Technical Note

Retrospective dosimetry using fingernail electron paramagnetic resonance response

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ABSTRACT

Human fingernails were used to estimate the radiation dose via electron paramagnetic resonance measurements of radiation-induced radicals. The limiting factors in this research were mechanically induced electron paramagnetic resonance signals due to the mechanical stress during the preparation of the samples. Therefore, different treatment methods of fingernails were used to reduce the mechanically induced signals. The results demonstrate that the mechanically induced and radiation-induced signals have apparently different microwave power saturation behaviors. In addition, the mechanically induced signal shows a fading evolution over time and reaches a constant value. Chemical treatment using the different reagents showed that the minimum mechanically induced signal was obtained using the dithiothreitol reagent. The dose—response curves of the samples treated with dithiothreitol for 30 minutes demonstrated a greater linearity than those of samples treated for 5 minutes. Therefore, to find an unknown absorbed dose in a fingernail sample using a calibration curve, we recommend adopting the mentioned chemical treatment procedure to reduce the uncertainty.

1. Introduction

Since the second half of the 20th century, a variety of releases of radioactive materials from industrial facilities and military program activities or overexposure of persons by the improper use and disposal of radiation sources has been experienced. These events resulted in a broad range of ionizing radiation exposure to a considerable number of people. Experience has proved that despite all precautions, radiation accidents occur. According to the Radiation Emergency Assistance Center/Training Site Radiation Accident Registries, in the period of 1944—2004, there were 421 major radiation accidents worldwide [1]. Therefore, the development of a noninvasive and reliable method that can produce results immediately after a radiation event is highly demanded for measuring radiation dose. Electron paramagnetic resonance (EPR) biodosimetry is a physical method based on measurement of the stable radiation-induced radicals in calcified tissues of the human body [2–7]. The application of this method for future assessment of radiation risk coefficients in epidemiological cohorts is relatively recent, having started 5—7 years ago. The individual dose can best be reconstructed using probes that are close to, or part of, the exposed individual. Therefore, human tissues are of special interest to retrospective dosimetry. Tooth enamel and bone are known for radiation dosimetry [8–12]. Tooth enamel is the most fully mineralized tissue of a human body. It has been used for more than three decades as a detector for *in vivo* dosimetry [9]. The use of fingernails as an EPR radiation dosimeter has a number of potential advantages, including high sensitivity [13–16] with estimated dose limits as low as 1—2 Gy; much more facile sampling than hematologic-based biodosimetry sampling because it does not require drawing blood; and *in situ* measurement of the incident, which does not require transport of the sample to a different site, thus avoiding the considerable logistical problems of linking back the individual with the sample under disastrous conditions. The radiation-induced EPR signal persists for many hours and is dose proportional. If needed, the signal can be preserved indefinitely by storage at low temperatures [17]. Moreover, there is a problem that cutting fingernails generates mechanically induced signal (MIS). In 1995, some researchers presented evidence that the dominant MIS species is a sulfur-centered radical [13,18,19]. The MIS has its own spectral parameters such as shape, g-factor, and line-width that are

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quite similar to the radiation-induced signal (RIS) [15,19–21]. Calculations that do not take these characteristics into account obviously overestimate the absorbed dose, e.g., the MIS could give a dose offset up to 10 Gy.

There are several possible approaches to try to solve this problem: annealing of the sample to accelerate fading of the MIS; numerical deconvolution of the fingernail spectrum to determine the radiation response from the complex spectrum; chemical treatment of the cut fingernails to oxidize the radical further; or a chemical reduction of mechanically induced signals.

MIS can be separated into two signals: MIS1 and MIS2. MIS1 is dominant during the first few hours after cutting and decreases drastically. MIS1 is thought to be originated from the elastic change in the fingernail structure because of the cutting process. MIS2 is very small at the first stage but becomes dominant after 24 hours. MIS2 is reported to stem from the plastic change of the fingernail structure. So, MIS2 is generally maintained for few weeks after the cutting process. Therefore, owing to the large MIS, the estimation method using the dose–response curve has been found to be arduous when trying to ensure reliability below 5 Gy [18].

