Touch of Grey

A 20-Year Retrospective on Radiation Dosimetry

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Harshaw TLD Users' Group Symposium
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Oh well, a Touch Of Grey
Kind of suits you anyway.
That was all I had to say;
It's all right.

- “Touch of Grey” by
  Robert Hunter & Jerry Garcia

Even a blind hog finds an acorn every once in awhile.

- Roscoe Hall

ACORN 1

The Impact of DOE's Laboratory Accreditation Program and Harshaw's Development of the Model 8800 TL System on Personnel Radiation Dosimetry

Personnel Dosimetry at DOE/OR pre-DOELAP

The 1980s Oak Ridge DOE Complex

- Three major facility types
  - Gaseous Diffusion Plants
    - Paducah KY (U_{nat} to ~1% ^{235}U)
    - Oak Ridge K-25 Plant (1% to 5% ^{235}U)
    - Portsmouth OH (5% to 97% ^{235}U)
  - Oak Ridge National Laboratory (multi-program energy R&D laboratory)
  - Oak Ridge Y-12 Plant (nuclear weapons fabrication)
- One Operating Contractor
  - Union Carbide until 1985
  - Martin Marietta/Lockheed-Martin 1985-1999

Gaseous Diffusion Plants External Radiation Hazards

- Uranium & daughter emissions – pretty significant in drained cylinders & pigtails
  - Hard β^- from ^{234m}Pa (E_{max}= 2.29 MeV)
  - Soft but plentiful β^- from ^{99}Tc in reactor returns
  - X & γ from both U & daughters
- Some neutron emissions (0.2 – 4 mrem/h) in UF_6 from $^{19}F(",n)^{22}Na$
- Industrial radiography sources
- Potential for accidental nuclear criticality

Oak Ridge Gaseous Diffusion Plant

Y-12 Weapons Components Fabrication External Radiation Hazards

- Uranium emissions (tons of U metal)
  - Low-Energy X, γ
  - Energetic β^- from ^{238}U daughters (^{234m}Pa)
- Industrial radiography sources
- Analytical neutron generators
- X-ray diffraction units
- Potential for accidental nuclear criticality (had one in 1957!)
- I can tell you, but then I'll have to shoot you. (Special projects)

