■ PAPER ■

PRELIMINARY STUDY OF WATER CONTENTS AND SIGNAL BEHAVIOR IN FINGERNAIL/EPR DOSIMETRY

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Heating method using electric heater was used to reduce water content in fingernail clippings. Authors found that low water content is helpful to measure EPR signal of fingernail sample with enhanced variability. Generally, natural dehydration happens in normal room condition at least one month and needs much time for using in experiment. So, artificial heating method is applied in this study for time savings. Fingernail samples were tested to find effect of water content to the EPR signal on microwave power levels for dosimetry. Low water content in fingernail reduced variability of EPR signal and makes it possible to measure accurate EPR signal. It also made it possible to measure constant movement of EPR signals on several microwave power levels. Although this method was difficult to apply directly in fingernail/EPR dosimetry, we, authors, believe that this heating method would be useful to differentiate MIS2 and RIS which are generally located at the same g-factor and almost impossible to be identified with each other.

Keywords: EPR, Fingernail, Water, RIS, MIS, Dosimetry

1. INTRODUCTION

There is growing recognition of the need to develop effective dosimetry methods to assess unexpected radiation exposure in the event of a large-scale emergency situation [1]. One of physically-based dosimetry methods, Electron Paramagnetic Resonance (EPR) spectroscopy has been studied to perform retrospective radiation dosimetry using extracted samples of tooth enamel and nail, following radiation accidents and exposures. But, teeth have a great difficulty in getting sample for its extraction from human. In case of fingernail, it is much easier to collect than teeth. Fingernails are largely composed of a keratin, which consists of α-helical peptide chains, twisted into a left-handed coil and strengthened by disulphide cross links [2]. Generally, ionizing radiation generates free radicals in the keratin matrix, and these radicals are stable over a relatively long period (days to weeks). Most importantly, the number of radical is linearly proportional to the magnitude of the dose over a wide dose range (0～30 Gy) [3-6]. However, relatively large background signal (BKS) converted from mechanically induced signal (MIS) after cutting process of fingernail, normally overlaps with the radiation induced signal (RIS) and makes it difficult to estimate accurate dose [2]. MIS can be separated into two signals, MIS1 and MIS2. MIS1 is dominant at first few hours after cutting and decrease drastically. MIS1 is thought to be originated from elastic change in fingernail structure from cutting process. MIS2 is relatively very small at first stage but become dominant after 24 hours. MIS2 is reported to come from plastic change of fingernail structure. So, MIS2 generally maintained for few weeks after cutting process. Therefore, due to these large MIS, estimation method using dose-response curve was difficult to ensure reliability below 5 Gy. So many studies about reduction or exclusion of MIS have been tried. Soaking method using water as remover of MIS induced in cutting edge of fingernail parings is one of them. After several pilot experiments using a water contents as remover, we found that soaking method is not efficient to differentiate RIS from MIS and difficult to be used in quan-

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titative dose evaluation process. Such soaking methods required severe control of samples and experimental procedure to reduce uncertainties. Generally, the measurement sensitivity is affected by two factors, water content in fingernail parings and mechanically induced radicals induced by cutting process. Two kinds of radicals from mechanical stress and irradiation may be different in response on microwave power levels, even though their spectrum shape and g-factor are same in X-band spectroscopy system. But, this difference could not be obtained by earlier experiment. Maybe, various water content which fulfilled microstructure of fingernail clippings could be the main cause of distortion of data. In this study, to get stable experiment result, we reduce water level in fingernail parings through heating process. The dehydration process induced evidently the deformation of fingernail structure and aging process which increase the intensity of EPR (Electron Paramagnetic Resonance, MIS) signal. So, to use these dehydrated fingernail samples for dosimetry, alternative method using power saturation behavior should be considered in the future.

