

# Ultraviolet and Infrared Spectral Analysis of Irradiated Polyethylene Films: Correlation and Possible Application for Large-Dose Radiation Dosimetry

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**ABSTRACT:** A detailed study was performed to develop the dosimetric characteristics of commercial low-density polyethylene film (LDPE), which is, by far, the most commonly used plastic for food and for many other-purpose packaging film, to be used as a film dosimeter for large-dose  $\gamma$ -radiation dosimetry. The useful dose range extends up to 880 kGy. Correlations were established between the absorbed dose of  $\gamma$ -radiation and the radiation-induced changes in LDPE measured using Fourier transform infrared (FTIR) and ultraviolet (UV) spectrophotometry. The results showed a significant dependence of the response on the selected readout tool of measurements whether FTIR (at  $1716\text{ cm}^{-1}$ ) or UV (at 220 and 270 nm) as well as on the quantity used for calculation. The radiation-chemical yield of the ketonic carbonyl group produced in irradiated LDPE film was found to be  $0.7\ \mu\text{mol}/\text{J}$ . The assessment of the random uncertainty associated with the measurement of the dose response and the effect of relative humidity during irradiation on the dosimeter performance as well as the postirradiation stability at different storage conditions are discussed. © 1998 John Wiley & Sons, Inc. *J Appl Polym Sci* **67**: 1837–1851, 1998

**Key words:** ultraviolet spectral analysis; infrared spectral analysis; polyethylene films; large-dose radiation dosimetry

## INTRODUCTION

Polyethylene, being one of the simplest polymers, has been the subject of a large number of investigations.<sup>1–6</sup> Irradiation of polymers induces crosslinking and degradation; in the case of polyethylene, crosslinking is the predominate effect upon irradiation in the absence of oxygen, while oxidative degradation and chain scission occurs also in the presence of oxygen.<sup>7</sup> Polyethylene gives a number of products on radiation-induced oxidation including  $\text{H}_2\text{O}$ ,  $\text{CO}_2$ , peroxides, and carbonyl and carboxyl compounds.<sup>8,9</sup>

Clear, transparent plastic films are available in large batches and are often used for routine high-dose monitoring. The radiation-induced response is coloration or fading of color in different parts of the ultraviolet (UV), visible, or infrared (IR) spectrum, usually analyzed by a spectrophotometer or densitometer.<sup>10</sup> The radiation-induced chromophores consist chiefly of side-chain unsaturation (e.g.,  $\begin{array}{c} \diagdown \\ \text{C}=\text{O} \\ \diagup \end{array}$  groups in polymethacrylate, polycarbonate, polyethylene, and polypropylene) and new main-chain unsaturation polyene groups (e.g.,  $-\text{C}=\text{C}-$  in vinyls, polyolefins, etc.).<sup>9,11</sup>

In polymeric film dosimeters, the relationship between the radiation-induced signal and the dose depends on the absorbed dose of ionizing radiation by the dosimeter and may also depend on

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the dose rate or fractionation of the dose,<sup>12</sup> on the temperature during the irradiation and handling, on the presence or absence of oxygen in the surrounding atmosphere or in the dosimeter,<sup>13,14</sup> and on the ambient humidity or, rather, on the amount of water in the dosimeter.<sup>15–21</sup>

There are only a few reports that were concerned with the use of LDPE films for radiation dosimetry. Wenxiu et al. (1980)<sup>22</sup> described briefly the dose-response functions of LDPE film in the dose range from 10 to 1000 kGy using UV absorption spectra. Infrared analysis of irradiated polyethylene films has been used for dosimetry in the dose range 100–1000 kGy, typically by measuring the absorption bands of the carbonyl groups at 1724  $\text{cm}^{-1}$  and the *trans*-vinylene groups at 943  $\text{cm}^{-1}$ .<sup>23,24</sup> These results deal only with the dose-response function of LDPE film; however, there are no available data concerned with the assessment of the uncertainty associated with the dose measurement or the environmental effects during irradiation on the response, e.g., temperature and humidity during irradiation.

The objective of the present work was to investigate the possible use of LDPE film in high-dose  $\gamma$ -radiation dosimetry using UV and Fourier transform infrared (FTIR) spectrophotometry. The overall uncertainty of the dose measurement, the radiation-chemical yield, the effect of relative humidity during irradiation on the film response, and the postirradiation stability of films under different storage conditions were also investigated.

## EXPERIMENTAL

### The Film and Its Analysis

The film used in this investigation was low-density polyethylene (LDPE) film (commercial product of Medical Packing Co., Egypt) of density 0.915, fusion index above 350°C, crystallinity 38%, melt flow index (MI) about 1.25 g/10 min, and thickness  $75 \pm 5 \mu\text{m}$ .

A Uvikon 860 spectrophotometer was used for scanning the absorption spectra and measuring the optical density at  $\lambda_{\text{max}}$  of the different film dosimeters. A Mattson 1000 Fourier transform infrared (FTIR) spectrometer (Unicam) was used for measuring and scanning the infrared absorption spectra at resolution of 4  $\text{cm}^{-1}$ . High signal-to-noise spectra were obtained by the collection of

100 scans for each sample. The resultant digitized spectra were stored for further data processing.

### Irradiation Procedure

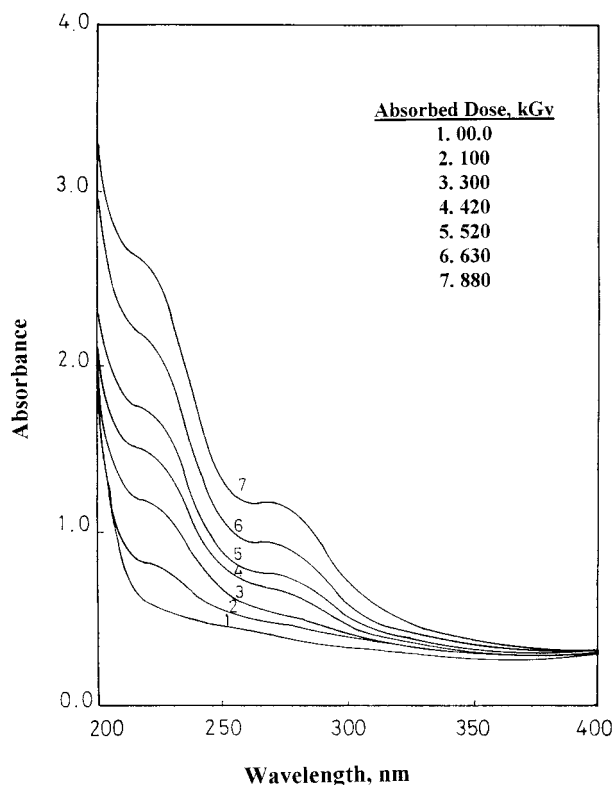
Irradiation was carried out in a <sup>60</sup>Co gamma chamber 4000 A (product of India). A dose rate of 3.76 kGy/h was used as checked by Fricke dosimetry.<sup>25</sup> To establish a good reproducibility and accuracy of the absorbed radiation doses at the gamma cell, a specially designed rack made from polystyrene was used to hold the film dosimeter at the central spatial position of the sample chamber. The design of the rack ensures that all irradiated film dosimeters are exposed to the same radiation field in the most homogeneous region of the irradiation chamber, i.e., when the polystyrene rack holding the film dosimeters is fixed inside the irradiation chamber, the film dosimeters are on an isodose cylindrical surface concentric with the cylindrical irradiation chamber.<sup>26</sup>

Before use, the LDPE films were conditioned in a glove box in air at 33% RH and at a temperature of  $25 \pm 2^\circ\text{C}$ . For irradiation, five LDPE films at each dose were grouped together and sandwiched between two poly(methyl methacrylate) (PMMA) plates of 3 mm thickness to maintain electronic equilibrium. These plates were enclosed in and sealed by aluminum polyethylene laminate foil. This procedure is usually performed in the glove box used for preconditioning, and, thus, a defined humidity condition could be maintained even during irradiation. These covered plates, containing the film dosimeters, were then irradiated at the cavity of the polystyrene rack in the gamma cell.

