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Preparation-induced Errors in EPR Dosimetry of Enamel: Pre- and Post-crushing Sensitivity

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Polyakov *et al.* (1995) showed errors in dose estimation as a function of grain size for enamel grains given beta irradiation after crushing. We tested the effect of gamma irradiation applied to the specimens before and after crushing.

We confirmed Polyakov's observations and found that post-crushing irradiation altered the slope of the dose-response curve of the hydroxyapatite signal and produced a grain-size-dependent offset. No changes in the slope of the dose–response curve were seen in enamel caps irradiated whole before crushing.

Introduction

Tooth enamel of various origins is a promising material for dating geological materials and retrospective accident dosimetry (Grün *et al.*, 1990; Ikeya *et al.*, 1986; Ishii and Ikeya, 1990; Nishiwaki and Shimano, 1990; Pass and Aldrich, 1985; Rink and Schwarcz, 1994; Shimano *et al.*, 1989; Stringer *et al.*, 1989; Tatsumi-Miyajima and Okajima, 1985). Major advantages of enamel include near absence of organic material with its associated competing EPR signal. Among the disadvantages are preparation-induced signals which interfere with measurement of the main radiation-sensitive signal.

There are two major signals as well as several smaller ones which are induced by mechanical trauma (Desrosiers *et al.*, 1989). One of these has a Lande value of $g = 2.0020$ and may be due to mechanically-induced amplification of the main hydroxyapatite signal, or a new signal which is almost exactly superimposed on the radiation-sensitive hydroxyapatite signal. The magnitude of this signal may be the equivalent of several Gy of experimental irradiation. Desrosiers *et al.* (1989), reported generating this signal by preparing granules with a dental drill.

Generation of the $g = 2.0020$ signal can be avoided if a gentler method of sample preparation is used, i.e. grinding with a mortar and pestle (Desrosiers *et al.*, 1989). In this case, a small signal is generated at $g = 2.0038$ (Desrosiers *et al.*, 1989; Pass and Aldrich, 1985; Tatsumi-Miyajima and Okajima, 1985).

Polyakov *et al.* (1995), recently demonstrated that the magnitude of the $g = 2.0038$ signal is increased by exposure to ^{90}Sr beta radiation. Further, their findings indicate the possibility of overestimation of doses with small grains and underestimation with large grains. Their data indicated that at an average grain size of approx. 200 μm, the effects were canceled.

In an effort to further elucidate the problems associated with mechanical trauma and grain size on enamel, and to extend the work to include gamma irradiation, we tested the effects of gamma rays applied to enamel before and after crushing.

Experimental

The design of the experiment is shown in Fig. 1. An enamel cap from a molar was weighed and irradiated with 5.6 Gy. The cap was then split into two halves which were weighed and designated as sides C and H, named for their state at the time of subsequent irradiation, H for half and C for crushed. C was crushed and sieved into four aliquots of sizes > 250, 106–250, 75–106 and < 75 μm. The 75–106 μm aliquots were discarded from both the H and C groups because of insufficient sample. EPR spectra were then taken for each of the three C aliquots separately. These three aliquots were then recombined into one conglomerate which was then reirradiated to a total of 11.2 Gy (5.6 + 5.6). Following this, the grains were resieved back into the same three aliquots and each was individually scanned. H was reirradiated prior to crushing, to a total of 11.2 Gy. It was then crushed and sieved into the same sizes as was C. The three aliquots from B were then scanned. The masses of the granules are given in Table 1.

For purposes of mass normalization, we had previously determined the EPR response to enamel

