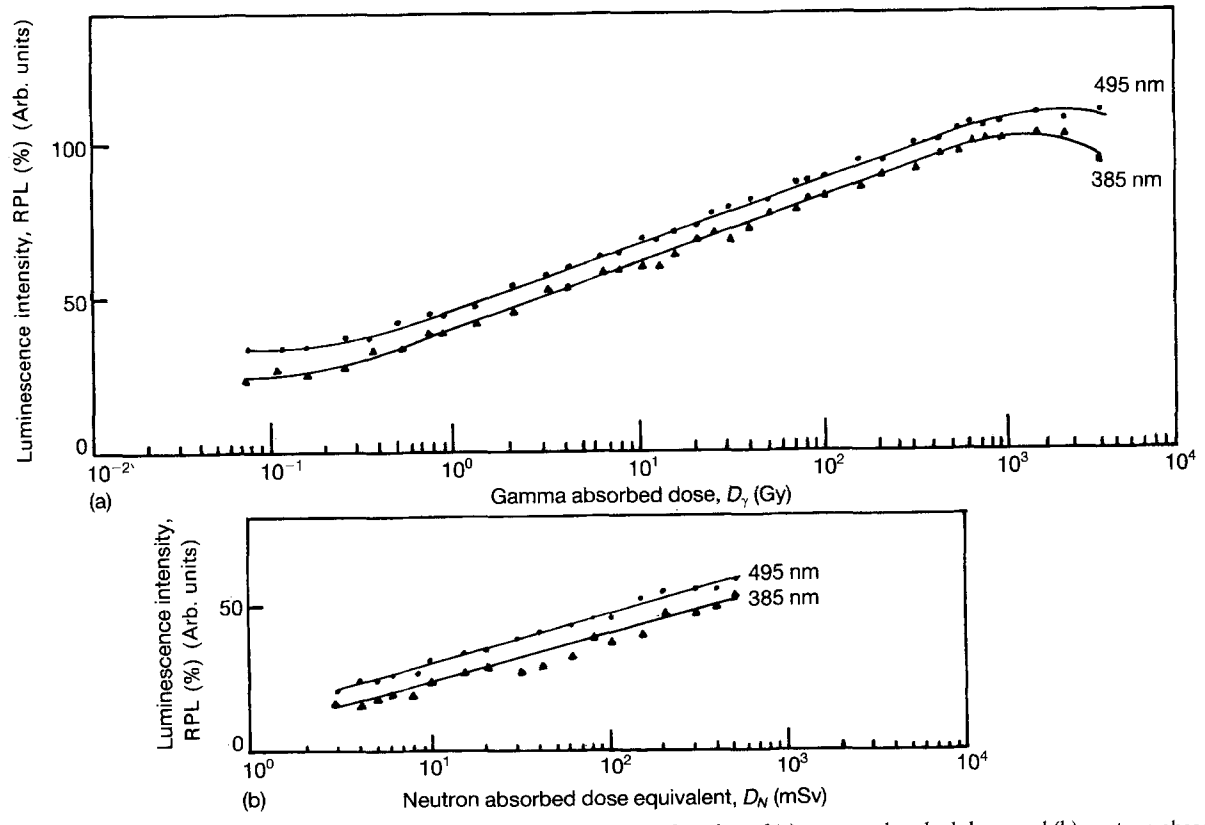

Figure 4 Radiophotoluminescence emission intensity, RPL, of gelatine as a function of (a) gamma absorbed dose, and (b) neutron absorbed dose.

TABLE II Interlaboratory comparison between the sensitivity of gelatine with other data noted in the literature [17–24]

Detector type	Measuring procedure and reference[a]	Response to radiation	Dose sensitivity range	Minimum detectability response	Remarks[a]
Gelatine (present work)	RPL EC H	Gamma thermal neutron	0.1–10^3 Gy 40–400 mSv	0.1 Gy 40 mSv	Present study
Sand	TL [17] EC [19]	Gamma	1–10^4 Gy 10–10^5 Gy	1 Gy 10 Gy	DT NDT
Quartz	TL [18] EC [19]	Gamma	0.7–10^4 Gy 12–10^5 Gy	0.7 Gy 12 Gy	DT NDT
Thin film [20]	TL RPL EC O.D. Cal.	Gamma "General review on the physical behaviour of different detectors type"	10–10^8 Gy	10 Gy	DT NDT NDT NDT NDT
Perspexs [21]	RPL EC H	Gamma	3–8 × 10^3 Gy 0.8–10^4 Gy 1–10^4 Gy	3 Gy 0.8 Gy 1 Gy	NDT NDT NDT
Mixed alkali	EC	Fast neutron	8–700 mSv	8 mSv	NDT
Silicate glass [22, 23]	RI	Fast neutron	8–700 mSv	8 mSv	NDT
Lithium borosilicate glass [24]	RI D H	Fission neutron	7–7000 mSv	7 mSv	NDT

