DOE/NV/10805--T4

Dosimetry using Environmental
and Biological Materials.

Final Report February, 1998

DOE Contract DE-FC08-89NV10805

E. Haskell, G. Kenner, R. Hayes

Center for Applied Dosimetry
University of Utah
729 Arapen Drive #205
Salt Lake City UT 84108-1218

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED
ph

MASTER

• **Dosimetry using Environmental and Biological Materials.**
Radiobiology Division, University of Utah

E. Haskell, G. Kenner, R. Hayes

Introduction

This report details a five year effort to improve the sensitivity and reliability of retrospective dosimetry methods, to collaborate with laboratories engaged in related research and to share the technology with startup laboratories seeking similar capabilities. Because of our experience in the development and application of dosimetry techniques for accident situations, we have been in a unique position to contribute to the development, coordination and implementation of quality assurance programs in laboratories currently, or soon to be, engaged in the measurement and dissemination of dosimetry information.

• **Accomplishments**

Our research program has focused on validation of EPR as a dosimetry tool and on optimization of the technique by reducing the lower limits of detection, simplifying the process of sample preparation and analysis and speeding analysis to allow greater throughput in routine measurement situations. This research has resulted in a total of 27 manuscripts which have been published, are in press, or have been submitted for publication. Of these manuscripts, the contents of 20 of them were reported on and manuscript copies were submitted in a report to DOE in September, 1996 also entitled "Dosimetry using Environmental and Biological Materials." The interested reader is referred to this report for the manuscripts which are not included here.

Synthetic Hydroxyapatite

• We have investigated the dosimetric signal of hard tissues in enamel, deorganified dentin, synthetic carbonated apatites and synthetic hydroxyapatite. We found that the dosimetric signal magnitude / carbonate content ratio maximizes at about 2.5% carbonate which is comparable to the concentration found in enamel (3%) but less than that found in bone. Further, the number of radicals responsible for the dosimetric signal diminishes by 80% following deorganification but the precursor ions are unaffected or increase in number (**Appendix 1**).

Collaborative studies with investigators from the Ukraine elucidated some of the changes seen when enamel is heated. Specifically, it was shown that when the isotropic signal attributed to CO₂(-) in the organic matrix is reduced by heating, there is a corresponding but smaller increase in the magnitude of the dosimetric signal at about 200° C (**Appendices 2 and 3**).

Light Effects

Factors influencing the accuracy of dosimetry with enamel have also been investigated. We observed the growth of a large signal in enamel near $g = 2.0018$, upon exposure to UV irradiation as well as sunlight (**Appendix 2**, 1996 report). We later, more systematically investigated the phenomenon (**Appendix 4**). and discovered that 1) UV increases the radiation sensitive component of enamel, 2) UV increases the native signal of enamel and 3) UV induces additional signals that might prove useful in assessing and making corrections for the total UV dose received. A dose depth profile indicated that the penetration depth of the effect is approximately 100 μm .

Grain size effects

On a visit to our laboratory, Dr. V. Polyakov of Tallinn, Estonia observed a phenomenon which indicated that the size of the enamel grain being used for analysis was a factor in the sensitivity of the EPR signal to radiation (**Appendix 4**, 1996 report). We subsequently investigated the effect further and discovered that the effect was not only grain size dependent, but also was a function of whether the sample was irradiated before or after the tooth had been reduced to a powder for analysis. Samples which had been irradiated after crushing showed a different sensitivity to radiation than those irradiated prior to crushing. The grain size range of 250 to 600 μm was shown to minimize the effect (**Appendix 5**, 1996 report). We again addressed this effect during the visit by Dr. Sholom from the Ukraine. In this case we treated all grains thoroughly in concentrated NaOH. The effect was still observed, however the magnitude of the sensitivity change was drastically reduced. The maximum change was seen to occur for grains less than 75 μm and the effect was approximately 10%.