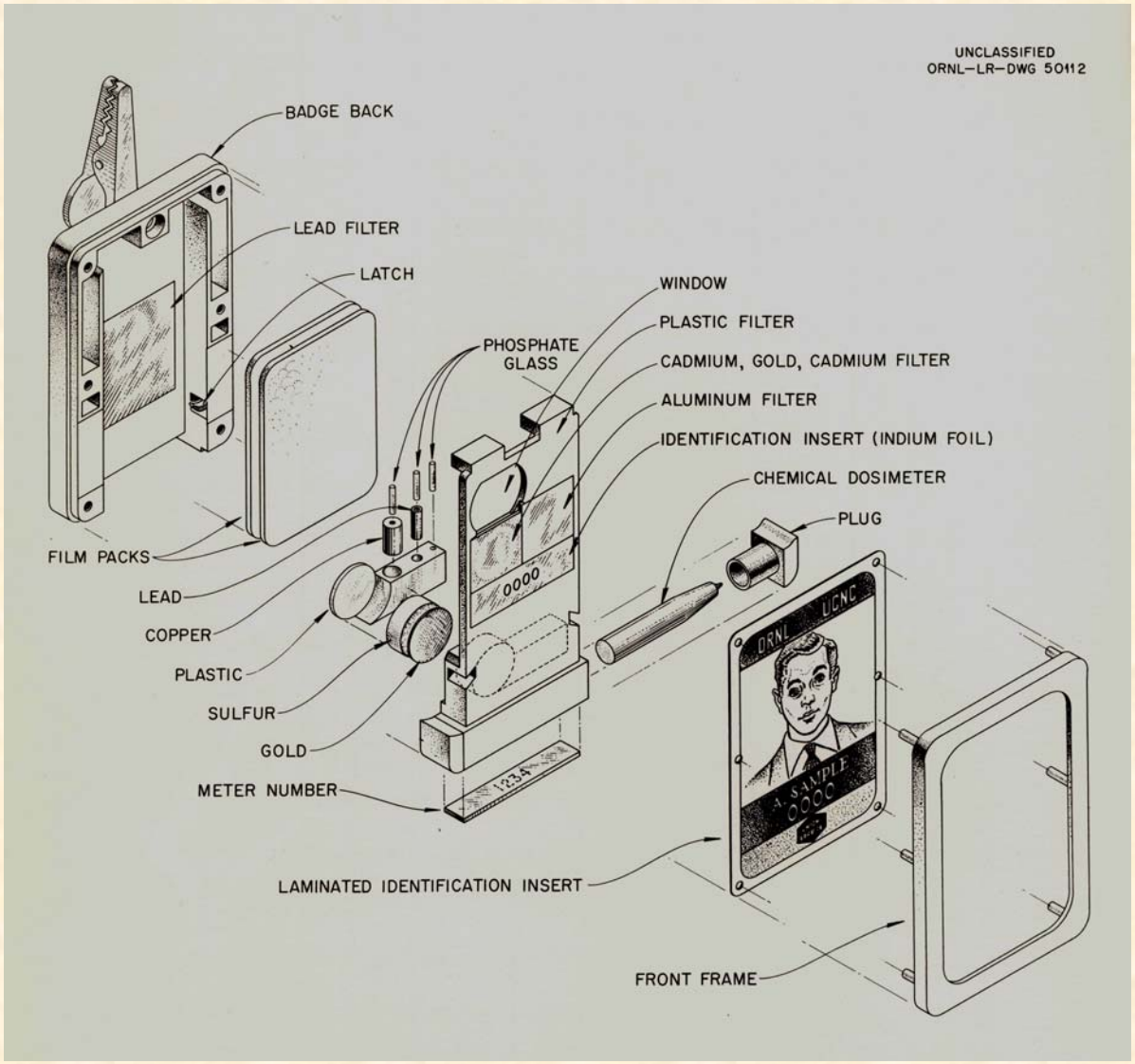
Oak Ridge Y-12 Plant

ORNL External Radiation Hazards

- Every radioactive isotope on the periodic chart, including *Euphorium*
  - some in large quantities (^{60}Co, ^{137}Cs, $^{232/233}U$)
- Nuclear reactors, many prototypes
- Particle accelerators up to 25 MV
- All kinds of analytical devices, like X-ray diffractometers & neutron generators
- Potential for accidental nuclear criticality
- Guys whose socks don't match