In this article, attempts are made to achieve some experience in our laboratory of the factors affecting the EPR response of fingernail samples and to determine how we can refine the RIS from affecting factors such as MIS.

2. Experimental

2.1. Sample preparation

Using a normal fingernail clipper, fingernail samples were cut from three persons. The collected samples were cut into small clippings 2-mm wide and about 10-mm long. An ordinary electric heater was used for the heating process of the fingernail samples.

2.2. Chemical treatment

Freshly cut fingernails from the right/left hands of the donor (~100 mg) were pooled and split into different portions, and each portion was treated for 5 minutes with 500 μL aqueous solutions of the different reagents (Table 1). One of the samples was not treated and was kept as the control. All the samples were rinsed with 500 μL deionized water for 5 minutes, separated by microfiltration, treated with 500 μL acetone for 3 minutes, and separated again. The samples were then dried in a vacuum oven for 50 minutes at room temperature.

2.3. Irradiation

The samples were covered with plastic and weighed. Irradiation was carried out with the two ^{60}Co γ-ray source facilities of a Picker V9 (USA). Correction was made for decay of ^{60}Co

radionuclide and a subsequent decrease of the dose rate. The samples were irradiated to doses of 0.1 Gy, 0.25 Gy, 0.75 Gy, 1, 3 Gy, 5 Gy, 7 Gy, and 10 Gy.

2.4. EPR measurement

The samples were put into quartz thin-wall EPR tubes, 4-mm in diameter, and were measured with a Bruker EMS-104 spectrometer operating in X band. The EPR signal intensities were measured as peak-to-peak height for the most intense EPR lines, viz., the first derivatives of the absorption spectra per sample mass. To ensure the reproducibility of the EPR signal intensity, the samples were examined in the same instrument settings. The EPR spectrometer parameters used for this study were 100 kHz modulation frequency, 0.05 mT modulation amplitude, variable microwave power, 40 msec time constant, 21 s sweep time, and variable number of scans.

3. Results and discussion

3.1. Effect of physical treatment

In fact, an EPR spectrum with a low level of moisture is more stable than that of the normal fingernail samples. Nevertheless, reducing the water content in the sample is time-consuming under normal room conditions. On the other hand, reducing the water content induces unwanted increase of MIS.

As can be seen in Fig. 1, after the cutting process, the shape transformation of the EPR signal is very rapid, and at least 20 hours is needed for stabilization. Therefore, it is impossible to make any measurement and comparison during the first day after the cutting period. To shorten the period of reduction, an artificial heating process using an electric heater for 3 hours at 70°C was used in our experiment. After the heating process, the water content of the sample evaporated, and the final EPR signal became similar to the one after 20 hours, as can be seen in Fig. 1. Therefore, the fingernail clippings were heated and measured for MIS and then were exposed to 5 Gy of gamma ray and measured for RIS. Fig. 2 shows the characteristic behavior of the EPR signals against the square root of microwave power. Indeed, the water content of the fingernail samples prevents the exact observation of the power saturation behavior in MIS or RIS. After the dehydration process, EPR signal increment shows a difference between MIS and RIS, as can be seen in Fig. 2. This difference can be interpreted as a result of MIS and RIS having apparently different microwave power saturation behavior, as can be observed in the figure.
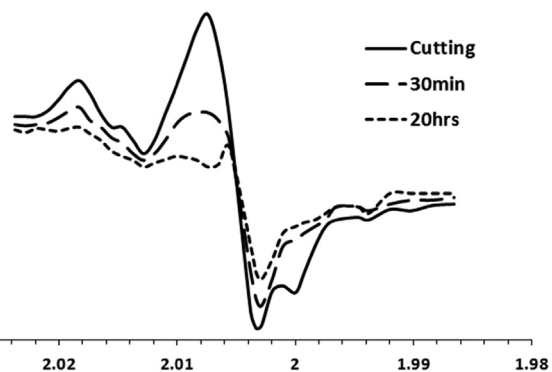

Fig. 1. Transformation of EPR signal spectrum of MIS after cutting process without heating.

