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Fingernail electron paramagnetic resonance dosimetry protocol for localized hand exposure accident

Jae Seok Kim [a, b, *], Byeong Ryong Park [a], Minsu Cho [a], Won Il Jang [a], Yong Kyun Kim [b]

[a] National Radiation Emergency Medical Center, Korea Institute of Radiological and Medical Sciences, 75 Nowon-ro, Nowon-gu, Seoul, Republic of Korea
[b] Department of Nuclear Engineering, University of Hanyang, 222 Wangsimni-ro, Seongdong-gu, Seoul, Republic of Korea

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ABSTRACT

Exposure to ionizing radiation induces free radicals in human nails. These free radicals generate a radiation-induced signal (RIS) in electron paramagnetic resonance (EPR) spectroscopy. Compared with the RIS of tooth enamel samples, that in human nails is more affected by moisture and heat, but has the advantages of being sensitive to radiation and easy to collect. The fingernail as a biological sample is applicable in retrospective dosimetry in cases of localized hand exposure accidents. In this study, the dosimetric characteristics of fingernails were analyzed in fingernail clippings collected from Korean donors. The dose response, fading of radiation-induced and mechanically induced signals, treatment method for evaluation of background signal, minimum detectable dose, and minimum detectable mass were investigated to propose a fingernail-EPR dosimetry protocol. In addition, to validate the practicality of the protocol, blind and field experiments were performed in the laboratory and a non-destructive testing facility. The relative biases in the dose assessment result of the blind and field experiments were 8.43% and 21.68% on average between the reference and reconstructed doses. The results of this study suggest that fingernail-EPR dosimetry can be a useful method for the application of retrospective dosimetry in cases of radiological accidents.

1. Introduction

The primary objective of dosimetry in the early stage of a radiological accident is to evaluate the radiation dose of individuals and assist in determining the initial medical treatment by providing exposure information to medical staff. Various techniques and human samples have been used for diagnostic purposes in clinical medicine and forensics and offer opportunities for evaluating acute exposure dose [1,2]. There are multiple approaches toward retrospective dosimetry for patients with acute exposure to ionizing radiation, including biological dosimetry, physical dosimetry, bioassays, and neutron activation [3]. These methods can be applied selectively based on the exposure condition and radiation type. In physical dosimetry methods using electron paramagnetic resonance (EPR), the dose delivered to the human body internally and externally is evaluated using biological materials (e.g., tooth enamel, nail, or hair) or objects and devices possessed by the exposed patients (e.g., cell phone and clothing) [4]. EPR dosimetry is a practical method for providing exposure information in radiological accidents [1].

The exposed dose in the hands of nuclear medicine staff working as medical radiation workers has been managed using ring dosimeters [5]. However, the read-out values do not accurately represent the exposed dose to the hand [6]. The value of the exposed dose measured at the fingertips has been reported to be two to six times the read-out values obtained from the ring dosimeter [7]. In case of localized hand exposure accident during the nondestructive testing (NDT), the EPR dosimetry using fingernail was applied to evaluate the exposed dose in the fingertips [8,9]. These cases imply the need for a method of evaluating the extremity dose in the hands of exposed patients. Among the potential biological samples for EPR, fingernails are an appropriate choice for evaluating the extremity dose in the fingertip owing to the ease of collecting them and their sensitivity to radiation [10].

Human nails exhibit three kinds of EPR signals: radiation-induced signal (RIS), mechanically induced signal (MIS), and background signal (BKS) [11–13]. The generated EPR signals in

* Corresponding author. 75, Nowon-ro Nowon-gu, Seoul 01812 Korea, Republic of Korea.
E-mail addresses: kimzgo@kirams.re.kr (J.S. Kim), ykkim4@hanyang.ac.kr (Y.K. Kim).

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exposed nails have different causes that overlap in a similar g-factor range [14,15]. To evaluate the exposed dose in the nail, the RIS must be distinguished in the complex spectrum [16]. To decrease the dose analysis time and ensure the accuracy and reliability of results, a protocol of dose assessment based on the dosimetric characteristics of the nail is required. Certain radiological accidents have been investigated using the fingernail-EPR dosimetry protocol [8,9,16,32]. However, the validation method was not presented for the result of dose assessment of fingernails in previous studies. The microwave bridges of X-band (~9.7 GHz) and Q-band (~34 GHz) were used in dosimetric research on radiological accidents. Using Q-band for RIS5 in fingernail is advantageous as it tends to be more sensitive than X-band for small mass samples (<10 mg) and provides a better spectral resolution for these materials [17]. Moreover, it has been demonstrated that RIS5 exhibits thermal and time stability which is not affected by the physical contact of fingernail with water [10]. On the other hand, the advantages of using the X-band for RIS2 in fingernails are higher reproducibility and sensitivity of signal above the appropriate amount (>10 mg), short analysis time, and wide dose range compared to Q-band. In particular, the low dose limit is 2 Gy, which is a noticeable advantage compared to Q-band (>10 Gy) [4,10,17]. However, the RIS2 stability is strongly influenced by temperature and humidity [17,18]. Therefore, to use RIS2 for dose assessment, the exposed fingernail should be collected without contact with water as soon as possible after a radiological accident. Recently, due to the increased interest in radiation exposure in radiation workers, strengthening of safety regulations, and advances in the personal dosimeter performance, it is possible to determine the occurrence of radiation exposure accidents in relatively shorter time. The most appropriate dose assessment method is needed for the situations involving radiological accidents.

The National Radiation Emergency Medical Center, the responding agency for radiological accidents in Korea under the Korea Institute of Radiological and Medical Sciences (KIRAMS), applied the protocol of fingernail-EPR dosimetry based on the dosimetric characteristics of fingernails to provide exposure information in a localized hand exposure accident. The dose response of fingernails was evaluated according to irradiation doses for the characteristics of dose linearity and variation between individuals. The variation in RIS and MIS after exposure and cutting was analyzed over time. A treatment method comprising soaking and drying processes was evaluated for the BKS as an intrinsic signal in fingernails. The minimum detectable dose (MDD) and minimum detectable mass (MDM) were evaluated to obtain reliable dose assessment results. In addition, dose reconstruction tests in blind and field experiments were performed in the laboratory and an NDT facility to validate the proposed protocol of fingernail-EPR dosimetry, which provides detailed procedures.