Transient radiation induced signals

Transient radiation induced signals reported in archaeological specimens of teeth prompted us to investigate these effects in modern enamel (**Appendix 5**). We discovered that the effect was significant on the $g = 2.0018$ signal normally used for dosimetry, that the signal reduced to near

background levels after approximately 1 month, and that the transient signals could be removed by heating to 90 °C for 2 hours.

Dosimetry using dentin

Our observation that removal of the organic component of bone could be achieved with the use of Soxhlett treatment in diethylenetriamine, with reduction of the normally obscuring native signal by several orders of magnitude (**Appendix 7**, 1996 report) led us to propose the use of dentin in conjunction with enamel for dosimetry of internal emitters (**Appendix 8**, 1996 report). We have also proposed (**Appendix 9**, 1996 report) a more extensive investigation of the utility of dentin as a dosimeter of internal emitters and have suggested that comparison of enamel and dentin of teeth exposed to injected or inhaled radionuclides as part of completed, long range beagle studies could be used for correlation with known organ doses for both lifetime and serial sacrifice data. We have collected several hundred teeth from beagles from the Utah studies for use in such a project.

Goniometry

The variation in EPR signal intensity and position resulting from anisotropies in enamel have been virtually eliminated with our use of a constant rotation goniometer during sample measurement (**Appendix 10**, 1996 report). This procedure has resulted in our being able to use large portions of enamel and even whole deciduous teeth (**Appendix 6**). The dose response characteristics indicate that doses of less than 100 mGy can now be measured. The ability to use whole deciduous teeth opens the possibility for the use of EPR as a screening technique of milk teeth in the event of a future nuclear accident on the scale of that at Chernobyl (**Appendix 12**, 1996 report).

Optimization of measurement protocol

Because of the time consuming nature of the additive dose method of EPR analysis, where a sample is repeatedly measured, dosed and remeasured, we have developed a mathematical optimization method which specifies the size of the doses to be applied and the number required to achieve a desired level of precision. (**Appendix 13**, 1996 report). This procedure should greatly reduce the time required for routine EPR analysis. The widespread utility of the method for systems showing a linear response prompted us to prepare a condensed version for general application for a mathematics journal (**Appendix 14**, 1996 report).

Other EPR dosimeters

In an effort to expand the utility of EPR dosimetry we investigated the dosimetric properties of a number of common materials. The most promising of those is common sheetrock which can be used for dosimetry over the course of approximately 6 months following irradiation (**Appendix 15**, 1996 report). A short letter was published in Health Physics to quickly get this information to the radiation protection community (**Appendix 16**, 1996 report).

Curve fitting.

We have developed computerized routines which allow us to simulate the native signal of enamel as well as the radiation induced signal. This approach is promising for future dosimetry and we have submitted a paper on the suitability of the Marquardt-Levenberg method for analyzing TL/OSL/EPR dose response data (**Appendix 10**).

Blind EPR Intercomparisons.

We have participated in two blind EPR intercomparisons. The first was organized by ECP-10, the dosimetry research program of the European Union. This intercomparison involved 9 laboratories which were given a homogeneous mixture of enamel from over 40 teeth. The grains were irradiated after crushing and the test did not address some of the effects we have described above. The results were as follows:

- 1000 mGy samples: $\pm 25\%$ for 6 labs
- 500 mGy samples: $\pm 25\%$ 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 $\pm 10\%$ for analysis of the 100 mGy samples. The report of the Utah groups findings in the process of participating in this effort are summarized in **Appendix 17** (1996 report).

The second was a bilateral intercomparison between our laboratory and that of V. Chumak and S. Sholom in the Ukraine. This intercomparison involved measurement of teeth which were irradiated prior to crushing. The results (**Appendix 18**, 1996 report) showed that at dose levels of approximately 250 mGy, doses could be measured with an accuracy of

- approximately 25%. Methods were used by both laboratories to quantify the dose from dental x-rays that the teeth may have received.

