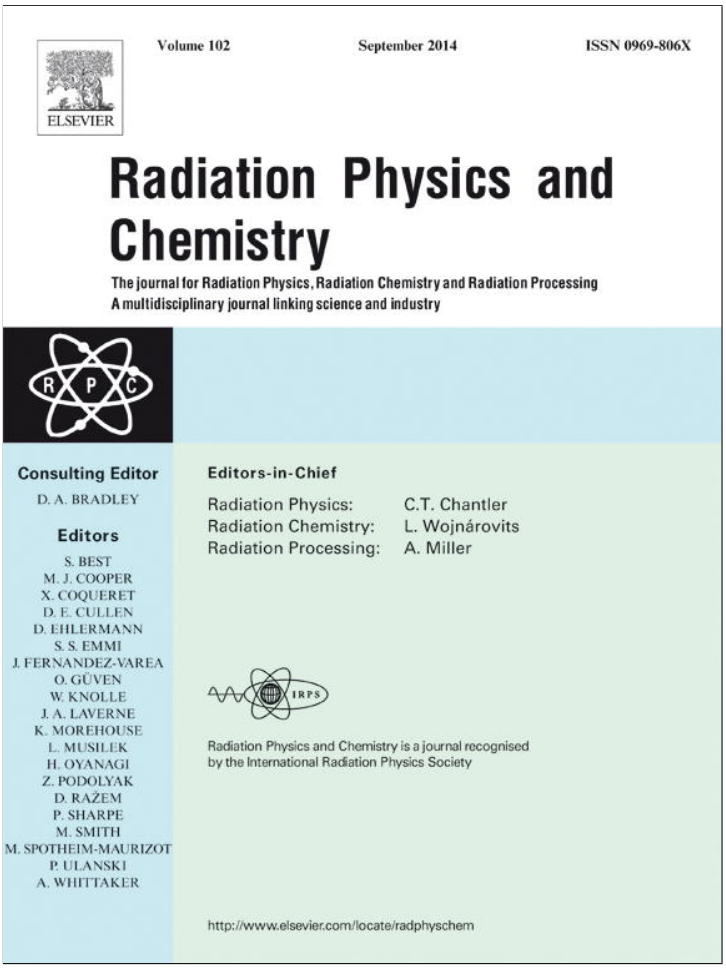

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EPR dosimetric properties of 2-methylalanine pellet for radiation processing application

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HIGHLIGHTS

- Preparation of 2-methyl alanine pellets for high-dose dosimetry (1–100 kGy).
- The dosimeter response is humidity independent in 33–76% relative humidity range during irradiation.
- The temperature coefficient equals 0.96%/°C in the range of 21–60 °C.
- Overall uncertainty of the dosimeter not exceeds 6.9% at 2σ.

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ABSTRACT

The dosimetric characteristics of γ-radiation induced free radicals in 2-methylalanine (2MA) pellet dosimeter are investigated using electron paramagnetic resonance (EPR) in the high-dose range of 1–100 kGy. The EPR spectrum of γ-irradiated 2MA exhibits an isotropic EPR signal with seven lines. The dosimeter response is humidity independent in the range of 33–76% relative humidity. The manufactured dosimeter is typically adipose tissue equivalent in the energy level of 0.1–15 MeV. The overall uncertainty (2σ) of the dosimeter is less than 6.9%.

1. Introduction

Irradiation of solid amino acid L-alanine by γ-rays produces a free radical exhibiting a significant EPR signal (Regulla and Deffner, 1982). The EPR signal of irradiated polycrystalline alanine is mainly due to a single radiation-induced radical species assumed to be CH_3–C•H–COOH radical, which is produced by deamination of neutral alanine (Miyagawa and Gordy, 1960; Kuroda and Miyagawa, 1982; Arber et al., 1991; Lund et al., 2002). One of suggested mechanism of radiation-induced free radical in alanine is by reductive route (protonation followed by deamination process) (Sagstuen et al., 2004). While, the second is an oxidative route (deprotonation followed by intermolecular hydrogen transfer) producing CH_3–C•NH_2–COOH radical.

