CR-39 as a gamma dosimeter: dielectric and infrared studies

M. EL-SHAHAWY, A. HUSSEIN

Department of Physics, Faculty of Science, Almonofeya University, Shebeen Al-Koom, Egypt

A. TAWANSI

Department of Physics, Faculty of Science, Mansoura University, Almansoura 35516, Egypt

The relative permittivity, ε' , and dielectric loss factor, tan δ , were measured in the frequency range 50–10⁵ Hz for the solid state track detector CR-39 irradiated with γ -rays (5–100 kGy). The variation of ε' and tan δ with γ -dose have been interpreted in terms of structural changes as emphasized from infrared spectra. A mathematical model has been introduced describing the use of CR-39 as a dielectric dosimeter.

1. Introduction

The importance of CR-39 (allyl diglucol carbonate) arises from its use as a solid state track detector. Since its introduction [1], intense efforts have been made to confirm its properties [2–4] and to develop its use in numerous fields of application [5–7]. This plastic is thermoset, cross-linked, amorphous and very sensitive to heavy-ion damage. In addition to the general thermoset characteristics, CR-39 is a high grade optical plastic whose refraction index is just slightly less than that of crown glass. In fact, it is used in making lightweight eyeglass lenses.

The CR-39 monomer has the following structure

$$\begin{array}{c} O \\ CH_2 - CH_2 - O - \begin{array}{c} O \\ CH_2 - CH_2 - O - \begin{array}{c} O \\ CH_2 - CH_2 - O - \begin{array}{c} CH_2 - O - CH_2 - CH = CH_2 \end{array} \\ O \\ CH_2 - CH_2 - O - \begin{array}{c} C \\ CH_2 - CH_2 - O - CH_2 - CH = CH_2 \end{array}$$

Because of the presence of two allyl groups, the monomer cannot only polymerize but also cross-links to form a three-dimensional network structure.

Application of this polymer in the field of dosimetry was investigated by measurements of latent damage trails using an etching technique [8, 9]. To our knowledge, no work has been reported regarding measurements of its dielectric properties. In the present investigation the effect of gamma rays on the dielectric permittivity, ε' , and dielectric loss factor, tan δ , of CR-39 is explained in terms of structural changes induced by irradiation. Also, the use of these measurements in the field of dosimetry is presented.

2. Experimental procedure

CR-39 homopolymer in the form of sheets of uniform thickness (3.0 mm) (Homalite, Wilmington, DE, USA) were used for dielectric measurements. For infrared measurements, thin sheets (250μ m) of CR-39 homopolymer (Pershore Mouldings Ltd, UK) were used.

The relative permittivity, ε' , and dielectric loss factor, tan δ , of the samples before and after being irradiated up to 100 kGy were measured using NF Dekameter type DK-05 having a frequency range of $30-10^5$ Hz at room temperature. The accuracy of the instrument for ε' was $\pm 0.3\%-0.5\%$ and for tan δ was $\pm 3\%-5\%$.

A 60 Co gamma cell type J-6500 at the National Centre for Radiation and Technology, Cairo, was used for irradiating the samples at a dose rate of 1.75 Gy s⁻¹ at room temperature in air.

The infrared spectra were obtained using a Fourier transform infrared spectrometer (Perkin Elmer 1430) in the wavenumber range of $200-4000 \text{ cm}^{-1}$.

3. Results and discussion 3.1. Infrared spectra

Fig. 1 shows the plots of the infrared absorption spectra for CR-39 in the spectral ranges $3700-1800 \text{ cm}^{-1}$ and $800-500 \text{ cm}^{-1}$ where spectral features are observed. In the region of $1800-800 \text{ cm}^{-1}$, the spectra of all CR-39 samples are quite similar. The observed features can be summarized as follows.