2. Materials and methods

2.1. Sample preparation and storage

Fingernails grown over two weeks were collected from Korean donors aged 30–50 years. The mass of the samples, depending on the position of the finger and donor, was 5–20 mg. The thickness of each fingernail was 0.5–1 mm, typically 0.6 mm approximately. Fingernail clippers were used to collect fingernail clippings that were 3–6 mm long and 1–2 mm wide. The collected samples were stored in a dark box under ambient conditions with 35% ± 2% relative humidity at 23 °C ± 1 °C (room temperature) [11].

2.2. Sample irradiation

Each sample was irradiated using a ^{137}Cs source (BioBeam 8000, STS Steuerungstechnik & Strahlenschutz GmbH, Germany) at a dose rate of 2 Gy min^{-1}. The fingernail samples were irradiated with the absorbed doses to the water-equivalent. The water-absorbed dose was monitored using a radio-photoluminescence glass dosimeter (RPLGD) (GD-352 M, ASAHI Techno Glass Corporation, Japan). The measurement uncertainty of the RPLGD reader system with a coverage factor of $k = 1$ was evaluated to be 4.08% [19].

2.3. EPR measurement

EPR measurements were conducted on a Bruker E500 EPR spectrometer equipped with a Bruker SHQE 4122 cavity resonator in the X-band. All measurements were performed under ambient conditions with 35% ± 5% relative humidity at 23 °C ± 1 °C at room temperature with a central magnetic field of 3530 G. The sweep width was 100 G, the receiver gain was 42 dB, the modulation amplitude was 4 G, the microwave frequency was 9.85 GHz, the number of scans was 10, the sweep time was 30.72 s, the conversion time 30 ms, and the microwave power was 1.002 mW. The EPR signal intensity in the fingernail was corrected based on the Bruker reference marker and the sample mass. Each sample was measured using quartz thin-wall EPR tubes with a diameter of 5 mm. EPR measurements were repeated three times, and the sample in the tube was shaken between consecutive measurements.

2.4. Dosimetric characteristics of fingernails

2.4.1. Dose response

Twenty-five samples were used to evaluate the individual dose response of the fingernail. The mass of the samples was 15–20 mg. The collected samples were stored in plastic vials for 24 h to decrease the effect of MIS interference. Twenty-five samples collected from five donors were irradiated at doses of 10, 20, 30, 40, and 50 Gy using ^{137}Cs γ-rays to evaluate the dose response for different individuals. Each set was composed of five samples collected from a single donor. The RIS spectrum overlapped with the BKS at an identical g-factor of 2.004. Therefore, to remove the BKS in the spectrum, the subtraction method was applied to evaluate the peak-to-peak RIS intensity. Regression curves were obtained using linear fitting, and the coefficient of determination (R^2) was calculated.

2.4.2. RIS fading

Twelve samples collected from three donors were used to evaluate the RIS fading. Each set of four samples collected from a single donor was irradiated with doses of 2, 10, 25, and 50 Gy using ^{137}Cs γ-rays. The irradiated samples were measured immediately and on days 1, 2, 6, and 10, and they were stored in vials between measurements. The variation in RIS was evaluated as the average of the relative standard deviation for 10 days for each set of irradiated samples.

2.4.3. MIS fading

Samples measuring 20 mg were cut enough to generate MIS in the cutting plane of the fingernail [20]. The size of each cut sample was 2–3 mm^2. The g-factor of the BKS in fingernails was considered when analyzing the intensity of the MIS. The MIS of the cut sample was measured using quartz thin-wall EPR tubes with a diameter of 5 mm for 400 h. In addition, the MIS of the samples had varying spectra depending on the cutting direction [10]. Therefore, the EPR measurements were repeated thrice after shaking the sample to

decrease the effect of cutting direction.

2.4.4. Treatment method for determining BKS

Twenty-four samples from eight donors were used to evaluate BKS in the exposed fingernail. The analysis procedure was performed in the following order: BKS measurement, irradiation of sample, RIS measurement, soaking and drying of sample, and sample measurement. To obtain the RIS in fingernails, three samples from one donor were irradiated with 30, 40, and 50 Gy. The RIS was then measured for each sample. To remove the RIS in each sample, the samples were soaked in distilled water for 15 min and then dried in a plastic vial with silica gel for 15 h [10,21]. The soaking and drying times were determined to be sufficient for the RIS to be removed by water and to result in a mass of fingernails that was similar to that before soaking, respectively [22].

2.4.5. Minimum detectable dose

The International Organization for Standardization (ISO) provides a guideline for performing ex vivo measurements of human tooth enamel samples by X-band EPR for dose assessment [23]. We adopted ISO 13304−2(2020) to evaluate the MDD in the fingernails. Samples from seven donors were used to evaluate the MDD. Table 1 shows that the fingernail mass of each sample was 15−20 mg.

The unexposed fingernails were measured 20 times to obtain the average and standard deviation of the BKS of the fingernail. Then, they were irradiated with 2, 5, 10, 20, 30 Gy as the additive dose to obtain the response curve. Each dose response curve was determined using the linear fitting described in Equation (1):

$$I = b_0 + b_1 D \tag{1}$$

where I is the EPR signal intensity, D is the irradiation dose, b_0 is the intercept, and b_1 is the slope. ISO 13304−2(2020) suggests applying the difference between the critical level (CL) and the detection limit (DL). The CL is the reference value used to distinguish between the measured signal and background noise. If the measurement result was higher than the CL, then the measurement was determined to detect a physical effect [24]. The CL of the dose was calculated using Equation (2):

$$D_{\mathrm{CL}} = \frac{I_{\mathrm{CL}} - b_0}{b_1} \tag{2}$$

where D_{CL} is the dose of the CL, and I_{CL} is the CL intensity. To evaluate the MDD (D_{DL}) of the fingernails, the dose of the DL (D_{DL}), was calculated using Equation (3), based on the ISO 13304−2(2020) approach [22].

$$D_{DL} = 2D_{CL} \tag{3}$$

The MDD was assumed to be twice the dose of the critical level (D_{CL}). Based on this assumption, the MDDs for seven fingernail samples from different donors were calculated.

