DOE/NV/10805--T3

EPR Dosimetry of Teeth in Past and Future Accidents: A Prospective Look at a Retrospective Method

E. Haskell[1], G. Kenner[1], R. Hayes[1], V. Chumak[2] and S. Shalom[2]

[1] Center for Applied Dosimetry, University of Utah
[2] Scientific Center for Radiation Medicine, Kiev

Accurate assessments of doses received by individuals exposed to radiation from nuclear accidents and incidents such as those at Hiroshima and Nagasaki, the Nevada test site, Cheliabinsk and Mayak are required for epidemiological studies seeking to establish relationships between radiation dose and health effects. One method of retrospective dosimetry which allows for measurement of cumulative gamma ray doses received by exposed individuals is electron paramagnetic resonance spectroscopy (EPR) of tooth enamel. Tooth enamel stores and retains, indefinitely, information on absorbed radiation dose. And teeth are available in every population as a result of dental extraction for medical reasons including periodontal disease and impacted wisdom teeth. In the case of children, deciduous teeth, which are shed between the ages of 7 and 13, can be a very important dosimetric source if documented collection is implemented shortly following an accident.

Current status

EPR of tooth enamel is a relatively new technique for retrospective dosimetry but in the past two years increasing effort has been put into its development and evaluation. Efforts have centered on determining the accuracy which may be achieved with current measurement techniques as well as the minimum doses presently detectable. One study conducted through the joint European Union (EU)/Former Soviet Union (FSU) project, ECP-10, involved the blind measurement of doses delivered to enamel grains by a Cs-137 laboratory source. The intercomparison involved eight laboratories from the EU and FSU and one from the United States. Grains from 40 teeth were combined to form a homogenous sample. Five dose levels ranging from 0-1 Gy were applied prior to distribution of the grains to the participants. Each

laboratory was free to process the grains further if desired and to apply their normal methods of analysis. The results were as follows:

1000 mGy samples:	± 25% for 6 labs
500 mGy samples:	± 25% for 5 labs
250 mGy samples:	±100 mGy for 5 labs
100 mGy samples:	±100 mGy for 4 labs
0 mGy samples:	±100 mGy for 4 labs

No laboratory achieved ±10% for analysis of the 100 mGy samples.

This first intercomparison used enamel grains irradiated after crushing and could not be considered a "real life" test of the technique since it did not allow for examination of the effects of irradiation on whole teeth nor the effects of crushing on dose estimation. However, an observation emerged from the results concerning the value of sample purification on dose measurement. The laboratory which used the most rigorous procedure for sample purification and evaluation of purity produced measurements most consistent with the applied doses (Fig. 1). The procedures used by that laboratory are summarized in a later section. The study also revealed that for the methods of analysis used, doses of 10 mGy could not be measured with high accuracy.

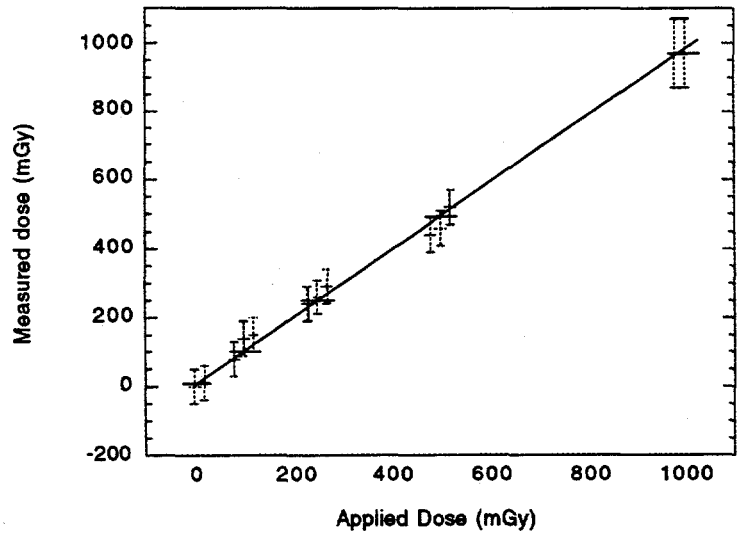

Figure 1. Measurement results for laboratory with stringent sample purification criteria. (From Chumak, et al, 1996A).