(a) For the nonirradiated sample (Curve a), the intense absorption bands at 2910 and 2750 cm⁻¹ can be assigned to saturated antisymmetric and symmetric C-H stretching vibration, respectively. The absorption band which appears at their shorter wavelength side ($\sim 3030 \text{ cm}^{-1}$) can be assigned to unsaturated = C—H stretching vibration [10]. It can be seen (Fig. 1) that increasing the γ -dose causes a gradual decrease in the intensity of this latter band which completely disappeared with irradiation dose of 100 k Gy. This behaviour indicates a cross-linking process, because it is usually possible to follow the course of cross-linking by monitoring the residual degree of unsaturation using infrared spectroscopy.

(b) It is obvious from Fig. 1 that irradiation of CR-39 with γ -rays causes a sharp increase in the intensity of absorbance at about 2330 and 650 cm⁻¹.

The most radiation sensitive group in CR-39 is the carbonate group which decomposes into CO_2 and CO on irradiation and is responsible for CR-39 track recording properties. Carbon dioxide absorbs at 2350 and 667 cm⁻¹, and the carbon monoxide band extends from about 2040–2220 cm⁻¹ [11, 12]. The presence of these molecules trapped in the glassy CR-39 network could be responsible for the increase in the intensity of the observed bands at about 2330 and 650 cm⁻¹. The absorbance of unirradiated CR-39 in this region may be attributed to the presence of some carbon dioxide even in the unirradiated sample as a by-product of decomposition of peroxydicarbonate which was used as the polymerization initiator.

3.2. Relative permittivity, ε'

Fig. 2 shows the relative permittivity of CR-39 as a function of frequency for different γ -doses up to 15 kGy. At all γ -doses the relative permittivity decreases with increasing frequency as a result of the



Figure 1 Infrared spectra of CR-39 obtained before and after γ -irradiation: (a) 0, (b) 5, (c) 25, (d) 100 kGy.



Figure 2 Relative permittivity as a function of frequency for different γ -doses-: (\bullet) 0, (\triangle) 5 kGy, (\times) 10 kGy, (\bigcirc) 15 kGy.

dielectric dispersion which arises from the lag of CR-39 macromolecules behind the variation of the applied field. On the other hand, ε' decreases with increasing γ -dose to 15 kGy over the entire range of frequencies studied.

Increasing the γ -dose beyond 15 kGy and up to 100 kGy shows a marked increase in ϵ' especially at low-frequency values as depicted in Fig. 3. The variation of the relative permittivity with gamma irradiation could be explained as follows.

(a) At low radiation doses, further cross-linking of the resin occurs and this restricts dipolar orientation, thereby reducing the relative permittivity. The occurrence of such a cross-linking process is indicated by the decrease in the intensity of the unsaturated = C—H stretching vibration band at 3030 cm⁻¹ as discussed above.

(b) The high values of relative permittivity at high doses may be attributed to the formation of the easily orientable low molecular mass (oxygen-containing molecules) due to the oxidative degradation.

Fig. 4a depicts a linear relation between ε' and dose value, *D*, up to 15 kGy at different frequencies. The highest slope is that at 100 Hz which can be represented numerically by

$$\varepsilon' = 5.466 - 0.037 D$$
 (1)

where D is in kGy. However, for the frequency 50 Hz, the relationship is linear between log D and ε' up to 25 kGy (Fig. 4b).



Figure 3 Relative permittivity as a function of frequency for different γ -doses:-(×) 25 kGy, (**■**) 30 kGy, (∇) 40 kGy, (**▼**) 60 kGy, (\bigcirc) 100 kGy.



Figure 4 Relative permittivity dependence of CR-39 on gamma doses (0–15 kGy) at different frequencies; (a) (\blacktriangle) 10⁵ Hz, (\bigtriangleup) 60 kHz, (\Box 30 kHz, (\times) 10 kHz, (\blacklozenge) 100 Hz; (b) (\bigcirc) 50 Hz.



Figure 5 Relative permittivity dependence on gamma doses (15–100 kGy) at 10 kHz; (\bigcirc) theoretical, (×) experimental, for CR-39.

In the higher γ -dose region (15–100 kGy) the relation between the relative permittivity and dose value is represented by a polynomial function as shown in Fig. 5 for ϵ' measured at 10 kHz. The empirical equation satisfying this relation is given by

$$\varepsilon' = 3.9174 + 1.456 \times 10^{-2} D$$

- 0.078 × 10⁻³D² (2)

An alternative relation which is more satisfactory but covers only the range 25-100 kGy, is shown in Fig. 6.



