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A Study on the Characteristics of Enamel to Electron Spin Resonance Spectrum for Retrospective Dosimetry

Daeseok HONG and Kunjai LEE

Korea Advanced Institute of Science and Technology
373-1, Guseong-dong, Yuseong-gu, Daejeon 305-701, Republic of Korea

Abstract

Electron Spin Resonance (ESR) spectroscopy is one of the methods applicable to retrospective dosimetry. The retrospective dosimetry is a part of dose reconstruction for estimation of exposed dose occurred years before the estimation. A tooth can be separated as enamel, dentine and cementum. Among the three parts, enamel is known as to show the best sensitivity to the absorbed dose and is most widely used. Since the later 80s, ESR dosimetry with tooth enamel has been studied and applied for the retrospective dosimetry. There are some factors affecting the sensitivity of enamel to absorbed dose. One of the factors is a size of enamel. Grain size of the 1.0mm~0.1mm range is commonly used and 0.6mm~0.25mm is recommended in other study. But the sensitivity can be varied by the grain size. In this study, the granular effect of enamel to the sensitivity is examined for application to retrospective dosimetry. In the enamel separation, to minimize the physically induced ESR spectrum, only chemical separation method was used. Separated enamels were divided by their size. The sizes of each sample is 1.0mm~0.71mm, 0.5mm~0.3mm, and below 0.1mm, respectively. All enamel samples show ESR spectrum related to the absorbed dose and the ESR spectrum shows linearity to the absorbed dose. The sensitivities are similar for each sample. But the enamel of size below 0.1mm shows poor characteristics relative to other enamel size. So, it is not recommended to use enamel samples below 0.1mm.

I. Introduction

ESR was discovered by Zavoisky(1945) for transition metal ions in salts and developed after World War II. Since mid 1950s, ESR has been used to measure radiation induced radicals in biological materials, but its potential for radiation dosimetry of bone and tooth enamel was not explored until mid-1960s. Interest in ESR dosimetry became apparent for accidental or emergency dosimetry using clothes, personal objects, and particularly tissues of the human body. In the later field the ESR technique found an early access to the reevaluation of individual doses, e.g. from A-bomb survivors of Hiroshima and Nagasaki[1)]. Today, ESR dosimetry with tooth enamel becomes a leading method for retrospective dosimetry of individual radiation exposures[2)]. But a lot of methods have been used for the separation of enamel from teeth and this may influence the dose evaluation[3)]. Also, it is known that various grain size samples show different regression coefficients to each other[4)]

In this study, we report on the effect of enamel size to ESR spectrum caused by radiation dose. In that viewpoint, the effect on background spectrum, the annealing process and the sensitivity of ESR spectrum is investigated.

II. Materials and Methods

1. Separation of Enamel

At the experiment, only molars or premolars which had been stored in ethanol were used. The surfaces of all teeth were clean and showed no signs of disease. But all the teeth might have been exposed to dental X-ray at least one time.

Using dental saw, the tooth crowns were separated from the root. As the separation process using dental drill or saw can induce additional ESR center by localized heating, no further mechanical working was applied. And each tooth crown was treated by ultrasonic bath in 2N NaOH solution. During ultrasonic treatment, NaOH solution was replaced at every 2 hours. After 2 times of ultrasonic treatment, all the samples were cleaned with distilled water and dried in oven at about 60℃. At the end of the drying process, remained dentines were scraped off with dental drill. The whole processes were repeated only pure enamels remained.

Before the experiment, all enamels were mixed up and crushed into powders to minimize the individual difference. Then the enamels were divided into 9 samples. Among them, 3 samples were used for the non-irradiated standard samples for each size group while the others were divided into 3 groups with 2 samples for each group. The enamel sizes were 1.0~0.71mm(S1), 0.5~0.3mm(S2) and smaller than 0.1mm(S3), for each group respectively.

2. Irradiation and ESR spectrum measurements

At irradiation, the source was Cs-137 and the exposure rate was 8.77(±6%) cGy/sec. The irradiator was IBL 437C model in RHRI (Radiation Health Research Institute). For the measurement of ESR spectra Bruker EMX model in the microwave of X-band was used. The parameters for the signal acquisition were such as the magnetic field sweep of 8mT, modulation amplitude of 0.5mT, time constant of 2.56ms and microwave power of 20.12mW. The other parameters (sweep field, time constant, conversion time) were set in order to assure a correct recording of the signals. To reduce the effect of transient signal, enamel samples were annealed at a temperature of about 92℃ for 2hours5).

To estimate radiation-induced signal, spectrum subtraction method was used. In the method, the ESR spectrum of a non-irradiated reference sample is subtracted from the spectrum of the irradiated sample[2)]. Also, using the g-factor of DPPH standard sample, g-factor of each spectrum was determined.

III. Results

1. Comparison of background signal

Background ESR spectrum of each sample was measured 3 times without annealing. After the first measurement, the second measurement was performed after 3 days and the third measurement was one after 3 months later. The change in peak-to-peak (PTP) amplitude of spectrum between the first measurement and the second one was within about 10% for S1 and S2 but about 30% for S3.