An additional intercomparison is planned which will examine effects associated exclusively with different methods of sample preparation and analysis (**Appendix 19**, 1996 report).

Dental x-rays

- Our analysis of teeth removed at a local dental clinic indicates that x-ray doses may not be as high as we had previously feared (**Appendix 18**, 1996 report). The use of a lead sheet behind the x-ray film as well as the localized exposures makes it unlikely that doses in excess of 10 mGy will be delivered to a given tooth during a routine series of x-rays. Since adult teeth are currently used for analyses, x-rays given prior to ingrowth of secondary teeth will not be a consideration. This coupled with the fact that dental visits tend to decline significantly in adulthood means that lifetime
- doses to adult teeth should rarely reach 50 mGy.

Problems identified with EPR analyses

The transient radiation induced signals, light induced signals, and the grain size effect are all potential sources of error in EPR analysis. Fortunately, with a knowledge of these effects, errors may be avoided by appropriate preanneal in the case of the transient signals, using only molars or wisdom teeth in the case of the light induced signals, and treating the sample with NaOH prior to analysis, or using whole teeth or large portions of teeth in the case of the grain size effect.

Dosimetry of marine animals

- An EPR dosimetry study of walrus cementum was done to test their suitability as biomarkers (**Appendix 7**). Walrus teeth are attractive because their cementum is laid down in annual rings over an approximately 20 year period. This research was funded largely through contract B29174 W-7405-ENG-48 with Lawrence Livermore National Laboratory.

Optimization of measurement protocol

We continued work in this area by publishing two detailed papers on the methods we use in our laboratory to measure the EPR properties of hard tissues (Appendices 8 and 9).

Alanine dosimetry

With slight modification, it was found to be possible to use the techniques developed for quantitating radiation in hard tissues, for performing similar measurements in alanine (**Appendix 11**).

Distribution of Information

The results of our studies and the current status of retrospective dosimetry using EPR and/or TL have been reported to the radiation measurement community through invited talks at IRPA96 (**Appendix 20**, 1996 report), the 2nd Hiroshima Semipalatinsk Workshop (**Appendix 12**), the International Workshop on Radiation Exposures by Nuclear Facilities (**Appendix 21**, 1996 report), (**Appendix 22**, 1996 report) and the Space Radiation Damage and Biodosimetry Workshop (**Appendix 13**). A collaborative paper summarizing chromosomal as well as solid state methods has been submitted to Radiation Research (**Appendix 14**).

Luminescence Research

Alpha Dose Measurements in Ceramic Toilet Tanks

In an effort to address the need for retrospective dosimetry of alpha and beta emitters in previously contaminated water supplies we have investigated methods for measuring doses deposited on the inner surface of toilet tanks using luminescence techniques. Because the range of alphas and low energy betas may be only micrometers, traditional thin section methods for obtaining dose versus depth profiles are not feasible. We instead developed a method which involves removal of the outer portion of thin sections by surface polishing. Measurements can then be made on a number of sections from which successively deeper layers have been removed. We have thus been able to generate a dose versus depth profile from artificially irradiated porcelain tanks which corresponds to the absorption profile calculated from measurement of thin sections removed from the exposed surface. Development of the technique was the subject of a Master's thesis, a draft of which is included in **Appendix 25** (1996 report).

Automation of TL Imaging and Analysis Software

Many luminescence laboratories throughout Europe involved in joint dosimetry efforts with the Former Soviet Union use Riso TL readers manufactured in Denmark. Our laboratory uses custom manufactured TL readers made through the joint effort of Daybreak Nuclear and Medical Systems and the University of Utah. Over the past 10 years we have developed an extensive suite of software tools to control our data collection equipment and to perform a variety of types of analysis for retrospective measurements. As a part of this study we have modified our software allowing control of laser input and control of our TL and OSL imaging systems. At the suggestion of researchers in laboratories which use the Riso equipment we have adapted the software for compatibility with the Riso system. This will greatly simplify the training procedures for those laboratories in the FSU which will obtain Riso luminescence system. The software capabilities are described in **Appendix 26**.

