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Preparation of New ESR Dosimetry System for High Dose Radiation Processing

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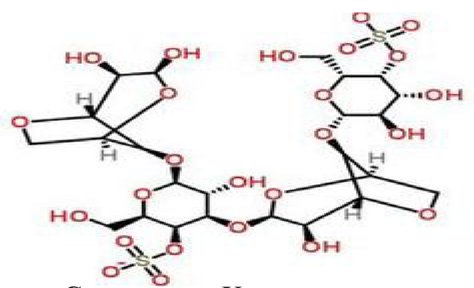
THE main objective of this work is to study the possibility of using the radiation sensitive carrageenan substance for routine process control in irradiation processing applications. The free radicals generated in the prepared carrageenan rods were investigated using the Electron Spin Resonance Spectrometer. The effect of relative humidity during irradiation on the response curve as well as short term and long term stability were investigated. The obtained data suggested the possibility of using the carrageenan substance in dose monitoring ranging from 0.5 to 50 kGy.

Keywords: Carrageenan, Electron Spin Resonance (ESR), Radiation Dosimetry, Irradiation processing.

Introduction

Ionizing radiation (gamma rays from Co^{60} and Cs^{137} sources, x- ray and electron beam) has been used for a long time in order to sterilize medical devices or to improve hygienic quality of food [1] or for sterilization of drugs and cosmetics [2,3]. Ionizing radiation dose assessment using an electron spin resonance ESR spectrometer increases widely. It provides accurate easy and fast procedure for dose estimation without seeking for complicated long practical procedures. Alanine/ ESR is considered the main dosimetry system for industrial applications since 1962 [4,5]. Although the application of ESR to radiation dosimetry yields several materials for use in transfer dosimetry at high and intermediate dose such as alanine and others, alanine possesses a complex time dependence [6]. In addition to its complicated spectrum which is attributed to not less than three radicals present [7-9], and its limited sensitivity at low doses that leads to large uncertainties [10]. Efforts to use ESR dosimetry in health physics have resulted in a minimum detectable dose of 10 mGy [11,12]. In the current study we investigated food grade, carrageenan substance for dose monitoring using the ESR technique.

Experimental



Carrageenan-Kappa

Scheme 1

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Preparation of Carrageenan Rods

An equal weight mixture of paraffin wax (molecular weight 282, congealing point 65 - 71^{0}C, BDH) and (ethylene vinyl acetate copolymer, EVA, hot-melt adhesive (molecular weight 164.01, Power Adhesive Limited, England) was melted in a round bottle at 85-95^{0}C in a water bath. EVA shows a complete compatibility with paraffin wax. Three concentrations of 20, 30, and 40 % fine powdered carrageenan material, "Carrageenan-Kappa, Molecular Weight 788.65764, Philippines source, .

Molecular formula is C_{24} H_{36} O_{25} S_2 " was added to the hot mixture solution and mechanically stirred for about 15 minutes at the same temperature to obtain a homogeneous mixture. The hot solution was pipetted into polypropylene tubes (inner diameter 3 mm) and was left to solidify by cooling. Carrageenan mixture rod was obtained by removing the polypropylene tube.

ESR Spectra Measurements and Dose Response Function

The free radical created in carrageenan rods by radiation in the range of 0.5-130 kGy at room temperature (21-25^{0}C) was detected by X-band EMX spectrometer (Bruker, Germany) using a standard rectangular cavity of ER 4102. Samples were inserted in ESR tubes, the bottom of each tube was adjusted at a fixed define position to ensure reproducible and accurate positioning of the rods in the sensing zone of the cavity. The stability of the ESR spectrometer and sensitivity were checked before and after each series of measurements using reference alanine dosimeter. Finally, the dose response curve was established in terms of correlated signal intensity against the absorbed doses. Three prepared carrageenan rods were irradiated at each dose point and analyzed by the ESR spectrometer.

Establishment of ESR Parameters for Irradiated Carrageenan Rods

Several ESR parameters such as microwave power, modulation amplitude, sweep width, time constant and conversion time were studied. The main reason is to choose the suitable parameters for establish good signal to noise ratio with highest amplitude, good reproducibility and resolution.

a) Microwave Power (MP)

Microwave power is one of the most important parameters in ESR measurements. It has a great effect on the spectrum shape, resolution and amplitude of signals. The intensity of ESR signal changes linearly with square root of MP till 2 $MP^{1/2}$ (1,4), then deviates from this linearity as shown in Fig (1). To obtain better sensitivity and good reproducibility, MP is determined from upper limits of the linear part of Fig. (1) and addressed in Fig. (2).