As a follow-up of the first intercomparison and to test the feasibility of a method of intercomparison which would allow for assessment of preparation effects on teeth, a bilateral study was performed on teeth irradiated prior to crushing. The protocol of this intercomparison involved the bi-section of adult molars followed by the irradiation of one half of each bisected tooth. The other half remained unirradiated and three matched pairs with blind dose levels known to be in the range of 10 to 50 mGy were given to both laboratories for analysis. Samples T1, T2 and T3 were supplied to one laboratory, samples T4, T5 and T6 were supplied to the other. The initial results of the study (given prior to disclosure of the applied doses) are shown in Table ___. These results were of the dosed teeth only, since time constraints precluded detailed analysis of the undosed pairs. Subsequent to disclosure of applied doses both laboratories performed additional measurements using the unirradiated teeth. One laboratory used a new method of analysis called the differential microwave power method (Romanyukha, et al., 1995; Serzhenkov, et al., 1995) which is described below. The results of these analyses are given in Table 2. The other laboratory performed analyses on additionally bisected portions of the teeth in an attempt to assess the magnitude of prior dental x-ray exposure (see below).

Group	Laboratory added dose, mGy	Measured dose, mGy
Group 1	171	
T1		190±50
T4		230±50
Group 2	256	
T2		180±50
T5		250±50
Group 3	200	
T3		190±100
T6		300±70

Table 1. Results of blind intercomparison intercomparison. (From Haskell, et al., 1996B).

	With paired Background Subtracted			No Subtraction	
Sample code	2mW	25mW	Differential	Differential	Applied (mGy)
T1E	190	180	170	310	171
T2E	140	200	250	300	256
T3E	10	110	70	210	200

Table 2. Results of differential power method. (values in mGy). (From Haskell, et al., 1996B).

Some factors influencing the accuracy of EPR dosimetry of enamel.

One factor effects the accuracy of EPR dosimetry of enamel more, perhaps, than any other. A large background signal, the so called "native" signal overlies the region of the radiation induced signal. Fig. __ shows a spectrum of enamel given a dose of 1.5 Gy while **Fig** __ shows a doconvoluted spectrum of enamel given a 100 Gy dose. The two components of the radiation induced signal, the parallel and perpendicular together with the native signal are clearly shown in Fig __ but at the lower dose the radiation sensitive signal is large obscured by the native signal. In many dosimetry methods, the signal being measured begins at zero and increases with increasing dose. The same is apparently true for the radiation induced EPR signal of enamel, however, because it is superimposed upon an underlying signal, its zero value is impossible to determine directly. Instead the zero must be determined using one of several methods. It is possible to 1) measure a standard unirradiated sample having only the native signal and assume its similarity to the native signal of the dosed sample in question, 2) model the native signal using curve fitting methods and apply this generated spectrum to the sample in question or 3) eliminate the native signal using sample measurements taken at two, well selected, microwave powers. Each of these methods is discussed further below.

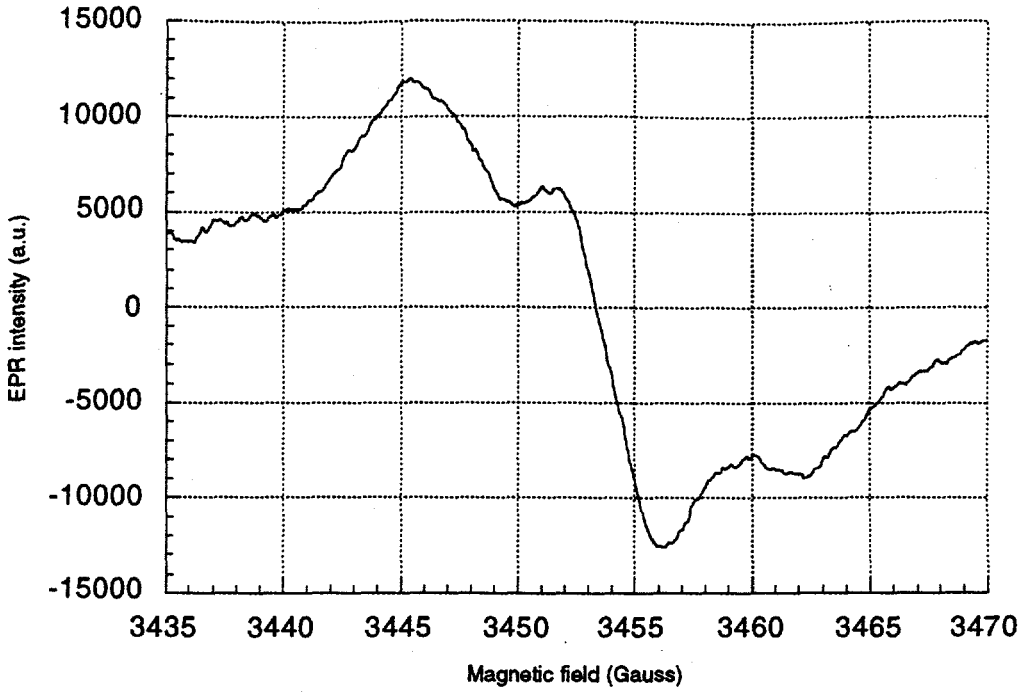

Figure. EPR spectrum of enamel sample given 1.5 Gy dose.

