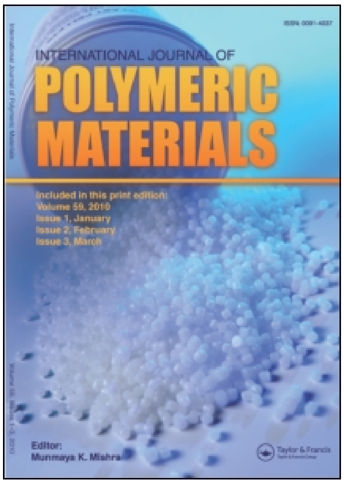

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NEW ALANINE/EPR DOSIMETER USING EVA COPOLYMER/PARAFFIN AS A BINDER FOR HIGH-DOSE RADIATION DOSIMETRY: PERFORMANCE CHARACTERIZATION

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NEW ALANINE/EPR DOSIMETER USING EVA COPOLYMER/PARAFFIN AS A BINDER FOR HIGH-DOSE RADIATION DOSIMETRY: PERFORMANCE CHARACTERIZATION

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Alanine/EPR rods ($3 \times 10\,mm$) for routine use in high-dose radiation applications have been prepared by a simple technique in the laboratory where alanine powder was mixed with molten mixture of paraffin wax and ethylene vinyl acetate copolymer (EVA). The binding mixture EVA/Paraffin does not present interference or noise in the EPR signal before or after the irradiation. The rods show good mechanical properties for safe and multi-use handling. The rods can be used with good precision in the dose range from 1 to 125 kGy. The overall uncertainty for calibration of the EVAPA rod dosimeters at 2σ was found to be 4.56%. The dose response, influence of humidity and temperature during irradiation, energy dependence as well as post–irradiation storage at different conditions are discussed.

Keywords: alanine, ethylene vinyl acetate, radiation dosimetry, EPR

INTRODUCTION

The alanine/EPR dosimetry system provides a reliable means for measuring absorbed dose. It is based on the generation of specific stable radicals in crystalline alanine by ionizing radiation. Identification and determination of the concentration of the specific alanine radicals are performed by electron paramagnetic resonance (EPR)

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spectroscopy. The concentration of alanine-derived radical is a function of absorbed dose. EPR technique is non-destructive, so alanine dosimeters can be read out repeatedly and thus can be used for archival purposes.

Several laboratories have successfully developed alanine-EPR dosimeter using different binders [1–10]. Alanine-EPR has been selected by IAEA as the transfer dosimeter for use in the quality audit service IDAS (International Dose Assurance Service) for several reasons, for example: near-tissue equivalency, low sensitivity to ambient environment, broad useful dose range, non-destructive analysis, and little fading of the response with time. The disadvantages of alanine/EPR dosimetry system, at that time, were the limited experience with the system and the significant cost of the analysis equipment, namely electron paramagnetic resonance (EPR) spectrometer. As a matter of fact, almost every Primary Standard Dosimetry Laboratory (PSDL) and Secondary Standard Dosimetry Laboratory (SSDL) is using now alanine/EPR as a reference or transfer dosimetry system and the price of the EPR spectrometer has substantially decreased. These facts are encouraging many industrial irradiation facilities to use alanine/EPR in their routine dosimetry measurements.

In the present work, a new simple method for preparation of alanine-EPR pellets with a new binding medium has been carried out. The dose response, influence of humidity and temperature during irradiation, energy dependence as well as storage at different conditions are discussed.

EXPERIMENTAL PROCEDURES

Materials

L-α-alanine (99%, BDH), hot melt stick adhesive based on ethylene vinyl acetate copolymer (Tec-Bond 232/12, Power Adhesives Limited, England), paraffin wax (congealing point 65–71°C, BDH) were used. Alanine was grinded to fine powder and sieved to less than 200 μm particle sizes.

Preparation of Alanine Pellets

For use in routine dosimetry adequate mechanical properties of alanine pellets are required. The pellets available in the market are brittle because they contain a high content of alanine (80–90%) and only 10–20% binder (mainly paraffin wax). To avoid brittleness and to improve the mechanical properties, the alanine content is reduced and ethylene vinyl acetate (EVA) hot melt adhesive is added to paraffin wax.