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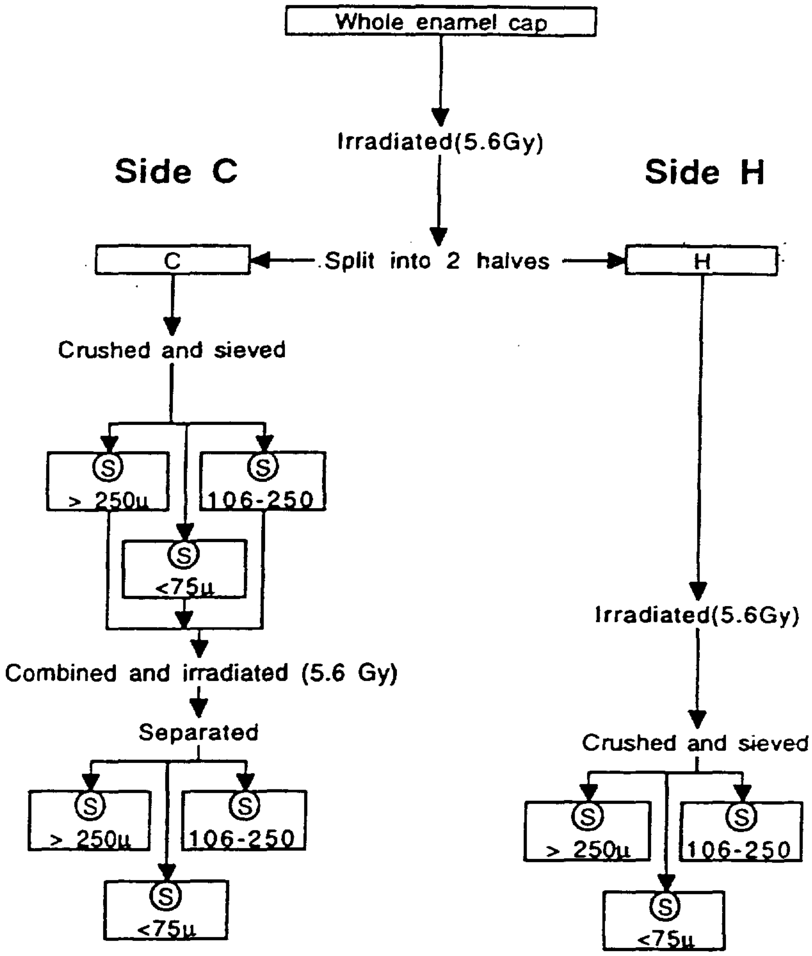

Fig. 1. Experimental protocol. Symbol (encircled S): specimen was scanned at 1 mW.

granules over a range of 0–100 mg in 5 mg increments. The results were linear with $R = 0.990$.

Dose responses were obtained at 5.6 and 11.2 Gy for the three C aliquots. This was done to test Polyakov's observation that there is a grain-size-sensitivity relationship.

The tooth used in this study was obtained from a routine extraction by the dental clinic of the Salt Lake City FHP (Family Health Plan) Hospital. Records of previous diagnostic X-rays were not available. Approximately the apical one-fourth of the tooth was removed with water irrigation using a 4" × 0.012" (10.2 cm × 0.03 cm) diamond saw blade mounted on a Buehler Isomet saw (Buehler Ltd, Lake Bluff, IL 60044). This removed most of the root of the tooth. The remaining dentine was removed with a dental drill using a ball bur. This was practical because the enamel could be visually differentiated from the darker dentine.

Table 1. Crushed sample masses (mg)

Specimen group	Grain size > 250 μ	106-250 μ	< 75 μ
Crushed sample C, 1st scan			
Mass	118.47	42.21	29.03
Crushed sample C, 2nd scan			
Mass	112.14	41.90	27.85
Crushed sample H			
Mass	103.56	65.17	52.54

The cap was divided in halves using the Buehler Isomet Saw. In this case, an effort was made at cutting which resulted in the specimen splitting neatly in half shortly after cutting began. Crushing was done with an agate mortar and pestle.

The specimens were irradiated with a ^{60}Co source in a volume irradiator (US Nuclear, Model GR9, Burbank, Calif.) with a dose rate to hydroxyapatite of 0.22 Gy min^{-1}. Calibration of the source was done with alanine EPR dosimeters (Albrecht Wieser Messtechnik, Munchen, Germany). The whole and half caps were irradiated in a copper cylinder (16 mm o.d. × 66 mm long, wall thickness 2 mm). The bottom 32 mm of the cylinder was filled with a plug of nylon glued in place with epoxy resin. A 45 mm long rod of nylon was used as a cap. The granules were irradiated in an aluminum holder (10.7 mm o.d., wall thickness of 2.7 mm). Confirmation of