If the only signals present in enamel were the native signal and the radiation induced signal, then these methods would produce consistent, reliable results. Unfortunately, additional signals, introduced environmentally, or during preparation and analysis, can be present, and their presence can be obscured by the large, overriding, native signal. These spurious signals include a mechanical signal introduced with excessive crushing, an organic signal which results from the presence, as the name implies, of organic impurities, a light induced signal which has components separate from, and coincident with, the radiation induced signal, and a transient radiation induced signals which changes with time following irradiation.

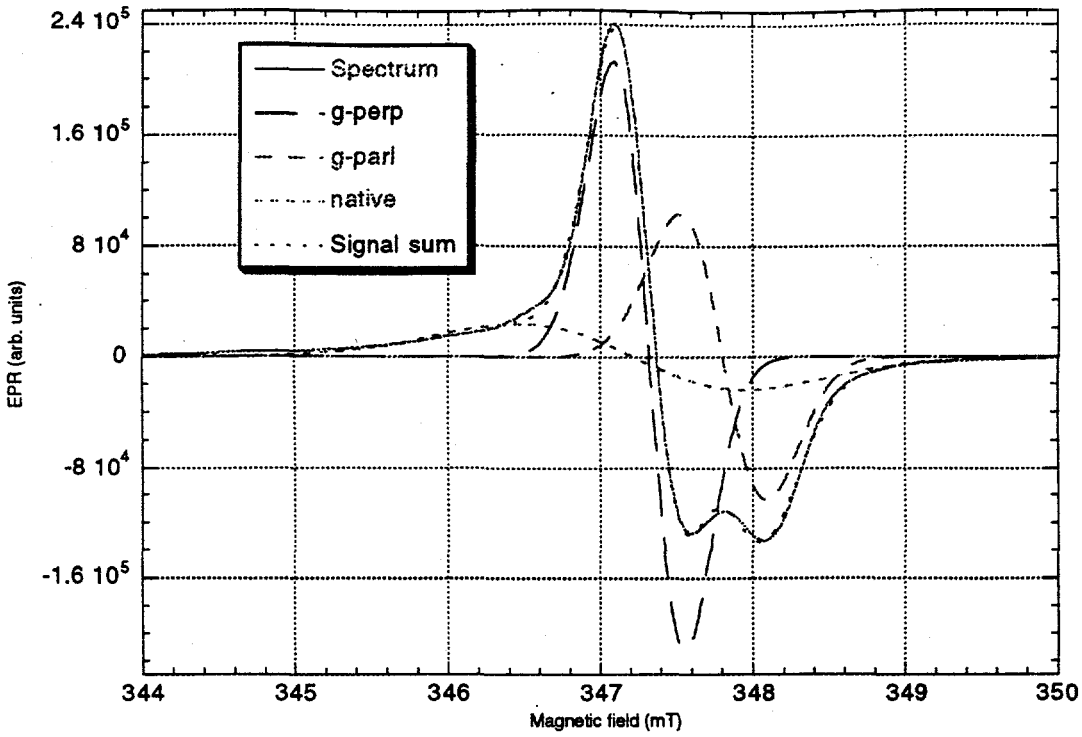

Figure 4 Deconvoluted spectra of enamel given 100 Gy dose.

Instrumental considerations

EPR signals increase as the square root of the microwave power until saturation slows and then reduces the signal intensity. Different signals saturate at different levels, and a large difference exists in saturation level for the native signal and for the radiation induced signal of enamel **(Fig __)**.