## RESULTS AND DISCUSSION

### Absorption Spectra

The UV spectrophotometric scan in the wavelength range between 200 and 400 nm of LDPE films was recorded before and after  $\gamma$ -irradiation for different doses and is shown in Figure 1. It can be seen that the absorption spectrum of unirradiated LDPE film (Fig. 1, curve 1) has no structure. Upon irradiation, two absorption bands at 220 and 270 nm wavelengths were developed. The amplitude of both absorption bands increases, with different sensitivity, with increase of the absorbed dose. The absorption band at 220 nm wavelength is related to the presence of a ketonic car-



**Figure 1** UV absorption spectra of LDPE films unirradiated and irradiated at different absorbed doses.

bonyl group due to the oxidation of polyethylene (PE) upon  $\gamma$ -irradiation in air, while the other absorption band at 270 nm wavelength is indicative of the presence of conjugated double bonds of polyenes.<sup>22</sup>

Structural changes of irradiated LDPE film may also be identified and quantified by Fourier transform infrared (FTIR) spectroscopy. The FTIR spectra of unirradiated and irradiated LDPE films for different absorbed doses are shown in Figure 2. The main obvious spectral change that occurs upon  $\gamma$ -ray irradiation is the appearance of an absorbance band at  $1716\text{ cm}^{-1}$ , which is attributed to the stretching vibration of ketonic carbonyl groups. The intensity of this absorbance band ( $1716\text{ cm}^{-1}$ ) increases with increasing the absorbed dose (see Fig. 2). Also, the effect of  $\gamma$ -ray irradiation on the LDPE film can be seen in the difference spectra obtained by absorbance subtraction of the unirradiated film from those obtained after irradiation to absorbed doses of 420, 520, and 710 kGy (see Fig. 3). In this difference spectrum, absorbance bands above the base line reflect an increase in a particular chemical species, while those bands below the base line

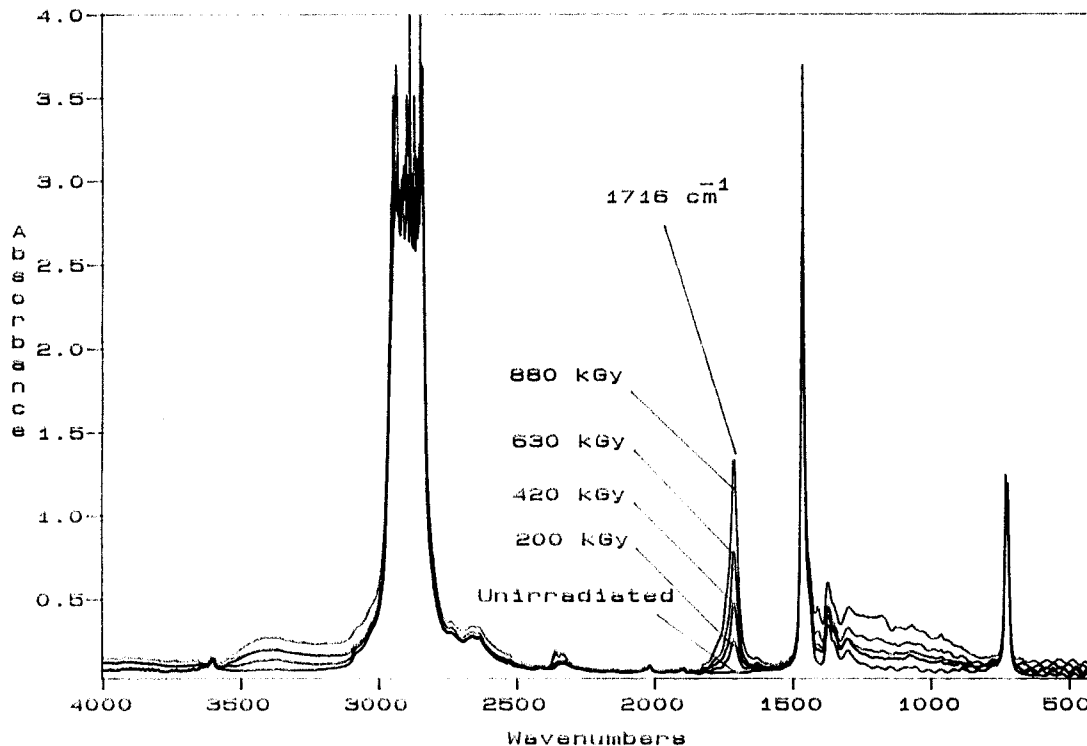
reflect a decrease in an absorption species due to the irradiation process.<sup>2</sup> The increase of the absorption bands at  $1716\text{ cm}^{-1}$  due to the irradiation process is more evident in the difference spectra. Moreover, in the difference spectrum, there are some weak observable spectral changes with irradiation which were also noticed. These spectral changes at wavenumbers 965, 1411, 1373, 1299, and  $1897\text{ cm}^{-1}$  are attributed to a *trans*-vinylene double bond (*trans* R—CH=CH—R'), methylene deformation influenced by an adjacent carbonyl group [RCH<sub>2</sub>—(CO)—CH<sub>2</sub>—R], symmetric methyl deformation, and amorphous and crystallinity absorbance bands, respectively.<sup>2,4,5,27,28</sup>

The FTIR subtracted absorbance values at some selected wavenumbers for irradiated LDPE film (absorbed doses of 60, 100, 200, 300, 420, 520, 710, and 880 kGy) are also presented in Table I. It appears quite clearly, from the obtained results in Figure 3 and Table I, that the  $\gamma$ -ray irradiation caused slightly observable changes at all selected wavenumbers except for  $1716\text{ cm}^{-1}$ , where the intensity of the absorbance increases clearly with increasing of absorbed doses. From the above obtained results, it can be concluded that the spectral change in absorbance at  $1716\text{ cm}^{-1}$  obtained by FTIR spectroscopy for irradiated LDPE film can be successfully used in the radiation dosimetry measurements.

### Response Curves

The response curves of LDPE film obtained by using the UV-spectrophotometric quantities ( $\Delta A\text{ mm}^{-1}$ ) and ( $A_i/A_0$ ) at 220 and 270 nm wavelengths as a function of the absorbed dose are shown in Figure 4, where ( $\Delta A\text{ mm}^{-1}$ ) is the change in absorbance before and after irradiation divided by the thickness of the film, and  $A_0$  and  $A_i$  are the optical densities at 220 or 270 nm wavelengths for the unirradiated and irradiated films, respectively.

It can be seen that the response curves of both ( $\Delta A\text{ mm}^{-1}$ ) and ( $A_i/A_0$ ) obtained at the 270 nm wavelength are linear while those at the 220 nm wavelength consist of two linear parts with an inflection point at an absorbed dose of 200 kGy. This behavior may be explained on the basis of the radiation-induced changes in the film represented by the increase in absorbance at the 220 and 270 nm wavelengths. The radiation-induced oxidation of the film, which was indicated by the increase in absorbance at 220 nm, was found to



**Figure 2** FTIR absorption spectra of LDPE films unirradiated and irradiated at different absorbed doses.

occur chiefly at the surface of the film.<sup>3,22</sup> At low doses, the reaction between oxygen and the film surface proceeds at a certain rate, forming an oxidation product layer on the film surface. The presence of this layer makes the diffusion of oxygen into the film more difficult. Accordingly, at higher doses, the rate of the reaction between the film and oxygen will slow down. On the other hand, the rate of the production of conjugated double-bond polyenes, which was indicated by the increase in absorbance at 270 nm, will proceed without change because it does not depend on the concentration of oxygen in the film. This explains why the response curve at 220 nm shows a change in slope at 200 kGy while that at 270 nm does not. Hence, the change in the slope of the response curves at 200 nm, which is indicated by the inflection point at 200 kGy, is attributed to the decrease in the rate of oxidation of the film at doses higher than 200 kGy.

