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Estimation of the Absorbed Dose in Gamma Irradiated Food Containing Bone by Electron Spin Resonance Spectroscopy

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Abstract

The use of electron spin resonance (ESR) spectroscopy to assess the absorbed dose to radiation-processed food containing bone is examined. The exposure of foodstuffs containing bone to a dose of ionizing radiation results in the formation of long lived free radicals that give rise to characteristic ESR signals in the bone. The yield of radicals was found to be proportional to absorbed dose. Additive re-irradiation of previously irradiated bone was used to estimate the original absorbed dose in the irradiated chicken bone. Simple non-linear rational equation was found to fit to the data and provide a good estimate of the initial dose in the range (1.0 - 4.9 kGy). Decay of the ESR signal intensity was monitored at a dose of 3.0 kGy up to 100 days. The absorbed dose in irradiated chicken (2.0 kGy) was assessed at 2, 6, and 12 days after irradiation. Relatively good results were obtained when measurements were made within the following days (up to 12 days) after irradiation. These data as well as the ability of the dose additive method to provide accurate dose assessments are presented.

Introduction

Ionizing radiations is used to reduce food losses and improve safety, wholesomeness, and nutritional quality of food products. Distinguishing irradiated from unirradiated food is a concern of consumers and regulatory officials (1, 2). Physical , chemical, and biological tests are being developed to routinely monitor and regulate trade of irradiated food. Also, it is important to verify quantitatively unspecified radiation doses to food, where the legal permissible dose to food may vary from one country to another (3). For meats containing bone , reliable dosimetry method has been reported using electron spin resonance (ESR) spectroscopy (4). The ESR method is based on the measurement of long-lived free radicals produced in bones as a result of irradiation. Additive re-irradiation of a bone sample generates a dose response which is used to estimate the initial dose. Different mathematical expressions such as linear or polynomial and exponential functions were used to describe the ESR response to the absorbed dose in bone. It was found that the exponential fit to the data provides improved accuracy of the estimated dose (5).

In the present study, the use of the exponential equation and another equation (Non linear- rational equation) to estimate the absorbed dose for previously irradiated chicken bones are evaluated. Post-irradiation stability of the radiation-induced free radicals in bone and effect of time after irradiation on the estimated dose were also investigated.

Experimental

Preparation of bone fragments

Chicken bones from local markets were scraped of excess meat, fractured to expose the marrow, dried over silica gel in a vacuum oven at room temperature and left for one week before irradiation. Bone fragments were cut to a height sufficient to fill the height of the microwave resonator (approximately 3.0 cm length).

Irradiation

Irradiations were carried out in the ^{60}Co gammacell 220. The absorbed dose rate was measured to be 12.7 kGy/h using Fricke dosimetry (6). All the dose values given are expressed in terms of absorbed dose in water.

ESR measurements

ESR spectra were performed with JEOL TE300 X-band ESR spectrometer, interfaced with computer data acquisition and analysis system, with the following settings: modulation amplitude 0.2 mT, microwave frequency 9.4 GHz and microwave power 12.5 mW. The values of the time constant and sweep rate were chosen to give an ESR signal with peak to peak line width of approximately 0.4 mT. Stability of the ESR spectrometer sensitivities was checked periodically using several alanine dosimeters irradiated to known doses. The peak-to-peak heights of the first derivative ESR spectra of irradiated bones were measured at room temperature. Positioning of the bone fragments was fixed always in the same direction in the cavity. The dose responses of bone were calculated from the measured peak-to-peak height per unit weight of bone, normalized to the gain of the ESR spectrometer. A computer software (Table curve, 2D, Jandel Scientific Co.) was used to fit the data.

Upon exposure to ionizing radiation, free radicals are produced in mineralized bone tissue and can be detected by ESR. The ESR signal of irradiated chicken bone is spectroscopically distinguishable from the unirradiated bone ESR signal. The ESR spectral features of irradiated and unirradiated chicken bones were previously discussed (7,8).