2.4.6. Minimum detectable mass

The limit of quantitation (LOQ) presented by the International Union of Pure and Applied Chemistry (IUPAC) was applied to evaluate the MDM in fingernails [25]. MDM is meaning of the minimum mass for dose assessment in fingernail. A sufficient analyte concentration must be present to produce an analytical signal that can reliably be distinguished from "analytical noise," the signal produced in the absence of an analyte [26]. LOQ is defined as the lowest mass of a sample that can be determined with acceptable precision and accuracy under the stated conditions of the test. It can be expressed by Equation (4):

$$\mathrm{LOQ} = \frac{10\sigma}{S} \tag{4}$$

where σ is the standard deviation of the response and S is the slope of the response curve according to mass [24]. The samples had masses of 1, 3, 5, 10, 20, and 30 mg. Samples were irradiated with a reference dose of 3 Gy to evaluate the MDM in fingernails.

2.5. Dose reconstruction test

2.5.1. Blind experiment in laboratory

Blind experiments were carried out to verify the protocol of fingernail-EPR dosimetry at the laboratory level. Three samples were collected from three donors. Blind doses were irradiated using ^{137}Cs γ-rays. Subsequently, each sample was cut to generate MIS in the fingernails. The cut samples were stored in a plastic vial with silica gel for 3 h to decrease the effect of MIS. To analyze the samples exposed to unknown doses, the initial EPR signal in the blind samples was measured before additive irradiation for sampling was performed. The additive dose method was used to obtain the dose-response curve of each sample [27]. Lastly, the BKS of the blind samples were measured using the treatment method, including soaking and drying.

2.5.2. Field experiment in NDT facility

A field experiment was conducted to verify the applicability of the proposed protocol under conditions that were consistent with the radiological accident. The exposure situation of localized hand exposure was simulated in an NDT facility. Fifteen samples collected from three donors were placed on the fingertips of a physical human phantom. Immediately before conducting the field test, the samples were collected from the donors. To reduce the influence of external contamination and drying, the samples were enclosed in a paraffin tape during the irradiation. Table 2 shows the mass of the fingernails according to their positions. Fig. 1 shows the experimental setup for localized hand exposure in the NDT facility. The ^{60}Co γ-ray source (98.6 Ci) was located at the tip of the guide tube for 15 min when the non-destructive irradiator was turned on. The whole-body dose was set to 3 Gy through the pre-irradiation plan using a Monte Carlo simulation. After the irradiation, the paraffin tape was peeled off from the fingernail, stored in sealed vials, and moved to the laboratory. The exposed fingernails were stored in sealed vials. The physical human phantom was developed based on the mesh-type reference computational phantoms of the International Commission on Radiological Protection (ICRP) [28]. An L-α-alanine dosimeter (Aerial CRT, France) and an RPLGD were located next to the fingernail for comparison with the exposed dose evaluated by the proposed fingernail-EPR protocol. The measurement uncertainty of the alanine dosimeter with a coverage factor of $k = 1$ was evaluated to be 2.33% [29]. The exposed fingernails were

Table 1
Fingernail mass (mg) according to donors.

Donor 1	Donor 2	Donor 3	Donor 4	Donor 5	Donor 6	Donor 7
19.29	16.50	17.45	19.57	17.61	15.7	19.98

Table 2
Fingernail mass (mg) according to position.

Sample	Thumb	Index	Middle	Ring	Little
Donor 1	15.68	10.73	13.38	8.45	6.61
Donor 2	16.17	9.72	11.85	10.44	7.06
Donor 3	13.48	6.08	7.48	5.26	4.68

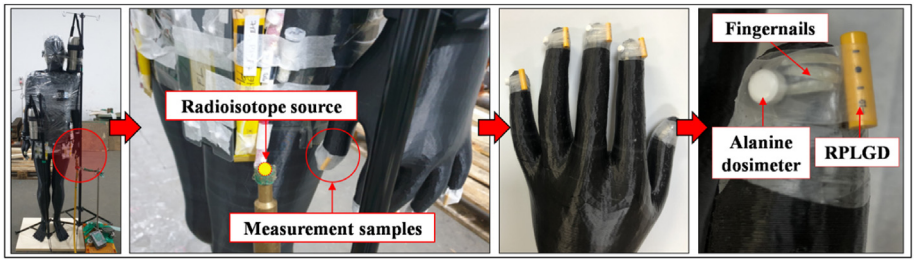

Fig. 1. Experimental setup for the localized hand exposure in the NDT facility.

evaluated using the proposed fingernail-EPR dosimetry protocol.

3. Results and discussion

3.1. Dosimetric characteristics of the fingernail

3.1.1. Dose response

The dose response from 1 to 100 Gy in fingernails can be used to evaluate the exposed dose on the fingertip over a wide dose range [30]. In a localized exposure situation, the patient is exposed to a relatively high dose compared to the whole-body irradiation dose [9]. Fig. 2 shows that the dose response curves for fingernails have a good linear relationship for irradiation doses in the range of 10–50 Gy. Regression curves were obtained using linear fitting, and R^2 exceeded 0.97 for each set of fingernail samples. However, the exposed samples exhibited different dose response curves for different individual donors. These results show that rather than using a standardized dose response curve for dose assessment, fingernails of the same person should be used.

3.1.2. RIS fading

Fig. 3 shows that the RIS fading was evaluated for ten days using the twelve samples from three donors. The variation of RIS over ten days was evaluated using the average of the relative standard deviation as 9.34% for the irradiation dose range from 2 to 50 Gy. The variation between the measurement and irradiation for signal stabilization was included in the uncertainty of RIS fading. In this study, the RIS fading was evaluated by minimizing the effects of water and heat. In an actual radiological accident, the variation in RIS is difficult to estimate before the collection of fingernails from an exposed patient. The exposed fingernail must be collected as soon as possible to avoid the effects of water and heat, which can reduce the RIS in the fingernail [20,31].