Oak Ridge National Laboratory

Pre-DOELAP Dosimetry in Oak Ridge

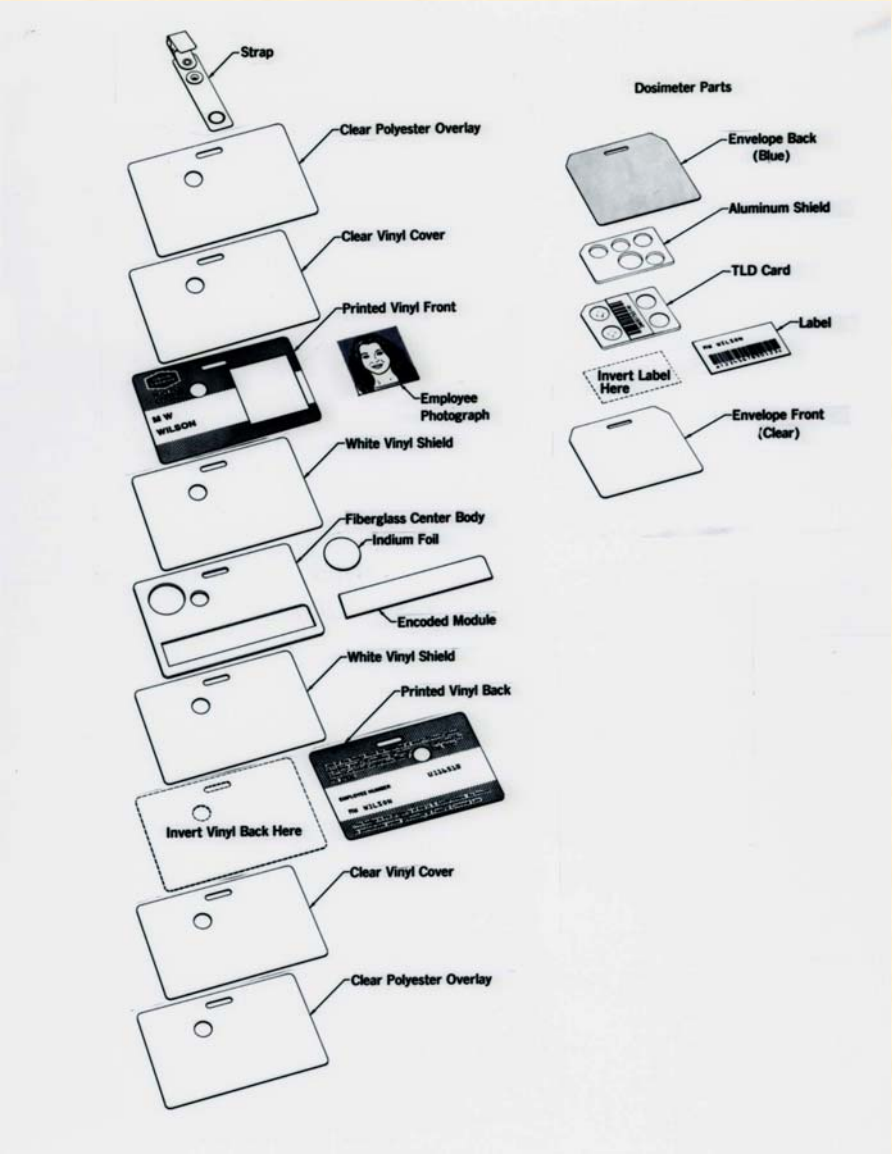


The ORNL badge-meter model II used from 1960-1980 at the 5 DOE-OR sites

WT Thornton, EM Davis & ED Gupton, *The ORNL Badge Dosimeter and Its Personnel Monitoring Applications*, **ORNL-3126**, 59 pp (1961).

One of the issues leading to the recall of this dosimeter was the problem of accounting for the gold.
- (CD Berger personal communication)

Note the intimate association of the dosimeter and employee ID – a recurring management issue.



UCC-ND employee meter badge used from 1980-1989 at the 5 DOE-OR sites

JD McLendon, ed., *External Radiation Monitoring of Union Carbide Corporation Nuclear Division Personnel*, **Y/DD-268**, 26 pp (1980).

The dosimeter components were assembled inside a little sealed plastic pouch that hung behind the security badge (which served as a filter for rudimentary energy discrimination).

Note that only 2 TL elements were used. The one for beta and soft X flopped around behind a hole in the security badge.

An additional dosimeter was assigned at ORNL to employees with neutron exposure potential.

Harshaw model 2276 automated reading system used with the UCC-ND employee meter badge TL card.

Logic Controller, picoammeter, reading module (from **Y/DD-268)**

Harshaw model 2276T transport system

(from **Y/DD-268**)

Development of the Harshaw Model 8800 TL System

- **1986 – DOELAP was coming**

  <u>from a December, 1985, memo, RS Lay to DC Parzyck</u>
  "The present two-unit system is inherently limited in its ability to differentiate between radiation energies, and it does not provide for neutron monitoring that is required at ORNL.

  … the present dosimeter is satisfactory for high-energy gammas [Co-60] (at low dose rates) and uranium slab betas.

  The dosimeter is probably unable to meet the DOELAP performance criteria. A pilot study conducted in 1980 at the University of Michigan indicates that the Harshaw TLD could pass only one out of five categories in a then-proposed ANSI standard. … The current DOELAP standard is more stringent and has 13 testing categories. … The present TLD was tested at ORNL for low-energy X-ray response and failed to perform satisfactorily at the 30 and 60 kVp energy levels… The purchase of a new, more capable dosimeter will be required in order to obtain DOELAP accreditation."