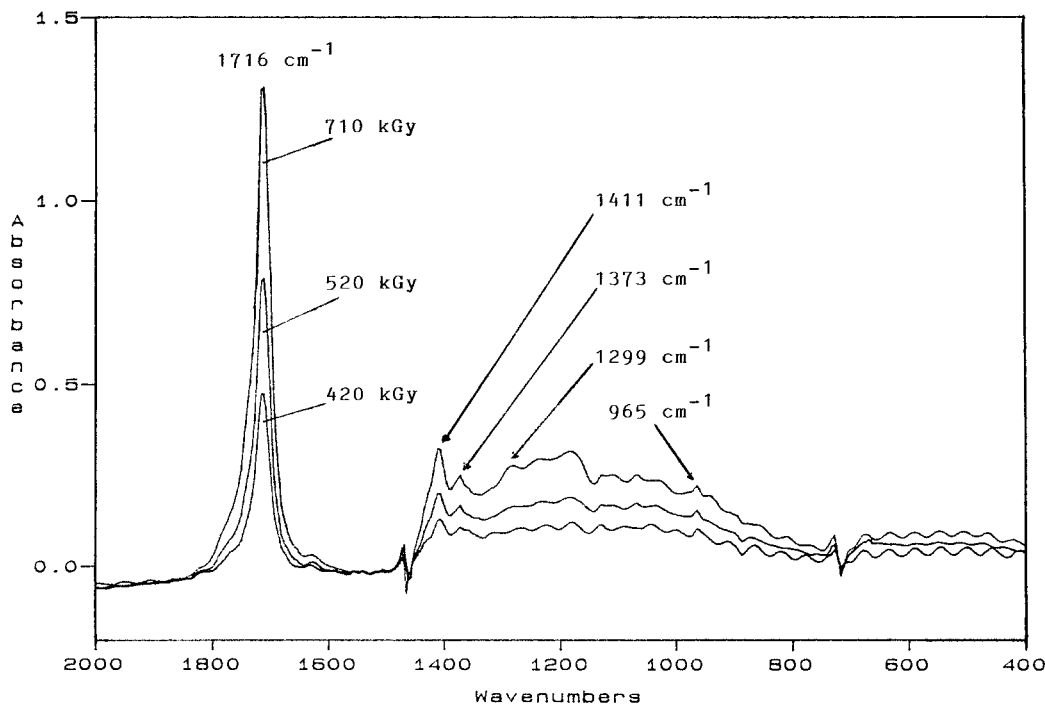
The linear response curves, obtained at both 220 and 270 nm wavelengths, can be represented by the following general equation:

$$y = a + bx \quad (1)$$

where  $y$  is  $(\Delta A \text{ mm}^{-1})$  or  $(A_i/A_0)$  at  $x$  kGy;  $x$ , the absorbed dose in kGy; and  $a$  and  $b$ , constants.

The constants  $a$  and  $b$  and the correlation coefficients ( $r^2$ ) for all response curves are summarized in Table II. From Table II, it can be noticed that the sensitivities, as indicated by the slopes of the second parts of the response curves, obtained with  $(\Delta A \text{ mm}^{-1})_{220}$  and  $(A_i/A_0)_{220}$ , are lower than those of the first parts.

It was previously assumed that the response curves obtained for LDPE film at 220 nm and 270 nm wavelengths are linear, and in the case of the 220 nm wavelength, the response is more sensitive and of minimum errors, as was stated in the literature.<sup>22</sup> However, our results showed that this assumed linearity is only an approximate one, especially when the 220 nm wavelength is used for UV absorbance measurements. To illustrate this more clearly, changes of UV absorbance  $(\Delta A \text{ mm}^{-1})_\lambda$  and the ratio  $(A_i/A_0)_\lambda$  values per unit dose, the  $K$  value, were plotted versus the absorbed dose and are shown in Figure 5  $\{K \text{ value} = [(\Delta A \text{ mm}^{-1}) - a]/\text{dose}$  or  $[(A_i/A_0) - a]/\text{dose}$ , where  $a$  is the intercept of the response curve with the  $Y$  axis}. It was observed that this type of representation shows that no linear relationship was



**Figure 3** FTIR difference spectrum of LDPE films irradiated at different absorbed doses (420, 510, and 710 kGy).

attained in the dose range up to 200 kGy. On the other hand, linear behavior is mostly obtained between  $(\Delta A \text{ mm}^{-1})_{\lambda}$  or  $(A_i/A_0)_{\lambda}$  and the absorbed dose for doses higher than 200 kGy (where a  $K$  value remains nearly constant). Therefore, it may be concluded that using LDPE film for measuring the absorbed dose below 200 kGy must be taken under specific reservation, i.e., measuring absorbed doses in this dose range is not appreciated under our experimental conditions.

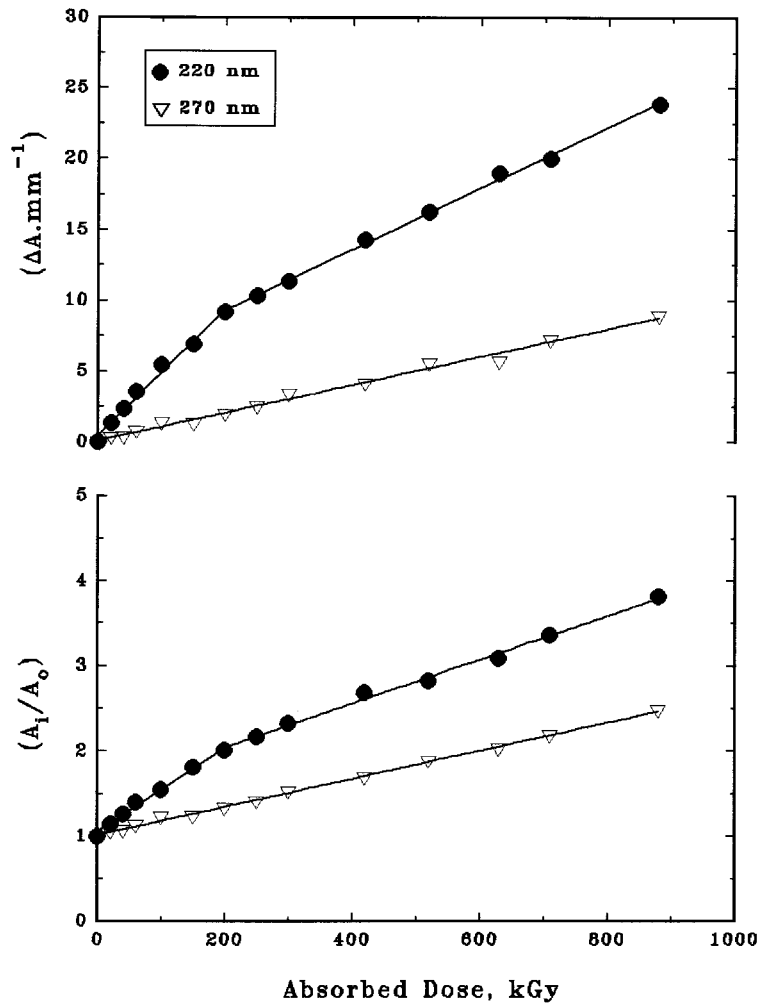
To establish the response functions of LDPE films for  $^{60}\text{Co}$   $\gamma$ -ray irradiation using FTIR spectroscopy, three different calculation quantities were used:

1. Radiation-induced peak absorbance at  $1716 \text{ cm}^{-1}$  divided by the thickness of the LDPE film,  $(\Delta A \text{ mm}^{-1})_{1716}$ .
2. Radiation-induced peak area from  $1585$  to  $1855 \text{ cm}^{-1}$  divided by the thickness of the LDPE film,  $(\Delta A \text{ mm}^{-1})_{\text{area}}$ .
3. The ratio of radiation-induced absorbance at  $1716 \text{ cm}^{-1}$ ,  $(A_i/A_0)_{1716}$ .