Scientific training and exchange

Our experience in the development and application of dosimetry techniques for accident situations has put us in a unique position to contribute to the development, coordination and implementation of quality assurance programs in laboratories currently, or soon to be, engaged in the measurement and dissemination of dosimetry information. During the course of the contract we have hosted visiting scientists from three regions of the former Soviet Union. Dr. Alexi Kondrashov from Obninsk visited the laboratory for a period of 2 weeks for preliminary training in TL techniques. Drs. S. Sholom and V. Chumak visited for two weeks to complete a blind bilateral intercomparison and Dr. Sholom visited again for a period of two months in the spring of 1996 for an extensive period of basic EPR research. We have hosted Mr. Nickolai Bougrov from the Urals for training in TL techniques on two occasions for periods of 2 months each.

Our luminescence software allows remote data collection and analysis over the internet. We are pilot testing this software by providing Nick Bougrov remote software for use in his institute in Russia. Samples left in our laboratory will be run remotely and his training in TL techniques can continue. Since many of the startup laboratories which hope to be engaged in TL measurements are not yet functional, this method may provide not only inexpensive training under our supervision, but also the analysis and characterization of field samples long before they would otherwise have been examined.

Collaboration with Dosimetry Efforts of the European Community.

In addition to ongoing collaborative efforts with individual laboratories in Europe and the former Soviet Union, we have accepted an invitation to participate in a collaborative dosimetry project under Framework IV (FIV) of the European Union (EU). The project involves laboratories throughout

- Western Europe and includes laboratories in the Former Soviet Union (FSU) under a complementary program known as Inco Copernicus. The purpose of these programs is to assess the feasibility of solid state and biological methods for retrospective dosimetry as well as modeling efforts which may be applied to accident sites in the FSU. The projects are built on two previous programs with which we have been involved, most recently the EU program ECP-10, and an IAEA Cooperative Research Program originally established through our initiative.

Our involvement with the FIV program provides the opportunity of full participation and significant input into the design and implementation of the research program. Funding is severely limited for all participants of the program, however funds to all workshops and contractors' meetings are being provided for our participation.

- The collaboration of our laboratory with others throughout Europe and the FSU involved in retrospective dosimetry provides an opportunity to leverage limited research funds and to pool the scientific expertise and analytical capabilities of established laboratories with those which are just
- entering the field. The inclusion of startup laboratories into the collaborative effort insures that the quality of research and routine dosimetric measurements which they will ultimately produce will conform to protocols and guidelines to be established through the FIV and Copernicus efforts. This quality assurance is also in the interests of ongoing bilateral epidemiological studies between the U.S. and FSU.

Our participation with the European Community dosimetry effort has involved close collaboration on both TL and EPR projects. We participated in a field trip from Moscow to Kiev in 1995 involving sample collection and training of FSU scientists in methods of sample collection and documentation. We are involved in ongoing intercomparisons with laboratories at Obninsk as well as laboratories in Kiev and the Urals.

Collaboration with Ukrainian Scientific Center for Radiation Medicine (USCRM).

We have worked very closely with Drs. Chumak and Sholom of the USCRM over the past several years. Both investigators visited our laboratory for research purposes, Drs. Chumak and Sholom in September of 1995 for a two week period, and Dr. Sholom again in April of this year for a period of two months. In December of 1995, Robert Hayes of our laboratory visited the USCRM for training in the sample preparation and analysis techniques in use at their facility. The collaboration has resulted in a number of publications involving potential problems and solutions to environmental and preparation effects associated with EPR dosimetry of enamel. It has also included a bilateral, blind intercomparison of artificially dosed teeth, as well as joint analyses of matching teeth from Chernobyl liquidators. The research has identified areas requiring further research to assess the potential impact on EPR dosimetry and we propose to continue the collaboration in the directions which have been identified. These areas include further investigation into the effect of solar radiation on tooth enamel, the effect of sample rotation on reducing lower limits of detection, the influence of transient irradiation effects on dose estimates, and the influence of annealing procedures on organic portions of enamel.