- **We were worried. So was Harshaw.**

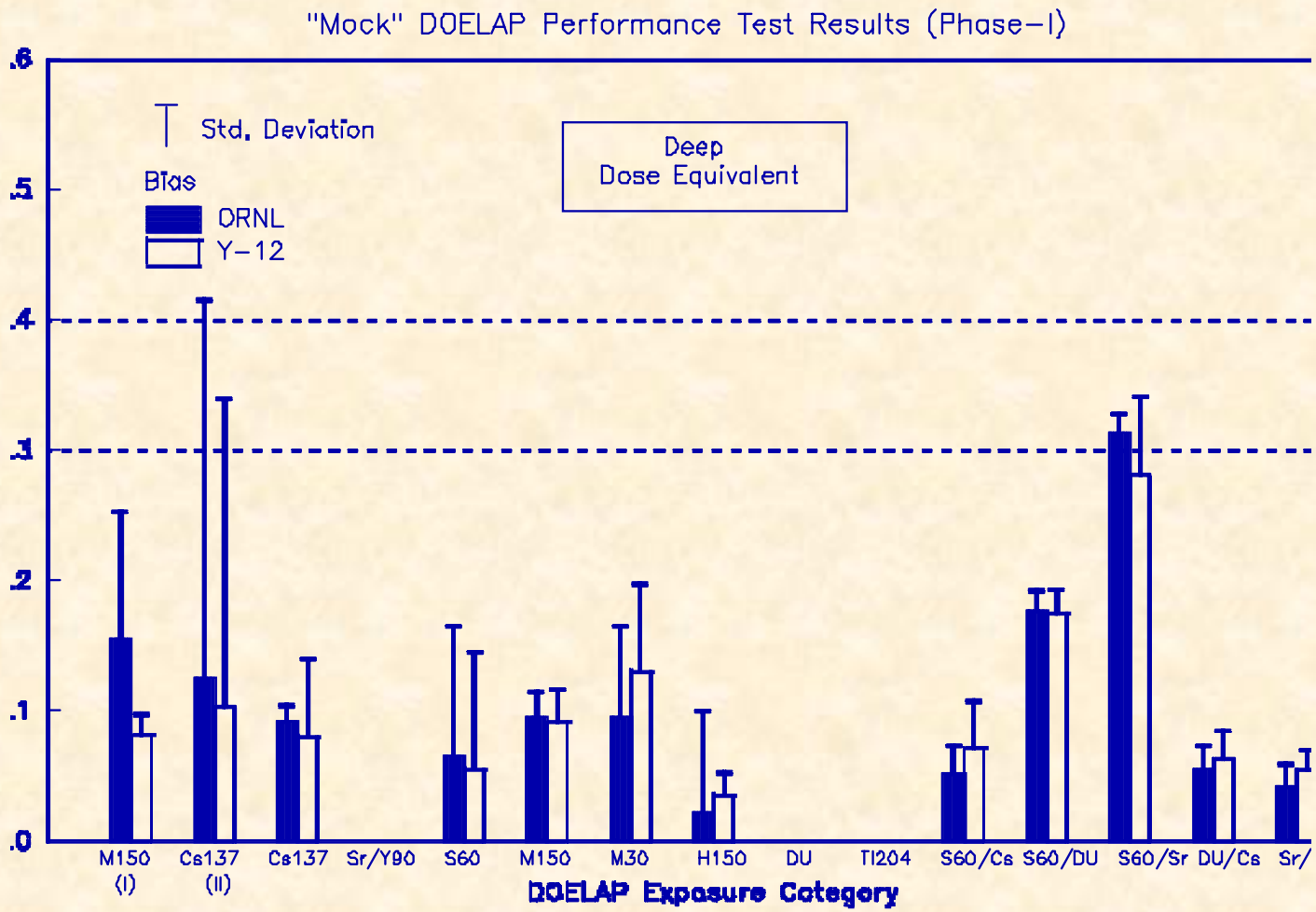
The Gauntlet is Thrown

- We decide to solicit bids for a new dosimetry system & Harshaw learns of this
- Harry Koppel & Terry Moore ask, “How do we win this contract?”
- Rhonda Lay, Jack Buddenbaum and I reply, “Pass DOELAP – all 13 categories!”

