

Evaluation of the Accuracy of the Additive Dose Method for Dose Assessment in Irradiated Refrigerated Chicken Bone

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Abstract. This paper reports an extensive study to evaluate the accuracy of the additive dose method to estimate the absorbed dose in irradiated refrigerated chicken bone. In this method additive re-irradiation of previously irradiated bone produces a reproducible dose response function which can be used to evaluate the initial dose by back-extrapolation. An exponential equation was found to fit the data and provide a good estimate of the initial dose in the range of 1.0- 10.0 kGy. The influence of the decay of the radiation-induced free radicals on the ESR bone dosimetry has been studied. The absorbed dose in irradiated chicken (1.0 to 10.0 kGy) was assessed at 14 and 21 days after irradiation. This data as well as the ability of the dose additive method to provide dose assessment is presented.

Introduction

Ionizing radiation is used to reduce food losses and improve safety, wholesomeness and nutritional quality of food products. Distinguishing irradiated food from unirradiated food is a concern of consumers and regulatory officials [1-4]. 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. For meats containing bone, reliable detection methods have been reported using electron spin resonance (ESR) spectroscopy [5-7]. In the present study, the use of the exponential equation to estimate the absorbed dose for previously irradiated chicken bones, post-irradiation stability of the radiation-induced free radicals in refrigerated bone and effect of time after irradiation on the estimated dose have been investigated.

Experimental

Refrigerated chicken purchased from local markets were scraped of excess meat and fractured to expose the marrow. Bone fragments taken only from the tibia were 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 (33 x 3 mm). On these bone fragments a mark was placed at one end so that it could be placed repetitively in the same direction in the ESR tube.

Irradiation

Gamma-ray doses were delivered at ambient temperature using ^{60}Co Gamma Cell 220 from MDS Nordion Inc., Canada (Absorbed dose rate in water = 12.7 kGy/hr as calibrated by Fricke dosimetry). The temperature of irradiation was kept constant at 5°C during all irradiation's by purging cold air inside the gamma chamber using air chiller (FTS Systems Inc., USA). This temperature was chosen to simulate the irradiation and storage temperatures of refrigerated chicken.

A plastic container made of polystyrene with 2 mm thickness was designed to irradiate bone samples under the conditions of electronic equilibrium and to ensure that the bone fragments are placed exactly at the center of the irradiation chamber. The container consisted of a flat circular base and cylindrical chamber attached to it through a vertical column. The chamber housed symmetrically and circularly arranged six holes in which bone fragments could be placed.

ESR measurements

Before carrying the ESR measurements, the chicken bones were dried, by using a freeze drier (FTS System Inc., USA), to prevent sensitivity losses in the ESR measurements. ESR spectra were recorded 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. The peak-to-peak heights of the first derivative of ESR spectra of irradiated bones were measured at room temperature. During the course of this investigation, the ESR amplitude was normalized by the spectrometer gain. After recording the ESR spectra on the day of irradiation, the samples were placed in closed glass bottles at a storage temperature of 5°C for 7, 14 and 21 days. Spectra were measured after each storage period and analyzed as described above.

The reproducibility of the ESR measurements of irradiated chicken bone was studied by removing and reinstating one bone fragment irradiated to 2 kGy in the ESR cavity in an arbitrary orientation and the 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).

A computer software (Table curve, 2D, Jandel Scientific Co., USA) was used to fit the data and calculate the estimated doses from the additive dose response.

Results and Discussion

The accuracy of the additive dose method to assess the initial dose for irradiated bones during storage was tested as a function of the post-irradiation time. Two sets of bone fragments (24 bones for each set) were prepared and irradiated to the doses in the range from 1 to 10 kGy (3 bones for each dose) at 5°C and stored at the same temperature. The signal amplitude for both sets of bones were measured after the storage periods and the estimated doses in bones were calculated by applying the additive dose (over the range 1-10 kGy) on each stored bone. The ESR readout and re-irradiation were performed all in the same day for each bone. Figure 1 shows the dose response of an irradiated bone (1 kGy) as obtained by measuring the ESR signal amplitude after 21 days and then re-irradiated with a series of additive doses up to 100 kGy with measurement signal amplitude at each dose.