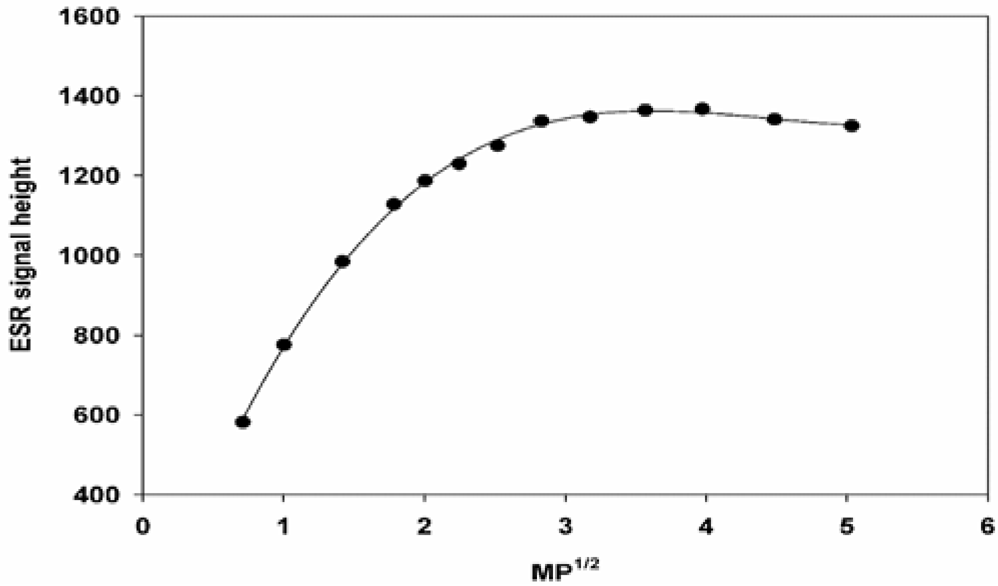


Fig. 1. Relationship between signal intensity of irradiated carrageenan and square root of microwave power ($MP^{½}$)

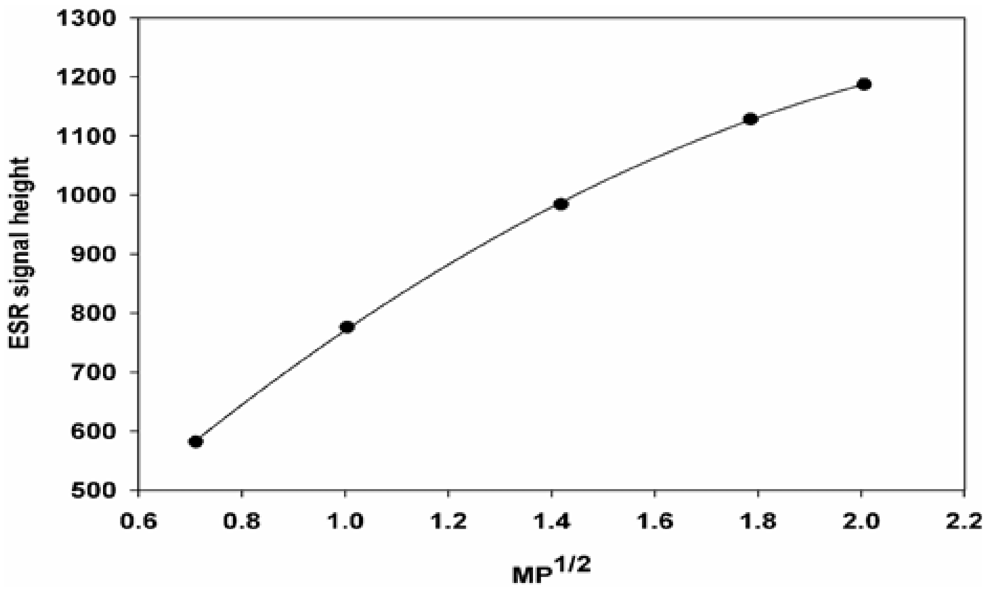


Fig. 2. Relationship between the upper limits of linear part in fig. 1 and Square root of microwave power ($MP^{1/2}$)

b) Modulation Amplitude, (MA)

The modulation amplitude is another important parameter that has great effect on shape, resolution and amplitude of the spectrum. The effect of the modulation amplitude on the ESR spectra has been studied by measuring the irradiated carrageenan rods that exposed to (5 kGy) at different modulation amplitude in the range of (0.5- 25 Gauss).