Figure 6 An alternative representation of ε' versus gamma doses at 10 kHz: (\bigcirc) theoretical and (X) experimental, for CR-39.



Figure 7 Dielectric loss factor, tan δ , versus frequency for CR-39 irradiated by different γ -doses: (\Box) 0, (\bullet) 5 kGy, (\triangle) 10 kGy, (\times) 15 kGy, (\blacksquare) 25 kGy, (\bigtriangledown) 30 kGy, (\blacktriangledown) 60 kGy, (\bigcirc) 100 kGy.



Figure 8 Tan δ plotted against γ -doses at different frequencies for CR-39: (\times) 50 Hz, (\Box) 100 Hz, (Δ) 200 Hz, (\bigcirc) 1 kHz, (\odot) 3 kHz.

This is represented by

$$\varepsilon' = 4.6976 - \frac{12.152}{D}$$
 (3)

3.3. Dielectric loss factor, $tan \delta$

Fig. 7 shows the dielectric loss factor, tan δ , of CR-39 versus frequency for different γ -doses. As described in the trend of the relative permittivity, tan δ decreases for low γ -doses, but the dose at which it starts to increase is higher than that exhibited for the relative permittivity. This may be explained by the need of more bond dissociation to enhance segmental and group mobility.

The decrease of the dielectric loss factor, $\tan \delta$, with irradiation in the low γ -dose region continues up to 30 kGy (Fig. 8). It is clear that $\tan \delta$ decreases linearly with γ -doses, for frequencies down to 200 Hz at which the highest slope is attained. This is represented by the equation

$$\tan \delta = 0.1707 - 0.315 \times 10^{-2} D \qquad (4)$$

where D is in kGy.

References

- B. G. CARTWRIGHT, E. K. SHIRK and P. B. PRICE, Nucl. Inst. Meth. 153 (1978) 457.
- 2. R. H. CASSOU and E. V. BENTON, Nucl. Track Det. 2 (1978) 173.
- T. DOKE, T. HAYASHI, I. MATSUMI, M. MATSUSHITA, H. TAWARA, K. KAWAGOE, K. NAGANO, S. NAKA-MURA, M. NOZAKI, S. ORITO and K. OGURA, in "Proceedings of the 12th International Conference on SSTDs". Acapulco (1983) p. 228.
- P. H. FOWLER, S. AMIN, V. M. CLAPHAM and D. L. HENSHAW, in "Proceedings of the 10th International Conference on Solid State Nuclear Track Detectors", Lyon (1979) 239.
- D. L. HENSHAW, A. P. FEWS and D. J. WEBSTER, *Phys. Med. Biol.* 24 (1979) 1227.
- S. A. NAJJAR, R. K. BULL and S. A. DURRANI, Nucl. Tracks 3 (1979) 169.
- P. B. PRICE, E. K. SHIRK, K. KINOSHITA and G. TARLE, in "Proceedings of the 16th International Conference on Cosmic rays", Kyoto (1979) p. 80.
- 8. M. FUJII, J. NISHIMURA and T. KOBAYASHI, Nucl. Inst. Meth. 226 (1984) 496.
- 9. D. L. HENSHAW, N. GRIFFITHS, O. A. L. LANDEN and E. V. BENTON, *ibid.* **180** (1981) 65.
- B. GEORGE and P. McINTYRE, in "Infrared Spectroscopy", edited by D. J. MOWTHORPE (Wiley, Chichester, 1987) pp. 312-20.
- 11. C. N. BANWELL, "Fundamentals of Molecular Spectroscopy", 3rd Edn (McGraw-Hill, New Delhi, 1983) pp. 91, 115.
- K. NAKANISHI and P. H. SOLOMEN, "Infrared Absorption Spectroscopy", 2nd Edn (Holden-Day, San Francisco, London, 1977) p. 23.

Received 24 July 1991 and accepted 11 March 1992