Figure 6 shows the change in the above-mentioned spectrometric quantities as a function of absorbed dose. From this figure, it can be seen that the response curve obtained by the spectrometric quantity  $(A_i/A_0)_{1716}$  consists of two linear parts with an inflection point at about 100 kGy,

**Table I** Observed Changes in Peak Absorbances of Irradiated LDPE Films

Frequency ( $\text{cm}^{-1}$ )	Absorbed Dose (kGy)							
	60	100	200	300	420	520	710	880
965	0.026	0.034	0.046	0.072	0.104	0.155	0.224	0.237
1299	0.220	0.210	0.258	0.289	0.324	0.404	0.463	0.503
1373	0.010	0.013	0.053	0.063	0.108	0.168	0.252	0.275
1411	0.017	0.011	0.054	0.077	0.132	0.201	0.324	0.348
1716	0.043	0.061	0.192	0.284	0.475	0.788	1.308	1.392
1897	0.063	0.056	0.074	0.088	0.096	0.098	0.098	0.099



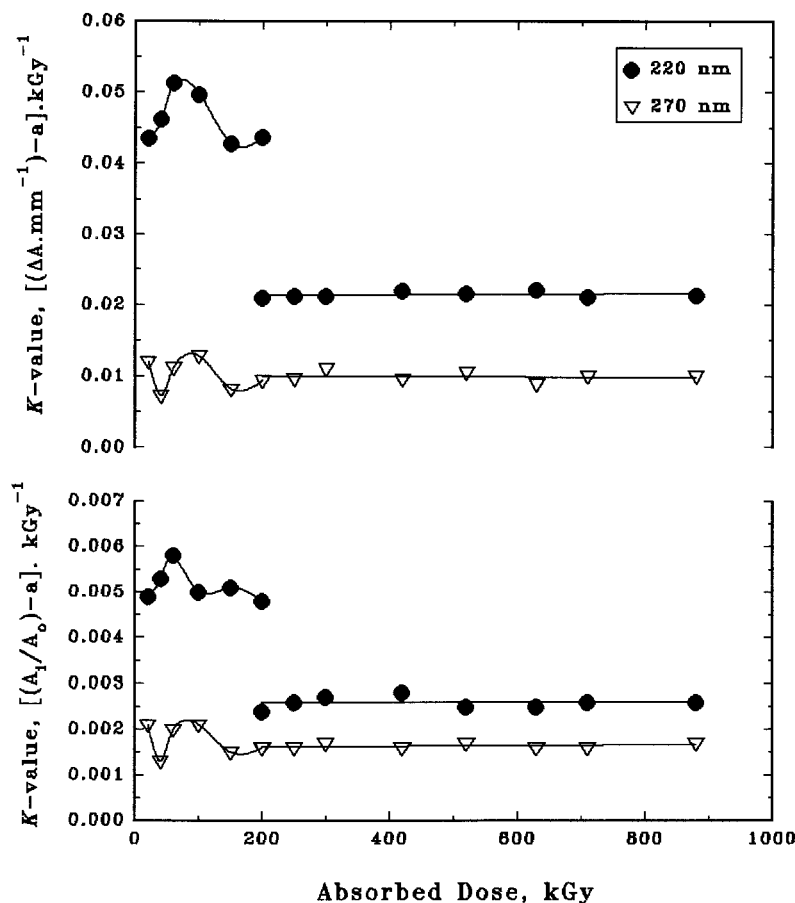
**Figure 4** Change of UV spectrophotometric quantities  $(\Delta A \text{ mm}^{-1})_{\lambda}$  and  $(A_i/A_0)_{\lambda}$  as a function of the absorbed dose ( $^{60}\text{Co}$   $\gamma$ -ray). Dose rate =  $3.76 \text{ kGy h}^{-1}$ . Irradiation temperature =  $35^{\circ}\text{C}$ . Wavelengths of the analysis are indicated.

while those obtained with the quantities  $(\Delta A \text{ mm}^{-1})_{1716}$  and  $(\Delta A \text{ mm}^{-1})_{\text{area}}$  also consist of two linear parts with an inflection point at 200 kGy, as in the case of the UV response curves, but with

higher sensitivity in the second part of the curves (see Fig. 4). The FTIR response curves can also be expressed by the above-mentioned equation [eq. (1)].

**Table II** Constants *a* and *b* and Correlation Coefficients (*r*<sup>2</sup>) of UV Response Curves

Constants	$(\Delta A \text{ mm}^{-1})_{\lambda}$			$(A_i/A_0)_{\lambda}$		
	220 nm		270 nm 0–880 kGy	220 nm		
	1st Part 0–200 kGy	2nd Part 200–880 kGy		1st Part 0–200 kGy	2nd Part 200–880 kGy	270 nm 0–880 kGy
<b>a</b>	0.000	5.040	0.000	1.070	1.525	1.020
<b>b</b>	0.044	0.021	0.0099	0.0047	0.0026	0.0016
<i>r</i> <sup>2</sup>	0.993	0.999	0.995	0.995	0.998	0.999



**Figure 5** Variation of  $K$  value,  $\{[(\Delta A \text{ mm}^{-1})_{\lambda} - a] \text{ kGy}^{-1}\}$  and  $\{[(A_i/A_0)_{\lambda} - a] \text{ kGy}^{-1}\}$ , as a function of the absorbed dose. Wavelengths of the analysis are indicated.

The constants  $a$  and  $b$  and the correlation coefficients ( $r^2$ ) of the FTIR response curves are also given in Table III. From this table, it can be noticed that the sensitivities, as indicated by the slopes of the second parts of the response curves obtained with the quantities  $(\Delta A \text{ mm}^{-1})_{1716}$  and  $(\Delta A \text{ mm}^{-1})_{\text{area}}$ , are higher ( $\approx 2.5$  times) than those of the first parts and this is the opposite case for the UV response curves obtained at 220 and 270 nm wavelengths.