2. MATERIALS AND METHODS

In this experiment, 20 fingernails from 8 persons were cut by normal fingernail clipper and used for experiment. When fingernail samples were collected from volunteers, all fingernail samples were used right after harvesting or stored in refrigerator at most one week. The collected fingernail samples were cut into small clippings which its size is about 1.5~2 mm wide and long. Cutting by normal fingernail clipper is special, because until now all cutting process was done by medical scissors to avoid big mechanically induced EPR signal. Toenail clippers were used to cut the harvested fingernail into small 10 pieces of clippings. After cutting, fingernail clippings were put into small plastic capsule without lid and heated by electric heater at 70℃ for 72 hours by muffle furnace (F0 100, Japan). From experiments, we found that only 3 hours of heating time was not sufficient for complete conversion from MIS1 to MIS2 and, at least, 72 hours of heating time was needed to get clear MIS2 signal. Remained little MIS1 signal was big obstacle to get EPR signal behavior at high microwave power levels. For measurement by EPR spectrometer, two quartz tubes composed of sample tube and alignment tube were prepared. Standard manganese source was fixed at the bottom of alignment tube inside and size of sample tube was chosen not to move easily in alignment tube, as shown in Fig. 1. EMX X-band EPR spectrometer (Bruker BioSpin, Germany) was used to measure EPR spectra. Measurement parameter is as follows: high-frequency modulation: 100 kHz; amplitude of modulation: 0.05 mT; Microwave power: variable; time constant: 40 ms; sweep time: 20 sec; number of scans: variable. Gamma irradiation to fingernail samples was done by ^{137}Cs blood irradiator (IBL437C, France) with dose rate of 6 Gy min^{-1} ±4%.

3. RESULTS AND DISCUSSION

After a series of pilot experiment, we found that EPR spectrum with low level of moisture is more stable than that of normal fingernail samples. But to reduce water level in fingernail naturally, under normal room condition, it takes quite a long time (about one month). Another weak point is that the reducing process induced unwelcomed increasing of EPR intensity, MIS. This uncontrolled increase of MIS intensity prevents accurate measurement, because each increment of MIS on each fingernail samples is different. Water content is decided from the hardness of microstructure and chemical elements of fingernails. To shorten the period of reduction, artificial heating by electric heater is chosen in this study. Three hours of heating seems to remove most of water in fingernails. But increased MIS

(a) *(b)* *(c)*

Fig. 1. (a). EPR spectrometer(EMX series), (b) Quartz and Cavity,
(c) Fingernail samples cut by conventional nail clipper and quartz tubes for EPR measurement.

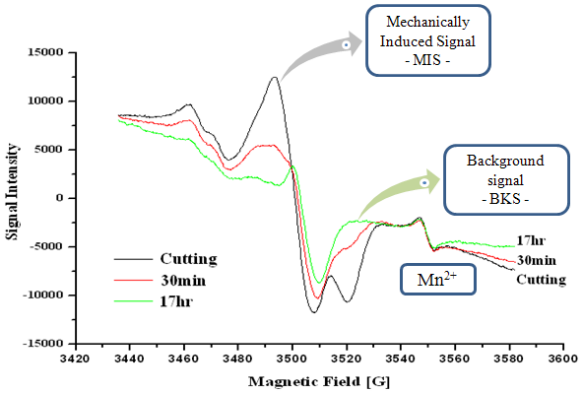

Fig. 2. Transformation of EPR signal spectrum of MIS and BKS (MIS) right after cutting process without heating.

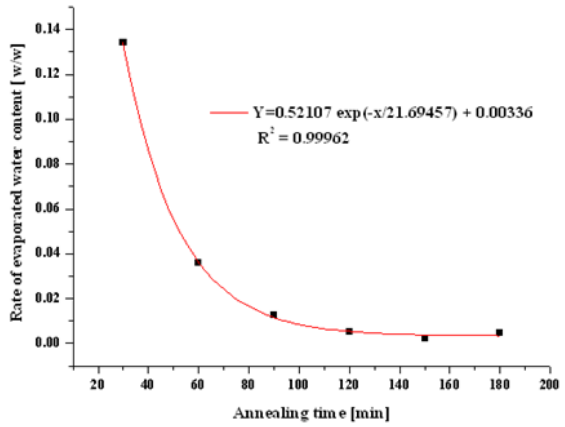

Fig. 3. Mass change rate on heating time for three hours at 70℃.