Results and Discussion

Reproducibility of the ESR measurements

The reproducibility of the ESR measurements of irradiated chicken bone was studied by removing and reinserting one bone fragment irradiated to 2 kGy in the ESR cavity in arbitrary orientation and ESR signal amplitude was measured 50 times. The results indicate a measurement uncertainty of 4.6 % (2σ) . When the sample was left in the ESR cavity between measurements (50 times) the uncertainty was found to be 0.9 % (2σ).

ESR response and fitting analysis

It was demonstrated previously (9) that the ESR response of irradiated bone tissue can be described by an exponential function, namely,

$$S_D = S_\infty \, [1-\exp(-(D_o + D`)/D_{37})] \ldots\ldots\ldots\ldots\ldots (1)$$

Where, S_D is the ESR signal intensity at the added dose D`, S_∞ is the saturation value, and D_{37} is the reciprocal of the dose at 63 percent of the saturation value. D_o is the original (unknown) radiation absorbed dose.

Another simple non-linear rational equation (2) was used here and found to correlate well with the data, although the saturation characteristics of this equation are not specified :

$$S_D = a + cD / 1 + bD \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots \quad (2)$$

Where, S_D is the ESR signal intensity at the added dose D and a, b and c are constants.

To test the accuracy of the dose additive method in order to assess the initial dose for bones, forty bone fragments were prepared and grouped into 4 sets (10 bones for each set) and irradiated to four doses: 1, 2, 3 and 4.9 kGy. The signal amplitude for each bone was measured immediately after irradiation and the bones were then re-irradiated with a series of additive doses (up to 70 kGy). The ESR spectra of irradiated bones were recorded at each dose, with measurement of signal amplitude for each bone, and both exponential and rational functions were applied to the data.

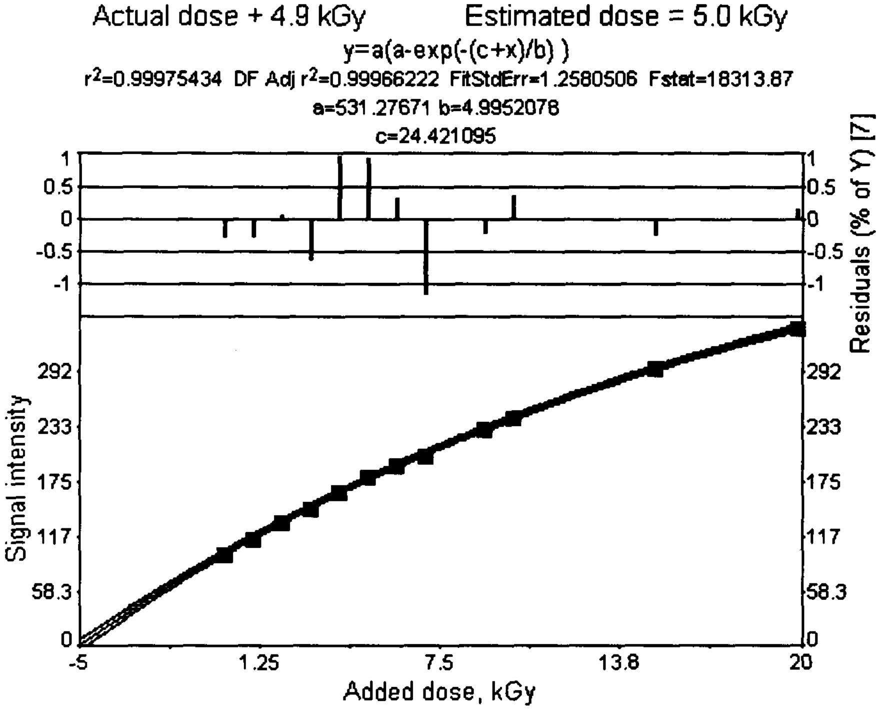

Fig. 1 Relationship between ESR signal amplitude / amplification and added absorbed dose (kGy), for irradiated chicken bone (4.9 kGy). (The curve is a computer generated fit using equation (1). Lines around the fitted curve represent the 95% confidence intervals).

