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Retrospective Dosimetry using EPR and TL Techniques: A Status Report

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Methods of retrospective dosimetry, including luminescence and electron paramagnetic resonance spectroscopy (EPR), rely on measurement of accident dose absorbed by naturally occurring materials - ceramics in the case of both thermoluminescence (TL) and optically stimulated luminescence (OSL) and organic materials and bio-minerals in the case of EPR. Each of these methods relies on measurement of radiation defects resulting from accidental exposure. Since defects also result from natural sources of radiation over the lifetime of a sample, analysis is usually restricted to materials for which the natural dose may be determined and subtracted from the measured cumulative dose.

The transient, or accident radiation dose 'D_X' is determined as follows

$$D_X = D_{TL} - (R_\alpha + R_\beta + R_\gamma + R_\chi)A$$

Where

D_{TL}	=	TL measurement of total accrued dose
A	=	Sample Age
R_α	=	alpha dose rate
R_β	=	beta dose rate
R_γ	=	gamma ray dose rate
R_χ	=	cosmic ray dose rate

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Luminescence dating techniques rely heavily on an accurate assessment of cumulative dose from natural radiation sources, and dating research has provided us with the bulk of our knowledge in this area [1]. Virtually all of the work on natural dose determination can be directly applied to retrospective techniques. With EPR techniques the cumulative dose from diagnostic x-rays is also of importance.

Electron Paramagnetic Resonance Techniques

EPR analysis of tooth enamel can provide measurements of cumulative dose to teeth. The attraction of enamel as an individual dosimeter lies in the ubiquity of the material and the fact that wisdom teeth are routinely removed and others are extracted as a result of age related periodontal disease.

Interlaboratory Comparisons of EPR Techniques

EPR dosimetry of tooth enamel is in a rapid stage of development. Two interlaboratory comparisons have produced recent validation of different aspects of the technique. The first, performed as part of ECP-10, was a blind study [2] involving 9 laboratories from the Former Soviet Union (FSU), Europe and an invited laboratory from the U.S. The results of this intercomparison showed marked variation in accuracy produced by individual laboratories, however one laboratory produced results which exceeded expectations (Fig. 1). The study was designed to assess the reliability of different methodologies under ideal conditions. The intercomparison involved the preparation of a homogeneous mixture of grains from unirradiated teeth which were irradiated after crushing. The project did not examine effects which may be associated with the preparation procedure or effects associated with irradiation of whole teeth versus crushed teeth. The results are currently being assessed by the individual members of the project, and the analytical procedures used by the most successful laboratory are being adopted in whole or in part.

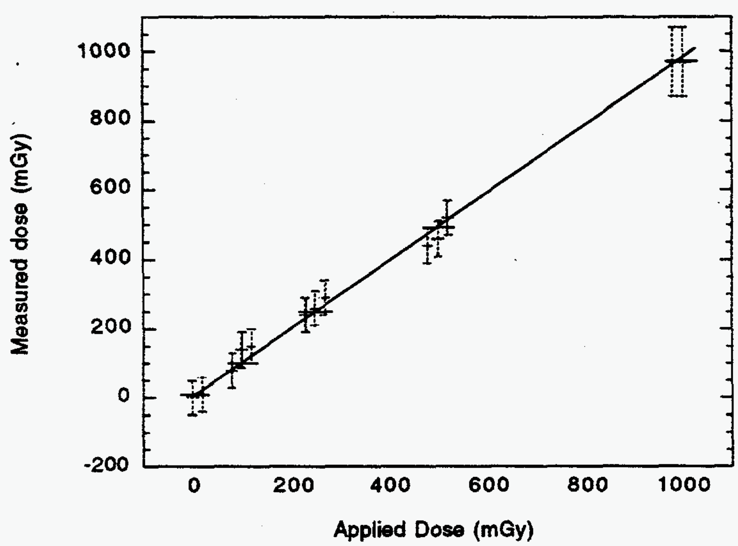

Fig. 1. Most accurate results of the ECP-10 intercomparison

A second, bilateral intercomparison [3], performed by the University of Utah and the Ukrainian Scientific Center for Radiation Medicine (USCRM), was designed as a pilot study for future multilaboratory comparisons. This study involved irradiation of teeth prior to crushing. The design of the blind study allowed assessment of applied dose as well as dose from prior x-rays. Results from both laboratories showed mean deviations of measured versus cumulative dose (including x-rays) of approximately 20% for doses on the order of 0.2 Gy.

DISCLAIMER

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APPENDIX 20.