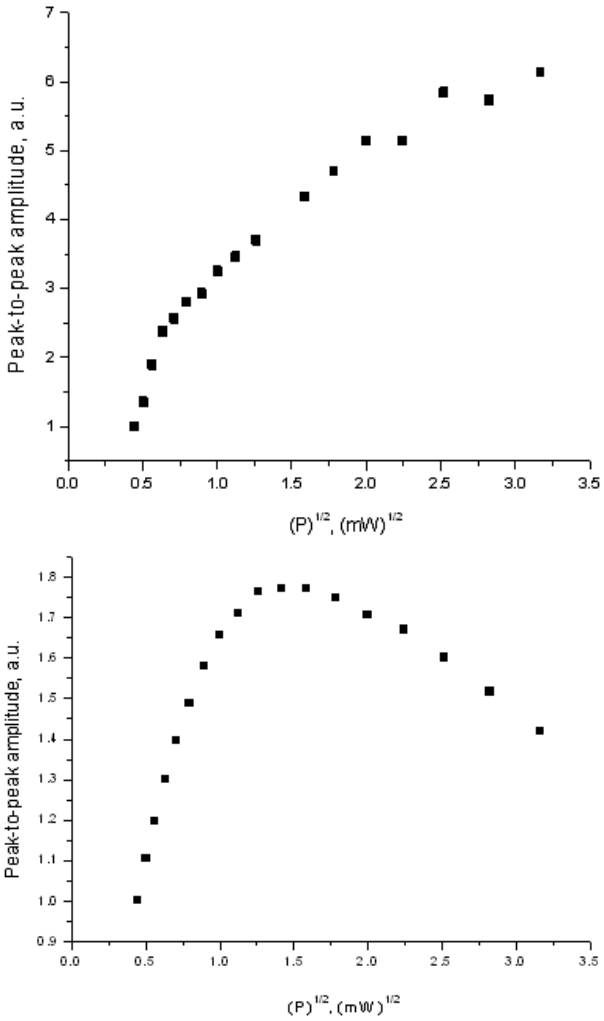

Fig. 4. A representative MIS2(upper) and RIS(lower) on consecutive microwave power level changes. At first, fingernail clippings were heated by electric heater at 70℃ for 72 hours and measured for MIS2. Then, it was exposed to 5 Gy of gamma source and measured for RIS .

intensity, another big obstacle needs another method for dosimetry. To evaluate exposure dose, conventional method which measure only peak-to-peak amplitude should be changed into some different measurement techniques depending on physical properties like power saturation behavior of two different signals, MIS and RIS.

And, as you can see in Fig. 2, at first stage of cutting process, the shape transformation of EPR spectrum is very rapid and the transformation period is at least 24 hours for stabilization. So it is impossible to do any measurement and comparison in this first one day after cutting period. After the first stage, relatively slow change is observed. So, for this period, artificial heating process was performed in this study. As you can see from Fig. 3, after artificial heating process for 3 hours at 70℃, we could observe that the mass change drastically thorough the evaporation process. It can be interpreted that water content inside fingernail structure got sufficient heat energy for relatively short time. After such a heating process, fingernail samples were measured by EPR spectrometer. Fig. 4 showed us characteristic behavior of EPR signals on power level increase. This stable shape of signal movement could not be observed before dehydration. Uncertainty was so high to get constant shape of peak-to-peak amplitude of EPR spectrum by use of normal (unheated) fingernail samples. This relation between water content and fixed shape of EPR signal change on microwave power level is possible only after the heating process. The various quantity of water content seems to prevent exact observation about power saturation behavior of MIS or RIS. As a result of dehydration process, EPR signal increment showed very special difference between MIS2 and RIS as in Fig. 4. This difference can be interpreted that MIS2 and RIS have apparently different microwave power saturation behavior. Actually this difference is apparently observed at two microwave ranges which is very high and low microwave power ranges. But, EPR signal at high level of microwave powers seems not to be useful for its instability of signals value as observed in Fig. 4. Low level of microwave power was relatively stable. From above experiment in this study, we believe that if we reduce water content by any means to certain level, it could be easy for determining some relationship between radiation exposure dose and EPR signals in future study. It means that