Figure (3) shows the ESR signal height as a function of modulation amplitude values. It can be noticed that the intensity of the ESR signal increases linearly up to 15 Gauss, then gradually increases up to 25 Gauss.

The operating parameters adopted throughout the experiment are microwave power =2.012 mW, modulation amplitude = 5.00 Gauss, modulation frequency = 100 kHz, sweep width = 600, microwave frequency = 9.70 GHz, time constant = 81.92 ms and sweep time = 20.97 s/scan.. The detection limits of ESR technique depend on the type of sample, sample size, detector sensitivity, frequency of the incident microwave radiation, and the electronic circuits of the instrument. All measurements of carrageenan rods were recorded at two orientation (0 and 90 degrees) and the mean of the two orientations was used.

Results and Discussion

Effect of gamma radiation on carrageenan rods

Three different concentrations of carrageenan namely 20, 30, 40% were irradiated to a dose of 30 kGy. The ESR spectra were shown in Fig. (4), The figure reveals increase in the peak intensity with increase of Carrageenan concentration in the rods.

Fig. (5) represents the ESR spectra of un irradiated and irradiated carrageenan rods (40%) recorded at a series of absorbed doses from 0.5-130 kGy. The figure (5a) elucidates that the ESR signals were developed upon irradiation and its amplitude increases with the absorbed dose of gamma ray photons without any change in its shape till 130 kGy, Figure (5b) showed the calculated change difference in the range 0.5 to 50 kGy (40% concentration) reach to about 15 times increase in signal height. On contrary the change difference in the range 50 to 130 kGy was 1.87 times increase in signal height only.

Dose response curve for carrageenan rods

Figure (6) represents the response curves of 3 sets of Carrageenan concentrations (20, 30, and 40%) irradiated to different absorbed dose range from 0.5 - 130 kGy. The response curves obtained for the irradiated samples expressed in terms of average peak to peak amplitude normalized to dosimeter mass (peak height/mass) versus the absorbed dose were recorded. It can be observed that the radiation sensitivity increases with the increase of carrageenan percentages in the rods. This radiation sensitivity boost is attributed to the increase of free radicals resulting from exposure to gamma photons.

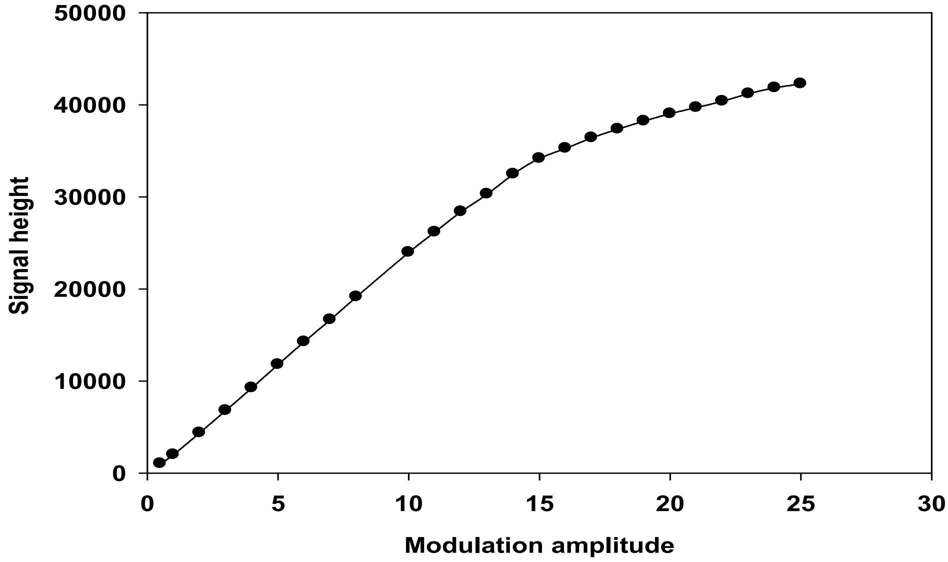


Fig. 3. ESR signal hight for an irradiated sample at (5 kGy) as a function of modulation amplitude in the range (0.5- 25 Gauss).