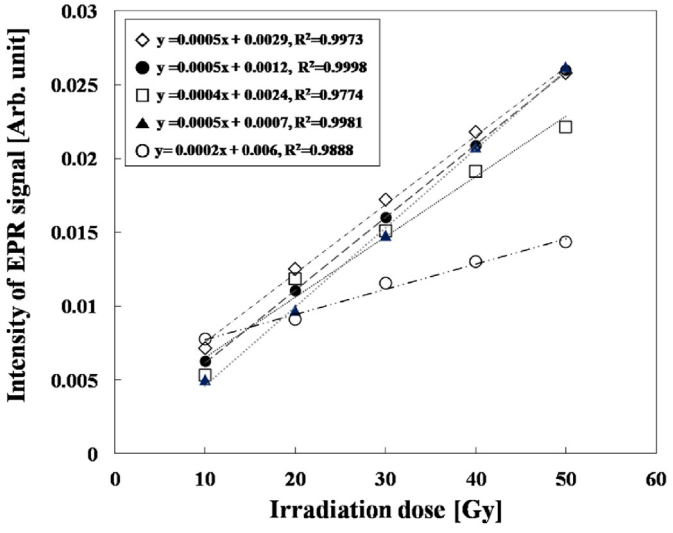

Fig. 2. Linear relationship between the irradiation doses and intensity of the EPR signals according to individual.

3.1.3. MIS fading

In a radiological accident, MIS occurs during fingernail collection from the exposed patient. To minimize the effect of MIS on the fingernail EPR signal, MIS fading after the cut time was analyzed. Fig. 4 shows the variation in the measured MIS over 400 h.

The g-factor of MIS was measured as 2.0035 using the EPR spectrometer. The RIS and MIS in the fingernail-EPR spectra have peak-to-peak intensities at different g-factors. The MIS interfered with determining the peak-to-peak intensity of the RIS, and this interference effect decreased as time after cutting increased. The exposed fingernails have irrelevant EPR signals from MIS caused by the collection process, and it is expected that measurement of samples at least 3 h after collection will produce more accurate dose evaluation results than RIS immediately after collection. The time required to minimize the effect of MIS was similar to that reported by Sholom and McKeever (2016) [32].

3.1.4. Treatment method for determination of BKS

In a radiological accident, measuring the BKS immediately after collecting samples is difficult. The treatment method should be used to analyze the BKS of exposed fingernails. The results of EPR signals were measured according to the treatment step and irradiation doses as shown in Fig. 5. Each intensity of EPR signals was evaluated for the fingernails collected from eight donors. The difference between the intensity of BKS in unexposed and treated samples was evaluated using the average relative bias of 14.67% for the irradiation dose range of 30–50 Gy. The RISs were a signal excluding the BKSs. The BKS after soaking and drying was evaluated as less than the initial BKS. The difference between the initial BKS and soaking and drying of BKS would be occurred due to the residual MIS2 in the collected fingernail [10]. In addition, the residual water in the fingernail can reduce the sensitivity of EPR measurements owing to dielectric loss [33]. Therefore, the appropriate soaking and drying times are important for determining the BKS. To determine the BKS in the fingernail through treatment method, the soaking and drying times were determined to be 15 min and 15 h, respectively.

3.1.5. Minimum detectable dose

The standard deviation of repeated BKS measurements and the dose response curve of each sample were used to evaluate the MDD in fingernails. The MDD calculated for seven fingernail samples from different donors was 1.68 ± 0.26 Gy. However, because the MDD can differ depending on individual dosimetric characteristics of the fingernails, these results suggest that the MDD of fingernails can be evaluated statistically. The reliability of evaluated exposed dose in fingernail can be confirmed preferentially by using the

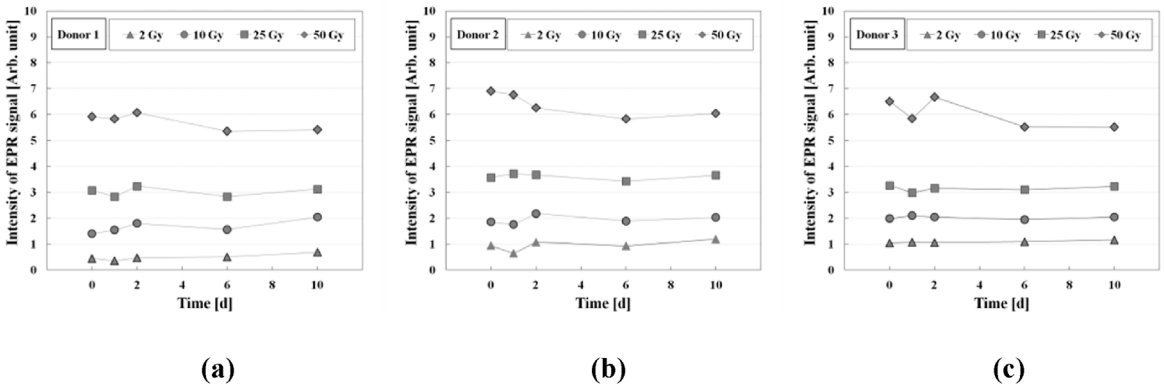

Fig. 3. Variation of RIS in fingernails collected from three donors over ten days from 2 to 50 Gy: (a) Donor 1, (b) Donor 2, (c) Donor 3.

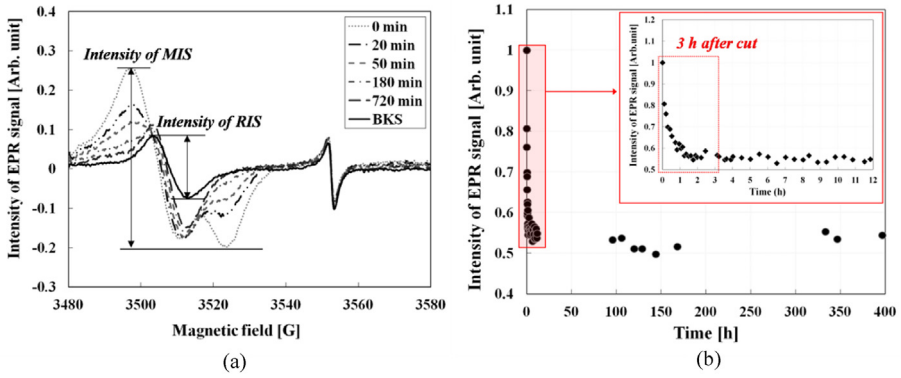

Fig. 4. MIS in fingernail after cut: (a) variation of EPR spectrum, and (b) normalized intensity of the MIS in fingernail according to time.

