Fingernail dosimetry using electron spin resonance for radiation disaster response

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Abstract. This study investigates the use of electron spin resonance (ESR) signals from human fingernails for retrospective dosimetry as part of radiation disaster response, focusing on the variabilities of individual responses to radiation. Samples of fingernails were collected from 7 adult donors (Asian type) and irradiated to 35 Gy and 70 Gy of gamma-rays from a Cs-137 source at a dose rate of 0.857 Gy/min. All irradiated fingernails were measured for 39 days with an X-band ESR spectrometer and stored in darkness inside the vacuum desiccator (30% humidity, 20°C) in between measurements at all times. All samples were harvested using one specific nail cutter and given no other special treatments. It was observed that the measured radiation-induced signals faded on about 10-12% after 1 day of exposure. Though the signal intensities showed a significant difference among the donors, stronger linearities in the dose responses were observed in the samples of younger donors. From the results obtained in this study, it is expected that fingernails would be a useful tool for retrospective dosimetry in case of an unexpected radiological accident or medical treatment error associated with exposure in therapeutic dose range, as far as the individual-based calibration curves were available. Further investigations will be made to clarify the reason for the different responses by using the fingernail samples taken from a greater number of donors of different ages and lifestyles.

1. Introduction
Electron spin resonance (ESR) spectroscopy has been long utilized as a useful, preferable technique for retrospective dosimetry. It is a physical dosimetry technique used to measure stable free radicals or point defects in matter caused by reactions with ionizing radiations under the assumption that the
measured radical densities in the matter are linearly related to the radiation absorbed dose. The intensity of the ESR signal from these radicals, known as the radiation-induced signal (RIS), can be detected in a wide range of materials [1–6]. The most advanced ESR dosimetry technique is with the use of tooth enamel [7,8], which has been verified to be a useful and reliable retrospective dosimeter through several interlaboratory comparisons [9–13]. Whereas, the drawback is that tooth enamel demands invasive surgery from the subjects. This fact, in turn, highlights the potential use of fingernails as they can be collected with a non-invasive procedure without causing discomfort.

In the last years, there has been a growing interest using fingernails as a potential dosimetric tool and thus as a source of ESR measurement rather than tooth enamel. Recent research has brought about significant developments in the application of fingernail dosimetry [14–16]. See also a recent review by Marciniak and Ciesielski [17]. One of the latest attempts has been published on in vivo ESR fingernail dosimetry during total body irradiation (TBI) procedures [18]. However, the data are limited in medical applications that require fundamental researches. Additionally, there are still main factors responsible for technical limitations on the application of ESR fingernail dosimetry. The complex structure of the ESR signal in fingernails was reported to be affected by mechanic-induced signal (MIS) [19] and its native background signal (BKG) [4]. Sholom and McKeever [20,21] also described that the confounding of the ESR spectrum in fingernails is the sensitivity to environmental humidity and light exposure. The signal stability is also considered a highly critical issue since the decay rate is 50% within 24 hours [4] and this remains to be an immense challenge for the researchers.

In the present study, we investigate the medical application of ESR dosimetry using human fingernails by examining the signal stabilities linked with storage conditions and other influential factors, focusing on the variability in individual dose responses of the ESR signals from the fingernails of different age/sex donors.

2. Materials and Methods

2.1. Sample preparation

Samples of fingernails were voluntarily provided by seven adults (3 male and 4 female, Asian type) for this study. The donors performed the collection of samples by themselves during regular hygienic procedures using one specific nail cutter. Right after clipping, the samples from the right/left hands of each donor were pooled and placed in sealed small plastic bags before sending to the research laboratory. The samples were then stored in darkness at room temperature (20°C) inside the vacuum desiccator with built-in pump (VE-ALL 1-8989-01, AS ONE, Osaka, Japan). Most of the harvested samples were approximately 1-2 mm wide and 4-5 mm long. Each sample mass consisted of several pieces totaling 20 mg. The samples were given no additional cuts or any special treatment between harvesting and irradiation and were always prepared on the same day as the irradiation experiment.