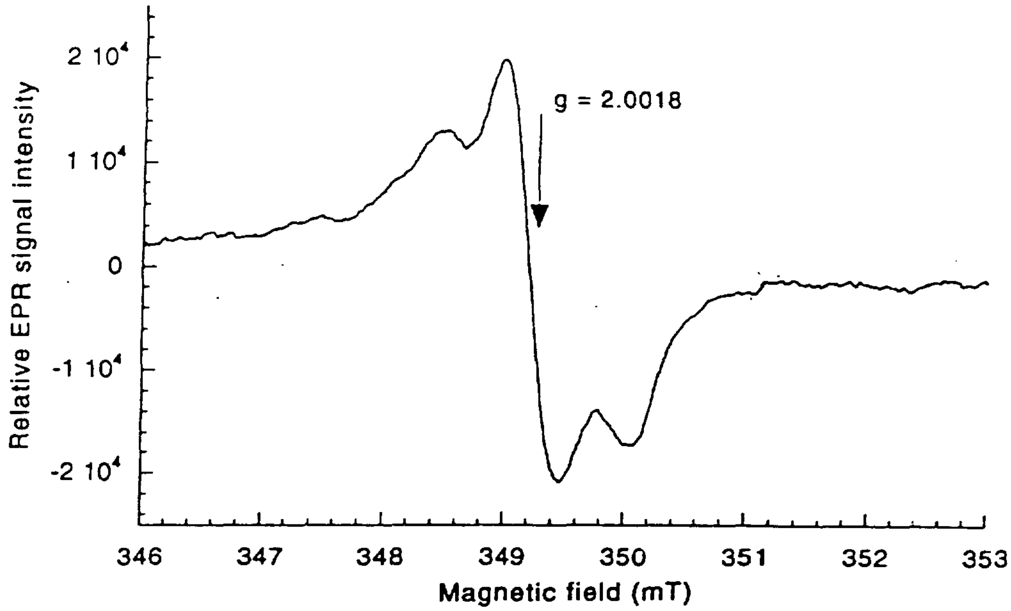

Fig. 2. Typical EPR spectrum for enamel. Post-irradiated specimen. 106–250 μm. 40 sweeps. Power 1 mW.

electron buildup and equivalence of absorbed dose to hydroxyapatite with the two capsule configurations was made using a commercial program for modeling dose deposition (PHOTCOEF, AIC Software, Grafton, MA 01519).

The EPR analysis was done using a conventional X band spectrometer (Bruker Instruments, Billerica, Mass.). Parameters used were power 1 mW, field modulation frequency 100 kHz, modulation amplitude 0.5 mT, time constant 164×10^{-4} s, receiver gain 1×10^5, room temperature. The microwave resonance frequency was approx. 9.7 GHz. Scan width was 7.5 mT. Weak pitch was used as the reference standard. Sweep rate was 1 sweep/0.43 min. Granules > 250 μm were scanned twice for 10 sweeps each. 106–250 μm and < 75 μm were scanned once for 40 sweeps, except for the measurement made on the 106–250 μm granules at 11.2 Gy of the C specimens which were scanned three times for 40 sweeps each. The powder was placed in a 0.495 cm o.d. × 17.8 cm long quartz EPR sampling tube (Wilmad Glass, Bena, NJ 08310). All spectra were mass normalized (using a linear mass vs signal size relationship) to a 100 mg sample size. This was done by multiplying the spectra by a factor of 100 divided by the sample's actual mass. The samples were stored at room temperature and low (ambient) humidity.

Measurements of the EPR signals were made peak-to-peak. The peaks were smoothed over 0.01 mT. The background signal was not subtracted from the spectra. Statistics were done using standard methods (Spiegel, 1961).

Results

Figure 2 shows a typical EPR spectrum for enamel.

Figure 3 shows the dose–response of the postirradiated specimens (first dose to whole cap, second dose to grains). The specimens shown at 5.6 Gy were irradiated as a whole enamel cap, split into two halves, one of which was crushed and then scanned. The resulting granules were then given an additional dose of 5.6 Gy and rescanned. The granules decrease in sensitivity with decreasing grain size as reported by Polyakov and his coworkers.

Figure 4 shows the dose–response of the preirradiated specimens (both irradiations applied prior to crushing). The points at 5.6 Gy (same as in Fig. 3) and the 11.2 Gy (re-irradiation of remaining half cap, 5.6 Gy + 5.6 Gy) were all irradiated before crushing. The sensitivities are similar for all grain sizes. An increasing negative offset is seen with decreasing grain size.

Polyakov and his coworkers reported that there appeared to be a crossover point at about 200 μm where the mechanical signal in large grains enhanced the sensitivity of the hydroxyapatite signal while its effect in small grains was to decrease the sensitivity. Figure 5, which is a plot of the estimated dose to the specimen vs grain size, suggests that the crossover point occurs between 200 and 500 μm.