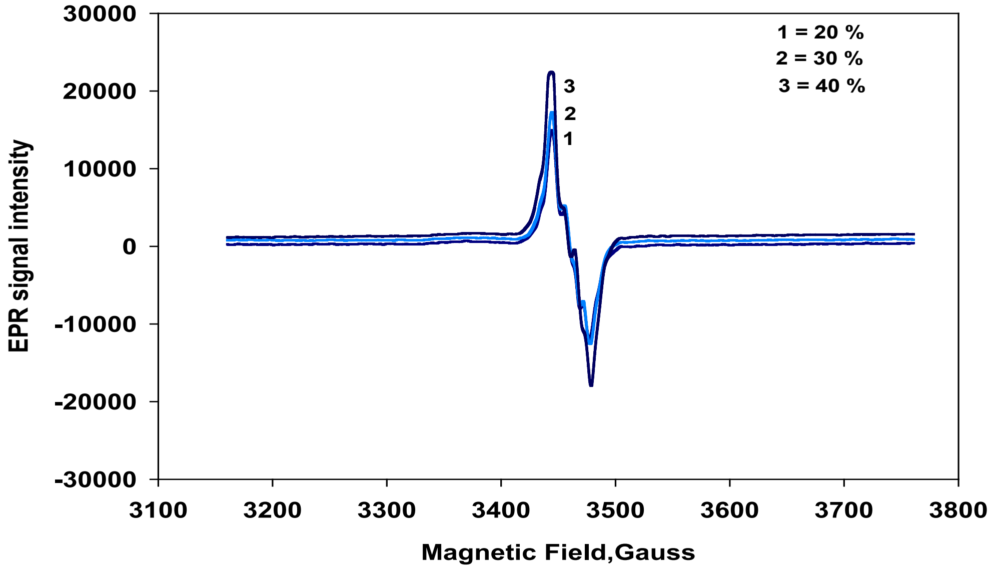


Fig. 4. ESR spectra recorded for carrageenan rods irradiated to a dose of 30 kGy at different concentrations.

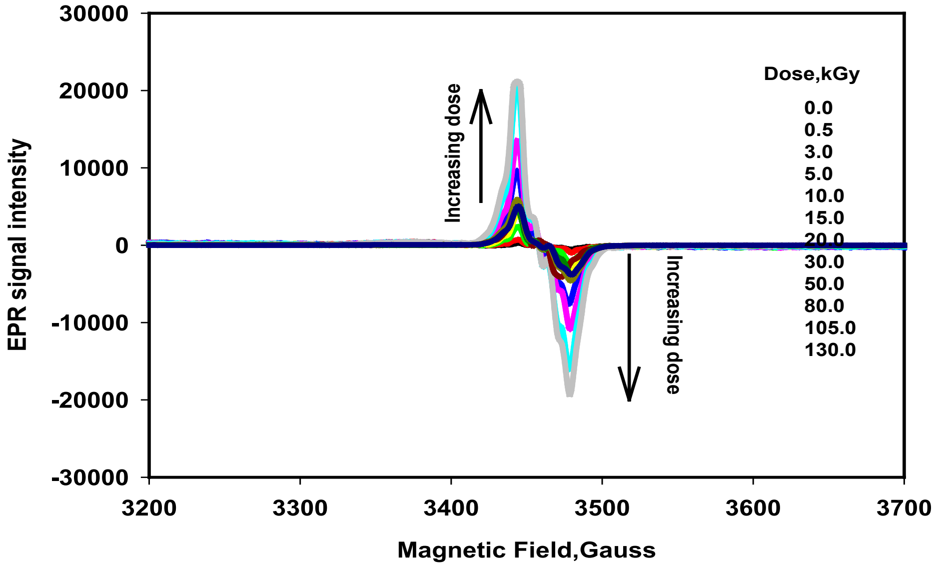
Post irradiation stability of carrageenan rods

a) Short – term post irradiation stability

Short term post irradiation stability test was carried out for carrageenan rods (concentration 40%) to a dose of 10 kGy, The test continued for six hours. Each of ESR signals recorded through the designed test period was divided on the immediately recorded value after irradiation Fig. (7) shows the obtained fading behavior of ESR signals. As shown the intensity values of the relative ESR signals are instable during the first 30 minutes and stabilize after an hour. So we can suggest a "cool down" period one hour before proceed any following measurements.