L-alanine has attracted considerable interest for use in radiation dosimetry (Regulla and Deffner, 1982; Ciesielski and Wielopolski, 1994) and has been formally accepted as a secondary standard for high dose dosimetry (Nette et al., 1993). In addition, several studies were initiated to find new dosimetric materials with good sensitivity for EPR dosimetry to substitute alanine dosimeter (Olsson et al., 2002; Lund et al. 2002, 2005; Gancheva et al., 2006; Beshir et al., 2012; Soliman and Abdel-Fattah, 2012; Lelie et al., 2013). One of those materials is 2MA, which is 70% more sensitive than L-alanine and is considered similar to tissue-equivalent material above energy of 100 keV (Olsson et al., 2002). The authors tested this material for clinical purposes in the dose range of 0.5–100 Gy and reported the linearity of its dose response function in this range. Gamma irradiation of 2MA produces a significant EPR signal (nearly isotropic signal) with seven lines. The intensity of the signal decreases with ~1% in 2 h after irradiation and then increases slowly to some extent above the starting value. This material was previously investigated using K-band EPR spectrometer for radiotherapeutic dose range of 0.5–30 Gy (Chen et al., 2007). The overall uncertainty of this dosimeter is less than 5%.

In spite of its good dosimetric properties, 2MA has not been totally investigated yet to high-dose dosimetry in radiation sterilization, unlike L-alanine. One of the goals of the present study was to investigate the EPR dosimetric characteristics of 2MA-manufactured pellets for high-dose range 1–100 kGy applications. In addition, the energy dependence and overall uncertainty of the dosimeter were calculated.

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2. Experimental

2.1. Dosimeter manufacturing

The dosimeter pellets were manufactured from a blend of 80% (w/w) 2-methylalanine ($C_4H_9NO_2$, 99% purchased from Merck) fine powdered and 20% (w/w) binder of low-density polyethylene/paraffin wax. The pellets were obtained by mechanical pressing this blend at 130 °C in stainless steel cast manufactured specially for such purposes. The nominal dimensions for a pellet were 4 mm length and 3.8 mm diameter.

2.2. Radiation source

The manufactured pellets were irradiated to 1–100 kGy range in a ^{60}Co Gamma Cell GC-220 Excel (MDS Nordion, Canada) using a specially designed polystyrene holder to ensure electronic equilibrium during irradiation. The dose rate (to water) of GC was calibrated using alanine by National Physical Laboratory (NPL), UK (NPL traceability). The dose rate at the period of analysis was ~2.47 kGy h^{-1}. The radiation source is equipped with a temperature control unit manufactured, calibrated and installed in the source cavity by NPL to maintain a constant temperature in the GC during establishing the response curve and to study the effect of temperature during irradiation on dose–response curve. The calibration of the thermometers used in this unit was carried out by NPL and checked during initiating this work using another calibrated thermometer.

2.3. EPR spectrometer

The free radicals created in 2MA pellets by radiation were recorded at room temperature (21–25 °C) by X-band EMX spectrometer (Bruker, Germany) using a standard rectangular cavity of ER 4102. The operating parameters adopted throughout the experiment are: microwave power, 1.594 mW; modulation amplitude, 4.00 Gauss; modulation frequency, 100 kHz; sweep width, 300 Gauss; microwave frequency, 9.779 GHz; time constant, 81.92 ms and sweep time, 20.972 s. The EPR tube used in this work is adjusted at a fixed position during all measurement into the cavity. We recorded all measurements of 2MA pellets at two orientations (0 and 90°) and the mean of the two orientations was used. The variation in signal height between the two orientations was reported to be less than 0.14% (1σ). In addition, we used irradiated alanine rod to check the sensitivity of the instrument and to correlate the signal height for each pellet sample. This rod of reference alanine was analyzed at the EPR spectrometer cavity before and after each series of measurement under identical analysis conditions (the same EPR resonance parameter and positioning in the cavity) of 2MA pellets. The mean of all reference measurements was used for correlating the peak height of 2MA dosimeter. Finally, the dose–response curve of the pellet dosimeter was established in terms of correlated signal height (peak height divided by pellet mass, peak height of reference alanine, and gain) against absorbed doses. Three pellets of 2MA dosimeters were used at each dose point to establish the dose response function. Then, all pellets were analyzed at the EPR spectrometer after 24 h from irradiation time under the same EPR analysis conditions.