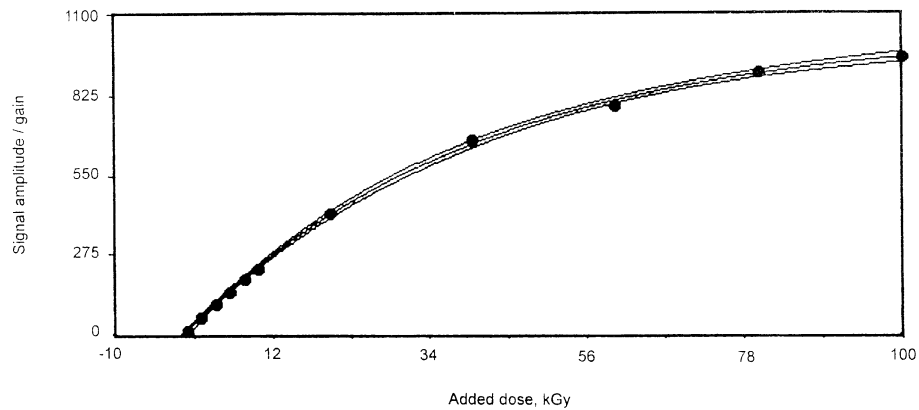


Fig.1. Relationship between (ESR signal amplitude/gain) and added absorbed dose (kGy), for irradiated (1 kGy) chicken bone. (The curve is a computer-generated exponential fit. Lines around the fitted curve represent the 95% confidence intervals).

The measured signal amplitudes were corrected for the spectrometer gain at each dose and plotted vs. added doses. The exponential function ($S_D = S_\infty [1 - \exp(-(D_0 + D) / D_{37})]$), where S_D is the ESR signal intensity at the added dose D ; S_∞ is the signal saturation value, D_{37} is the dose at 63% of the saturation value and D_0 is the original radiation absorbed dose[6], was applied to the data as seen in Fig. 1. The initial dose for

the bone fragment was determined by back-extrapolation of the response curve to the negative dose axis (abscissa) (Fig. 1). The above mentioned procedure was repeated for all bones stored for 14 and 21 days.

Table 1 shows the estimated doses in bones stored for 14 and 21 days and the uncertainty limits. It can be observed that the estimated doses at both storage periods are lower than the actual doses. Estimates obtained on post-irradiation storage for 14 days averaged lower than those stored for 21 days with the exception of samples estimated at 9 and 10 kGy. The results indicate that the decay of free radicals in bone have a marked influence on dose assessment and lead to an underestimation of the dose.

Table 1. Estimated doses calculated by additive dose method in irradiated bones (1-10 kGy) after storage for different time periods at 5°C

Actual dose (kGy)	After 14 days of storage estimated dose (kGy)	After 21 days of storage estimated dose (kGy)
1	0.47 ± 0.07	0.54 ± 0.22
2	1.08 ± 0.66	1.60 ± 0.11
3	1.76 ± 0.56	2.64 ± 0.14
4	2.56 ± 0.33	3.52 ± 1.22
5	3.53 ± 0.88	4.82 ± 1.10
6	4.49 ± 0.69	5.97 ± 1.36
7	6.09 ± 1.65	7.01 ± 1.10
8	6.34 ± 0.93	7.76 ± 0.99
9	8.78 ± 1.35	7.83 ± 0.95
10	10.45 ± 1.60	7.43 ± 1.58

Note: Data fit to a function of the form $S_D = S_\infty [1 - \exp(-(D_0 + D) / D_{37})]$ [6]. Estimated uncertainty limits are expressed at the 95% confidence level.

This conclusion is in agreement with the results of Onori and Pantaloni [8]. They suggested the use of correction factor for long term decay by measuring the ESR signal amplitude of irradiated bone after it reached stability (10 days). The reasons for over estimation in the case of 10 kGy is yet to be unraveled. In order to correct the estimated doses for long term decay of free radicals, the stability of the radiation-induced free radicals in bone was studied by irradiating 8 sets of bones (5 bones for each set) to different doses (1 to 10 kGy) and then stored at 5°C after irradiation. The ESR signal amplitude of the radiation-induced signal in bones was measured at different intervals of time (7, 14 and 21 days) during the post-irradiation storage period. The percentage decrease in normalized signal amplitude as a function of storage time is shown in Table 2. It can be observed that the radiation-induced free radicals decay rapidly in the first 7 days after irradiation and then stabilize. It can also be observed that the fading is dependent on the dose and this conclusion is in agreement with the results of Onori, *et al.* [9]. The estimated doses in Table 1 were corrected for long-term decay by applying the correction factors derived from the data presented in Table 2.