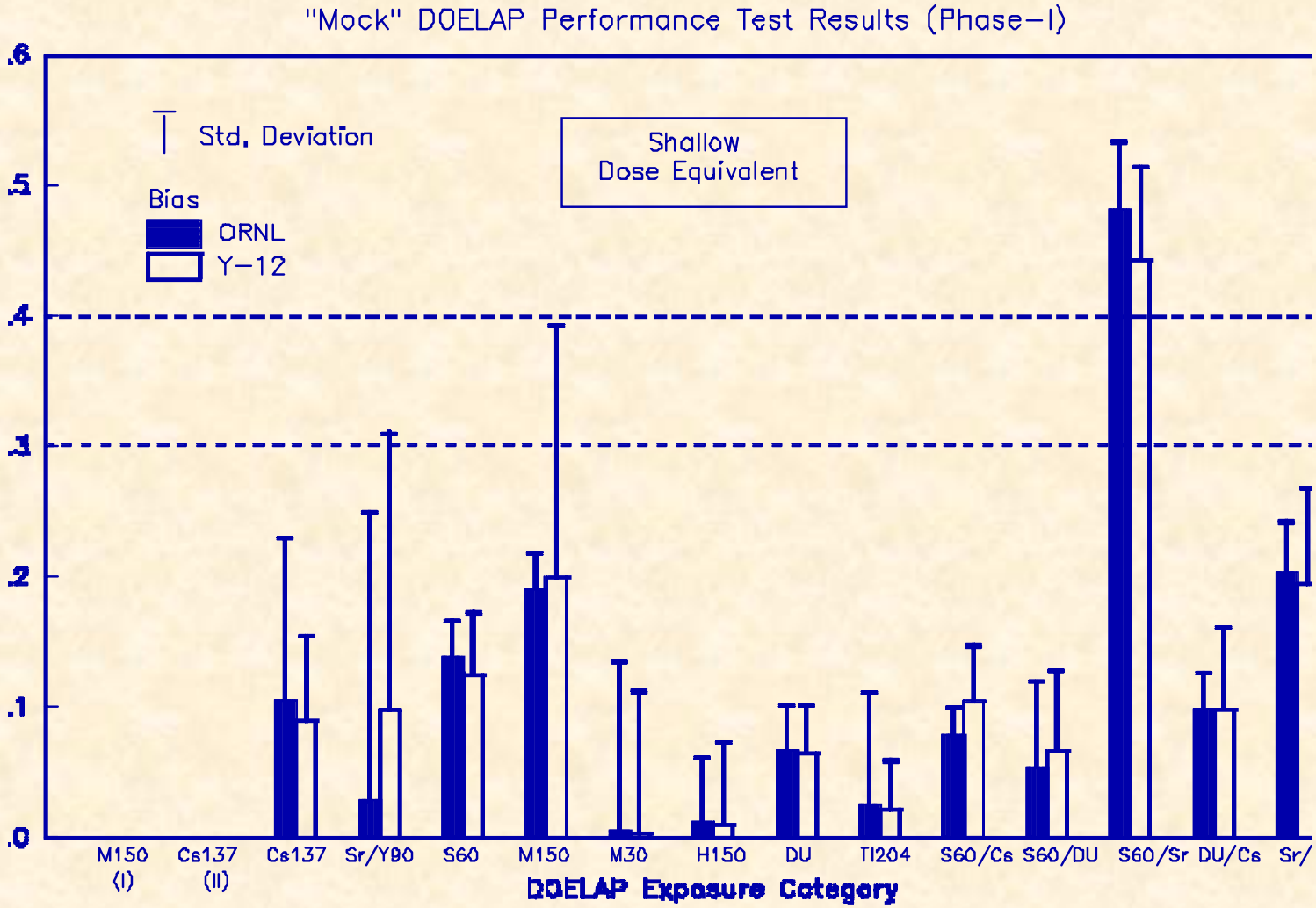
The Gauntlet is Taken Up

- Harshaw wins the bid
  - Cited a successful NVLAP design based on work for the Navy
  - Convinced us that they could pass DOELAP
- But there's still work to do
  - We specified a thin chip for the shallow dose
  - And there's all those additional exposure categories
- Harshaw suggests a partnership to develop the final product, & we agree