An equal weight mixture of paraffin wax and EVA hot-melt adhesive was melted in a round bottle at 95°C in a water bath. EVA shows a complete compatibility with paraffin wax. 5, 10, or 20% fine powdered L-α-alanine was added to the hot mixture solution and mechanically stirred for about 10 min at the same temperature to obtain a homogeneous mixture. The hot solution was sucked into polypropylene tubes (inner diameter 3 mm) and was left to solidify by cooling. Alanine mixture rod was obtained by removing the polypropylene tube, and then cutting into rods 3×10 mm dimensions). Three different types of rods were prepared depending on alanine concentration, namely EVAPA5, EVAPA10, and EVAPA20 containing 5, 10, and 20% of alanine, respectively. The average mass of the rods in the three concentrations was found to be 46.7 ± 1.5 mg (1σ).

Irradiation

Irradiations were carried out with gamma radiation in the ^{60}Co gamma chamber (model Issledoleev, product of Russia), which was calibrated using dichromate dosimeters (supplied and measured by National Physical Laboratory, England). The absorbed dose rate at the time of irradiation was about 7.5 kGy/h. Five rods were irradiated together at the central position of the sample chamber using a specially designed holder made from polystyrene to ensure electronic equilibrium.

EPR Measurement

EPR spectra were recorded at ambient temperature with a Bruker EMX spectrometer (X-band); the cavity used was the standard Bruker ER 4102 rectangular cavity. The operating conditions for the EPR spectrometer were as follow: sweep field: 3480 ± 100 G; microwave frequency: 9.72 GHz; microwave power 4 mW; modulation frequency 100 kHz; modulation amplitude 1 G; time constant: 163.48 ms; sweep time 0.35 min/scan. The bottom of the EPR tube was adjusted at a fixed position to ensure reproducible and accurate positioning of the pellets in the sensing zone of the cavity.

EPR spectra were recorded at two orientations of each pellet in the EPR cavity (0° and 90 degrees). The dose responses of dosimeters were calculated in terms of the average peak-to-peak heights of the two orientations (h_0 and h_{90}) per unit weight of dosimeter and normalized to the receiver gain of the EPR spectrometer. Stability of the EPR spectrometer sensitivity was checked before and after each series of measurement using reference alanine dosimeters irradiated to known doses.

RESULTS AND DISCUSSION

Dose Response

The prepared rods have good mechanical properties adequate for easy and safe handling because they contain relatively low concentrations of alanine. Increasing alanine content increases the brittleness of the pellets. Nevertheless, the reduced alanine concentration in the rods has limited effect on the sensitivity of the dosimeter for doses higher than 1 kGy at the used EPR parameters. Figure 1 shows the EPR spectra of EVA/Paraffin without alanine and EVAPA20 rods irradiated to a dose of 25 kGy and measured at the same parameters where the spectrum of the EVA/Paraffin is magnified by 10. It can be seen that almost no signal has been detected in the irradiated EVA/Paraffin mixture reflecting its suitability as a binding material for the alanine powder.

Figure 2 shows the calibration curves obtained for the irradiated EVAPA5, EVAPA10, and EVAPA20 rods in terms of average peak-to-peak amplitude normalized to dosimeter mass and receiver gain [peak height/(gain × mass)] versus the absorbed dose over the range from 1 to 125 kGy. Different polynomial functions were tested to fit the different response curves given in Figure 2. Based on the correlation