The change of the FTIR spectrometric quantities  $(\Delta A \text{ mm}^{-1})_{1716}$ ,  $(\Delta A \text{ mm}^{-1})_{\text{area}}$ , and  $(A_i/A_0)_{1716}$  per unit of absorbed dose ( $K$  value) were plotted versus the absorbed dose, as in the case of the UV spectrophotometric quantities (see Fig. 7). It can be seen that, in the case of the quantity  $(A_i/A_0)_{1716}$ , a linear response function was obtained only at doses above about 100 kGy. On the other hand, below or above the inflection dose point at about 200 kGy, there are approximate linear relationships between the absorbed dose and both  $(\Delta A \text{ mm}^{-1})_{\text{area}}$  and  $(\Delta A \text{ mm}^{-1})_{1716}$ . In

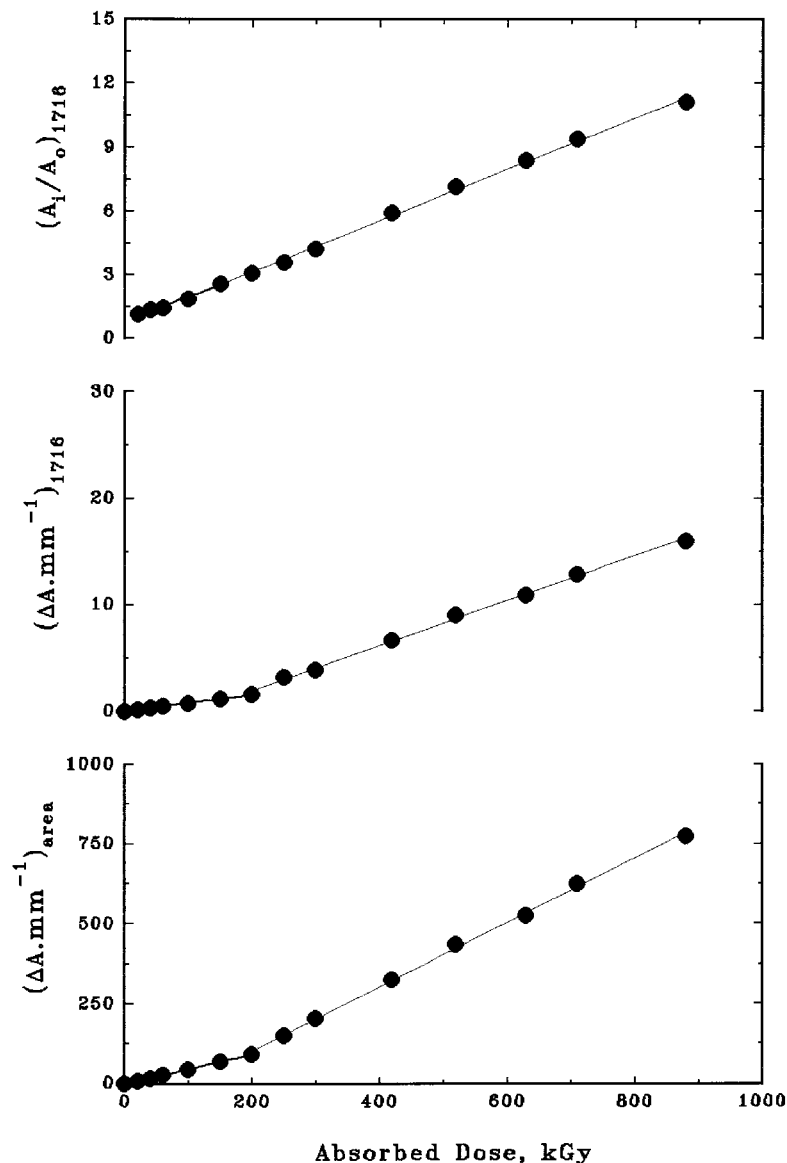
the case of the quantity  $(\Delta A \text{ mm}^{-1})_{\text{area}}$ , the linearity of the response is of a lesser extent than that for the quantity  $(\Delta A \text{ mm}^{-1})_{1716}$ . This result is due to the sensitivity of the peak area measurement to any change in the base line of the FTIR spectra; therefore, the spectrophotometric quantity  $(\Delta A \text{ mm}^{-1})_{\text{area}}$  is not recommended for quantitative measurements in radiation dosimetry.

#### Radiation-Chemical Yield

The radiation-chemical yield ( $G$  value) is defined as the number of moles of a substance produced or degraded by the absorption of one joule of energy (mol/J). The  $G$  value is calculated from the general relation<sup>18</sup>

$$G \text{ value} = \frac{\Delta A}{D \cdot \varepsilon \cdot \rho \cdot b} \text{ (mol/J)} \quad (2)$$

where  $\Delta A$  is the change in the absorbance at  $\lambda_{\text{max}}$ ;



**Figure 6** Change of FTIR spectrometric quantities  $(\Delta A \text{ mm}^{-1})_{1716}$ ,  $(\Delta A \text{ mm}^{-1})_{\text{area}}$ , and  $(A_i/A_0)_{1716}$  as a function of the absorbed dose ( $^{60}\text{Co}$   $\gamma$ -ray). Dose rate = 3.76 kGy  $\text{h}^{-1}$ . Irradiation temperature = 35°C. Wavelengths of the analysis are indicated.

$b$ , the optical path length (cm);  $\epsilon$ , the linear molar extinction coefficient at  $\lambda_{\text{max}}$  ( $\text{L mol}^{-1} \text{ cm}^{-1}$ );  $\rho$ , the density of the dosimeter ( $\text{g cm}^{-3}$ ); and  $D$ , the absorbed dose (Gy).

The radiation-chemical yield of the ketonic carbonyl group produced in irradiated LDPE film (thickness  $\cong 0.075$  mm) was evaluated from the increment of the FTIR absorbance ( $\Delta A$ ) at  $1716 \text{ cm}^{-1}$  and the reported molar extinction coefficient  $\epsilon_{1716} = 220 \text{ L mol}^{-1} \text{ cm}^{-1}$ .<sup>29,30</sup> It was found to be  $0.4 \mu\text{mol/J}$  for absorbed doses lower than 200 kGy and  $1.0 \mu\text{mol/J}$  for absorbed doses higher than 200 kGy.

The average  $G(\text{C=O})$  value over all the absorbed dose range was found to be  $0.7 \mu\text{mol/J}$ , which is comparable to the  $G(\text{C=O})$  value ( $0.65 \mu\text{mol/J}$ ) obtained by Dole<sup>10,11</sup> for LDPE film having a thickness of 0.064 mm.

#### Uncertainty of Dose Measurements

The overall uncertainty (Type A) (at two standard deviations, i.e.,  $2\sigma$ , approximately equal to



**Table III** Constants **a** and **b** and Correlation Coefficients ( $r^2$ ) of FTIR Response Curves

Constants	$(\Delta A \text{ mm}^{-1})_{1716}$		$(\Delta A \text{ mm}^{-1})_{\text{area}}$		$(A_i/A_0)_{1716}$ 0–880 kGy
	1st Part 0–200 kGy	2nd Part 200–880 kGy	1st Part 0–200 kGy	2nd Part 200–880 kGy	
<b>a</b>	0.000	–2.290	0.000	–99.060	0.781
<b>b</b>	0.0079	0.021	0.460	1.006	0.012
$r^2$	0.999	0.999	0.999	0.999	0.999

a 95% confidence level) arising during the calibration of LDPE film was calculated, where the measurements were made under conditions of repeatability.<sup>31</sup> Five replicate measurements of radiation-induced change were made at each value of the absorbed dose (13 doses were applied in the range 20–880 kGy, i.e., 65 replicates). By pooling the sets of the UV spectrophotometric quantities [ $(\Delta A \text{ mm}^{-1})_\lambda$  and  $(A_i/A_0)_\lambda$ , at 220 and 270 nm] and the sets of the FTIR spectrometric quantities [ $(\Delta A \text{ mm}^{-1})_{1716}$ ,  $(A_i/A_0)_{1716}$ , and  $(\Delta A \text{ mm}^{-1})_{\text{area}}$ ], a single value for the uncertainty for each set was found. The overall (Type A) percent uncertainties (at 1 standard deviation,  $1\sigma$ ) associated with the measurement of the dose response of LDPE films are calculated using the following equation<sup>31</sup>:

$$CV\% = \sqrt{\frac{\sum_i (n_i - 1)(\sigma_{i-1}/\bar{\mathbf{X}}_i)^2}{\sum_i (n_i - 1)}} \times 100 \quad (3)$$

where  $\sigma_{i-1}$  is the sample standard deviation of a spectrophotometric quantity for  $i$ th set of data;  $(n_i - 1)$  is the degrees of freedom for  $i$ th set of data;  $\bar{\mathbf{X}}_i$  is the average value of a spectrophotometric quantity for  $i$ th set of data; and  $n_i$  is the number of replicate measurements for  $i$ th set of data.