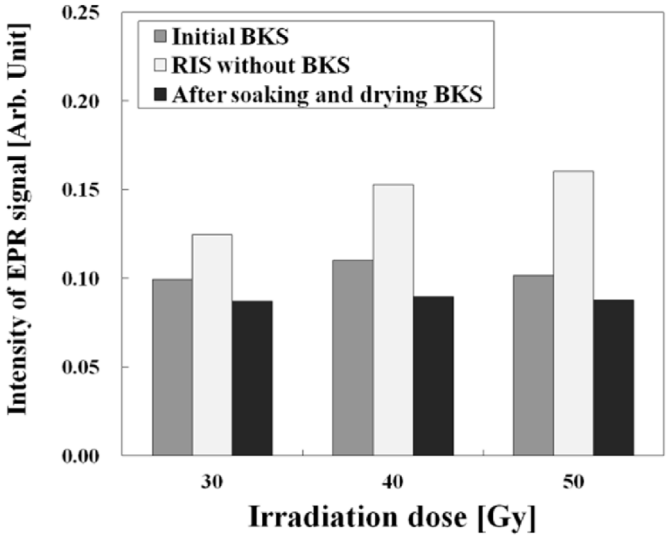

Fig. 5. Intensity of EPR signal according to the treatment step and irradiation dose.

results of MDD results when there is no reference dosimeter.

3.1.6. Minimum detectable mass

The mass of the fingernail is an important factor in evaluating the exposed dose. Difficulty in collecting a sufficient mass of fingernail samples is anticipated in a localized hand exposure accident. The MDM was evaluated to establish the minimum sample mass required to obtain reliable dose assessment results. The slope was calculated from the intensity of the RIS according to the mass of the fingernails. Subsequently, Equation (4) was applied to evaluate the MDM through statistical analysis. The MDM was 3.11 ± 0.54 mg at an irradiation dose of 3 Gy. In other words, the result of the evaluation refers to the amount of fingernail sample that could be statistically analyzed when the exposed dose of fingernails was 3 Gy. The MDM was evaluated by the particular irradiation dose at the adjacent MDD. We can estimate that the slope of the response curve according to mass increased as the exposed dose in the fingernail increased. These results show that the MDM can be determined differently based on the exposed dose in the fingernail.

3.2. Protocol of fingernail-EPR dosimetry

The proposed fingernail-EPR dosimetry protocol is based on the dosimetric characteristics of fingernails obtained in this study. The protocol is summarized in Fig. 6 [18]. It was assumed that the dose assessment was performed on fingernails collected immediately after the localized hand exposure accident. The additive dose method was adopted to evaluate the exposed dose in fingernails because the fingernail has a limited mass available for collection from exposed patients [23,27]. To evaluate the accurate the BKS in fingernail, the conditions of fingernail treatment were same as the dose additive methods and BKS evaluation. It took 24 h to complete the proposed protocol for dose assessment of three samples.

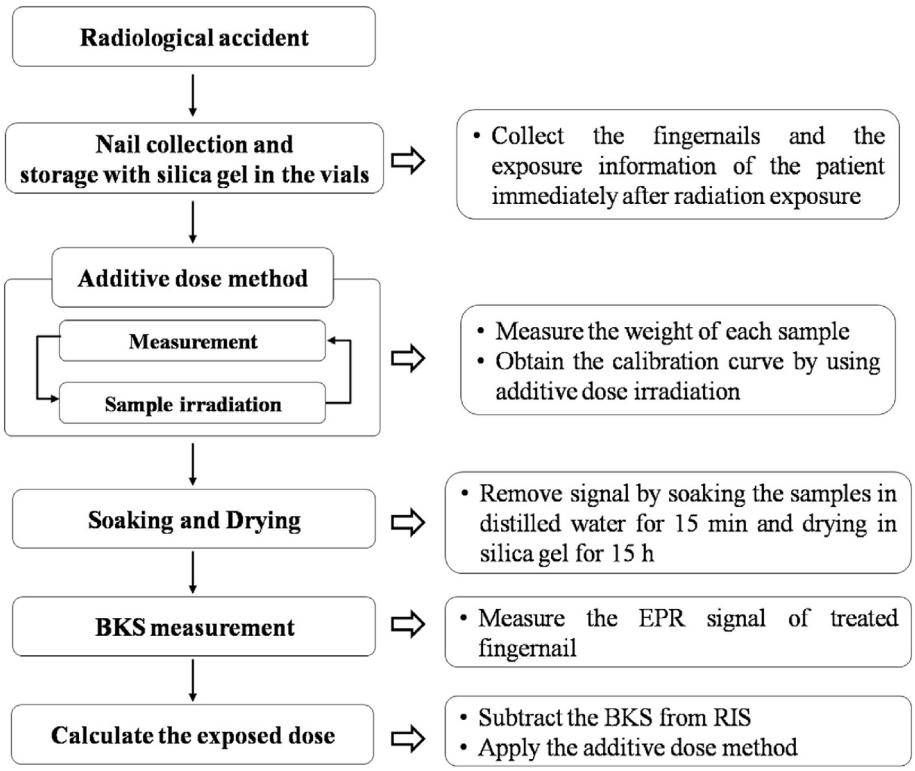

Fig. 6. Workflow diagram for the protocol of fingernail-EPR dosimetry.

Table 3
Results of dose assessment for blind experiment.