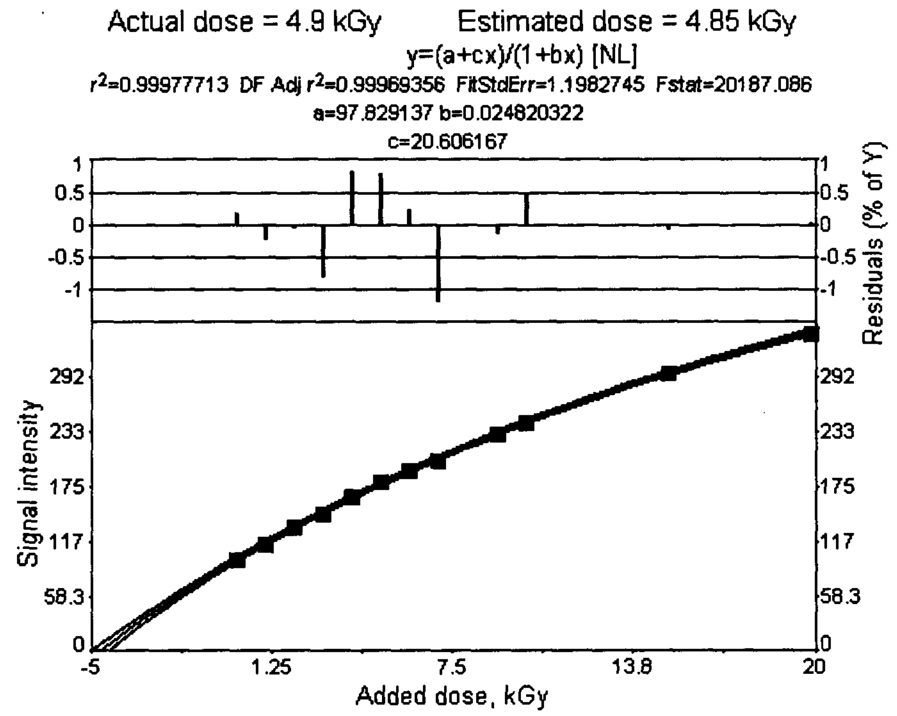

Fig. 2 Relationship between ESR signal amplitude / amplification and added absorbed dose (kGy), for irradiated chicken bone (4.9 kGy). (The curve is a computer generated fit using equation (2). Lines around the fitted curve represent the 95% confidence intervals).

Figures 1 and 2 show the obtained responses of an irradiated bone fragment (4.9 kGy) with the respective fits, 95 % confidence bands (lines around the curve), and residuals for equations 1 and 2, respectively. It can be observed that the 95 % confidence bands are very tight and are very close to the actual fitted curve, even in the region of extrapolation for both equations. The goodness of fit is evaluated by several parameters that are obtained as a part of the fitting process. Two of such parameters are the correlation coefficient, r^2, and F-statistic. It can be seen that the correlation coefficient is almost the same for both equations, while F-statistic value for equation (2) is higher than that for equation (1), which indicates that relatively better fit to the data could be obtained by using equation (2). Also, it can be observed that the obtained small percentage differences (residuals %) between the actual values of signal height and those predicted by the curves indicate a good fit for both equations. The initial doses (unknown) for all bone fragments were determined by back-extrapolation of the response curves to the negative dose axis (abscissa) (see Figs.1 and 2). The dose estimates and the associated uncertainty (at 95 % confidence limit) at each dose provided by both equations are in good agreement as seen in Table (1).

Table 1 Estimation values of initial dose (in kGy) for different mathematical fits to the bone ESR dose response

Actual Dose (kGy)	Estimated Dose (Exponential Fit)	Estimated Dose (Rational Fit)
1.0	**1.02 ± 0.07**	**0.94 ± 0.24**
2.0	**2.40 ± 0.40**	**2.30 ± 0.30**
3.0	**2.88 ± 0.50**	**2.90 ± 0.60**
4.9	**5.10 ± 0.30**	**4.80 ± 0.40**

Estimated uncertainty limits are expressed as the 95 % confidence level

Post-irradiation stability and the effect of time of evaluation on the accuracy of the estimate.