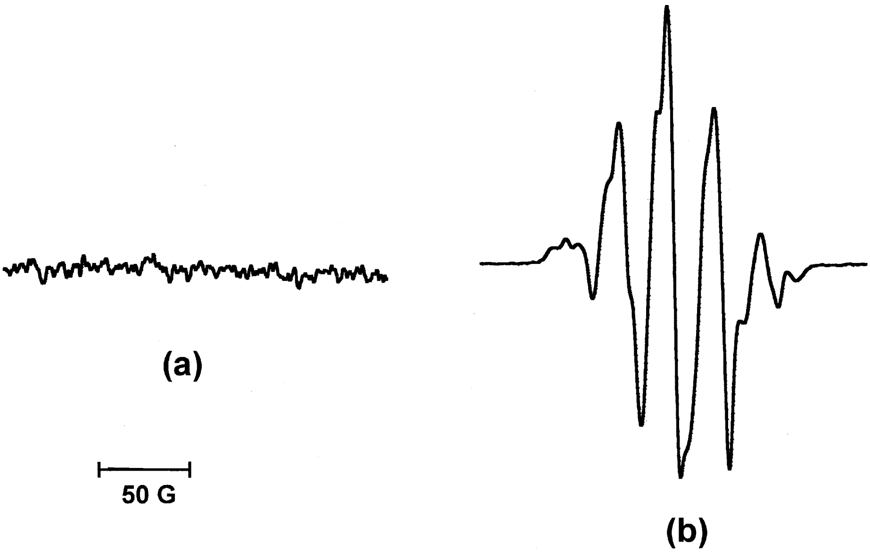

FIGURE 1 (a) EPR spectrum of EVA/paraffin pellets without alanine irradiated to 25 kGy. (b) EPR spectrum of EVAPA20 pellets irradiated to 25 kGy.

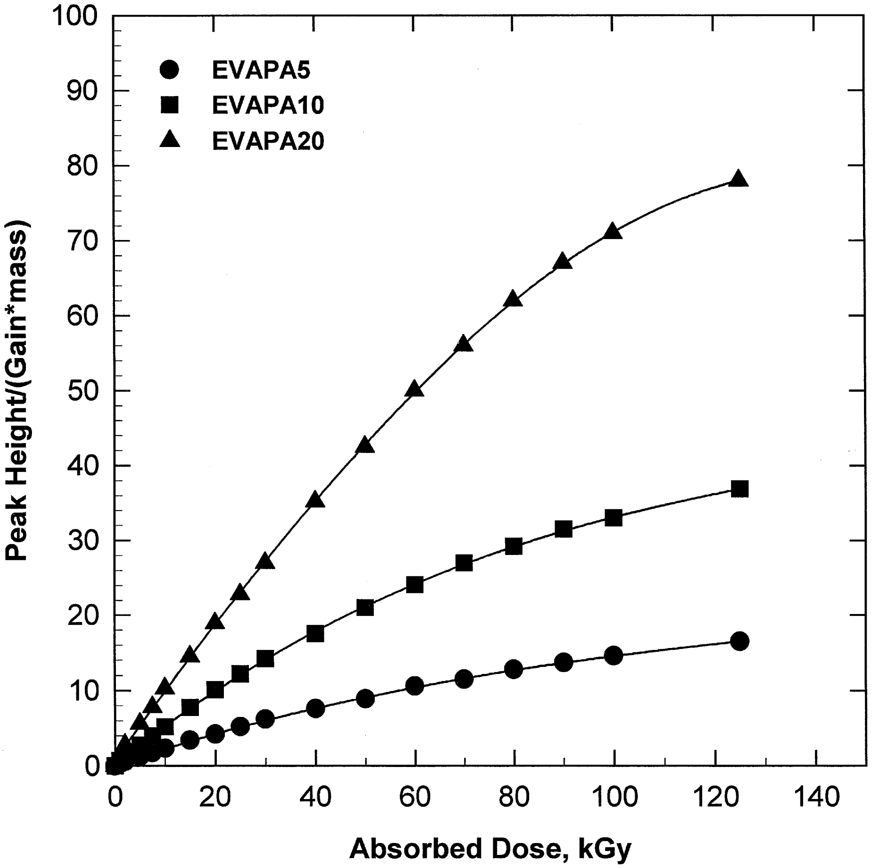

FIGURE 2 Dose-response curves of the EVAPA5, EVAPA10, and EVAPA20 pellets.

coefficients and *F*-statistics values, the best fit was found with the 3rd order polynomial equation. Table 1 shows the constants a, b, c, d, and correlation coefficients (r^2) as well as *F*-statistics values.