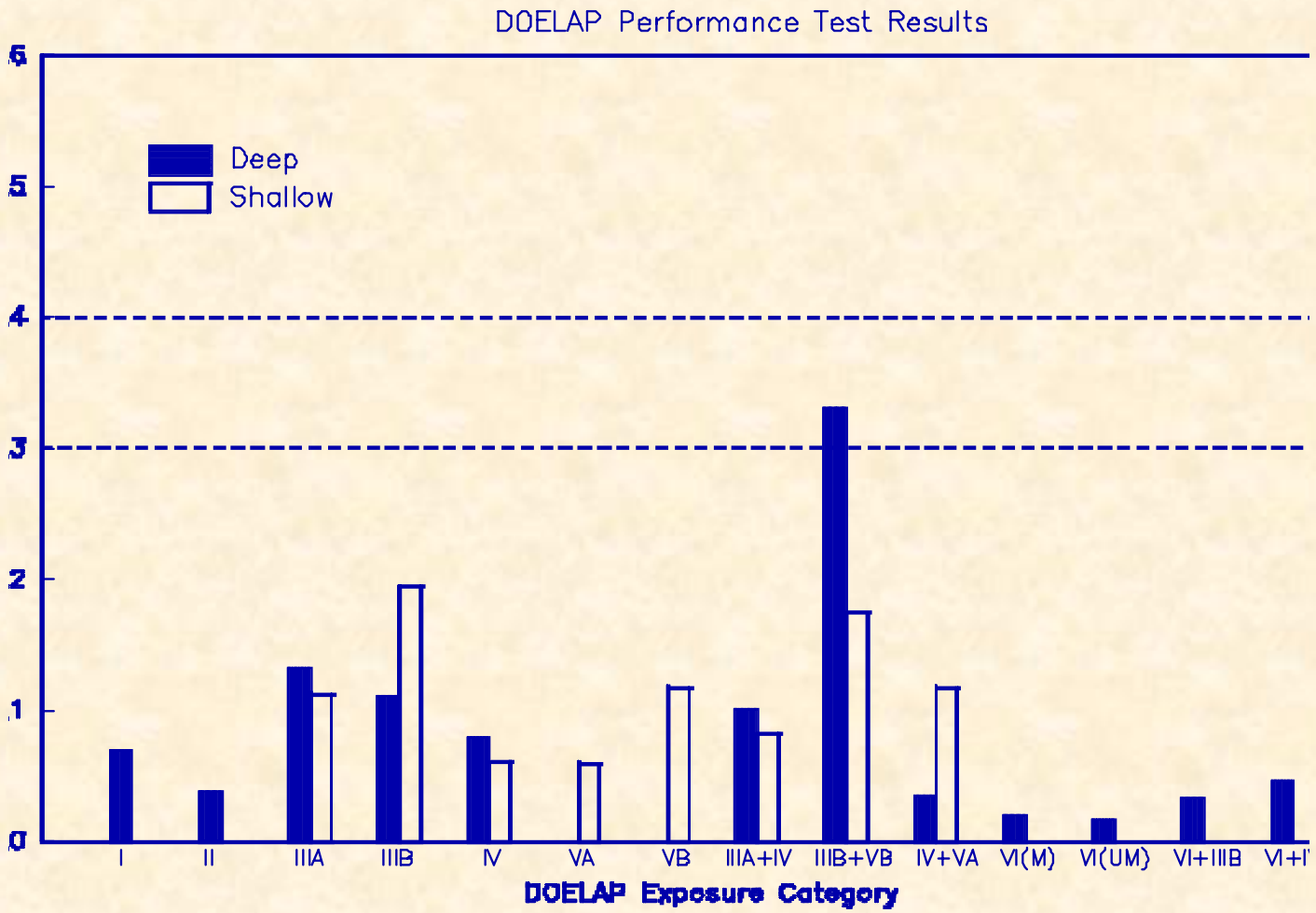
Results of First Mock DOELAP Performance Test (Deep DE)



Results of First Mock DOELAP Performance Test (Shallow DE)



Results of Second Mock DOELAP Performance Test (Improved Algorithm)



And the Sweet Savor of Success

Memo from TH Row to BD Walker, October 1989

"We have been notified informally by Doug Carlson, administrator of the DOE Laboratory Accreditation Program (DOELAP) for personnel dosimetry in Idaho Falls, that the Energy Systems Centralized External Dosimetry System (CEDS) has successfully met the performance criteria for DOELAP accreditation…

This accomplishment is particularly important to the Department of Energy because it represents the *first instance that performance testing was successfully completed by an applicant in all the available DOELAP exposure protocols*. The capability was developed by very close cooperation between Englehard Corporation and Energy Systems health physics staff. Englehard supplied the dosimeters, readers, and dose assessment algorithms; our staff communicated the performance requirements, tested and verified dosimeter and algorithm performance, and suggested system improvements."

(emphasis mine)

But, In Case You Labor Under the Misconception that Your Government Knows What It's Doing …

"Your November 9, 1989, memorandum to J. S. Bogard advising us of our success in the DOE Laboratory Accreditation Program (DOELAP) performance tests refers to our system as the "**ORNL Personnel Dosimetry System.**" The system is actually part of a Martin Marietta Energy Systems corporate personnel dosimetry program, and the successful implementation of the program depended on significant contributions from radiation protection staff at the Oak Ridge Y-12 Plant, Oak Ridge Gaseous Diffusion Plant, and Paducah Gaseous Diffusion Plant, as well as staff at Oak Ridge National Laboratory and the corporate Environmental and Safety Affairs Office…

We would appreciate your changing the name of the dosimetry system in your files and correspondence to the *Martin Marietta Energy Systems Centralized External Dosimetry System (CEDS)*. This name more accurately reflects the corporate nature of the external dosimetry program, and it recognizes the contributions from across Energy Systems."