Sample number	Blind dose (Gy, $k = 1$)	Reconstructed dose (Gy, $k = 1$)	MDD (Gy)
1	15.13 ± 0.61	13.41 ± 2.63	1.28
2	10.68 ± 0.43	10.26 ± 1.65	3.42
3	16.91 ± 0.68	15.22 ± 1.69	2.42

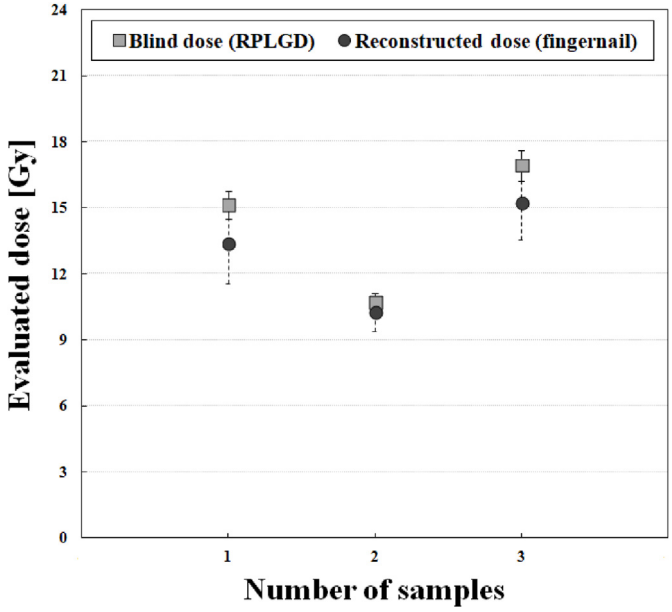

Fig. 7. Results of the blind experiment of three evaluated doses in blind samples.

3.3. Dose reconstruction tests

3.3.1. Blind experiment for verification of the protocol

The results of the blind experiment are presented in Table 3 and Fig. 7. The relative biases were determined to be 11.36%, 3.93%, and 9.99% for the three exposed doses of 15.13, 10.68, and 16.91 Gy. The measurement uncertainty of the protocol was determined by the sample irradiation, treatment method for BKS, EPR measurement, and regression curve of the additive dose method [34,35]. The results of the reconstructed dose were underestimated compared to those of the blind dose. In the predicted results, the reconstructed doses in fingernail would be evaluated larger than the blind dose due to that could not removed the MIS completely [10]. However, the reconstructed doses were not larger than the blind doses. It was estimated that the BKS was overestimated by the treatment method for determining the BKS. The treatment process to obtain the BKS forms the largest portion of measurement uncertainty of protocol. The individual character of the water content in the fingernail may influence the results of the reconstructed doses. However, all blind doses were achieved using the range of measurement uncertainty of the reconstructed dose. The reconstructed doses were higher than the MDD of the blind samples.

Table 4
Results of dose assessment for field experiment according to finger position and sample.

Position	Dosimeter		Fingernail		
	RPLGD	Alanine	Donor 1	Donor 2	Donor 3
	(Gy, k = 1)				
Thumb	45.81 ± 3.73	42.80 ± 2.41	14.37 ± 4.92	14.63 ± 5.00	17.36 ± 5.38
Index	12.36 ± 1.01	15.91 ± 0.89	12.32 ± 4.05	12.36 ± 6.34	9.31 ± 1.98
Middle	8.37 ± 0.68	11.04 ± 0.62	9.69 ± 3.70	10.72 ± 2.73	13.33 ± 3.36
Ring	8.66 ± 0.70	6.90 ± 0.38	10.29 ± 3.97	6.57 ± 2.05	6.81 ± 1.75
Little	6.63 ± 0.54	5.82 ± 0.32	7.25 ± 2.75	4.87 ± 1.59	5.73 ± 1.52

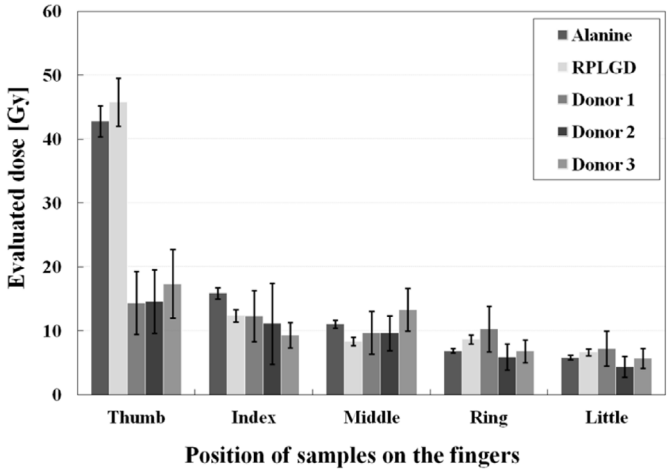

Fig. 8. Results of evaluated doses in RPLGDs, alanine dosimeters, and fingernails according to finger position.

3.3.2. Field experiment in NDT facility

The exposed samples of RPLGDs, alanine dosimeters, and fingernails were evaluated by each analysis system and the protocol of fingernail-EPR dosimetry according to the finger position, as shown in Table 4 and Fig. 8. The results of exposed dose in fingernail were evaluated as the conversion in terms of kerma in tissue. Relative biases between dosimeters and fingernails according to finger position averaged 65.11%, 22.70%, 12.24%, 1.33%, and 7.02%, for the thumb, index finger, middle finger, ring finger, and little finger, respectively. The average read-out value in dosimeters and the average reconstructed dose of fingernails were compared according to respective fingers.

The measurement uncertainty of the protocol was determined using the sample irradiation, RIS fading, treatment, EPR measurement, and regression curve of the additive dose method [34,35]. The reconstructed doses in the fingernails were higher than the MDD of each fingernail. The dosimeters and fingernails located on the thumb nearest to the source gave the highest exposed doses among the samples. However, the exposed dose in thumbnails was found to be relatively lower than the result of dosimeters on the thumb. Thumbnails were underestimated on the hand of the physical human phantom for several reasons. The fingertip did not have enough space to set the measurement samples in an identical position, and the reconstructed results could be affected by tiny differences in the position of the sample, despite the samples being close to each other. In addition, the exposed thumbnails were irradiated with additional doses exceeding the read-out value of the dosimeters because the additive dose method was applied to the exposed samples. The total exposed dose in thumbnails was higher than the irradiation dose researched by the treatment method for the evaluation of BKS. It was supposed that the BKS of fingernail exposed to more than 50 Gy has not been accurately evaluated. Therefore, additional research on treatment time to remove RIS higher than 50 Gy must be considered in further studies.