2.2. Irradiation

The collected samples were irradiated in vitro under controlled exposure conditions with doses of 35 Gy and 70 Gy gamma-rays using a Gammacell40 Exactor Low Dose Rate Research Irradiator (Best Theratronics Ltd., Canada) located at the Radiation Isotope Facility of Kasumi campus, Hiroshima University in Japan (See Figure 1). The equipment has dual Cs-137 sources with a total radiation activity of 178 TBq; it is regularly checked for its dose rate, and the dose uniformity is ± 7% in the whole area of the sample container. The size of the sample container is 260 mm in diameter and 100 mm in height. As of October 2018, the recorded dose rate was 0.855 Gy/min. It should be noted that high doses were used to generate a strong and clear shape of RIS that could dominate the ESR spectrum and to simulate an accidental exposure. Three sets of fingernails for each individual were irradiated in order to evaluate the reproducibility of the results and for further analysis. Moreover, one set of samples from each donor was kept unirradiated as control and measured at the same time as the irradiated sets.
Figure 1. Experimental set-up for irradiation of the fingernail samples with Gammacell40 Exactor Low Dose Rate Research Irradiator (left) and geometry of samples to be irradiated placed inside the sample container (right).

2.3. ESR measurement
The ESR spectra acquisitions were performed at room temperature with an X-band ESR spectrometer (JES-FA 100, JEOL Inc., Chiba, Japan) in the microwave frequency of ~9.4 GHz coupled with the ES-UCX2 standard cavity. The samples were measured in a 5 mm quartz sample tube with a covered black sheet and positioned at the center of the cavity. The acquisition parameters were as follows: microwave power of 1 mW; sweep width 10 mT; modulation width 0.25 mT; sweep time 60 s; time constant 0.03 s; total number of scans 10; total recording time 12 mins (which included sample loading and spectrometer tuning times). During spectra acquisition a MgO: Mn²⁺ standard sample was fixed at the bottom of the ESR cavity. Quantitative analysis of the spectra which included baseline corrections and measurements of the peak-to-peak amplitude of the central line (i.e. between the 3rd and 4th line of MgO: Mn²⁺) was carried out using the A-System Data Processing software (version 3.9.2.0, JEOL Resonance Inc., Chiba, Japan). All the peak-to-peak amplitudes analyzed in this work were considered as the magnitude of the ESR signal of the fingernails, with subtracted BKG signal (i.e. unirradiated samples, 0 Gy). Measurements were conducted at various times after irradiation: 5 mins, 6h, 12h, 24h, 3d, 5d, 7d, 14d, 21d, 28d, and 39d. It should be mentioned, however, that in periods between sample collection and all subsequent procedures (i.e. irradiations and ESR measurements), all the samples were always stored in darkness inside the vacuum desiccator (30% humidity) at room temperature.

3. Results and Discussion

3.1. Dose-response
The dose-response plots and linear regression curves in irradiated fingernails are shown in Figure 2. Note that the RIS intensities shown here and elsewhere in this paper were obtained by subtracting the BKG signals from the main singlet intensities. Each plot is an average of the data of three samples at the first measurement or 5 mins after irradiation. Error bars were included in the plot symbols. Data points were best fit with a second-order function \( y = ax + bx^2 \). Summary of the linear regression data including the fitting parameters, \( R^2 \), and slope values for fingernail samples are shown in Table 1. Though all \( R^2 \) values were good (0.90 to 0.99), it was observed that the samples taken from younger donors (i.e. donors 1-6) showed greater linearities in this dose range (up to 70 Gy) than from the samples from the older donor (i.e. donor 7). Significant variability was also found between the dose-response curves among the donors. It was noted during the study that the fingernails taken from donor 7 were thicker, more brittle, and yellowish in color than the samples collected from donors 1-6. This observed variation may be related to the age-related changes in the components of the fingernails that might have occurred due to the influences of environmental and chemical factors (sunlight, water from daily washing, nail cosmetics, etc.).