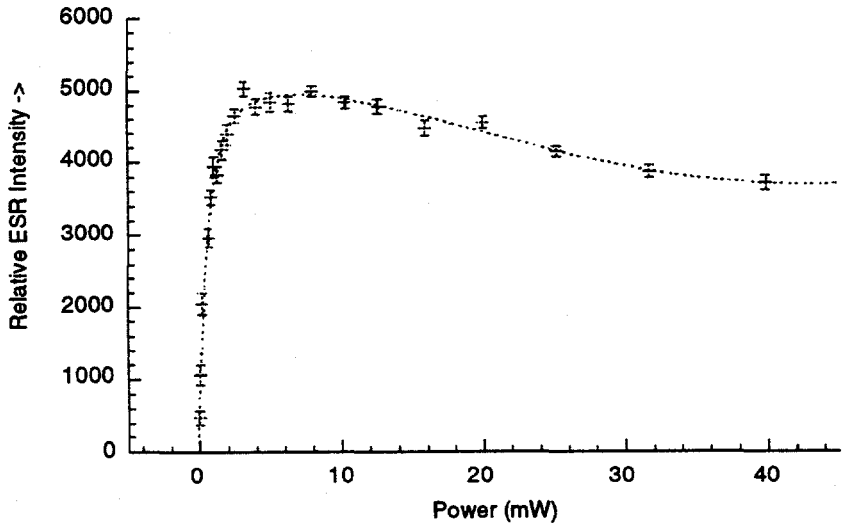

Figure. Microwave power saturation curve for native signal of enamel.

Since the ratio of the radiation induced signal to the native enamel signal increases even at microwave powers in excess of 200 mW, it would appear that measurement at the highest microwave powers would be desirable. Unfortunately, the noise level of the enamel spectrum, in general, also increases with power. The competing signals mentioned above each have their own saturation characteristics which can lead to complex variations in the shape of the spectrum as a function of microwave power (**Fig __**). Furthermore, anisotropic effects, variations in signal intensity which result from positional changes in sample orientation, are often enhanced at elevated powers. For these reasons, lower microwave powers have been preferred for EPR dosimetry of enamel, however recent innovations in preparation and instrumentation promise to change this approach. Rigorous purification methods drastically reduce anisotropies, while the use of a goniometer for sample rotation during spectral collection averages anisotropic effects (Haskell, et al., 1996A), to the point that whole, untreated teeth, can be accurately measured (unpublished data).

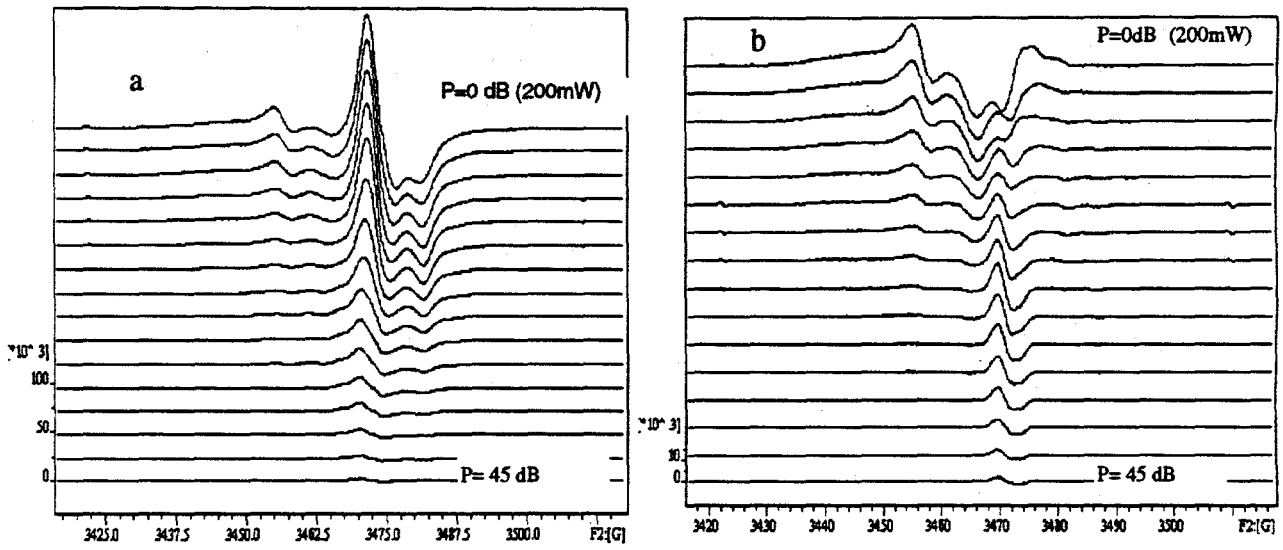

Figure. 3. Spectra of sample with dose of 100 Gy, recorded at different microwave power. The upper spectra in each parts a and b of Fig. 3 correspond to maximum microwave power 200 mW (attenuation 0 dB). The attenuation between each neighbor spectra is 3 dB, so as the bottom

spectra correspond c. 0.0638 mW power (45 dB attenuation). Set a are original spectra, set b - result of subtraction from a appropriate signals of sample Tr16 and consist of "pure" transient signals. (From Sholom, et al, 1996A).