b) Long term post - irradiation stability

The post- irradiation stability of carrageenan rods of concentration 40 % stored at (35 % RH) and at room temperature (25 ± 3^{0}C) under laboratory fluorescent light after irradiation was studied. Five rods were irradiated to dose of 25 kGy. The ESR signal height of the rods was recorded immediately after irradiation. The rods were investigated through the 70



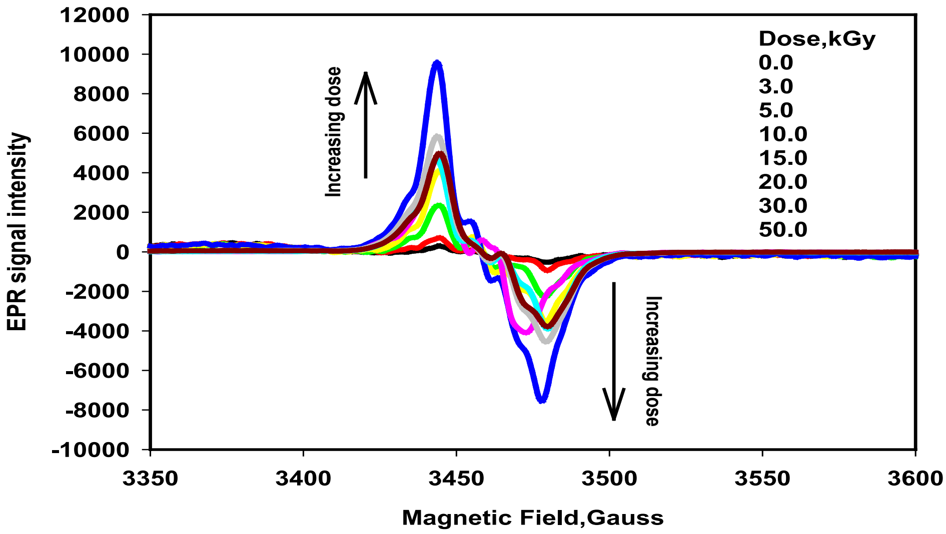


Fig. (5 a, b) ESR spectra of irradiated carrageenan rods (40%) at different absorbed doses.

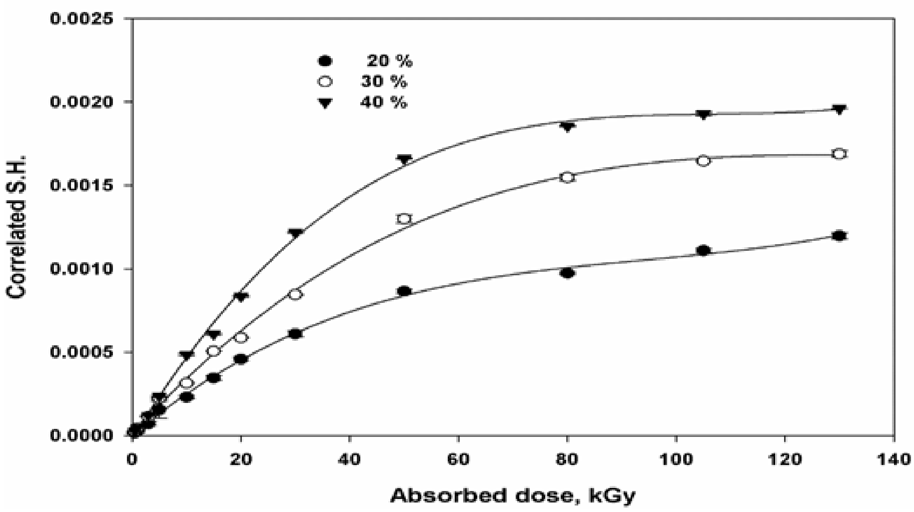


Fig. 6. Dose response curves for irradiated carrageenan rods at different concentrations.

days storage period. Fig. (8) shows dramatic decrease in relative signals response during the first two days, then gradual decrease till twenty days completion. So the optimal time for measurement is after just one hour and not exceeding 6 hours. Correction should be done if the measurement takes place in any periods out of the recommended time range expressed in this study.