4. Conclusion

We investigated the dosimetric characteristics of fingernails, including the dose response, RIS and MIS fading, treatment method for BKS, MDD, and MDM to propose a protocol for fingernail-EPR dosimetry, and validated the proposed protocol. The dose response of fingernails showed different EPR intensity at the same irradiation dose for different individuals. The RIS in the exposed fingernail was evaluated for variation of EPR over time. In a clipped fingernail, MIS generated in the cross section of fingernails decreased the accuracy of result evaluation of exposed doses. The time taken to minimize the effect of MIS was determined to be at least 3 h. The soaking and drying times were determined to be 15 min and 15 h, respectively, to determine the BKS in the fingernail through treatment. To evaluate the BKS of fingernails, using the control samples could be one of the treatment methods. If the control fingernail was not affected by exposure in a radiological accident, it could be applied to the evaluation of BKS. However, it is difficult to determine which nails to use as control samples. In the case of localized hand exposure, the control group is also highly likely to be exposed because it is close to the radioactive source. Therefore, soaking and drying methods were used for evaluation of BKS in the proposed protocol. In this study, a methodology for evaluating MDD and MDM that can provide reliable results for dose assessment using fingernails was presented. Because the MDD and MDM were dependent on each other, they were evaluated at random mass and dose. The obtained properties were applied to propose a protocol for fingernail-EPR dosimetry for determining localized exposed dose in the hand. Dose reconstruction tests were carried out to verify the validity and practicality of the fingernail-EPR dosimetry protocol. The results of a blind test demonstrated the applicability of fingernail-EPR dosimetry at the laboratory level. The average relative bias between blind and reconstructed doses was 8.43%. A field experiment was performed to evaluate the practicality of the protocol in an actual NDT facility. Exposed doses in the fingertip were evaluated by on-site simulation using fingernails, RPLGDs, and alanine dosimeters. Most of the reconstructed doses in fingernails were satisfactory compared with the results of RPLGDs and alanine dosimeters. The average relative bias between dosimeters and reconstructed doses was 21.68%. However, there was a large difference between the dosimeters and results obtained from nails located on the thumb. As the distance between the source and the samples decreased, the results of the exposed doses were significantly different depending on the different positions. Because the hand of the physical human phantom was not an even surface, the samples were not irradiated uniformly in the

field experiment. Furthermore, additional research on the treatment process of samples exposed to doses exceeding 50 Gy is required to obtain accurate BKS. Nevertheless, the results of the dose reconstruction tests were significant in most dose ranges. An exposure scenario in a specific situation of immediately collecting fingernails after the radiological accident was considered, the possibility of providing exposure information in high-dose localized hand exposure accidents has been confirmed by the validation of the proposed protocol for fingernail-EPR dosimetry. For application in radiological accident cases, a comprehensive dose assessment is needed not only using various dosimetry methods in addition to the fingernail-EPR dosimetry. The skin dose of the personal dose equivalent will be evaluated using the protocol for fingernail-EPR dosimetry and a ring dosimeter in future studies. This will provide useful operational quantity information in high-dose localized hand exposure situations.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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References