Retrospective Dosimetry using EPR and TL Techniques: A Status Report. (Presented at the International Congress on Radiation Protection. Vienna, Austria April 15-19, 1996)

Problems associated with EPR analysis of enamel

EPR of enamel is not without its problems, and additional research is needed to clarify and minimize uncertainties. The presence of organic and mechanically induced signals [4,5,6] superimposed on the radiation sensitive signal has been recognized as a potential limiting factor in the ultimate accuracy of the technique. The need for a zero dose background spectrum which is subtracted from the spectrum of an irradiated tooth means that variations in the large organic signal will be translated into significant errors in the overall dose estimate. The approaches to this problem have been threefold. The first involves examination of each spectrum generated from an irradiated tooth and comparison with a well prepared zero dose background tooth [7]. Deviations in the organic signal are, with experience, recognized and eliminated by treatment in NaOH. Once organic signals match, the spectra may then be subtracted. The validity of this approach is evidenced by results (Fig. 1) from the ECP-10 intercomparison. A second approach involves modeling of the EPR signal and computer isolation of the organic and radiation sensitive components. Progress in identifying and isolating the organic component from the mechanical portion has recently been made with the use of organic and synthetically grown apatites [8]. The third approach involves variations on the measurement procedure itself and utilizes differences in signal saturation as a function of microwave power. The differential power method [9,10] allows determination of cumulative dose without the need for a zero dose background signal. Additional work is needed with methods two and three above, however the implementation of the first method offers an immediate reduction in uncertainties due to the background organic signal.

Another recently recognized problem associated with EPR dosimetry of teeth involves the effect of light exposure on the EPR signal of teeth [11]. This problem was identified by observing large variations in dose estimates obtained by analysis of incisors and canine teeth which may be exposed to sunlight. This effect poses a serious limitation in the ability to perform EPR dosimetry on canines and incisors, however it appears to have no effect on analysis of molars or wisdom teeth. It does reduce the total number of collected samples which can be analyzed with confidence, and in some cases it may mean a retrospective deletion of previously measured samples..

Yet another potential source of error of EPR dosimetry of enamel concerns the effect of pre and post crushing sensitivity of the radiation induced signal. Recent research [12,13] indicates that an overestimate of dose may be obtained if grain sizes smaller than 100 µm are used to measure dose delivered prior to crushing. This study, while not yet confirmed by other laboratories also indicates that use of grain sizes in the range of 250 to 600 µm does not produce this overestimate. This effect also requires further examination and explanation.

Dental x-rays can represent a significant source of uncertainty in dose estimation due to the difference in energy dependence of hydroxyapatite versus that of soft tissue. A seven to one difference in deposited dose in enamel versus that in soft tissue at typical x-ray energies of 60 KeV represents a threat to the utility of EPR as a biological dosimeter. Fortunately, two factors reduce this uncertainty. The first is the purported rarity of dental x-rays in the FSU, an assertion which needs additional documentation, and the second is the differential dose deposition in the outer and inner portions of teeth due to attenuation of the x-ray beam. This effect has been used previously to distinguish dose due to x-irradiation [14,15,16] and was used by USCRM to determine x-ray dose in the teeth used in the bilateral intercomparison mentioned above [26]. Because of the potential overestimation of dose due to dental x-rays, it may be necessary to routinely measure interior and exterior portions of teeth. Molars or wisdom teeth, because of their large size, may again be required for this multiple analysis.

Riding on each of these uncertainties is another potential limitation to the accuracy and ultimate lower limit of detection of EPR techniques. This source of uncertainty is the non-isotropic response associated with measurement of crystalline grains. Theoretically grains sizes sufficiently small will result in an isotropic EPR response which is independent of sample orientation. In practice, use of such small grain sizes may present a problem of sensitivity difference mentioned above. Additionally, the effect of anisotropy is enhanced with increasing microwave power. This places limits on the effectiveness of the two power method discussed previously. A recent addition to the EPR dosimetry process, a constant rotating goniometer [17], reduces the effect of anisotropy making measurements at all microwave powers, and particularly the higher powers, significantly more accurate. Since the organic component of the EPR signal seems to be most effected by anisotropy, this procedure may well result in reduced limits of detection. Fig. 2 shows the result of three spectra obtained following separate shakings with no rotation and three spectra obtained following separate shakings with rotation during spectral measurement. The spectra have been passed through a low bandpass filter to remove high frequency noise and more clearly show the improved reproducibility.