Discussion

The postirradiation study (first dose to whole cap, second dose to grains) confirmed the observations of

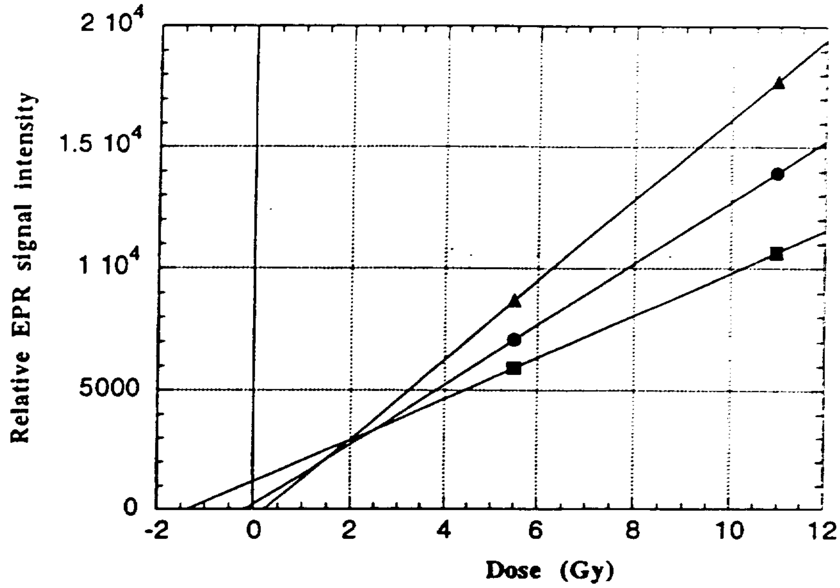

Fig. 3. Dose-response of post-irradiated specimens. The enamel cap was irradiated with 5.6 Gy and subsequently split in two. One half cap was crushed and the resulting granules were scanned. These are the points at 5.6 Gy. The granules were then given an additional 5.6 Gy and rescanned. These are the points at 11.2 Gy. Triangles: $> 250 \mu m$. $y = -338 + 1650x$. Circles: $106–250 \mu m$. $y = 238 + 1248x$. Squares: $< 75 \mu m$. $y = 1191 + 862x$. Granules $> 250 \mu m$ were scanned twice for 10 sweeps each (SD = 40.3–86.2). $106–250 \mu m$ and $< 75 \mu m$ were scanned once for 40 sweeps, except the measurement made on $106–250 \mu m$ granules at 12.2 Gy which were scanned three times for 40 sweeps each (SD = 918.2).

Polyakov *et al.* (1995) that there is a decrease in sensitivity of the radiation induced hydroxyapatite signal at $g = 2.0018$ with decreased grain size.

We interpret the results of the preirradiation study (both irradiations applied prior to crushing) to mean that the mechanical signal is not affected by irradiation received prior to crushing, and that the samples' sensitivities to radiation applied prior to crushing are likewise not grain size dependent. Although it appears that both methods produce errors in dose estimates depending on the size of the grains being analyzed, that is not necessarily the case since a background zero signal has not been subtracted in either method. In the postirradiated