-- RJ Forbes to RD Carlson, 20 Nov 1989

(bolded emphasis mine)

Finally, the certificate...

United States Department of Energy
Laboratory Accreditation Program
for Personnel Dosimetry Systems

DOELAP

Certificate of Accreditation

Martin Marietta Energy Systems'
Centralized External Dosimetry System

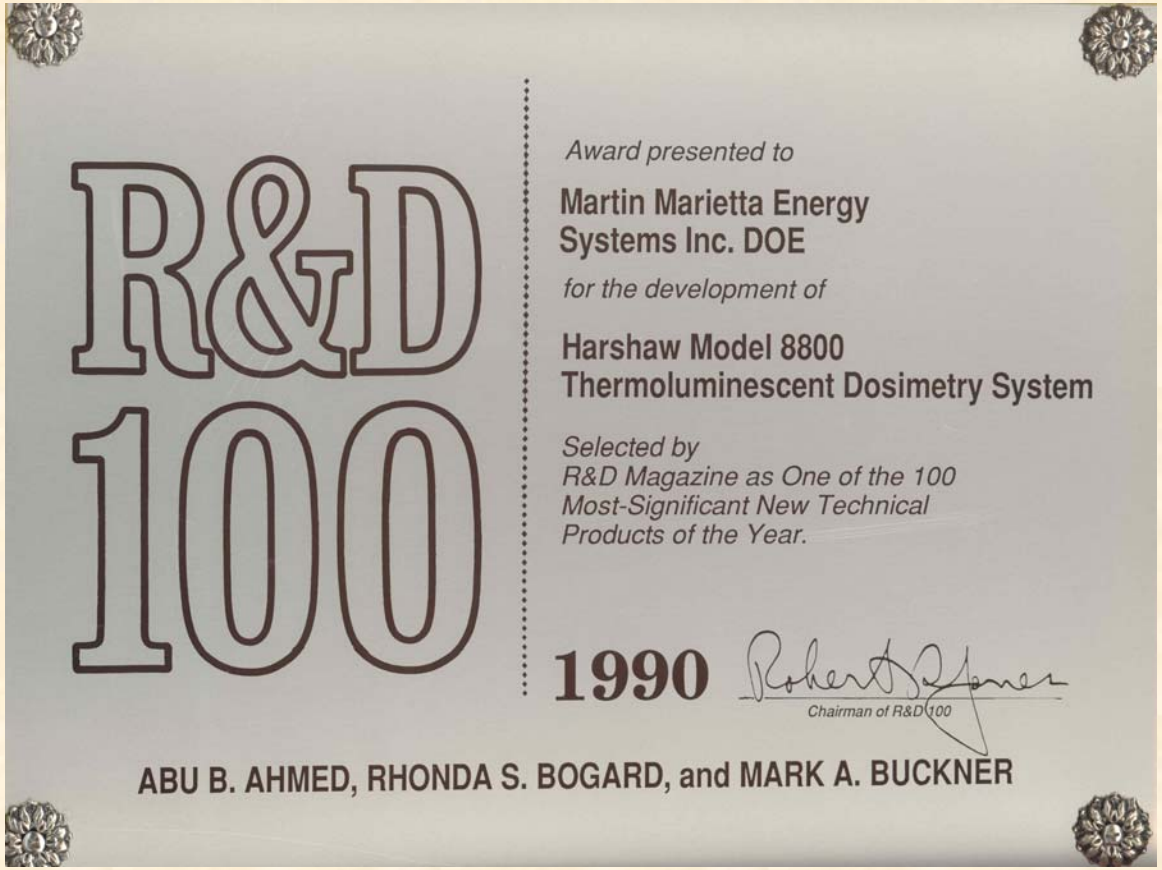
is recognized for demonstrating compliance with DOE performance criteria for personnel dosimetry systems. Accreditation is granted for the dosimetry systems and irradiation categories listed on the back of this certificate.