The stability of radiation-induced free radicals in chicken bone was studied by irradiating a bone fragment to 3 kGy and then stored in the laboratory at ambient temperature ~22°C and relative humidity of ~50%. The ESR signal amplitude of the radiation induced signal was measured at different intervals of time during the post-irradiation storage period of 3 months. The percentage decrease in signal amplitude

Table 2. Effect of the time after irradiation on the estimated Dose

Storage Time (day)	Actual Dose (kGy)	Estimated Dose (kGy)	
		Exp. Fit	Ration. Fit
2	2	2.12 ± 0.2	2.2 ± 0.3
6	2	1.8 ± 0.3	1.9 ± 0.2
12	2	2.3 ± 0.2	2.3 ± 0.1

Estimated uncertainty limits are expressed as the 95 % confidence level

It can be observed that no significant difference is seen among the dose assessed for bones stored to 2, 6, or 12 days after initial irradiation to 2 kGy. It is also observed that both equations (1 and 2) when applied to the data yielded similar results. In fact, the available data in the literature on the decay of radiation induced free radicals in bone, under different storage conditions, are limited. Studies on the decay of radicals in eggshell (apatite) suggests that corrections for decay should be applied to the ESR signal amplitude to obtain better estimate of the original dose (10). However, other studies on the effect of decay of radical on the accuracy of dose estimate in irradiated chicken bone revealed that the time of evaluation (up to 20 days post-irradiation) did not affect the estimate for 0.5 and 3.0 kGy bones, but moderately affect the 7.0 kGy estimates (11). Therefore, detailed studies of the effect of time of evaluation on the accuracy of dose estimate in irradiated chicken bones is needed.

Conclusions

The above results indicate that the ESR detection method is clearly able to distinguish between irradiated and unirradiated bones. Concerning the dose as a function of storage time is shown in Fig.3. It can be observed that the radiation induced free radicals decay rapidly in the first few days after irradiation and then stabilize over a long period of time. Over this storage period (3 months) the signal amplitude decreased by about 40 % of its initial value.

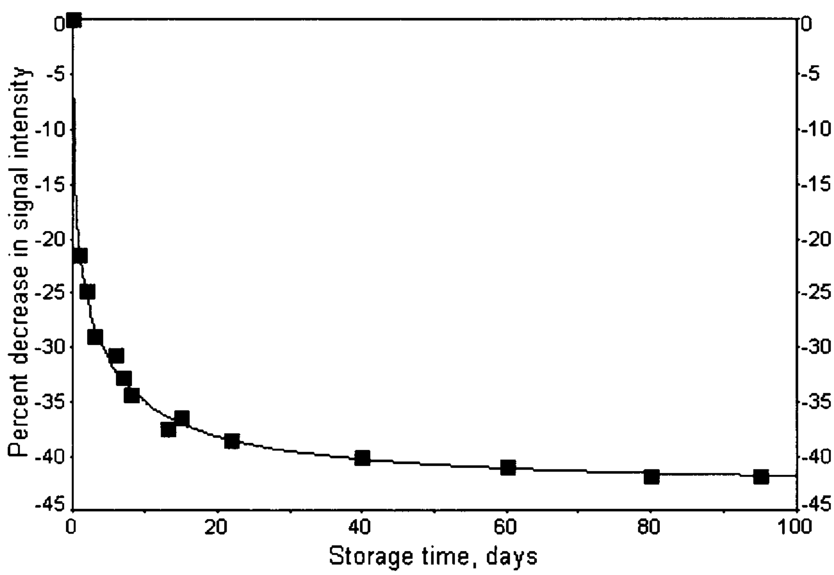

Fig.3 Percent decrease of ESR signal amplitude for chicken bone irradiated to 3 kGy as a function of storage time

The accuracy of the dose additive method to assess the initial dose for bones irradiated to 2 kGy was tested as a function of the post-irradiation time evaluation. The estimates for the dose level (2 kGy) derived from equations 1 and 2, at different storage times, are summarized in Table 2.

estimation, the preliminary results presented in this paper, using exponential and non-linear rational fits, are encouraging. Although reasonable evaluations of dose estimates are supplied by the rational fit up to 5 kGy, further studies will be made on large batches of bone fragments covering the whole dose range (1-10 kGy) permitted for radiation processing of chicken.

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