Two environmental factors are of concern to the ultimate accuracy of EPR dosimetry. The first is exposure to dental x-rays. The second is exposure to sunlight. Dental x-rays are of major concern due to the greater absorption of low energy photons in enamel. For x-rays of 60 keV, the deposition could be a factor of 6 greater than gamma rays of 1.5 MeV. Steps toward detection of x-ray doses and correction for them has involved differential measurement of the inner and outer portion of teeth. Such measurements were a part of the bilateral intercomparson, above, and are shown in figure ___.

Tooth #	Inner part (unexposed) mGy	External part (unexposed) mGy	Inner part (exposed), mGy	External part (exposed), mGy	Mean total dose of the exposed half, mGy	Mean "unknown" laboratory dose of the exposed half, mGy
T4	30	50	230	240	230±50	195±50
T5	20	50	230	270	250±50	215±50
T6	20	50	290	300	300±70	260±70

Table 3. The results of dose determination for different parts of teeth (SCRM). (From Haskell, et al., 1996B).

Light effects were first suspected of affecting dose measurements during a large scale study of teeth from populations in Russia exposed to the Chernobyl fallout (ref). Uncertainties in the dose measurements were reduced substantially when front teeth were removed from the study. Subsequent results have shown that UV irradiation can cause substantial errors and it is now recommended that only molars and wisdom teeth be examined.

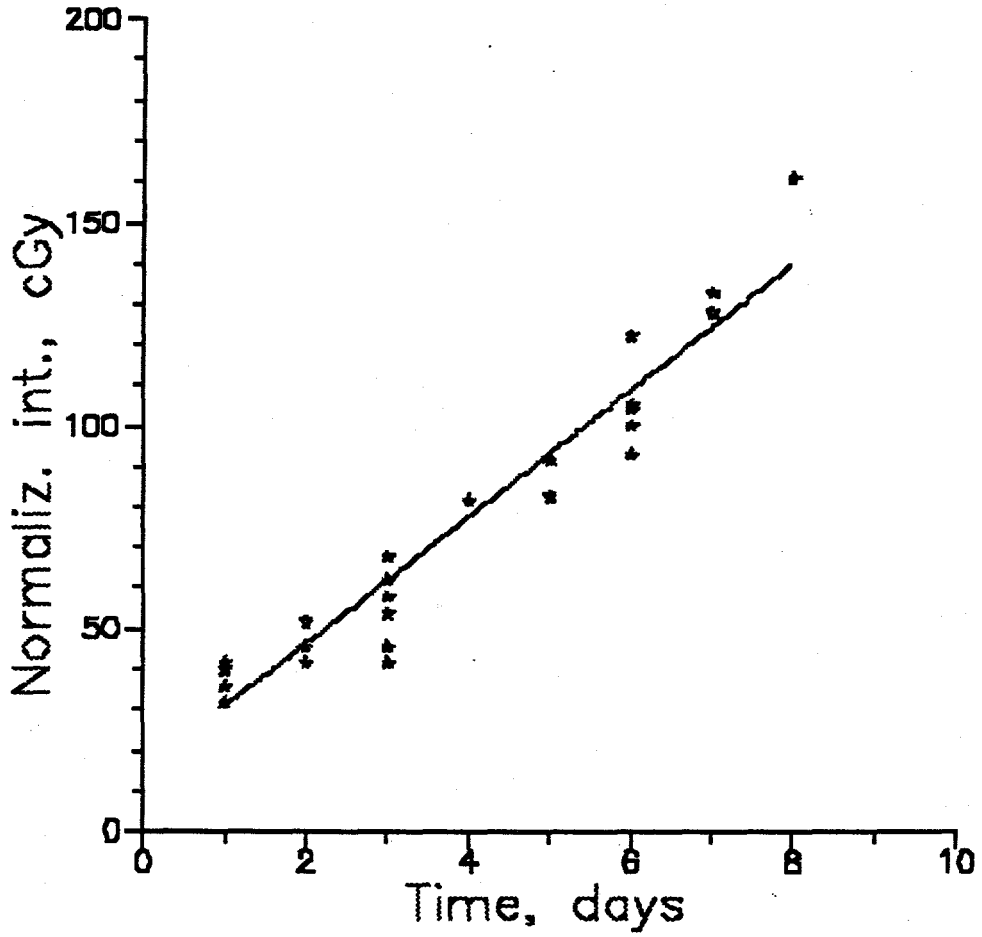

Fig. 4. Intensity of sunlight induced signal in enamel in dose equivalent units. (From Sholom, et al., 1996B).