Humidity during irradiation

Many materials used for dosimetry absorb water readily. The effects of humidity on the response of many high- dose dosimetry systems have been investigated [13] . Humidity effects may result in a different dose – response function, these effects can lead to important uncertainties in dose estimation. The response and stability of some systems can be optimized by a correct pre-

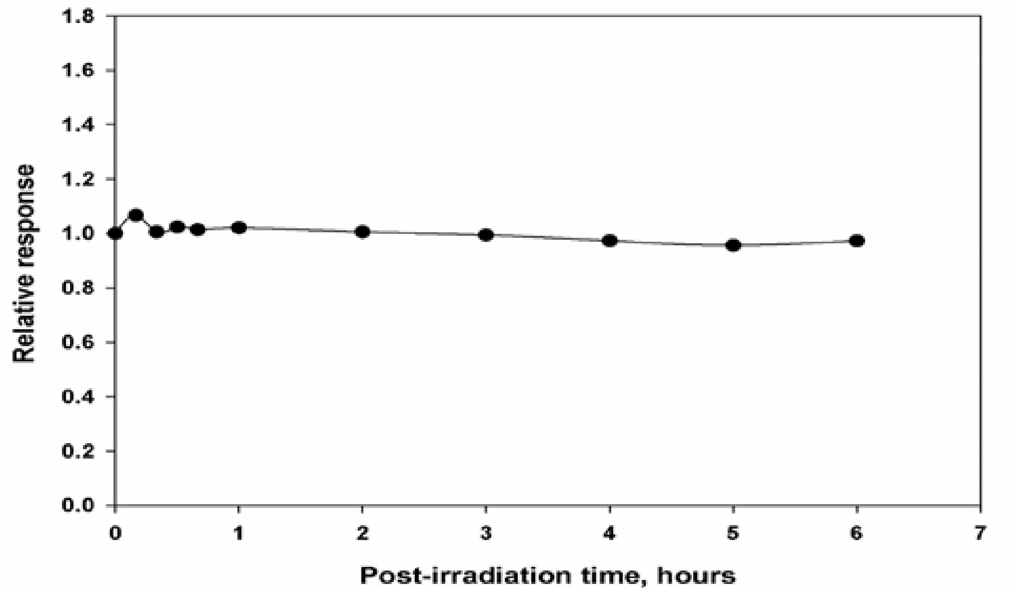


Fig. 7. Stability of ESR signal detected in carrageenan rods upon gamma irradiation to 10 kGy relative to a signal measured immediately after irradiation

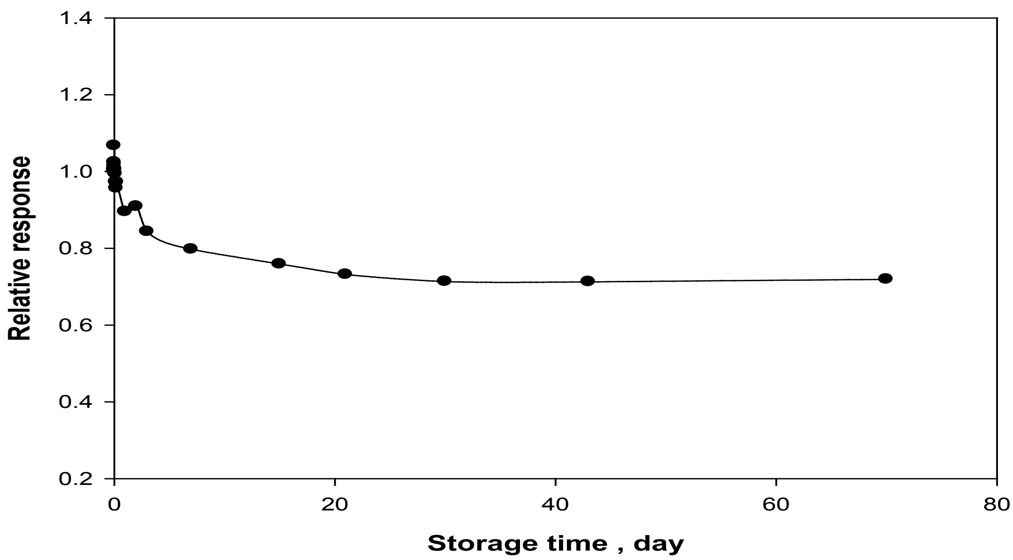


Fig. 8. Stability of ESR signal detected in Carrageenan rods (40%)nupon gamma irradiation to 25 kGy relative to signal measured immediately after irradiation as a function of storage time .

irradiation adjustment of the dosimeter moisture content.