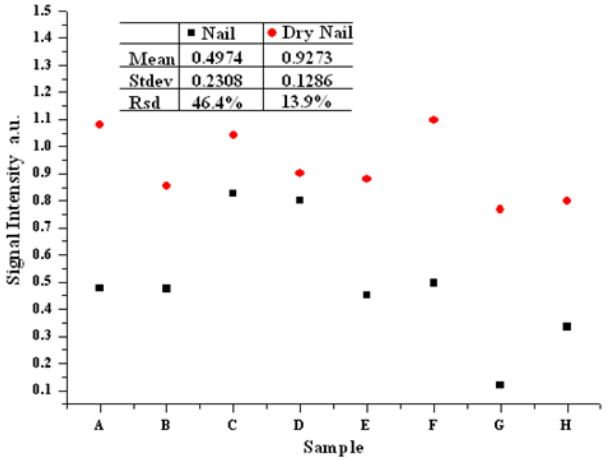

Fig. 5. EPR signals (peak-to-peak amplitude) of 16 fingernail samples from 8 different volunteer(two samples from each volunteer). Round one is the signal from 8 fingernail samples which was heated at 70℃ for 3 hours. Rectangular one is from the unheated normal fingernail samples. The signal deviation of heated group was smaller than that unheated normal groups.

EPR signal, absorption of microwave energy, didn't come from MIS or RIS radicals but from water contents inside fingernail clippings. Representative power saturation plot in Fig. 4 indicates that the power saturation behavior of the RIS and MIS2 was apparently different, especially at low microwave power levels of 0.3-0.8 mW. The fingernail samples exposed to gamma radiation showed a big curvature than that of the unexposed fingernail samples at low microwave power levels. The difference of the curvature of MIS and RIS in this study was distorted by effects of various water contents in each fingernail samples.

This effect of water content to stability of EPR signal is also tested by another method and the result is in Fig. 5. Standard deviation value seems to decrease after being heated for 3 hours apparently. We used the different samples from same volunteers for comparison. Apparent difference of uncertainty is observed between unheated and heated samples. Consequently, heated fingernail samples showed intrinsic physical properties of MIS and RIS. But the heating process necessarily induced uncontrolled increase of MIS. So, at low dose ranges under 5 Gy, conventional measurement method should be considered. As dose response value in 'dose-response curve', new response value should be considered for EPR dosimetry in near future.

4. CONCLUSION

In this study, we investigated the effect of water content and EPR signal on microwave power levels in fingernails. Low water content level in fingernail reduced variability of EPR signal and constant movement of EPR signals on microwave power levels. Although this method was difficult to apply in fingernail/EPR dosimetry, it is meaningful that we could find a method to differentiate MIS2 and RIS which are generally located at the same g-factor and almost impossible to be identified with each other. At low microwave power ranges, the curvature of RIS was apparently bigger than that of MIS2. In real situation, the curvature difference may not be so big, because exposed fingernails have both radicals related to MIS2 and RIS. This difference of curvature could be used for dosimetry, if curvature of MIS at unexposed state is not so fluctuated by heating process as being tested in this study. In future, this EPR signal curves can be fitted to some function and used for fingernail/EPR dosimetry.

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