Table 2. Percent decrease in normalized signal amplitude for bones irradiated to different doses and stored after irradiation for different periods at 5°C. Five bones for each dose were tested

Dose (kGy)	Day 7 (%)	Day 14 (%)	Day 21 (%)
1	16	18	18
2	15	16	16
3	15	16	17
4	16	16	17
5	12	14	13
6	12	16	14
7	7	11	10
8	9	13	13
9	10	13	13
10	8	13	13

Correction of short-term decay was not applied in this case since the ESR readout and re-irradiation was performed all in the same day [9]. The corrected estimated doses and uncertainty limits are summarized in Table 3. The data presented in this table indicate that better dose estimates can be obtained by applying correction factors for decay of free radicals.

Table 3. Estimated doses calculated by additive dose method and corrected for decay in irradiated bones (1-10 kGy) after storage for different periods at 5°C

Actual dose (kGy)	After 14 days of storage estimated dose (kGy)	After 21 days of storage estimated dose (kGy)
1	0.56 ± 0.05	0.66 ± 0.17
2	1.29 ± 0.50	1.91 ± 0.46
3	2.10 ± 0.43	3.18 ± 1.00
4	3.05 ± 0.25	4.24 ± 0.94
5	4.10 ± 0.66	5.54 ± 0.82
6	5.35 ± 0.53	6.94 ± 1.02
7	6.84 ± 1.19	7.86 ± 0.62
8	7.28 ± 0.68	8.91 ± 0.73
9	10.09 ± 0.99	9.00 ± 0.69
10	12.00 ± 1.17	8.93 ± 1.03

Note: Data fit to a function of the form $S_D = S_\infty [1 - \exp(-(D_0 + D) / D_{37})]$ [6]. Estimated uncertainty limits are expressed at the 95% confidence level.

Conclusion

From the data presented in this paper, it can be concluded that the dose additive method can be used for a reliable evaluation of the absorbed dose in refrigerated chicken bone. Also, the long term decay of the radiation-induced radical has a marked influence on dose assessment and leads to an underestimation of the dose. Therefore, in order to obtain more accurate dose estimates, correction factors accounting for the long-term

decay of the radiation-induced free radicals in bone must be determined and applied. In this case, the operator needs to know the exact decay behavior of the radicals and the influencing factors on decay in chicken bone.

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تقويم دقة طريقة الجرعة المضافة في تقدير الجرعة الإشعاعية في عظام الدجاج المبرد بعد التشعيع

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ملخص البحث. تعتبر طريقة الجرعة المضافة باستخدام جهاز الطنين الدوراني الإلكتروني (ESR) من أكثر الطرق نجاحاً في تقدير الجرعة الإشعاعية الممتصة في عظام الدجاج بعد التشعيع. تستعرض هذه الورقة نتائج دراسة مستفيضة لتقويم دقة الجرعة المضافة في تقدير الجرعة الإشعاعية الممتصة في عظام الدجاج المبرد بعد التشعيع. باستخدام هذه الطريقة يتم إعادة تشعيع عظام مشعة مسبقاً وذلك عدة جرعات إشعاعية بحيث تعطي هذه العظام إشارات (ESR) بتكرارية جيدة يمكن استخدامها في تقدير الجرعة الإشعاعية الأساسية بتمديد منحني الرسم إلى المحور السيني. كما أوضحت الدراسة دقة المعادلة الأسية (Exponential) في رسم نتائج التجارب ، حيث أعطت تقدير جيد للجرعة الإشعاعية الأساسية في مدى ١-١٠ كيلو جراى . كما تمت دراسة تأثير اضمحلال الشقوق الحرة التي تم توليدها بالإشعاع على تقدير الجرعة الإشعاعية حيث تم تقدير الجرعة الإشعاعية الممتصة في عظام الدجاج المعالج بالإشعاع بعد ١٤ يوماً و ٢١ يوماً من يوم التشعيع في مدى ١-١٠ كيلو جراى. تستعرض هذه الورقة جميع النتائج السابقة إضافة إلى دقة الطريقة المضافة في تقدير الجرعة الإشعاعية الممتصة في عظام الدجاج المعالج بالإشعاع.