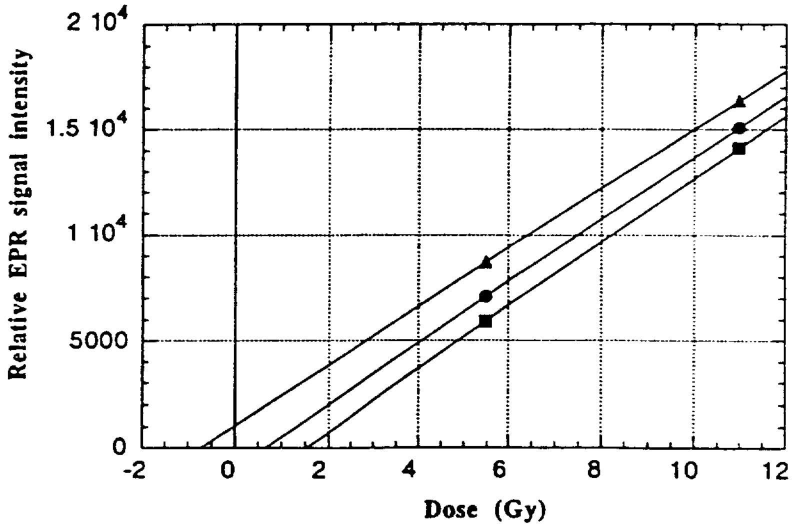

Fig. 4. Dose-response of pre-irradiated specimens. The points at 5.6 and 11.2 Gy were all irradiated before crushing. Triangles: $> 250 \mu m$. $y = 1029 + 1401x$. Circles: $106–250 \mu m$. $y = -925 + 1460x$. Squares: $< 75 \mu m$. $y = -2281 + 1496x$. The points shown at 5.6 Gy are the same as those in Fig. 3. In the case of the 12.2 Gy points, the granules $> 250 \mu m$ were scanned twice for 10 sweeps each (SD = 404.5) while the $106–250 \mu m$ and $< 75 \mu m$ granules were scanned once for 40 sweeps.

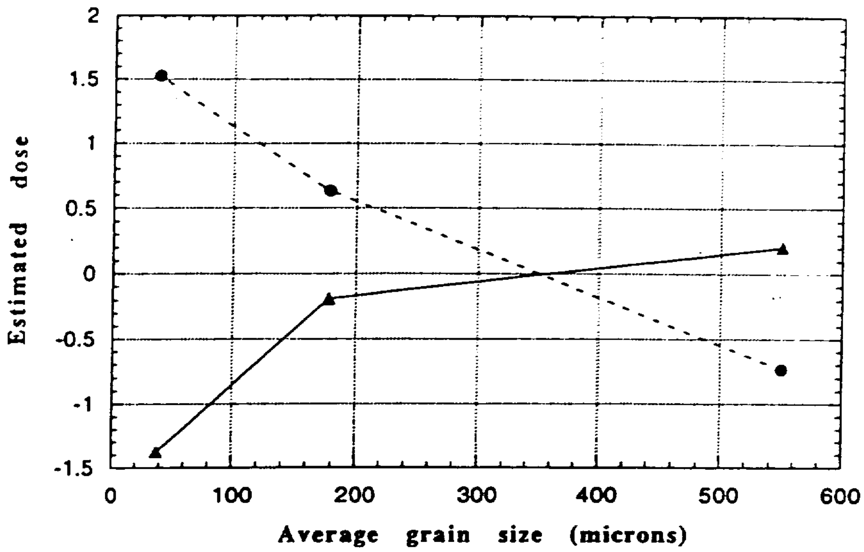

Fig. 5. Estimated dose vs average grain size. The estimated grain size was determined by averaging the maxima and minima of each grain fraction. The zero crossover points occur between 300 and 500 μm. Diamonds: estimate from post-irradiation specimens (Fig. 2). $y = 0.0027x - 1.14$, $R = 0.86$. Circles: estimate from pre-irradiated specimens (Fig. 3). $y = -0.00426x + 1.56$, $R = 0.99$.

specimens, subtraction of the background signal appropriate for the grain sizes in question would be expected to produce identical fits for each of the grain sizes since the slopes are similar. The preirradiation study, on the other hand, has errors which are a function of both background and sensitivity change, and subtraction of background signals would not eliminate the errors.

The decrease in sensitivity with decreasing grain size can be explained by postulating either a decrease in the number of, or an increase in competitors of, the radiation-sensitive hydroxyapatite centers. This could be a uniform volume effect with the smallest grains assumed to have the greatest stress and thus effect, or a surface to volume phenomenon with the surface of all grains affected equally. The increase of surface to volume ratio with decreasing grain size would then account for the sensitivity difference.

Another possible explanation for the effects observed in this study concerns increases in packing density (sample mass/sample volume) associated with decreasing grain sizes. Although packing density was not examined in this study, such an effect could produce apparent sensitivity changes due to normalization nonlinearities and could conceivably produce differential broadening of the mechanical vs the radiation sensitive signal resulting in the grain size dependent offsets observed here. We plan to address this issue in future work.

Conclusion

We have confirmed the observations of Polyakov *et al.* (1995) that the sensitivity of hydroxyapatite to radiation is dependent on the size of grains analyzed. Further, our results indicate that preirradiation does not affect the sensitivity of the hydroxyapatite dose response curve. Finally, this experiment suggests that the optimum grain size range for accurate dose estimation would be approx. 200–500 μm.

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