Temperature during Irradiation

The effect of temperature during irradiation on response of EVAPA20 rods was investigated by irradiating the pellets to a dose of 5 kGy at different temperatures (5, 15, 25, 35, 45, and 55°C) using thermal baths and isolated during irradiation using Styrofoam phantom. The pellets were kept at the same temperature for about 1 h prior to

TABLE 1 The Constants a, b, c, d, and the Correlation Coefficients (r^2) as Well as F-Statistics Values for the 3rd Polynomial Fit of EVAPA5, EVAPA10, and EVAPA20 Response Curves

	a	b	c	d	r^2	F-Statistics
EVAPA5	0.1026	0.2284	−0.0011	2.4×10^{-6}	0.9996	13527
EVAPA10	0.1601	0.537	−0.0026	5.1×10^{-6}	0.9998	31855
EVAPA20	0.6728	0.9366	−0.0014	-8.7×10^{-6}	0.9999	45785

irradiation to maintain thermal equilibrium in the samples during irradiation. Figure 3 shows the variation in response [peak height/(gain × mass)] as a function of temperature during irradiation relative to that at 25°C. It can be seen that the response increases linearly with the irradiation temperature in the studied range of temperature. The slope of the obtained straight line is equal to $0.0021°C^{-1}$. In other words the temperature coefficient at the studied dose level (5 kGy) is equal to $+0.21\%°C^{-1}$ for the experimental conditions, which is fairly

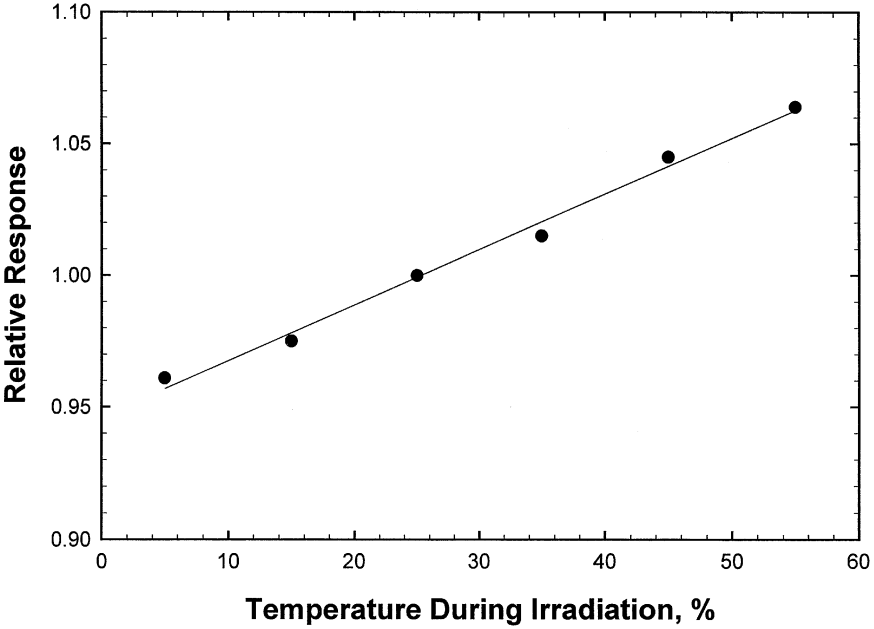

FIGURE 3 Relative variation in response of EVAPA20 as a function of temperature during irradiation relative to that at 25°C. Irradiation dose = 5 kGy.

in agreement with the values reported by different authors for different dosimeter compositions (0.15–0.3%°C^{-1}) [11–14].

Humidity during Irradiation

The effect of relative humidity (RH) during irradiation on response of EVAPA20 rods was investigated by irradiating the pellets to a dose of 50 kGy at different relative humidities (0, 12, 33, 54, 76, 92, and 100%) using saturated salt solutions. The rods were stored before irradiation for a 48-h period under the same RH conditions as when irradiated, so that equilibrium moisture content in the dosimeter could be established during irradiation. Figure 4 shows the variation in response [peak height/(gain × mass)] as a function of percentage relative humidity during irradiation relative to that at 54%. It can be seen that the response is almost flat from 0 up to 60% RH and shows a slight decrease at higher RH values. This result reflects the insignificant dependence of these rod dosimeters on the change of relative humidity during irradiation.