3. Results and discussion

3.1. EPR spectra and dose response function

Fig. 1 shows the EPR spectra of 2MA pellet dosimeters irradiated to series of absorbed doses. The spectrum of unirradiated

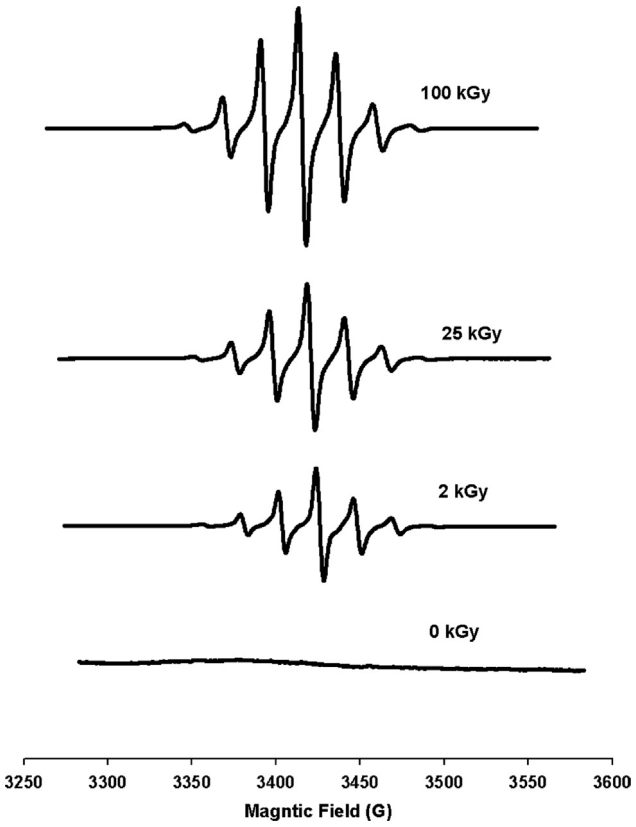

Fig. 1. EPR spectra of unirradiated and γ-irradiated 2MA pellet dosimeters.

2MA sample exhibits no EPR signal even at high microwave power and modulation amplitude. However, upon irradiation, the spectrum exhibits a nearly isotropic EPR signal with seven lines (Olsson et al., 2002), and its intensity grows with the radiation doses. The isotropic seven-lines in irradiated 2MA are mainly related to the interaction of unpaired electron with six equivalent protons of two methyl groups (Olsson et al., 2002; Chen et al., 2007). We selected the central line of the spectrum for further dosimetric investigations.

Fig. 2 shows the dose response function of the dosimeters investigated in the dose range of 1–100 kGy. The signal intensity increases with increasing absorbed doses with a non-linear trend over the whole dose range, however, the linearity is observed only in the low dose range of 1–11.4 kGy as seen in Fig. 3.

3.2. Effect of humidity during irradiation on dose response function

Fig. 4 shows the dependence of dosimeter response on relative humidity (RH) in the range of 0–95%. To adjust the RH, the peletts were suspended before irradiation in tightly closed jars over appropriate saturated salt solutions (Wexler and Hasegawa, 1954; Abdel-Fattah and Miller, 1996); except for 0 % RH, dried silica gel was used. The tightly closed jars were for 72 h to ensure equilibrium conditions, and then they irradiated simultaneously to 10 kGy. It is obvious from Fig. 4 that the dosimeter response is RH independent only in the range of 33–76%. However, out of this range, the dosimeter response is RH dependent. Thus, it is advisable to package the pellets during irradiation into sealed aluminum pouches in atmosphere with the range of 33–76% RH, to minimize errors of RH.