Transient radiation induced signals.

The measurement process of EPR dosimetry involves application of a laboratory dose on top of the dose being determined. This process assumes that no significant differences in signal induction will result from the laboratory applied dose versus the dose received prior to extraction. Olduvai and Sales reported the presence of a transient signal which could affect results of EPR measurements on archaeological teeth. Additional work has shown that this effect can also produce errors at lower applied doses. Fig. __ shows the change in transient signals as a function of microwave power and Fig. __ shows change in sensitivity of the radiation induced signal that can occur

over the period of one month following irradiation. Fortunately a post-irradiation anneal for 1 hour at 190°C appears to minimize this effect.

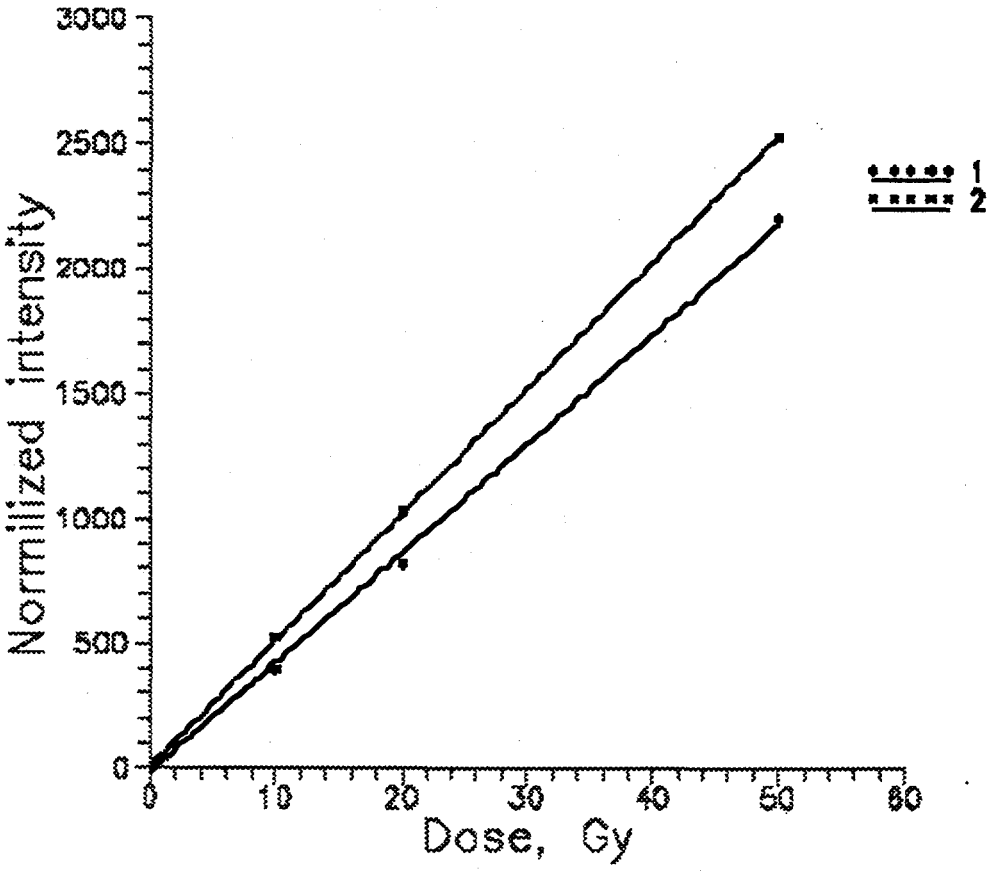

Fig. 3. Radiation sensitivity of enamel measured immediately after exposure (line 1) and one month after exposure (line 2). (From Sholom, et al., 1996B).

Sample preparation

The method which has currently been demonstrated to be most effective in removal or reduction of the native signal is that developed by Chumak et al. (1996B). This method involves separation of the enamel from the dentine using a water cooled saw, treatment of the enamel cap in acetic acid to soften the attached dentine; crushing of the sample followed by treatment of the grains in potassium hydroxide at 60 °C for twelve hours. Following this treatment the grains are scanned and the signal compared against a standard background spectrum. If there is not good correspondence of the native

portions of the two spectra the sample under preparation is again treated in potassium hydroxide. Subsequent scanning is again used as criteria for continuation of the treatment process, or continuation of the measurement method.