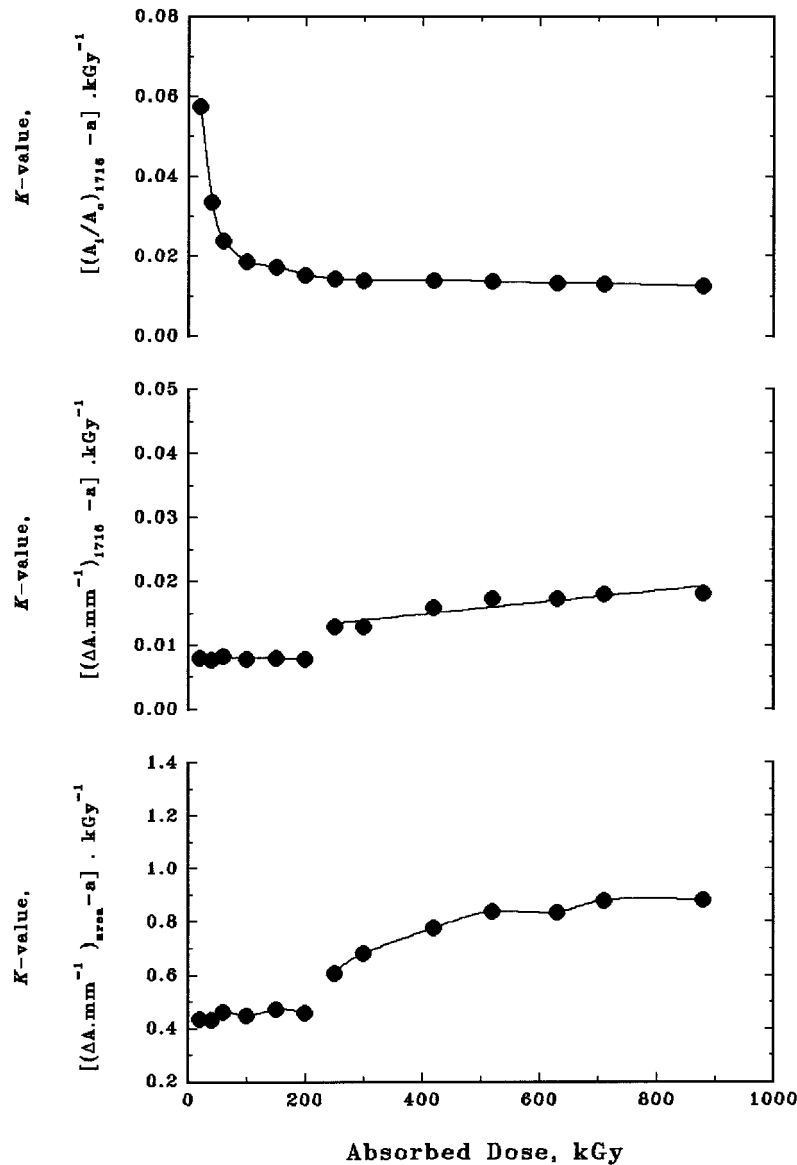
The percent overall (Type A) uncertainty at a confidence level of 95% (at 2 standard deviations, i.e.,  $2\sigma$ ) is obtained through multiplication of CV% by 2 and the results are given in Table IV. The values in Table IV indicate that the precision of assessing the absorbed dose using LDPE films depends largely on the used spectrophotometric quantity. It can be observed that the lowest uncertainty values in the dose assessment were obtained by using the UV spectrophotometric quantity  $(A_i/A_0)_{220}$  (4.3%) as well as the FTIR spectrometric quantity  $(\Delta A \text{ mm}^{-1})_{1716}$  (2.9%). On the other hand, the maximum uncertainty values

were obtained with the quantities  $(\Delta A \text{ mm}^{-1})_{\text{area}}$  (9.6%) and  $(\Delta A \text{ mm}^{-1})_{270}$  (9.8%). On the basis of the obtained results, it can be concluded that the spectrophotometric quantities  $(A_i/A_0)_{220}$  and  $(\Delta A \text{ mm}^{-1})_{1716}$  are more preferable than are the other quantities for the characterization of LDPE film in radiation dosimetry.

### Relative Humidity During Irradiation

To investigate the effect of relative humidity (RH) during irradiation on the response of LDPE films, the films were irradiated to an absorbed dose of 400 kGy (dose rate =  $3.76 \text{ kGy h}^{-1}$ ) at different relative humidities by suspending the films (five films per each RH) over different saturated salt solutions in a tightly enclosed glass tube,<sup>20,32</sup> except for the two extreme values of the RH. The 0% RH value was made with films suspended over dried silica gel and the 100% RH was made with films suspended over water. The mean temperature during  $\gamma$ -ray irradiation was approximately 35°C. The films were stored before irradiation for 48 h under the same RH conditions as when irradiated, so that equilibrium moisture in the LDPE film could be established before irradiation. Immediately after the irradiation, the film dosimeters were removed from the enclosed glass tubes and then read out spectrophotometrically at room temperature.

Figures 8 and 9 show the variation of response in terms of UV spectrophotometric quantities [ $(\Delta A \text{ mm}^{-1})_\lambda$  and  $(A_i/A_0)_\lambda$ ] and FTIR spectrophotometric quantities [ $(\Delta A \text{ mm}^{-1})_{1716}$  and  $(A_i/A_0)_{1716}$ ] relative to that value at 33% RH, as a function of % RH, respectively. It was found that the responses of all spectrophotometric quantities are not influenced by the change in the RH during irradiation. The response values change within  $\pm 2\%$  in all cases; therefore, these films can be used successfully in radiation dosimetry purposes without any correction.



**Figure 7** Variation of  $K$  value,  $\{[(\Delta A \text{ mm}^{-1})_{1716} - a] \text{ kGy}^{-1}$ ,  $[(\Delta A \text{ mm}^{-1})_{\text{area}} - a] \text{ kGy}^{-1}$ , and  $[(A_i/A_0)_{1716} - a] \text{ kGy}^{-1}\}$  as a function of the absorbed dose.

### Postirradiation Stability

The postirradiation stability of the LDPE films was investigated using two different procedures:

1. Films were irradiated, at ambient RH (35–40% RH), for an absorbed dose of 400 kGy and then stored under different storage conditions of either normal laboratory illumination plus daily incident daylight at ambient temperature or dark at three different temperatures (0, 27, and 40°C).
2. Films were irradiated at three different

relative humidities (12, 33, and 76% RH) for an absorbed dose of 400 kGy and then stored at the same RH as used during irradiation, while exposing them to diurnal cycles of daylight and darkness at ambient temperature.

These films were read out using UV and FTIR spectrophotometry at different intervals of time during the postirradiation storage period of 56 days.