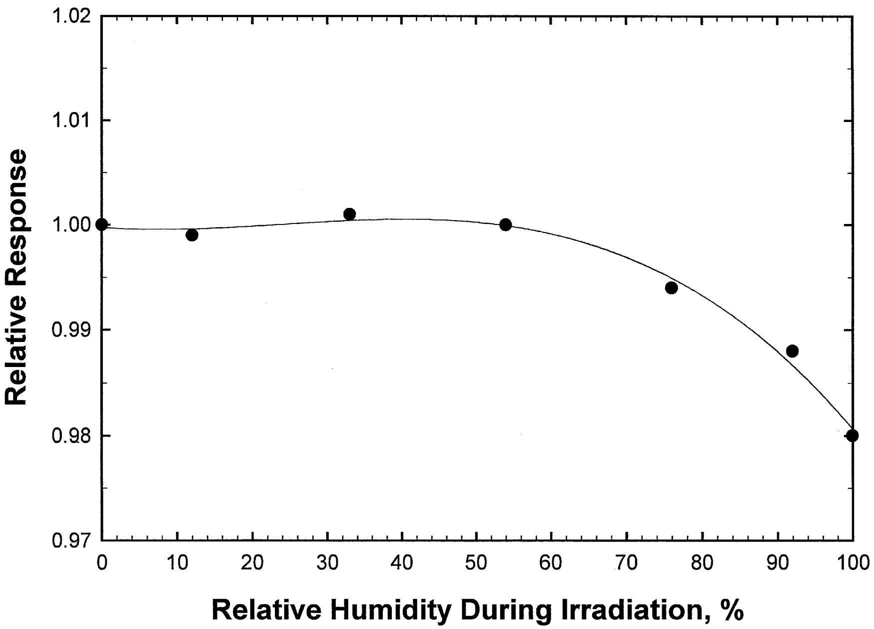

FIGURE 4 Relative variation in response of EVAPA20 as a function of relative humidity during irradiation relative to that at 54%. Irradiation dose = 50 kGy.

Uncertainty

Factors contributing to the total uncertainty may be separated into two types, type A and type B. Type A is evaluated by the statistical analysis of a series of observations and type B is evaluated by means other than the statistical analysis of a series of observations [15].

Sources of uncertainty for the measurement of absorbed dose by EVAPA dosimeters may include the uncertainty in the absorbed dose received by the dosimeters during calibration, analysis dosimeter response, uncertainty associated with measurement of response and fit of dosimetry data to calibration curve. Table 2 shows the overall uncertainty for calibration of EVAPA dosimeters. The overall combined uncertainty, U_c, was found to be 2.28% (1σ). The combined uncertainty (at two standard deviations, i.e., $2\ \sigma$, approximately equal to a 95% confidence level) is found by multiplication of U_c (at $1\ \sigma$) by two. Thus, the combined uncertainty at $2\ \sigma$ using EVAPA pellets is 4.56%.

Energy Dependence

The energy dependence of dosimeters may cause inaccuracies in the measurement of absorbed dose in a material of interest. Most such errors arise when a dosimeter is calibrated under specific conditions

TABLE 2 Overall Uncertainty for Calibration of EVAPA Dosimeters

Component of uncertainty	Type A	Type B
Absorbed dose received by dosimeters		
Response of dichromate dosimeters		1.1
Irradiation time		0.1
Half-life of ^{60}Co		0.02
Irradiation temperature		0.2
Correction to nearest day		0.03
Reproductivity of position of sample drawer and dosimeter holder		0.1
Analysis of dosimeters		
Vertical positioning of the dosimeter in EPR cavity	0.2	
Angular positioning of the dosimeter in EPR cavity	0.2	
Reproducibility of EPR measurements	0.2	
Precision of balance		0.2
Uncertainty associated with measurement of EVAPA dosimeters response (5 rods at each dose)	1.85	
Uncertainty from polynomial fit for response curve as determined from the variation of the residuals.	0.6	
Type A and Type B combined standard uncertainty	1.975	1.145
Overall combined uncertainty	2.28	
Expanded uncertainty (k = 2)	4.56	

with respect to radiation energy and irradiation geometry, and it is used later under conditions that are significantly different.