Table 1
Effect of different chemical reagents on the EPR response of the sample MIS. All samples were treated for 5 minutes with dithiothreitol.

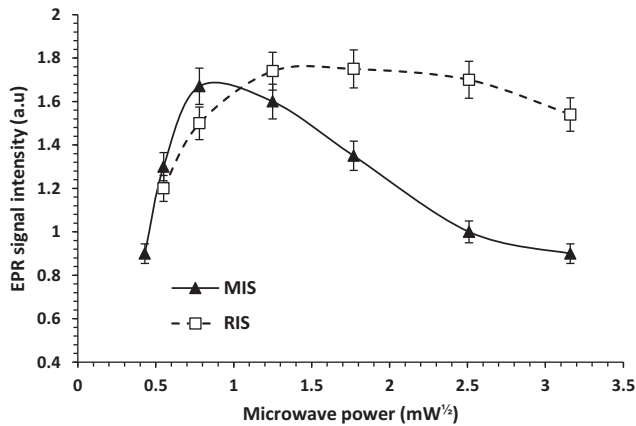| Chemical reagents	Mass (mg)	Mass-normalized EPR response (a.u.)
H_2O_2	9.8	0.55
Acetone	9.8	0.55
Hydroxylamine	10.5	0.50
Urea	9.8	0.33
Sodium thioglycolate	9.8	0.39
Dithiothreitol	9.4	0.24
None (control)	11.0	1.00

Fig. 2. Variation of MIS and RIS against the square root of microwave power.

From this experiment, we conclude that if we reduce water content, we can easily determine a relationship between the radiation exposure dose and the EPR signals. In other words, the EPR signal implies that the absorption of the microwave energy is not due to the MIS or RIS radicals but due to the water content inside the fingernail clippings.

3.2. Effect of cutting

Three different cutting lengths were applied to the irradiated fingernail sample selected from the same donor. The EPR measurements were performed immediately after cutting. Fig. 3 shows the EPR response variation of the irradiated and freshly cut fingernail samples with time after cutting. It can be concluded from this figure that the MIS shows fading evolution with time. In addition, the decrease in the signal intensity reaches 70% and then does not vary anymore, as evidenced in Fig. 1. On the other hand, the signal intensity of the sample with the higher cutting length decreases more rapidly. This means that the greater the cutting length is, the more mechanically induced radicals and the more rapid fading there will be.

Fig. 4 shows the variation of the EPR response after each cut and the humidification versus the cutting area of the irradiated fingernail samples. The humidification was localized on the edge of the cuts. As can be seen, the signal intensity decreases after each cycle of cutting and humidification. The decrease in the EPR response reaches 60% after about 5 mm² and then does not vary anymore. This result shows that the signals measured after
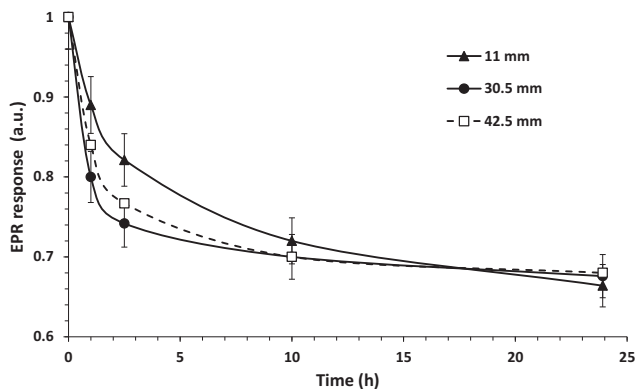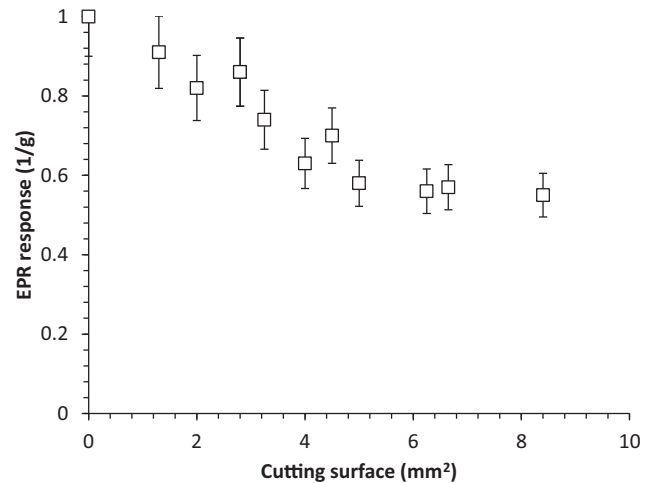