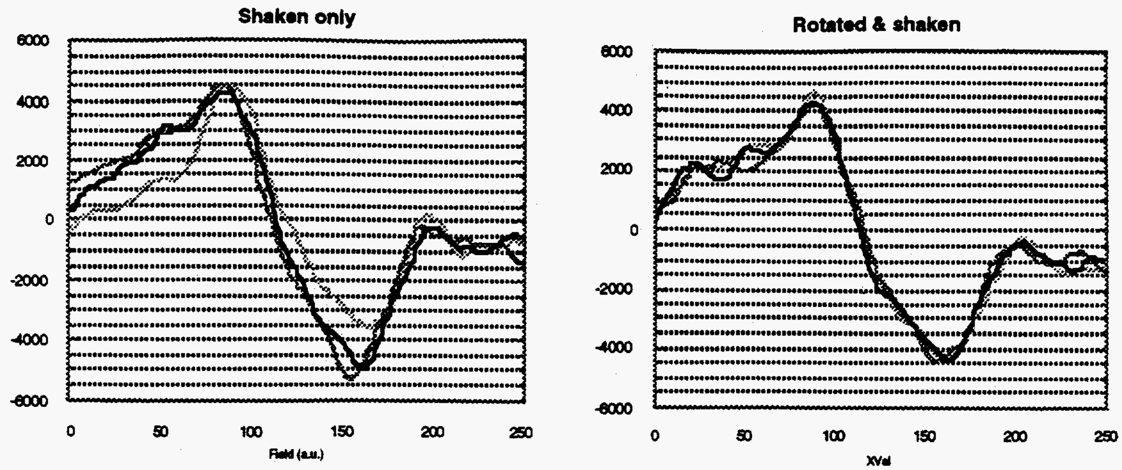

Figure 2. Effect of constant rotation goniometer on EPR reproducibility.

Many of the uncertainties addressed above are serious and can result in significant inaccuracies if not properly recognized and addressed. However, none of them seems insurmountable nor represents a fundamental limit to the utility or accuracy of the technique. Assessment of errors has received recent attention [18] and methods for time optimization of analysis are also being developed [19]. In fact, it seems highly likely that continued research will result in steady improvements in the speed, reliability, and lower limits of detection of teeth as an EPR dosimeter.

A related EPR technique involves measurement of deorganified dentine and comparison with measurements from enamel [20,21]. This technique has the potential of allowing differential measurements of dose due to external gamma rays as well as internal emitters such as Sr-90. An advantage to the use of dentine as a dosimeter is the fact that the organic signal present in enamel is eliminated in the deorganified dentine. Use of dentine also eliminates any possible effects of UV irradiation. At present the lower limit of detection of dentine dose does not reach that of enamel. However, this area of research is still in its infancy.

Luminescence Techniques

Luminescence dosimetry of environmental materials was first associated with accident dose reconstruction during the early 1960's. Thermoluminescence techniques were applied to the reconstruction of doses at Hiroshima and Nagasaki [22,23,24,25,26], the Nevada test site [27,28], and most recently, the regions downwind of the Chernobyl Nuclear power plant [29,30,31]. Several reviews of retrospective techniques used in these studies detail procedures, associated problems and experimental validations which have been conducted [32,33,34]. Perhaps the most extensive effort at technique validation prior to current efforts associated with the Chernobyl accident was the TL study of Hiroshima and Nagasaki conducted as a part of DS-86 [24]. Validations included tests of dose rate effects, neutron sensitivities, preparation effects and numerous intercomparisons and intercalibrations. A partially blind, multilaboratory study involving NBS-irradiated annealed quartz removed from a Nagasaki brick sample produced measurements of dose which agreed with applied values to within better than ±10% (1s) at each of 3 dose levels ranging form 82 mGy to 417 mGy . Comparison of identical tile samples, when measured by both the high temperature and pre-dose TL techniques, produced agreement of ± 10% [35]. In an intercomparison involving different tiles from the same sampling sites, the laboratories also produced dose estimates agreeing to within ± 10%. The TL results showed close agreement with theoretical calculations in the city of Nagasaki. Interlaboratory agreement was again very close at a site in Hiroshima examined by the laboratories, however the overall measurements in Hiroshima at a distance from the hypocenter of approximately 1.4 km was approximately 20% higher than theoretical calculations. Further measurements using both the pre-dose and high temperature techniques at even greater distances has tended to verify these results [36,37,38]. The reason for the discrepancy between the measured and calculated gamma ray dose values in Hiroshima remains unclear, as does an even greater discrepancy between theoretical and measured neutron fluences in that city [39,40,41].

Concurrent with the Hiroshima/Nagasaki effort was an evaluation of doses delivered to regions downwind of the Nevada Test Site [27,28]. This effort evaluated transient doses of less than 100 mGy and relied exclusively on the use of the predose TL technique. Because the doses of interest were low compared to natural dose accumulation, rigorous diagnostic tests [27,28,42,43] were developed and applied. The study allowed comparison of measured doses in building bricks versus evaluated exposure estimates based on soil sampling measurements, reevaluation of original monitors reports and a comprehensive review project.

The Chernobyl nuclear accident has lead to several recent applications [29,30,31,34] of TL techniques. Again collaborative efforts were employed allowing intercomparisons of separate techniques as well as similar techniques applied by different laboratories.

The multi-national effort sponsored by the European Community (ECP-10) in collaboration with the Former Soviet Union, continues to examine the feasibility of retrospective techniques applied to the Chernobyl accident. The program has contributed to the validation of more recently developed techniques, has incorporated measurement into Monte Carlo models at actual exposed locations has investigated correlation of measured dose depth profiles in ceramics with theoretical calculations and has mounted a series of studies designed to insure interlaboratoy reliability [44].

Perhaps the biggest question currently facing luminescence dosimetry efforts involves optimization of methods for integrating results into modeling efforts. This problem is being addressed by the ECP-10 effort and requires close cooperation of modelers and measurers at the earliest stages of reconstruction.

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