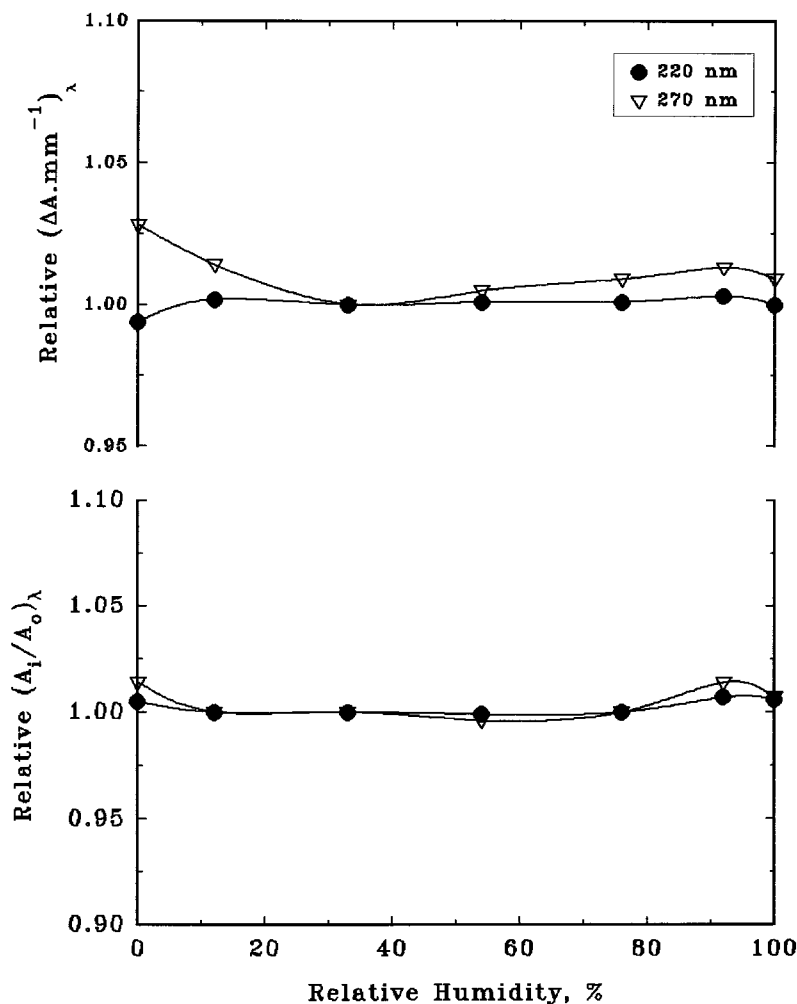
The results of conditions 1 and 2 are shown in

**Table IV** Percent Overall (Type A) Uncertainty ( $2\sigma$ ) of LDPE Films Using Different UV and FTIR Spectrophotometric Quantities

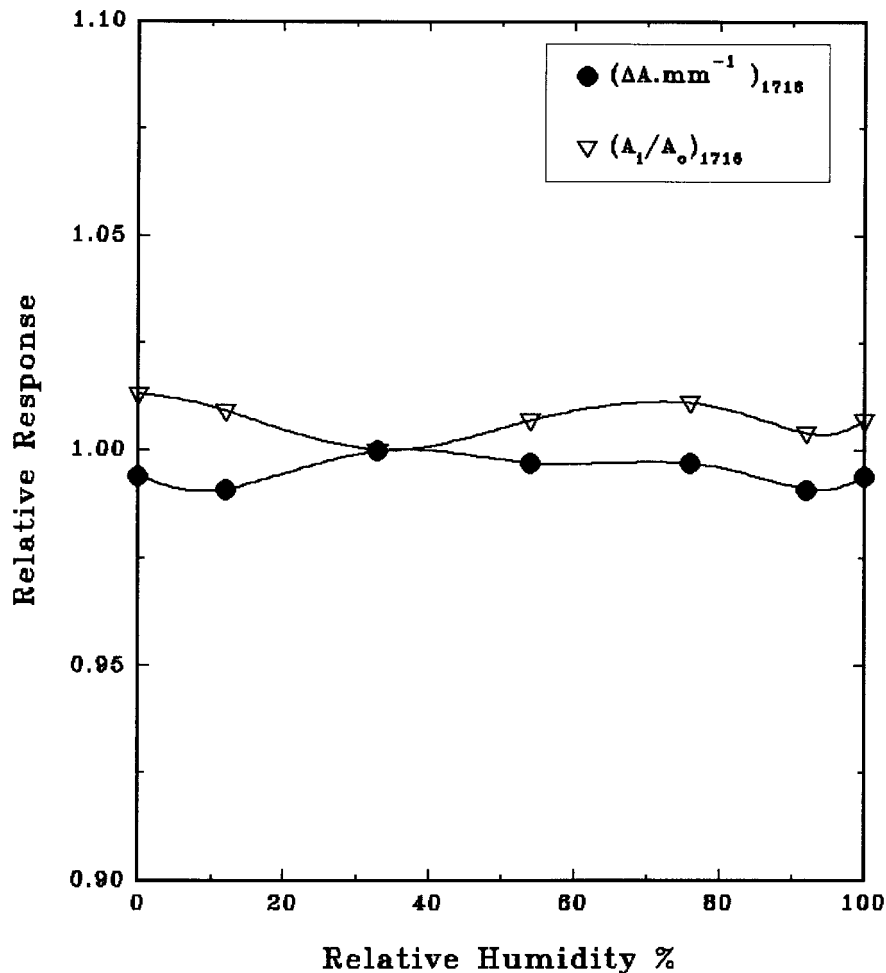
Uncertainty (%)	UV Spectrophotometry				FTIR Spectrophotometry		
	$(\Delta A \text{ mm}^{-1})$		$(A_i/A_0)$		$(\Delta A \text{ mm}^{-1})$		$(A_i/A_0)$
	220 nm	270 nm	220 nm	270 nm	1716 $\text{cm}^{-1}$	peak area	1716 $\text{cm}^{-1}$
	7.3	9.8	4.3	6.5	2.9	9.6	5.4

Figures 10 and 11, respectively, in terms of the relative change of the UV and FTIR spectrophotometric quantities as a function of storage time in days. From these figures, it can be seen that there is no significant change observed for all spectro-

photometric quantities at the different storage conditions during the storage period (56 days). Also, identical results were obtained with the films irradiated for an absorbed dose of 800 kGy and stored as in condition 1. These results agree



**Figure 8** Variation of UV spectrophotometric quantities  $(\Delta A \text{ mm}^{-1})_{\lambda}$  and  $(A_i/A_0)_{\lambda}$ , relative to that value at 33% RH, as a function of percentage RH during irradiation. Absorbed dose = 400 kGy.



**Figure 9** Variation of FTIR spectrometric quantities  $(\Delta A \text{ mm}^{-1})_{1716}$ ,  $(\Delta A \text{ mm}^{-1})_{\text{area}}$ , and  $(A_i/A_0)_{1716}$ , relative to that value at 33% RH, as a function of percentage RH during irradiation. Absorbed dose = 400 kGy.