Based on the comparison of the dose responses among different-age donors, it is suggested that calibration curves are not solely related to the age but dependent on individual physiological properties. For instance, there was 11%-33% difference in dose-response between the slopes of
Figure 2. Dose-response curves of vacuum-stored samples irradiated to gamma-rays from the Cs-137 source. Each data set is fitted with a second-order function of the form $y = ax + bx^2$.

Figure 3. Decay curves of the radiation-induced ESR signals from the samples exposed to (a) 35 Gy and (b) 70 Gy gamma-rays from the Cs-137 source. All experimental data points were normalized to the initial measurements for 35 Gy exposure.

Table 1. Summary of linear regression data including $R^2$ and dose-response (slope) values from irradiated fingernail samples.

| Donor number | Age | Linear equation | $R^2$ value | Parameters of the second-order function $y = ax + bx^2$ |
|--------------|-----|----------------|-------------|---------------------------------------------------|
| 1            | 27  | $y = 0.086x$   | 0.99        | $a = 0.12$ $b = -5.7 \times 10^{-4}$ |
| 2            | 27  | $y = 0.080x$   | 0.99        | $a = 0.10$ $b = -2.4 \times 10^{-4}$ |
| 3            | 27  | $y = 0.063x$   | 0.99        | $a = 0.08$ $b = -2.5 \times 10^{-4}$ |
| 4            | 26  | $y = 0.063x$   | 0.99        | $a = 0.07$ $b = -1.3 \times 10^{-4}$ |
| 5            | 30  | $y = 0.054x$   | 0.96        | $a = 0.09$ $b = -5.9 \times 10^{-4}$ |
| 6            | 45  | $y = 0.049x$   | 0.99        | $a = 0.07$ $b = -3.0 \times 10^{-4}$ |
| 7            | 63  | $y = 0.060x$   | 0.90        | $a = 0.14$ $b = -1.2 \times 10^{-3}$ |
donors of the same age (i.e. donors 1-3). It is thus desirable to continue further investigations for verifying these results and establishing a method for precise correction of the individual differences by using more fingernail samples from a greater number of people including children.

3.2. Signal Stability

Figure 3a and Figure 3b illustrate the variations in time of RIS intensities in vacuum-stored samples irradiated to 35 Gy and 70 Gy, respectively. The experimental data points are fitted in the function $y = a - bx^c$. The results presented here show that the measured RIS intensity faded on about 10-12% after 1 day of exposure and further decreased to 55-64% during its fifth week of measurement. Our preliminary results are also in agreement with the previous study of Sholom and McKeever [20] for water-treated vacuum-stored samples with 10% decreased in RIS intensity; it is attributable to the different storage time. Additionally, the storage in the vacuum desiccator in between sample collection following irradiations and ESR measurements showed improvement in the stability of RIS.

4. Conclusion

The individual responses and stabilities of the ESR signals in human fingernails of seven healthy-state adults exposed to different doses were investigated. In the dose-response data, it was seen that the linearity results tended to be stronger in the samples of younger donors. While, a significant variation in the signal levels was observed among the same-age donors. Also, patterns of signal reductions after irradiation were different among the samples of different-age donors. These results indicate the importance of making individual-based calibration curves for practical use of this dosimetry method.

From the findings obtained in this study, it is expected that human fingernails could be a potential retrospective tool for an unexpected radiological event or medical treatment error accompanied with radiation exposure in therapeutic dose range, as far as the dose response was established on an individual basis. Thus, we plan to collect and examine a greater number of fingernails from the people of different ages and lifestyles to clarify the reason of the different responses of the ESR signals (i.e. efficiencies of radical formations) and to establish the method for correcting such potential variations, which is critically important for practical applications of this method to real, complex situations.

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