As shown in Fig. 1, the change in PTP amplitude between the first and the third one increased to 50% for S3. Among the samples, S2 shows the least PTP change between the measurements.

2. Size effect to annealing process

From Fig. 2 to Fig. 4 show the change of ESR spectrum caused by annealing effect. As shown, each sample displays the change of ESR spectrum in radiation sensitive and insensitive signal. These differences increased as the enamel size decreased and exposed dose increased. Also, group S1 shows drastic changes in radiation insensitive signals during the annealing process.

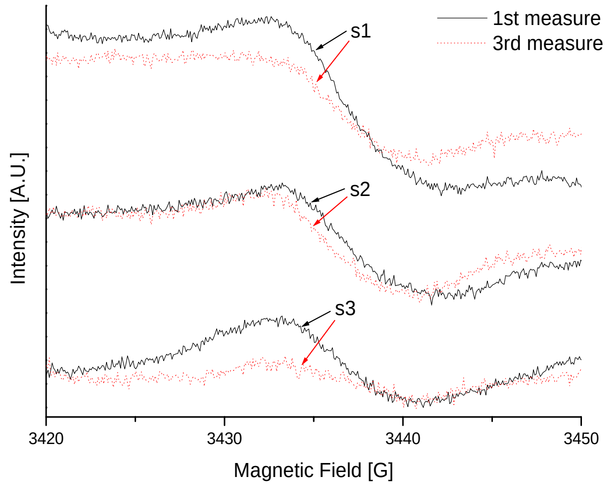

Fig. 1. Background ESR signal change

The changes in PTP amplitude of radiation sensitive signal was below 10% but the changes in PTP amplitude of radiation insensitive signal increased drastically as the irradiation time increased.

In the figures, the asymmetric ESR signal with $g\perp$ = 2.0018 and $g\parallel$ = 1.997 which is sensitive to radiation was determined using g-factor of standard DPPH sample.

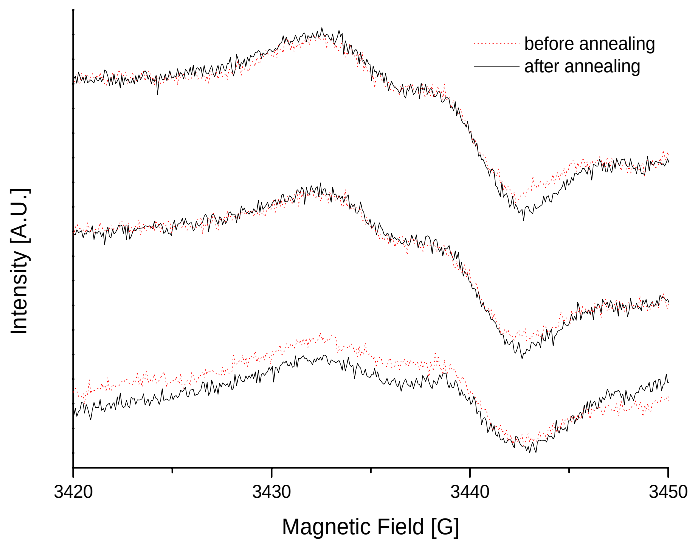

Fig. 2. Changes of ESR spectrum by the annealing process (irradiation time = 11sec)

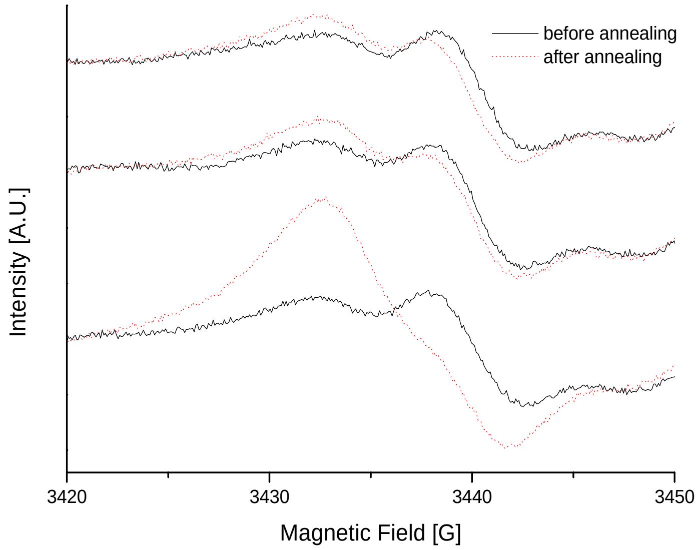

Fig. 3. Changes of ESR spectrum by the annealing process (irradiation time = 23sec)