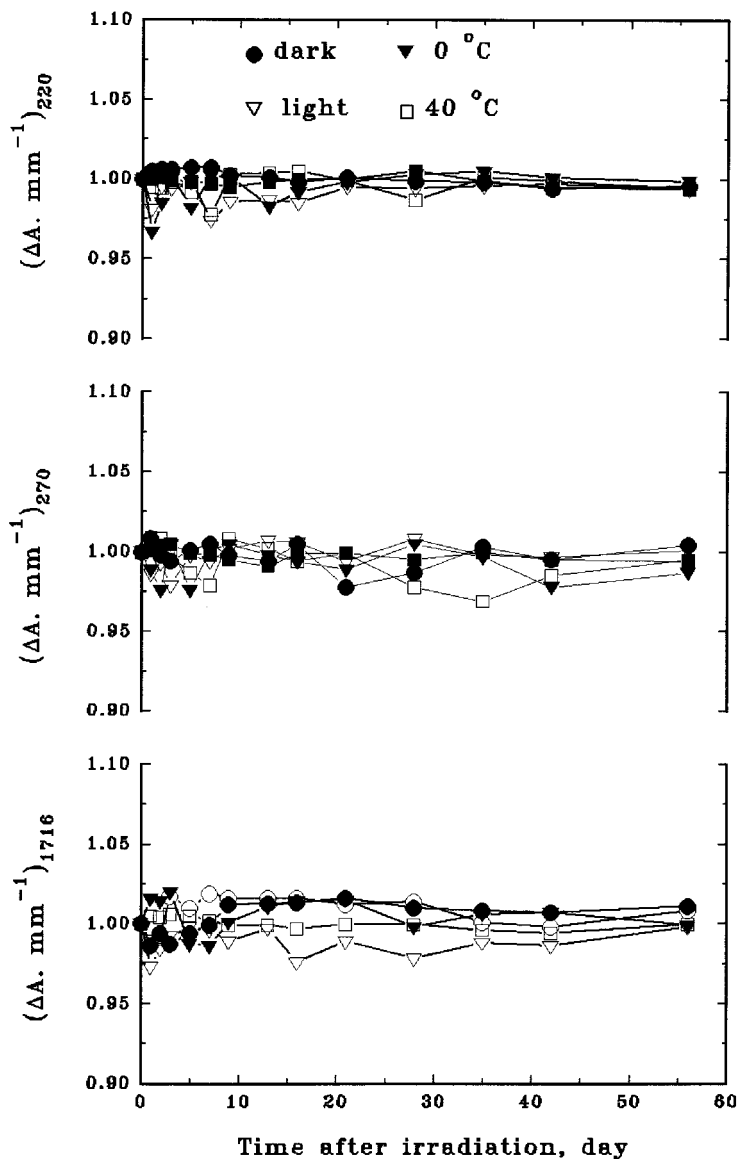
well with the results reported by Fallgatter and Dole<sup>6</sup> and Wenxiu et al.<sup>22</sup>

The postirradiation oxidation of polyethylene was explained by Seguchi et al.,<sup>33</sup> where the radicals trapped in the amorphous regions react rapidly with the oxygen that diffuses relatively easily into those regions of the polyethylene. In other words, the radicals formed during irradiation may interact relatively fast either during irradiation or during a very limited period of time after irradiation. However, a small number of radicals which are trapped mainly in the amorphous regions may interact with oxygen that has diffused after relatively longer periods of time after irradiation. Hence, the insignificant postirradiation oxidation of LDPE films was observed under our atmospheric conditions.<sup>22,28,34</sup>

## SUMMARY AND CONCLUSIONS

The results obtained in this investigation are summarized as follows:

1. The response curves obtained for LDPE films using both the UV spectrophotometric quantities [ $(\Delta A \text{ mm}^{-1})_{270}$ , and  $(A_i/A_0)_{270}$ ] are linear. On the other hand, the response curves obtained by using the spectrophotometric quantities  $(A_i/A_0)_{1716}$ ,  $(\Delta A \text{ mm}^{-1})_{220}$ ,  $(A_i/A_0)_{220}$ , and  $(\Delta A \text{ mm}^{-1})_{1716}$  consist of two linear parts, having different sensitivities, with an inflection point at the absorbed doses of about 100 kGy for the quantity  $(A_i/A_0)_{1716}$  and of about 200 kGy for all other quantities.

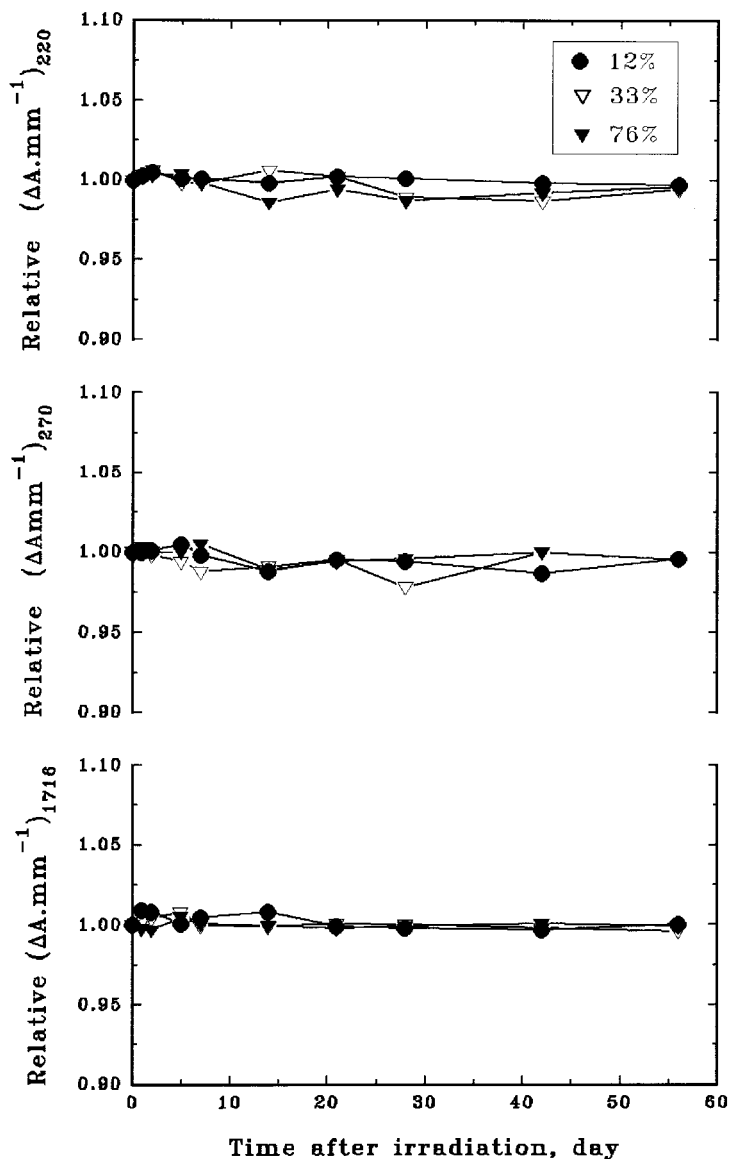


**Figure 10** Relative change of the UV and FTIR spectrophotometric quantities,  $(\Delta A \text{ mm}^{-1})_{\lambda}$ , as a function of storage time after irradiation of LDPE films (400 kGy) stored at different conditions. Wavelengths of analysis and different storage conditions are indicated.

2. The precision of assessing the absorbed dose and the overall uncertainty associated with the measurement of the LDPE film response depends largely on the selected readout tool of measurement whether FTIR (at  $1716 \text{ cm}^{-1}$ ) or UV (at 220 and 270 nm wavelengths) as well as on the quantity used for calculation. The lowest uncertainties associated with the measurement of the LDPE response were obtained with the FTIR spectrometric quantity  $(\Delta A \text{ mm}^{-1})_{1716}$  (2.9%) and the UV spectrophotometric quantity  $(A_i/A_0)_{220}$  (4.3%).

3. The radiation-chemical yield,  $G(\text{C}=\text{O})$ , of the LDPE film was found to be  $0.7 \mu\text{mol}/\text{J}$ , which agrees fairly well with that reported in the literature.
4. The response of the LDPE films was found to be independent of the change in relative humidity (RH) during irradiation.
5. The response of the LDPE films exhibits good postirradiation stability under different storage conditions.

On the basis of the present results, it can be con-



**Figure 11** Relative change of the UV and FTIR spectrophotometric quantities,  $(\Delta A \text{ mm}^{-1})_{\lambda}$ , as a function of storage time after irradiation of LDPE films (400 kGy) stored at different RHs. The different storage relative humidities are indicated.

cluded that LDPE films can be used successfully as a high-dose dosimeter for  $\gamma$ -radiation within a wide range, namely, from 10 to 880 kGy (sterilization and radiation material processing). On the other hand, using LDPE films for radiation dosimetry measurements in the lower dose range up to 200 kGy must be taken under specific reservation, i.e., measuring absorbed doses in this dose range is not appreciated under our experimental conditions.

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