The EVAPA20 rod dosimeters were studied for their energy sensitivity to ionizing photons in the energy range 10 keV to 20 MeV. The mass attenuation coefficient, μ/ρ, the mass energy-absorption coefficient, μ_{en}/ρ and the collision stopping powers, $(1/\rho\ .dE/dX)_{coll}$, have been calculated as a function of photon energy for the EVAPA20 rod dosimeters. These calculations were based on the data available online at the NIST physical reference data Internet web site [16–17]. Figures 5–7 show the calculated attenuation coefficients, absorption coefficients, and stopping powers of the EVAPA20 pellets, respectively, compared with the values of the adipose tissue published in the same web site. It can be seen that the ratios of $(\mu/\rho)_{Adipose}/(\mu/\rho)_{EVAPA20}$ and $(\mu_{en}/\rho)_{Adipose}/(\mu_{en}/\rho)_{EVAPA20}$ are almost equal to unity in the photon energy range from 0.1 to 20 MeV and show a noticeable decrease at lower photon energies. On the other hand the ratio of stopping power of adipose to EVAPA20 rods is almost unity overall the studied photon energy range (0.01–20 MeV). In conclusion, energy dependence of these rod dosimeters is insignificant over 100 keV.

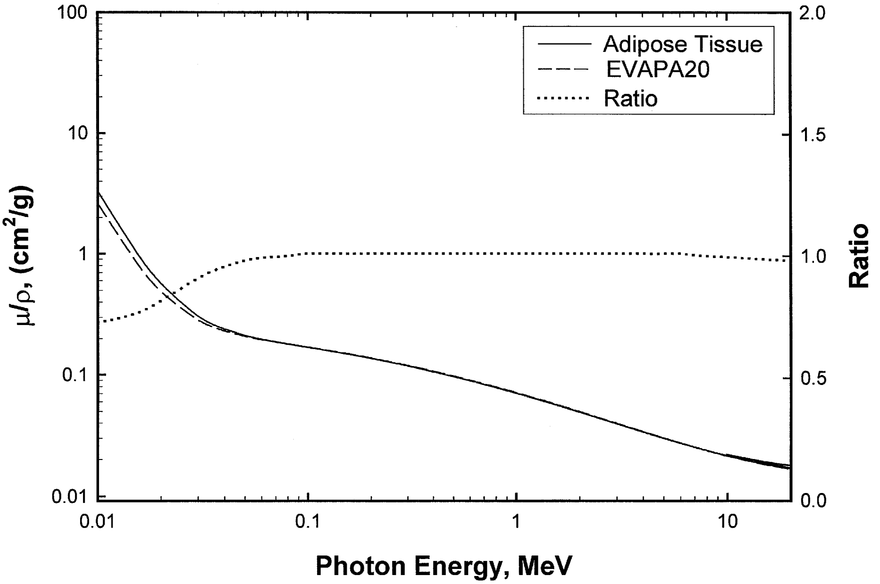

FIGURE 5 Left ordinate: Calculated mass-energy attenuation coefficients of EVAPA20 and Adipose tissue versus photon energy. Right ordinate: Ratio of attenuation coefficients versus photon energy.