The USCRM is also initiating TL measurements on ceramics and porcelain insulators taken from the contaminated areas as well as the power plant itself. The USCRM has little previous experience with TL analysis and we feel that it is important to provide training and oversight for these measurements. We propose bilateral training visits between our two laboratories and replicate measurements on actual field samples as well as intercomparisons on blind irradiated samples in a manner similar to that performed on irradiated enamel.

Collaboration with the Urals Research Center for Radiation Medicine (URCRM)

Nickolai Bougrov of the URCRM is presently visiting our laboratory for a period of two months for training in the techniques of TL preparation and analysis. His laboratory in the Urals is not yet established, however once equipped it will be involved in routine measurement of ceramic and porcelain samples removed from the contaminated regions of the Urals. We anticipate close cooperation during the training phase and during the establishment of the analytical capabilities of his laboratory.

Collaboration with the Urals Institute of Metal Physics.

Dr. Alex Romanyuhka has spent considerable time in the EPR laboratory of Dr. Albrecht Wieser at GSF in Germany and is developing considerable expertise in EPR analysis of enamel. We have collaborated with Dr. Romanyuhka and Dr. Wieser on development of methodology for EPR analysis of internal emitters using differential measurements of dentin in addition to enamel, and we anticipate continued collaboration on methodology and analysis of enamel samples collected from individuals in the exposed regions of the Urals.

Collaboration with the Medical Radiological Research Center (MRRC), Obninsk, Russia

Dr. Alexi Kondrashov from the MRRC visited our laboratory for training in TL techniques and for analysis of samples of electrical insulator brought from regions of Russia contaminated by Chernobyl fallout. In August of 1995 EH visited the MRRC and participated in the EU program of field work in Russia, Belarus and the Ukraine involving training in sample collection, background dosimetry and documentation of ceramic samples from contaminated regions.

*Removed for separate processing

Table of Appendices

1.

EPR Properties of Synthetic Apatites, Deorganified Dentine, and Enamel. (*Calcif. Tiss. Int.* in press).

Reprint *

2.

Heating Related Changes in the Characteristics of Paramagnetic Centers in Tooth Enamel using EPR Techniques. (*Mineralogical Journal*, 19:3-12; 1997).

Reprint *

3.

Alignment of CO₂- Radicals of Tooth Enamel with Heating. (*J. Mineralized Tissue Research*, submitted).

Reprint *

4.

Properties of Light Induced EPR Signals in Enamel and their Possible Interference with Gamma-Induced Signals. (*Radiation Measurements*, in press).

Reprint *

5.

Influence of Crushing and Additive Irradiation Procedures on EPR Dosimetry of Tooth Enamel. (*Radiation Measurements*, in press).

Reprint *

6.

EPR Dosimetry of a Whole Deciduous Tooth using a Constant Rotation Goniometer and Background Subtraction with Dentin and Enamel Standards. (*Health Physics*, submitted).

Reprint *

7.

An Assessment of the Levenberg-Marquardt Fitting Algorithm on Saturating Exponential Data Sets. (Ancient TL, submitted)

Reprint *

8.

EPR Dosimetry of Pacific Walrus (*Odobenus rosmarus divergens*) Teeth. (*Rad. Prot. Dosim.*, submitted).

Reprint *

9.

Preprint* Technique for Increasing Reproducibility in EPR Dosimetry of Tooth Enamel. (*Rad. Prot. Dosim.*, submitted).

10.