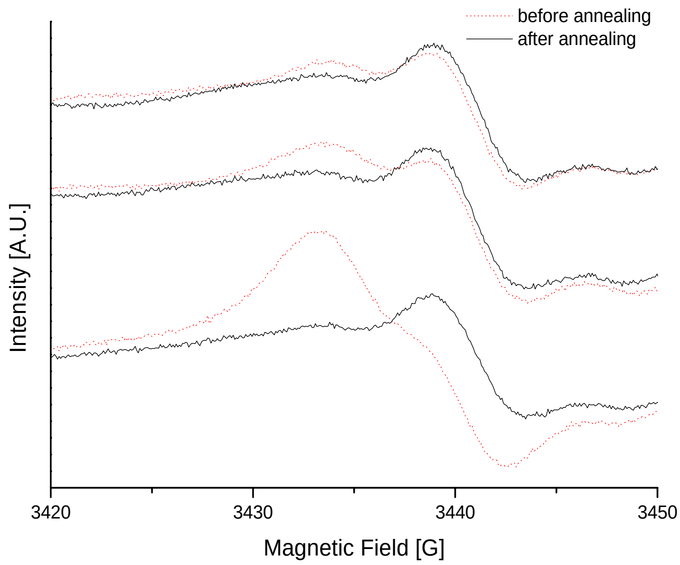

Fig. 4 Changes of ESR spectrum by the annealing process (irradiation time = 31sec)

3. Comparison of linear regression analysis

The PTP amplitude of each group for the exposed dose was analyzed using linear regression analysis. For the analysis, simple linear fitting function of Eq. (1) was used.

$$(\text{PTP amplitude}) = A + B\times(\text{exposed dose}) \qquad (1)$$

As listed in Table 1, every group showed linear relationship between exposed dose and ESR spectrum. Among 3 groups, group S3 shows poor correlation coefficient and linearity. Group S2 shows better properties for linear relationship.

Table 1. Results of linear regression analysis of each group
(R ; correlation coefficient, A & B ; refer to Eq. (1))

	R	A	B
S1 (1.00~0.71mm)	0.985	-409.27	29.47 ± 1.29
S2 (0.5~0.3mm)	0.983	-620.08	31.76 ± 1.51
S3 (below 0.1mm)	0.944	-755.20	26.76 ± 2.33

In Fig. 5, the result of linear fitting is plotted. In accordance with Table 1, group S2 and S3 shows better properties than other groups.

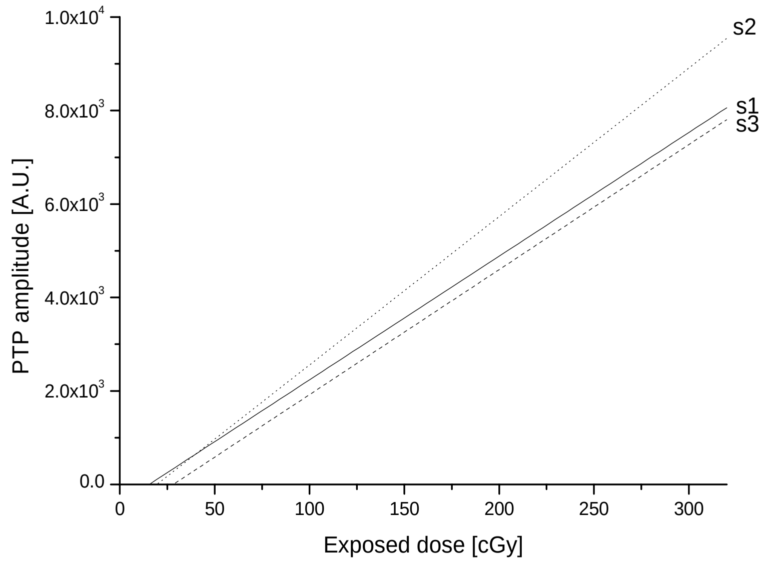

Fig. 5 Linearity of ESR spectrum to exposed dose for each group

IV. Discussion

As shown in the results, ESR spectrum of enamel shows linearity to radiation dose. But the size of enamel sample can affect the process of dose evaluation. Background signal, spectrum change during the annealing process and the sensitivity of ESR spectrum to exposed dose are affected by the size of enamel sample.

The result of background signal comparison shows that the transient ESR signal induced by the sample preparation process may last long time and the transient ESR changes a lot for the sample of fine size. This can be explained by the mechanical stress which generates additional radicals in enamel during the crushing process of enamel. So, S3 group shows large change in transient signal.

Annealing is required for dose evaluation to minimize the transient signal effect of ESR spectrum. Also, it causes a spectrum change in both radiation sensitive and insensitive signal and this can be reduced by choice of appropriate sample size.

Finally, the sample size shows the effect on the linearity of ESR spectrum to exposed dose. This should be reduced for the exact estimation of radiation dose. According to the results of experiment, sample size of 0.3mm~0.5mm shows better properties than other sample size ranges.

V. Conclusion

Enamels with the size of 1mm and below are applicable to radiation dosimetry. But the effect of sample size should be considered importantly. It can affect the dosimetry in the viewpoint of background spectrum, transient signal and sensitivity of ESR spectrum to exposed dose.

The results of this study show that enamels with the size range of 0.3mm~0.5mm is the best for dosimetry but enamels with the size of 0.1mm or below shows poor properties. It is also known that the spectra of samples with smaller grain sizes frequently have additional low-intensity ESR signals, but these signals usually possess random properties[6)]. So, it is not recommended to use enamel samples of below 0.1mm size for dosimetry.

References

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