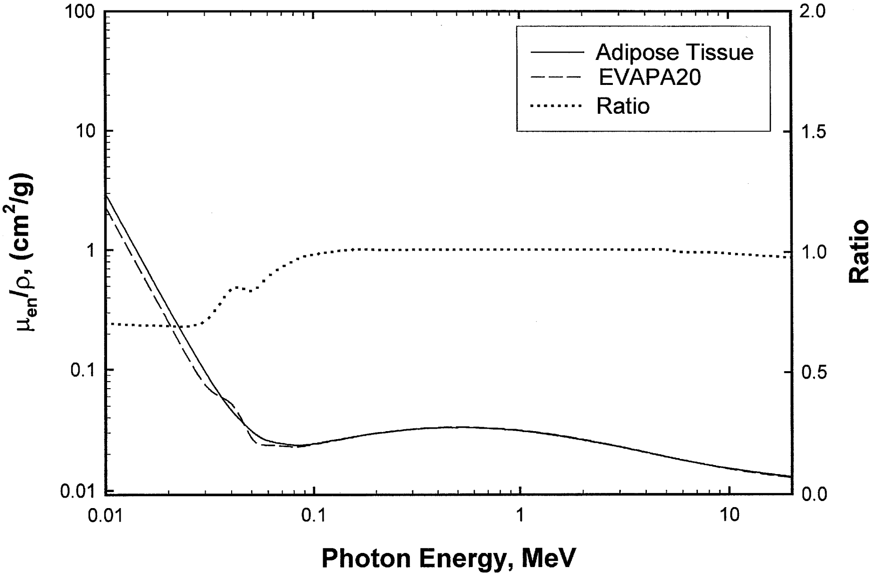

FIGURE 6 Left ordinate: Calculated mass-energy absorption coefficients of EVAPA20 and Adipose tissue versus photon energy. Right ordinate: Ratio of absorption coefficients versus photon energy.

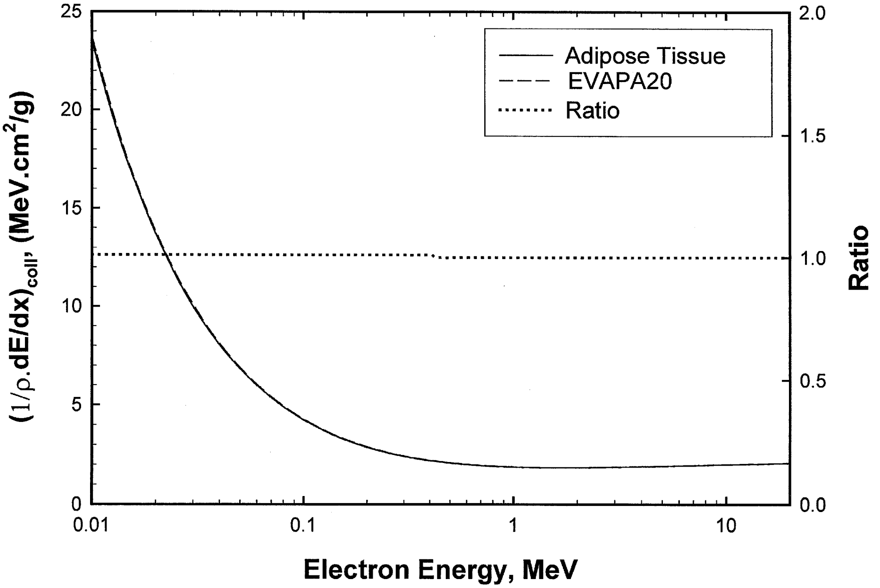

FIGURE 7 Left ordinate: Calculated collision mass stopping powers of EVAPA20 and Adipose tissue versus photon energy. Right ordinate: Ratio of stopping powers versus photon energy.

Stability at Different Storage Conditions

The post-irradiation stability has been studied for 9 sets of EVAPA20 pellets (each set consists of 3 rods) for a period of 180 days. 3 sets were irradiated to a dose of 25 kGy and stored in dark at different temperatures (namely, 0, 25, and 40°C) whereas the other 6 sets were stored in the dark at 25°C after irradiation to doses of 2, 5, 10, 25, 50, and 100 kGy. Figure 8 shows the relative peak-to-peak signal height as a function of post-irradiation storage time. It can be seen that these pellets are fairly stable after irradiation and show only little fading