The effect of relative humidity during irradiation on the response of carrageenan (40% concentration) was investigated by irradiating the rods to a dose of **25** kGy at different relative humidity values (0-95%). The rods were stored before irradiation for three days period over various saturated salt solutions in tightly closed vials seeking for establishing moisture equilibrium in the rod before and during irradiation. Fig. (9) shows the variation in ESR signal intensity as a function of relative humidity during irradiation with respect to the response value at 33 % relative humidity. It can be concluded that carrageenan rods almost have negligible humidity effect in the range of humidity from 0 to 95 %.

Uncertainty in the estimated dose value

Uncertainty of measurements may be defined as a parameter associated with the result of a measurement that characterizes the dispersion of the values that could reasonably be attributed to the measured. [14,15], The uncertainty may be grouped into two categories, type A (evaluated by statistical methods UA from a series of repeated observations) and type_B evaluated by non –statistical methods UB i.e. taken from manufacturer – supplied calibration. There are different sources that could contribute for uncertainty such as calibration of the irradiated facility, Reproducibility of EPR spectra measurement , Sensitivity variation of EPR measurement, Stability of EPR spectrometer Angular positioning (orientation effect), Balance used, all these sources contribute by ±3.1 at **(1σ).** The total estimated uncertainty at **(2σ)** (95% confidence limit) was 6.2%.

Conclusion

From the represented data in this study, the following conclusion can be addressed. Carrageenan rods can be suitable dosimeter in a dose range of 0.5-50 kGy. The dosimeter has insignificant dependence on the change of relative humidity during irradiation. The properties of the prepared carrageenan rods suggest their useful application as dosimeter for food irradiation and medical irradiation process with ± 6.2% uncertainty. The dosimeter is fairly stable after irradiation during the first 6 hours (excluding the initial first 30 minutes) and correction should be done for any measurements out of the specified range mentioned in this study. It is recommended to calibrate this dosimeter under the same conditions of use.

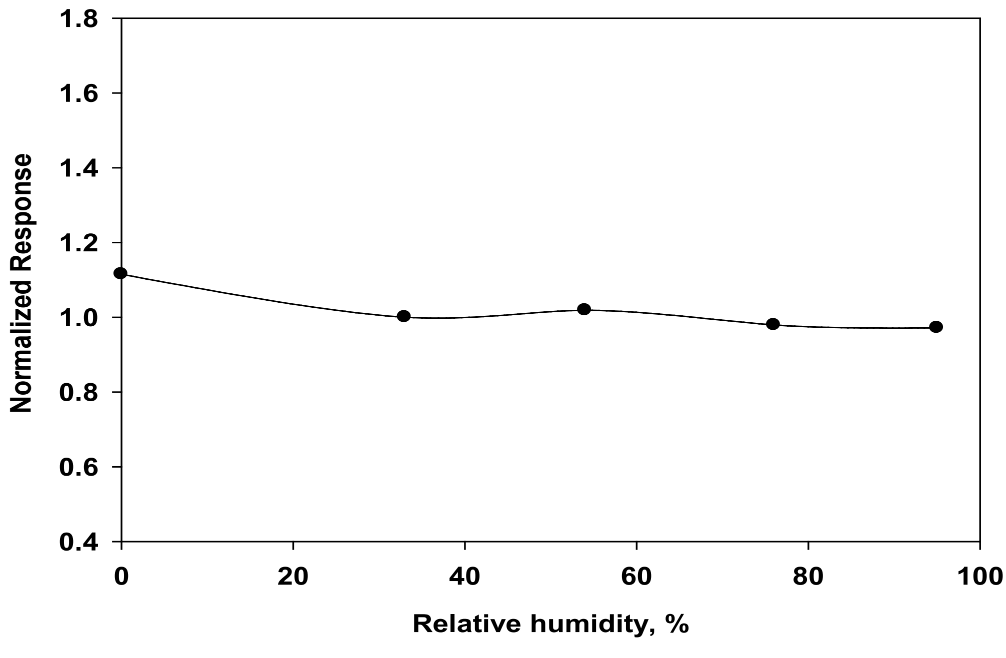


Fig. 9. Variation of ESR response of carrageenan rods (40%) as a function of relative humidity during irradiation at 25 kGy.

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