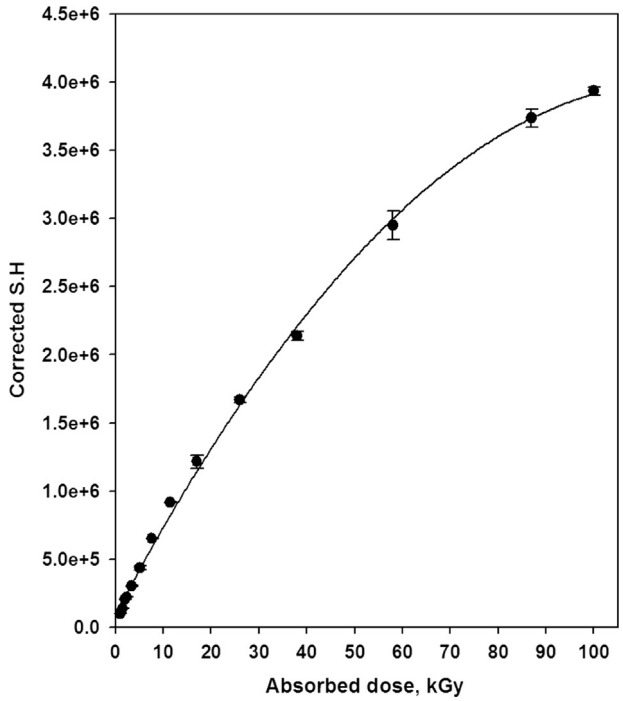

Fig. 2. EPR dose response of 2MA pellet dosimeter irradiated by ^{60}Co γ-rays in the dose range of 1–100 kGy. Error bars represent 2σ.

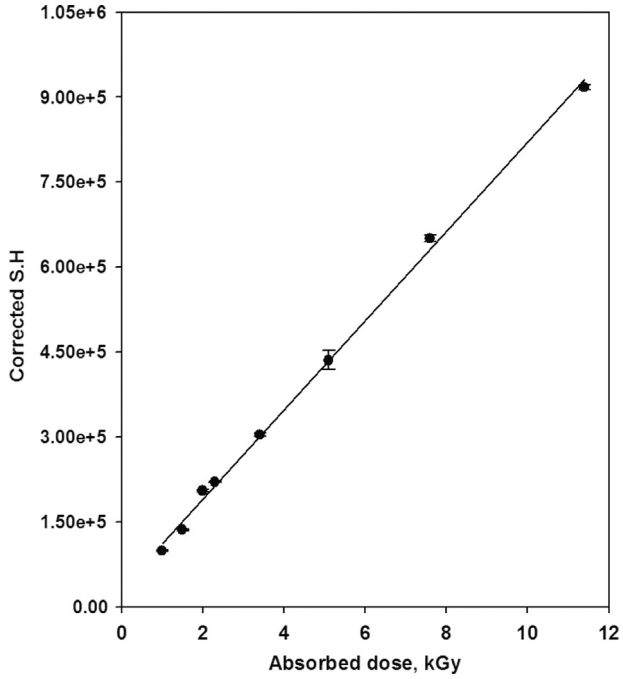

Fig. 3. EPR dose response of 2MA pellet dosimeter irradiated by ^{60}Co γ-rays in the dose range of 1–11.4 kGy. Error bars represent 2σ.

3.3. *Effect of temperature during irradiation on dose response function*

To study the effect of irradiation temperature on response, the dosimeters are maintained before irradiation in the temperature controller unit in the GC cavity at the required irradiation temperature for about 5 min. The dosimeters are analyzed by EPR spectrometer after 24 h from irradiation at room temperature. Fig. 5 shows the dependence of dosimeter response on irradiation temperature. The curve indicates that the dosimeter response

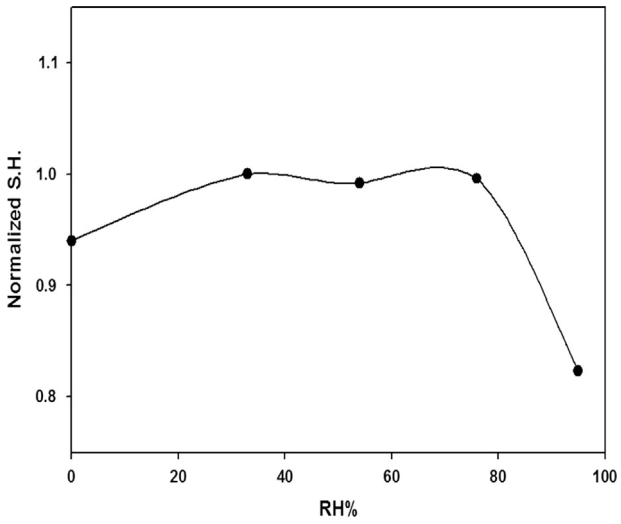

Fig. 4. Variation of normalized response of the dosimeter irradiated to 10 kGy as a function of RH during irradiation.