[a] RPL, radiophotoluminescence; TL, thermoluminescence; EC, electrical conductivity; D, density; H, hardness; RI, refractive index; OD, optical density; DT, destructive testing "erase the radiation information"; NDT, non-destructive testing "keep the radiation information".

induced effects of thermal neutrons on the human-tissue organs is mostly considered to be significantly twice that due to an equivalent dose of gamma radiation.

Gamma and thermal neutron effects on the gelatine network induce changes in the physical properties. These changes were quantitatively determined using three different non-destructive measurement procedures. The typical range of response to gamma radiation is extended in the range 10^{-2}–10^3 Gy, and to fission neutron from 7–7000 mSv. The minimum detectable sensitivity of the gelatine was experimentally found to be 0.01 Gy gamma radiation and 7 mSv for thermal neutrons. The present data show that (i) gelatine is more sensitive to gamma radiation, and (ii) there is a lower sensitivity to thermal neutrons, when compared with other literature reviewed [7–24] as noted in Table II.

The errors involved in the determination of gamma and thermal neutron absorbed doses using the three different measuring procedures under investigation arise from (1) statistical error due to short- and long-term electronic stability of the measuring devices [16], (2) statistical errors in the calibration of the gamma and thermal neutron radiation fields, (3) analytical errors in the fitted data, (4) non-uniformity of sample thickness, and (5) the effect of laboratory ambient conditions on the samples before, during and after irradiation.

The statistical errors arose from electronic non-stability daily "as short-term stability" and their average mean "as long-term stability" over 6 weeks. In addition, samples of gelatine detector were subjected to different room-temperature conditions between 22 and 40 °C at different seasons.

Fig. 5 shows the relation between the overall reproducible accuracy, σ_{all}, with D and D_N. The data showed that σ_{all} depend on D_γ and D_N.

4. Conclusions

The beneficial utilization of gelatine as gamma and neutron radiation sensitivity dosimeters is found to fulfil the following criteria.

1. Remarkable effects of gamma and neutron radiation were induced on the physical behaviour of the material. These induced physical changes were found to depend on the induced changes of material hardness, resistivity and, in addition, the increased probability of the colour centres by the effect of increased gamma and neutron absorbed doses. These induced physical parameters are easily and simply measured in the field of practice.

2. More stable, induced physical changes, were practically confirmed by the stability of the fading effect after storage for intervals of 1 week under laboratory ambient conditions, post-sample irradiation. Table III gives the experimentally determined data on fading effects.

3. The high induced gamma and neutron detection sensitivity extended over the range 10^{-2}–10^2 Gy for gamma radiation and 4–400 mSv for thermal neutron equivalent doses, with reproducible accuracy over the range 8%–15%.