Differential microwave power method

The differential microwave power method relies on the fact that the native signal of enamel saturates at lower powers (approx 8mW) than does the radiation induced signal (>200mW). The goal of the differential power method is to select two microwave powers for analysis both at the saturation level of the native signal but one sufficiently higher than the other so that the radiation induced signal is larger in one than the other. By subtracting the spectrum taken at high microwave power from that taken at lower power, the native signal will in theory be subtracted equally and the radiation induced signal isolated. The powers chosen for the two measurements were 2 mW and 25 mW in the bilateral intercomparison (Haskell, et al., 1996B) described above and were determined from microwave power saturation curves for the samples. The presence of additional signals with different saturation characteristics, can greatly effect the accuracy of this method.

Curve fitting

The use of curve fitting methods for generation of native spectral signals and for computerized subtraction is gaining increased attention. Methods developed range from fitting of the native signal only to fitting of the native, radiation induced, light induced etc. Figure __ shows the deconvolution of the native signal and the radiation induced signal from a single spectrum. Further deconvolution involving isolation of additional signals ,to be discussed below, is shown in Figure __.

Reducing Signal Anisotropy

Signal anisotropy is a problem which increases with microwave power. Anisotropic effects are observed with sample impurities and in large grains or whole pieces of enamel. Because of the anisotropic effect of large grains of enamel, the use of whole teeth for retrospective dosimetry has to this point been difficult and insensitive (Ishii and Ikeya, 1990). Rotation of the sample during measurement however, (Haskell, et al., 1996A) can effectively average out anisotropic effects in enamel. Figure __ shows the effects of rotation on a sample measured at a microwave power of ___. The upper figure shows three spectra taken without sample rotation, while the lower spectra are of three spectra obtained with sample rotation. Perhaps the greatest benefit of rotation is the fact that measurements may be taken at high microwave powers of large grains of sample or even of whole teeth as is demonstrated in Figure __. Figure ___ shows the spectrum of a deciduous incisor. The tooth is measured at a microwave power of 25 mW and although no preparation was performed on this sample, the spectrum compares well with a background spectrum generated from a powdered sample. Figure __ shows the background spectrum subtracted from the whole tooth which had been given an additional dose of 100 mGy prior to spectral measurement. The dose response curve of a powdered portion of sample performed at low added doses is shown in Figure __. This dose response curve indicates sensitivity and the lower limits of detection which may be achieved with sample rotation.

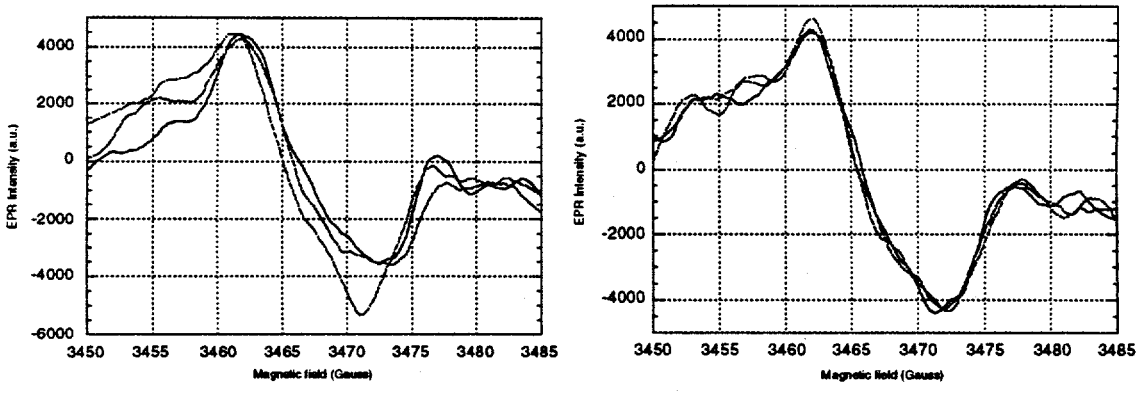

Figure . Powdered enamel sample shaken between spectra. Left, no rotation. Right, with rotation during measurement. Spectra are low bandpass filtered to show differences clearly. (From Haskell, et al., 1996A).

Reducing the Effects of Instrumental Fluctuations.