Fig. 4. EPR response via cutting area of the fingernail samples (normalized to the signal intensity of the first measurement). The cut edges were humidified to eliminate MIS after each cut.

humidification do not vary with the mechanical stress. In reality, the signal intensity should have remained constant, but as shown in another research work, the BKG signal is also affected by the humidification, but in a very different way than the MIS [19].

3.3. Effect of chemical treatment

Experimental results regarding the EPR response of the unirradiated sample (MIS) using the various chemical treatments from one donor (no.1) are presented in Table 1. The sample having the minimum MIS was obtained using dithiothreitol reagent for 5 minutes; this was the best result. This treatment reduced the MIS by a factor of about 4 in comparison with the control sample. Therefore, this chemical reagent was selected for treatment and measurement of the dose response in the irradiated fingernail samples. The results conform to those obtained by the other researchers [21].

Fig. 5 shows the dose−response results of the irradiated fingernail samples treated at various times by the same chemical reagent dithiothreitol. It can be seen that 20 minutes after the
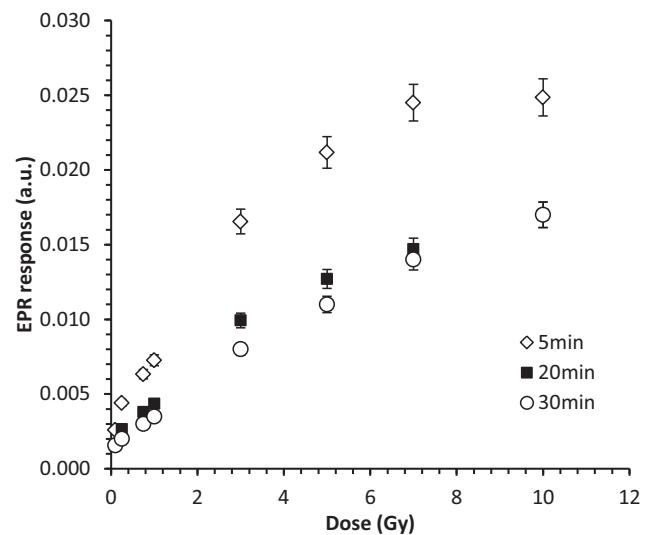

Fig. 3. Variation of the MIS via time in irradiated and freshly cut fingernail samples from the same donor with the different cutting length.

Fig. 5. Dose−response curves of the irradiated fingernail samples (donor no.1) treated at various times using the same chemical reagent dithiothreitol.

treatment, almost all samples saw a significant reduction of EPR response. In addition, an additional 10-min treatment resulted in more linearity than the previous case. Because the chemical treatment was able to reduce all EPR signals with different origins such as MIS and RIS, it can be concluded that the treatment principally reduces the MIS, although some reduction of RIS may also occur.