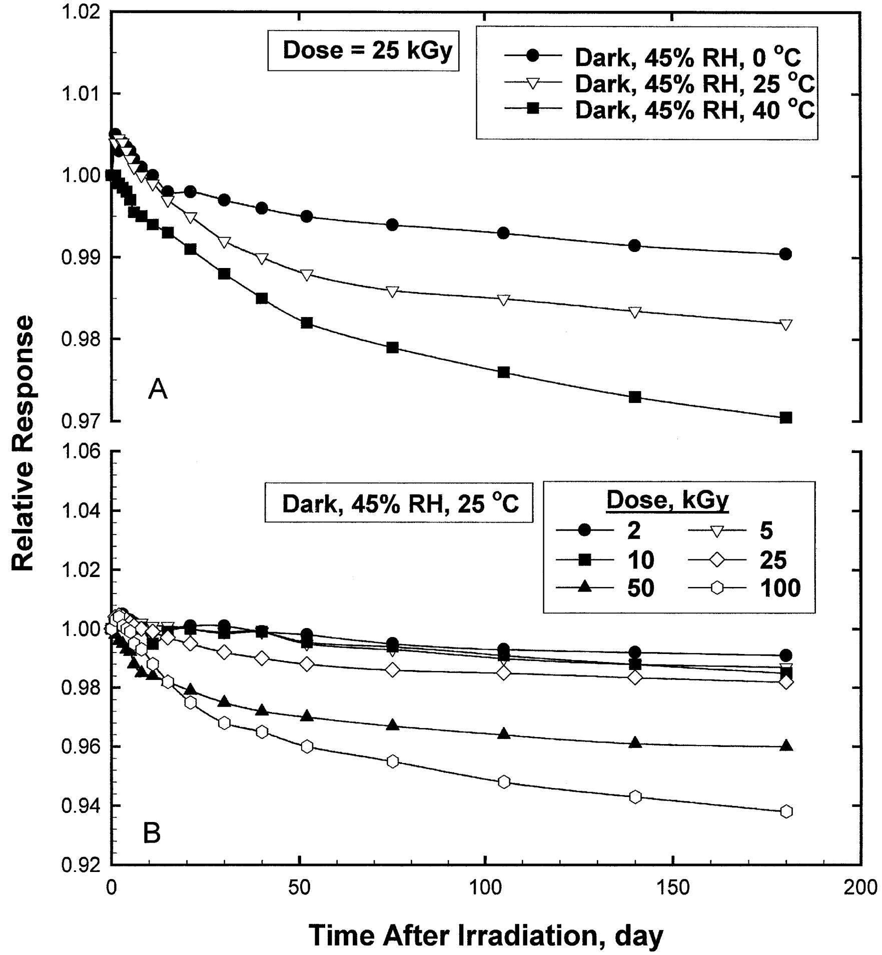

FIGURE 8 Relative peak to peak signal height as a function of post-irradiation storage time at different conditions. Storage conditions were indicated.

between 2 and 5% over the 180-day storage period depending on irradiation dose and storage condition where the fading percent increases gradually with the increase of the storage temperature and irradiation dose.

CONCLUSIONS

From the data presented in this article, the following conclusions can be drawn:

- A new alanine-EPR rod dosimeter (EVAPA) has been prepared by a simple technique in the laboratory using ethylene Vinyl acetate copolymer and paraffin wax as binding materials.
- The binding mixture of EVA/Paraffin does not present interference or noise in the EPR signal before or after the irradiation.
- The prepared rods have good mechanical properties adequate for easy and safe handing because they contain relatively low concentrations of alanine.
- The EVAPA rods can be used with good precision in the dose range from 1 to 125 kGy.
- The temperature coefficient was found to equal to $+0.21\%^{\circ}C^{-1}$.
- These rod dosimeters have insignificant dependence on the change of relative humidity during irradiation.
- The overall uncertainty for calibration of the EVAPA rod dosimeters at 2σ was found to be 4.56%.
- Energy dependence of these rod dosimeters is insignificant over 100 keV.
- These dosimeters are fairly stable after irradiation and show little fading between 2 and 5% over a 180-day storage period depending on irradiation dose and storage condition.

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