Instrumentation variations including fluctuation in gain as well as shift in magnetic field position are common problems with extended collection times. The problem can occur within a single collection run where spectra are collected and averaged over minutes or hours. Variations in a single spectrum or group of averaged spectra are not as critical as variations which occur in gain and field position between spectra. The latter is important because of the precision required for low dose measurements and the requirement that the background spectrum be subtracted. The use of reference spectra of standards such as manganese and zinc are useful for both field positioning as well as amplitude normalization. The method employed at both the Utah and Ukrainian laboratories involves an in cavity standard of manganese. The two lines of the manganese spectra at 1.981 and 2.034 g values are collected for every sample analyzed. During the process of spectrum subtraction the manganese lines are aligned with each other for both field width and amplitude. This method basically corrects for long term changes in gain and field width which occur between collection of a standard background and measurement of the spectrum with the radiation induced signals which may occur weeks, months or even years later. Unfortunately, this method does nothing to correct for short term shifts in gain or field during collection of a spectrum. Instrumentation which would be useful for correction of short term shifts is a field frequency log which would monitor the position of a standard and make frequency corrections dynamically during spectral collection. Another method for monitoring and correcting for machine variations is in use at the Bremen laboratory in Germany. This method has the advantage not only of correcting semi-dynamically for shifts in field position but also for maximizing the collection efficiency of the radiation induced signal. The method involves spectral collection of a chromium or zinc standard followed by rapid point to point collection of the field regions of the maximum and minimum of the radiation induced signals. Collections of over 2,000 points can be made over the period of one minute. By alternating measurements of the standard with peak to peak collections short term variations in field position may be corrected for by concentrating collection time only on those points of direct interest for

dosimetry. The collection time is optimized but at the expense of loss of resolution of spectral shape. Never the less, this method holds promise in cases where spectral shape has been verified using traditional sweeping methods and increased accuracy is desired for low dose measurements.

Acknowledgements -- Supported by the U.S. Department of Energy, Contract DE-FC08-89NV10805 and U.S. Department of Energy by Lawrence Livermore National Laboratory under contract no. W-7405-ENG-48.

REFERENCES

Chumak, V., Bailiff, I., Baran, N., Bougai, A., Dubovsky, S., Feedosov, I., Finin, V., Haskell, E., Hayes, R., Ivannikov, A., Kenner, G., Kirilov, V., Khlamidova, L., Kolesnik, S., Liidja, G., Likhtarev, I., Lippman, E., Maksimenko, V., Meijer, A., Minenko, V., Pasalskaya, L., Past, J., Puskar, J., Radchuk, V., Sholom, S., Skvortzov, V., Stepanenko, V., Vaher, U., Wieser, A. (1996) The first international intercomparison of EPR dosimetry with teeth: first results. Appl. Radiat. Isot. (in press).

Chumak, V., Pavlenko, J. and Sholom, S. (1996B) An approach to the assessment of overall uncertainty of determination of dose using ESR technique. Appl. Radiat. Isot. (in press).

Haskell, E.H., Hayes, R.B. and Kenner, G.H. (1996A) Improved accuracy of EPR dosimetry using a constant rotation goniometer. Radiation Measurements (submitted).

Haskell, E., Kenner, G., Hayes, R., Sholom, S. and Chumak, V. (1996B) An EPR intercomparison using teeth irradiated prior to crushing, Radiation Measurements (submitted for publication).

Ishii, H. and Ikeya, M. (1990) An electron spin resonance system for in-vivo human teeth dosimetry. Jap. J. Appl. Phys. 29:871-875.

Romanyukha A.A, Ignatiev E. A., Koshta A. A. and Wieser A. (1995) Selective saturation method of EPR Dosimetry with tooth enamel. Presented at the 4th International Symposium on ESR Dosimetry and Application, Munich, Germany May 15-19, 1995.

Serezhenkov V.A, Moroz I.A., Klevezal G.A. and Vanin A.F. (1995) Estimation of accumulated dose of radiation by the method of ESR-spectrometry of dental enamel of mammals. Presented at the 4th International Symposium on ESR Dosimetry and Application, Munich, Germany May 15-19,1995.

Sholom, S.V., Haskell, E.H., Hayes, R.B., Chumak, V.V., and Kenner, G.H. (1996A) Influence of crushing and additive irradiation procedures on EPR dosimetry of tooth enamel. Radiation Measurements (submitted).

Sholom, S.V., Haskell, E.H., Hayes, R.B., Chumak, V.V. and Kenner, G.H. (1996B) The properties of sunlight and UV light induced EPR signals in enamel. Radiation Measurements (submitted).