Preprint* Achievable Resolution and Accuracy in EPR Dosimetry of Tooth Enamel. (*Rad. Prot. Dosim.*, submitted).7

~~An Assessment of the Levenberg-Marquardt Fitting Algorithm on Saturating Exponential Data Sets. (Ancient TL, submitted).~~

11.

Preprint* A High Sensitivity EPR Technique for Alanine Dosimetry. (*Rad. Prot. Dosim.*, submitted).

12.

Conference*
paper
Reprint EPR Dosimetry of Teeth in Past and Future Accidents: A Prospective Look at a Retrospective Method. (*Proceedings of the Second Hiroshima International Symposium. Hiroshima University : Hiroshima, Japan, 1996*).

13.

Reprint* Electron Paramagnetic Resonance Techniques and Space Biodosimetry. (*Radiation Research*, 148:S51-S59, 1997).

14.

Reprint* Emerging Technological Bases for Retrospective Dosimetry. (*Stem Cells*, 15:183-193, 1997).

* Removed for separate processing

APPENDIX 1.

EPR Properties of Synthetic Apatites, Deorganified Dentine, and Enamel. (*Calcif. Tiss. Int.* in press).

Reprint - Removed for
separate processing

APPENDIX 2

Heating Related Changes in the Characteristics of Paramagnetic Centers in Tooth Enamel using EPR Techniques. (*Mineralogical Journal*, 19:3-12; 1997).

Reprint - removed for
separate processing

APPENDIX 3

Alignment of CO₂- Radicals of Tooth Enamel with Heating. (*J. Mineralized Tissue Research*, submitted).

Preprint- Removed for
separate processing

APPENDIX 4

Properties of Light Induced EPR Signals in Enamel and
their Possible Interference with Gamma-Induced Signals.
(*Radiation Measurements*, in press).

Reprint — removed for
separate processing

APPENDIX 5

Influence of Crushing and Additive Irradiation Procedures
on EPR Dosimetry of Tooth Enamel. (*Radiation
Measurements*, in press).

Reprint - removed for
separate processing

APPENDIX 6

EPR Dosimetry of a Whole Deciduous Tooth using a Constant Rotation Goniometer and Background Subtraction with Dentin and Enamel Standards. (*Health Physics*, submitted).

Preprint - removed for
separate processing

APPENDIX 7

An Assessment of the Levenberg-Marquardt Fitting Algorithm on Saturating Exponential Data Sets. (*Ancient TL*, submitted).

Preprint - Removed for
separate processing

APPENDIX 8

EPR Dosimetry of Pacific Walrus (*Odobenus rosmarus divergens*) Teeth. (*Rad. Prot. Dosim.*, submitted).

Preprint- Removed for
Separate processing

APPENDIX 9

- Technique for Increasing Reproducibility in EPR Dosimetry of Tooth Enamel. (*Rad. Prot. Dosim.*, submitted).

Preprint - Removed for
separate processing

APPENDIX 10

Achievable Resolution and Accuracy in EPR Dosimetry of
Tooth Enamel. (*Rad. Prot. Dosim.*, submitted).

Preprint - Removed for
separate processing

APPENDIX 11

A High Sensitivity EPR Technique for Alanine Dosimetry.
(*Rad. Prot. Dosim.*, submitted).

Preprint - removed for
separate processing

APPENDIX 12

EPR Dosimetry of Teeth in Past and Future Accidents: A Prospective Look at a Retrospective Method.
(*Proceedings of the Second Hiroshima International Symposium. Hiroshima University : Hiroshima, Japan, 1996*).

Reprint —
Conference paper - Removed for
separate processing

APPENDIX 13

Electron Paramagnetic Resonance Techniques and Space
Biodosimetry. (*Radiation Research*, 148:S51-S59, 1997).

Reprint - removed for
separate processing

APPENDIX 14

Emerging Technological Bases for Retrospective Dosimetry. (*Stem Cells*, 15:183-193, 1997).

Reprint - Removed for
separate processing