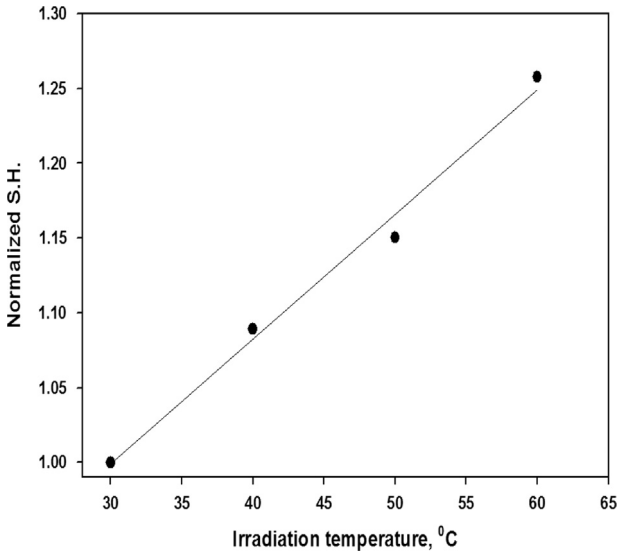

Fig. 5. Variation of normalized response of the dosimeter irradiated to 10 kGy as a function of temperature during irradiation.

increases linearly with increasing irradiation temperature with a temperature coefficient of ~0.96%/°C in 21–60 °C range. Thus, in order to minimize the effect of irradiation temperature on the dose response, calibration of pellet dosimeter in the production facility together with reference dosimeter is recommended.

3.4. *Energy dependence*

The energy dependence of dosimeter may cause errors in the absorbed dose estimation in the material of interest. Most many errors arise due to calibration of dosimeters under significantly different conditions of radiation energy. The mass energy-absorption coefficient, μ_{en}/ρ normalized to values of water versus photon energy in the range of 0.1–15 MeV are shown in Fig. 6 for 2MA powder, 2MA pellet dosimeters, alanine and adipose tissue. The present calculations were based on the data presented online at NIST physical reference data and in ICRU Report 44 (Hubbell and Seltzer, 2004; ICRU Report 44, 1989). It is clear that 2MA pellet dosimeter is nearly similar to 2MA powder, alanine and adipose tissue. Additionally, the manufactured 2MA pellets are nearly water equivalent in the photon energy level of 0.15–4 MeV;

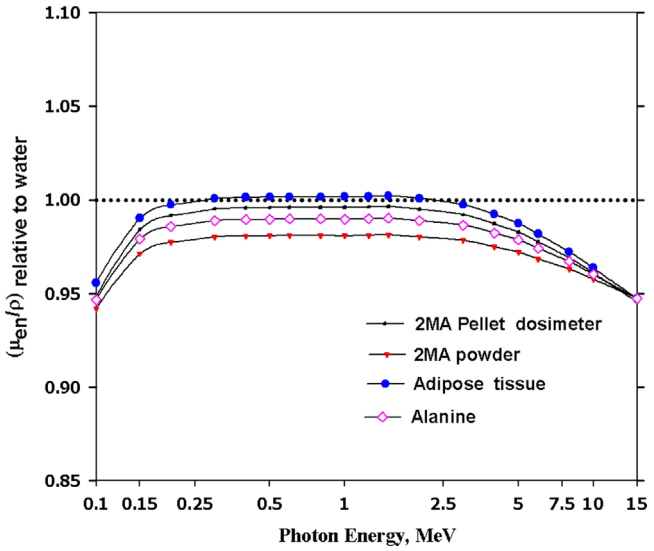

Fig. 6. The mass attenuation coefficient, μ/ρ, (a) and the mass energy-absorption coefficient, μ_{en}/ρ, (b) of 2MA pellet dosimeter normalized to adipose tissue versus photon energy in the range of 0.1 to 20 MeV.

Table 1
Uncertainty budget of 2MA pellet dosimeter in the dose range of 11.4–100 kGy, which can be suitable for radiation medical sterilization dose range.