4. The advantages of these techniques are their simplicity and direct measurements, inexpensive and

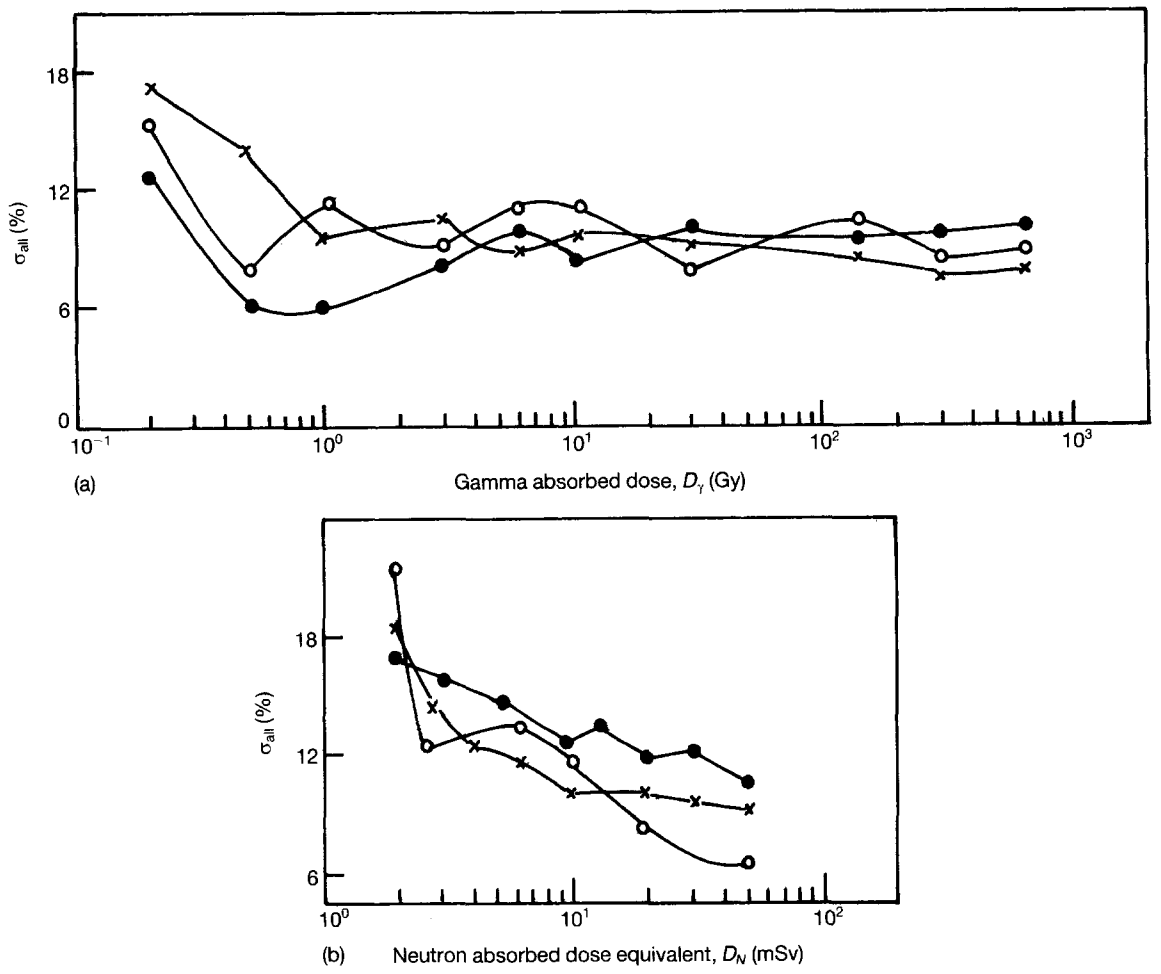

Figure 5 (a) Gamma absorbed dose; (b) Neutron absorbed dose. (×) Microhardness, (○) RPL, (●) Resistivity.

TABLE III Stability of radiation-induced effects on gelatine

Measuring procedure	Type of radiation	Fading[a], F_{av}, effects at different storage periods post-irradiation (days)				
		1	3	6	15	21
Electrical resistivity	Gamma	0.92 ± 0.007	0.88 ± 0.110	0.79 ± 0.120	0.78 ± 0.100	0.81 ± 0.122
	Neutron	0.94 ± 0.132	0.89 ± 0.116	0.81 ± 0.122	0.80 ± 0.096	0.79 ± 0.111
Microhardness	Gamma	0.89 ± 0.134	0.84 ± 0.109	0.76 ± 0.114	0.78 ± 0.109	0.77 ± 0.123
	Neutron	0.91 ± 0.100	0.83 ± 0.116	0.78 ± 0.117	0.79 ± 0.095	0.80 ± 0.105
RPL	Gamma	0.91 ± 0.118	0.84 ± 0.094	0.78 ± 0.117	0.80 ± 0.120	0.81 ± 0.105
	Neutron	0.89 ± 0.142	0.86 ± 0.103	0.81 ± 0.105	0.79 ± 0.119	0.76 ± 0.106

The stated error is the average mean of the total analytical error stated in the text and statistical error arising with F_{av}.
[a] Each predicted value of fading, F_{av}, is the average of five measured samples.

even gelatine is available in commercial sheets at low cost, and also of tissue-equivalent composition. Gelatine can also be beneficially used as a neutron and gamma detector in the low- and protection-radiation level.

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