In this stage, two sets of fingernails from two other different donors (nos. 2 and 3), in addition to sample No.1, were used. To achieve our aim, attempts were made to collect the samples with approximately the same pieces of fingernails, i.e., the same cutting areas. All three sets of samples were subjected to gamma-ray irradiation at different absorbed doses and treated with the selected reagent for 5 minutes. Fig. 6 demonstrates the comparative dose–response curves of the samples. According to this figure, the EPR response curve is strongly dependent on the sample type, i.e., on the donors. In fact, considerable differences are obvious
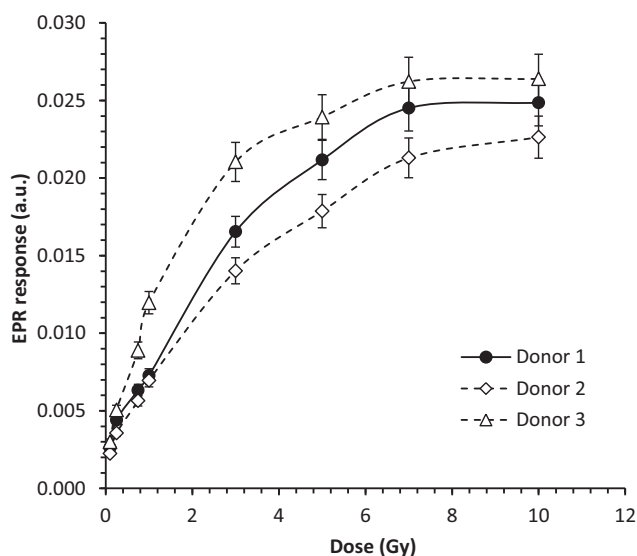
between the curves; in addition, all of them are saturated at about 8 Gy. This result originated from the creation of different MIS due to the various sets of the samples.

Further studies were carried out to see the effect of MIS on the sample's EPR responses. Fig. 7 shows the dose–response curves of the irradiated fingernail samples (3 sets), treated with the same chemical reagent dithiothreitol for 30 minutes. The obtained result demonstrates that the differences found between the curves and also the observed saturation seen in Fig. 6 are mostly due to mechanically induced radicals in the fingernail samples. Therefore, to determine an unknown absorbed dose in a fingernail sample using a calibration curve, the mentioned method is recommended to reduce the uncertainty.

4. Conclusion

The fading behavior of MIS was observed after 20 hours, and the EPR signal intensity did not vary anymore. Creation of more mechanically induced radicals in a fingernail due to greater cutting length caused more rapid fading in its EPR signal intensity. In addition, the EPR response of the humidified fingernail did not vary with the mechanical stress.

The dose–response curve strongly depended on the donors. Chemical treatment of fingernails using dithiothreitol reagent for 30 minutes reduced the difference between the dose–response curves of the different fingernails in comparison with the nonchemical treated samples, which is the novelty of this work in evaluating the absorbed dose value of an unknown sample. Therefore, to determine an unknown absorbed dose in a fingernail sample using a calibration curve, this method is an asset to reduce uncertainty.

All results are in agreement with those of other researchers investigating this domain, but the recently mentioned outcomes can considerably improve this field of study.

The authors of this article believe that by sampling a large number of donors, this claim can be proven more accurately in the future.

Conflict of interest

There is no conflict of interest.

Appendix A. Supplementary data

Supplementary data related to this article can be found at https://doi.org/10.1016/j.net.2018.01.014.

Fig. 6. Dose–response curves of the irradiated fingernail samples treated with the same chemical reagent of dithiothreitol for 5 minutes.
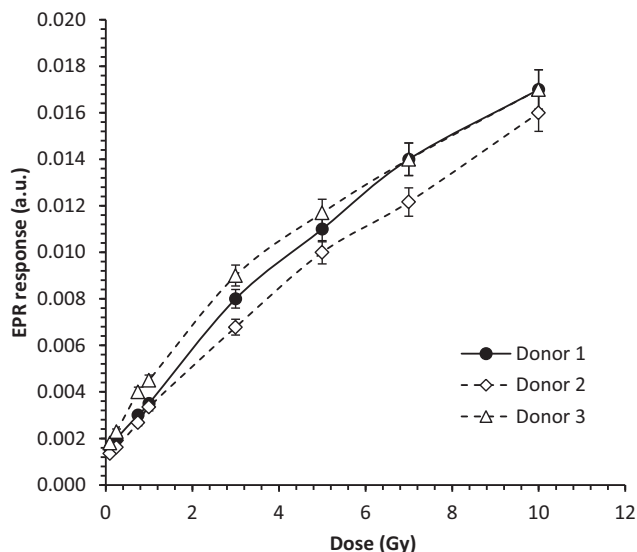

Fig. 7. Dose–response curves of the irradiated fingernail samples treated with the same chemical reagent of dithiothreitol for 30 minutes.

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