Source of uncertainty	Type of uncertainty	Standard uncertainty (%)
Calibration irradiation dose rate	B	1.15[a]
Irradiation facility	B	0.25
Sensitivity variation of EPR spectrometer[b]	A	0.20
Reproducibility of EPR spectrometer[c]	A	0.33
Uniformity of dosimeter	A	1.57
Uncertainty of calibration fitting	A	2.38
Temperature effect on response	A	1.11
Combined standard uncertainty (u_c), 1σ		3.30
Overall uncertainty (2σ)		6.60

[a] As quoted from calibration certificate.
[b] Estimated from measurements of EPR signal intensity (h_o & h_{90}) of an irradiated 2MA pellet dosimeter for hundred times, while the dosimeter was fixed in the rectangular cavity.
[c] Estimated from measurements of EPR signal intensity (h_o & h_{90}) of an irradiated pellet for many readings, while the dosimeter was taken out and returned to the rectangular cavity between each measurement.

however, out of this range, the variation from water equivalency is within 6%.

3.5. Uncertainty of routine dose estimation

There are different sources can contribute for uncertainty of absorbed dose estimation such as calibration of the irradiation facility, irradiation facility, uniformity of dosimeters, EPR spectrometer measurement (instrument sensitivity variation and reproducibility of measurements), temperature and RH during irradiation, and calibration of dosimeters. The uncertainties may be grouped into two categories (ISO/ASTM 51707, 2004; Taylor and Kuyatt, 1994) type A (evaluated by statistical methods u_A from a series of repeated observations) and type B (evaluated by non-statistical methods u_B i.e. taken from manufacturer—supplied calibration).

Table 1 presents the uncertainty budget of dose monitoring in the high dose range of 11.4–100 kGy. Some of uncertainty components are explained in the footnotes under Table 1, while the others are discussed below.

The uncertainty related to batch uniformity of pellet dosimeters was investigated by irradiating three sets of pellets to different doses under the same identical conditions and by analyzing them immediately after irradiation at the same EPR spectrometer (Sharp and Miller, 2009). The coefficient of variation (standard uncertainty %) of the results is ~1.57% at σ.

The uncertainty from applied equation of calibration curve fitting was calculated from the residuals analysis according to the detailed discussion in the literatures (Sharp and Miller, 2009; Abdel-Fattah et al., 2012). The standard uncertainty of curve fitting for the low (1–11.4 kGy) and the high (11.4–100 kGy) dose ranges was estimated as ~2.52% and ~2.38%, respectively. These estimated values were calculated using the following formula (Sharp and Miller, 2009):

$$u = \sqrt{\frac{\sum_i (n_i - 1)(\sigma_i)^2}{\sum_i (n_i - 1)}}$$

where n_i and σ_i are the number of dosimeters and the standard deviation of the dose measurements at a given dose level, respectively. The standard deviations are generally expressed as percentage of the mean dose at each dose level.

The irradiation temperature will also influence the dose response function. The temperature coefficient of 2MA pellet is recorded as 0.96%/°C. Assuming that the difference between the irradiation temperature during, calibration and, routine dose monitoring is ± 2 °C, and the temperature effect has a rectangular probability distribution, the uncertainty will be ~1.11% according to the following equation (Sharp and Miller, 2009).

$$u(1\sigma) = \frac{(2 \times 0.96)}{\sqrt{3}} = 1.11\%$$

A similar analysis showed that the overall uncertainty of the measured doses was ~6.81% and ~6.60% (2σ, 95% confidence level) for the dose ranges of 1–11.4 kGy and 11.4–100 kGy, respectively, reflecting its suitability for use as a routine dosimeter in wide spread applications in radiation process control. It is assumed here that the irradiation conditions during routine dose monitoring are the same as those during calibration of the dosimeter. If the conditions are different, an unknown error may be observed in the estimated dose value.

4. Conclusion

Gamma irradiation of 2MA pellet dosimeter exhibits an EPR signal of seven lines. The EPR signal intensity grows linearly with increasing doses up to 11.4 kGy. The response of the pellet dosimeter is RH independent in the range of 33–76% RH during irradiation. However, it is temperature dependent in the range of 21–60 °C (temperature coefficient ≈0.96%/°C). The overall uncertainty (2σ) of the dosimeter is less than 6.9%, reflecting its potentiality for use in some food irradiation applications, medical sterilization, and polymer modifications.

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