[1] International Commission on Radiation Unit and Measurements, (ICRU), Method for initial-phase Assessment of individual doses following acute exposure to ionizing radiation, ICRU 94 (2019).
[2] G.A. Alexander, et al., BiodosEPR-2006 Meeting: acute dosimetry consensus committee recommendations on biodosimetry applications in events involving uses of radiation by terrorists and radiation accidents, Radiat. Meas. 42 (2007) 972–996, https://doi.org/10.1016/j.radmeas.2007.05.035.
[3] L. Waldner, et al., The 2019–2020 EURADOS WG10 and RENEB field test of retrospective dosimetry methods in a small-scale incident involving ionizing radiation, Radiat. Res. 195 (2021) 253–264, https://doi.org/10.1667/RADE-20-00243.
[4] F. Trompier, C. Bassinet, A. Wieser, C.D. De Angelis, D. Viscomi, P. Fattibene, Radiation-induced signals analysed by EPR spectrometry applied to fortuitous dosimetry, Ann. Ist. Super Sanita 45 (2009) 287–296.
[5] M. Sans Merce, et al., Extremity exposure in nuclear medicine: preliminary results of a European study, Radiat. Protect. Dosim. 144 (2011) 515–520, https://doi.org/10.1093/rpd/ncq574.
[6] S. Papierz, Z. Kamiński, M. Adamowicz, M. Zmyślony, Assessment of individual dose equivalents Hp(0.07) of medical staff occupationally exposed to ionizing radiation in 2012, Med. Pr. 65 (2014) 167–171, https://doi.org/10.13075/mp.5893.2014.013.
[7] D. Adliene, B. Griciene, K. Skovorodko, J. Laurikaitiene, J. Puiso, Occupational radiation exposure of health professionals and cancer risk assessment for Lithuanian nuclear medicine workers, Environ. Res. 183 (2020), 109144, https://doi.org/10.1016/j.envres.2020.109144.
[8] F. Trompier, F. Queinnec, E. Bey, T. De Revel, J.J. Lataillade, I. Clairand, M. Benderitter, J.F. Bottollier-Depois, EPR retrospective dosimetry with fingernails: report on first application cases, Health Phys. 106 (2014) 798–805, https://doi.org/10.1097/HP.0000000000000110.
[9] A. Romanyukha, F. Trompier, R.A. Reyes, D.M. Christensen, C.J. Iddins, S.L. Sugarman, Electron paramagnetic resonance radiation dose assessment in fingernails of the victim exposed to high dose as result of an accident, Radiat. Environ. Biophys. 53 (2014) 755–762, https://doi.org/10.1007/s00411-014-0553-6.
[10] F. Trompier, A. Romanyukha, R. Reyes, H. Vezin, F. Queinnec, D. Gourier, State of the art in nail dosimetry: free radicals identification and reaction mechanisms, Radiat. Environ. Biophys. 53 (2014) 291–303, https://doi.org/10.1007/s00411-014-0512-2.
[11] A. Marciniak, B. Ciesielski, M. Juniewicz, A. Prawdzik-Dampc, M. Sawczak, The effect of sunlight and UV lamps on EPR signal in nails, Radiat. Environ. Biophys. 58 (2019) 287–293, https://doi.org/10.1007/s00411-019-00777-2.
[12] X. He, et al., Development and validation of an ex vivo electron paramagnetic resonance fingernail biodosimetric method, Radiat. Protect. Dosim. 159 (2014) 172–181, https://doi.org/10.1093/rpd/ncu129.
[13] C.A.B. Gonzales, J.E. Taño, H. Yasuda, An attempt to reduce the background free radicals in fingernails for monitoring accidental hand exposure of medical workers, Appl. Sci. 10 (2020) 1–12, https://doi.org/10.3390/app10248949.
[14] M.C.R. Symons, H. Chandra, J.L. Wyatt, Electron paramagnetic resonance spectra of irradiated finger-nails: a possible measure of accidental exposure, Radiat. Protect. Dosim. 1 (1995) 11–15, https://doi.org/10.1093/oxfordjournals.rpd.a082591.
[15] G. Strzelczak, M. Sterniczuk, J. Sadlo, M. Kowalska, J. Michalik, EPR study of γ-irradiated feather keratin and human fingernails concerning retrospective dose assessment, Nukleonika 58 (2013) 505–509.
[16] S. Hirota, C.A.B. Gonzales, H. Yasuda, Behavior of the electron spin resonance signals in X-ray irradiated human fingernails for the establishment of a dose reconstruction procedure, J. Radiat. Res. 62 (2021) 812–824, https://doi.org/10.1093/jrr/rrab027.
[17] A. Romanyukha, F. Trompier, R.A. Reyes, M.A. Melanson, EPR measurements of fingernails in Q-band, Radiat. Meas. 46 (2011) 888–892, https://doi.org/10.1016/j.radmeas.2011.04.004.
[18] F. Trompier, L. Kornak, C. Calas, A. Romanyukha, B. Leblanc, C.A. Mitchell, H.M. Swartz, I. Clairand, Protocol for emergency EPR dosimetry in fingernails, Radiat. Meas. 42 (2007) 1085–1088, https://doi.org/10.1016/j.radmeas.2007.05.024.
[19] J.S. Kim, et al., Measurement uncertainty analysis of radiophotoluminescent glass dosimeter reader system based on GD-352M for estimation of protection quantity, Nucl. Eng. Technol. 54 (2) (2021) 479–485, https://doi.org/10.1016/j.net.2021.08.016.
[20] H. Choi, S.A. Choi, B. Lee, Preliminary study of water contents and signal behavior in fingernail/EPR dosimetry, J. Radiation Protect. Res. 38 (2013) 185–188, https://doi.org/10.14407/jrp.2013.38.4.185.
[21] R.A. Reyes, A. Romanyukha, C. Olsen, F. Trompier, L.A. Benevides, Electron paramagnetic resonance in irradiated fingernails: variability of dose dependence and possibilities of initial dose assessment, Radiat. Environ. Biophys. 48 (2009) 295–310, https://doi.org/10.1007/s00411-009-0232-1.
[22] S. Hirota, C.A.B. Gonzales, H. Yasuda, I. Yamaguchi, S. Toyoda, Electron spin resonance signal of human nails: increase after irradiation, J. Radioanal. Nucl. Chem. 328 (2021) 1369–1373, https://doi.org/10.1007/s10967-020-07540-8.
[23] International Organization for Standardization (ISO), Radiological Protection-Minimum Criteria for Electron Paramagnetic Resonance (EPR) Spectroscopy for Retrospective Dosimetry of Ionizing Radiation-Part 2: Ex Vivo Human Tooth Enamel Dosimetry, ISO 13304 2, 2020.
[24] P. Fattibene, et al., The 4th international comparison on EPR dosimetry with tooth enamel Part 1: Report, Radiat. Meas. 46 (2011) 765–771, https://doi.org/10.1016/j.radmeas.2011.05.001.
[25] E. Bernal, Limit of detection and limit of quantification determination in gas chromatography, Adv. Gas Chromato. (2014).
[26] A. Shrivastava, V. Gupta, Methods for the determination of limit of detection and limit of quantitation of the analytical methods, Chronicles Young Sci. 2 (2011) 21–25, https://doi.org/10.4103/2229-5186.79345.
[27] M. Nenoi (Ed.), Current Topics in Ionizing Radiation Research, IntechOpen, 2012, https://doi.org/10.5772/2027.
[28] International Commission on Radiological Protection (ICRP), Adult mesh-type reference computational phantoms, ICRP 145, 2020.
[29] B.R. Park, et al., The first KREDOS-EPR intercomparison exercise using alanine pellet dosimeter in South Korea, Nucl. Eng. Technol. 52 (2020) 2379–2386, https://doi.org/10.1016/j.net.2020.03.025.
[30] F. Trompier, A. Romanyukha, L. Kornak, C. Calas, B. LeBlanc, C. Mitchell, H. Swartz, I. Clairand, Electron paramagnetic resonance radiation dosimetry in fingernails, Radiat. Meas. 44 (2009) 6–10, https://doi.org/10.1016/j.radmeas.2008.10.005.
[31] A. Marciniak, B. Ciesielski, A. Prawdzik-Dampc, The effect of dose and water treatment on EPR signals in irradiated fingernails, Radiat. Protect. Dosim. 162 (2014) 6–9, https://doi.org/10.1093/rpd/ncu207.
[32] S. Sholom, S.W.S. McKeever, Emergency EPR dosimetry technique using vacuum-stored dry nails, Radiat. Meas. 88 (2016) 41–47, https://doi.org/10.1016/j.radmeas.2016.02.014.
[33] M. Sueki, G.A. Rinard, S.S. Eaton, G.R. Eaton, Impact of high-dielectric-loss materials on the microwave field in EPR experiments, J. Magn. Reson. A. 118 (1996) 173–188, https://doi.org/10.1006/jmra.1996.0025.
[34] International Organization for Standardization (ISO), Practice for Calibration of Routine Dosimetry Systems for Radiation Processing, 2013. ISO/ASTM 51261.
[35] International Atomic Energy Agency (IAEA), Use of Electron Paramagnetic Resonance Dosimetry with Tooth Enamel for Retrospective Dose Assessment, vol. 1331, IAEA-TECDOC, 2002.