January 31, 1992

Effective until

DOE Laboratory Accreditation
Program Administrator

And the Blush on the Rose, an R&D-100 Award



The Oak Ridge Award Team

The Harshaw Award Team

Bill Bruml
Charles Martis
Marko Moscovitch
Tony Rothermel
Andy Szalanczy
Rod Tawil
Ken Velbeck

The 8800 System Project Investment

- Oak Ridge
  - 5 years, 10-30 dedicated staff
  - > $1.5M
  - Significant Risk -- Unproven Technology
- Harshaw (*my guess)*
  - 25-50 staff-years
  - <u>Significant</u> financial risk

The 8800 System Project Payoff

- Oak Ridge
  - Second-to-none personnel dosimetry
  - Significant dosimetry program improvement
    - Technical
    - QA/QC
  - Leadership in the DOE family
- Harshaw
  - Most successful dosimetry system worldwide
  - Market leaders

Final Note on Neutron Dosimetry

- We still use a separate dosimeter for rad workers with neutron exposure potential
- An unexpected twist from being associated with the Oak Ridge DOE facilities:

  Jerry Hunt recalls that we had all the free world's ^{6}Li – Harshaw had to get it from us!
  - Used years ago by Y-12
  - Stored at ORGDP
  - Distributed by ORNL Isotope Sales

ACORN 2

Continuing collaboration with Marko Moscovitch to develop an energy-independent neutron dosimeter

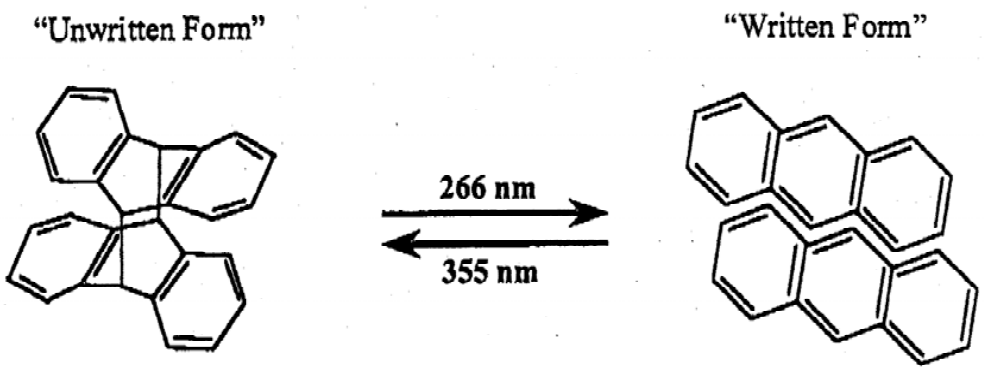
3D OPTICAL RANDOM ACCESS MEMORY (3D ORAM) TEAM

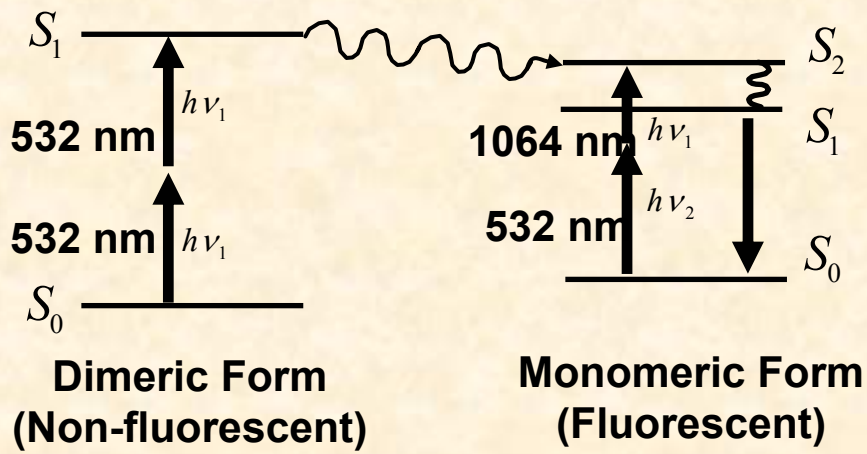
Jim Bogard	Oak Ridge National Laboratory
Marko Moscovitch	Georgetown University
Gary Phillips	Georgetown University
Tuan Vo-Dinh	Oak Ridge National Laboratory

Optical Memory Material Response to Neutrons and Charged Particles

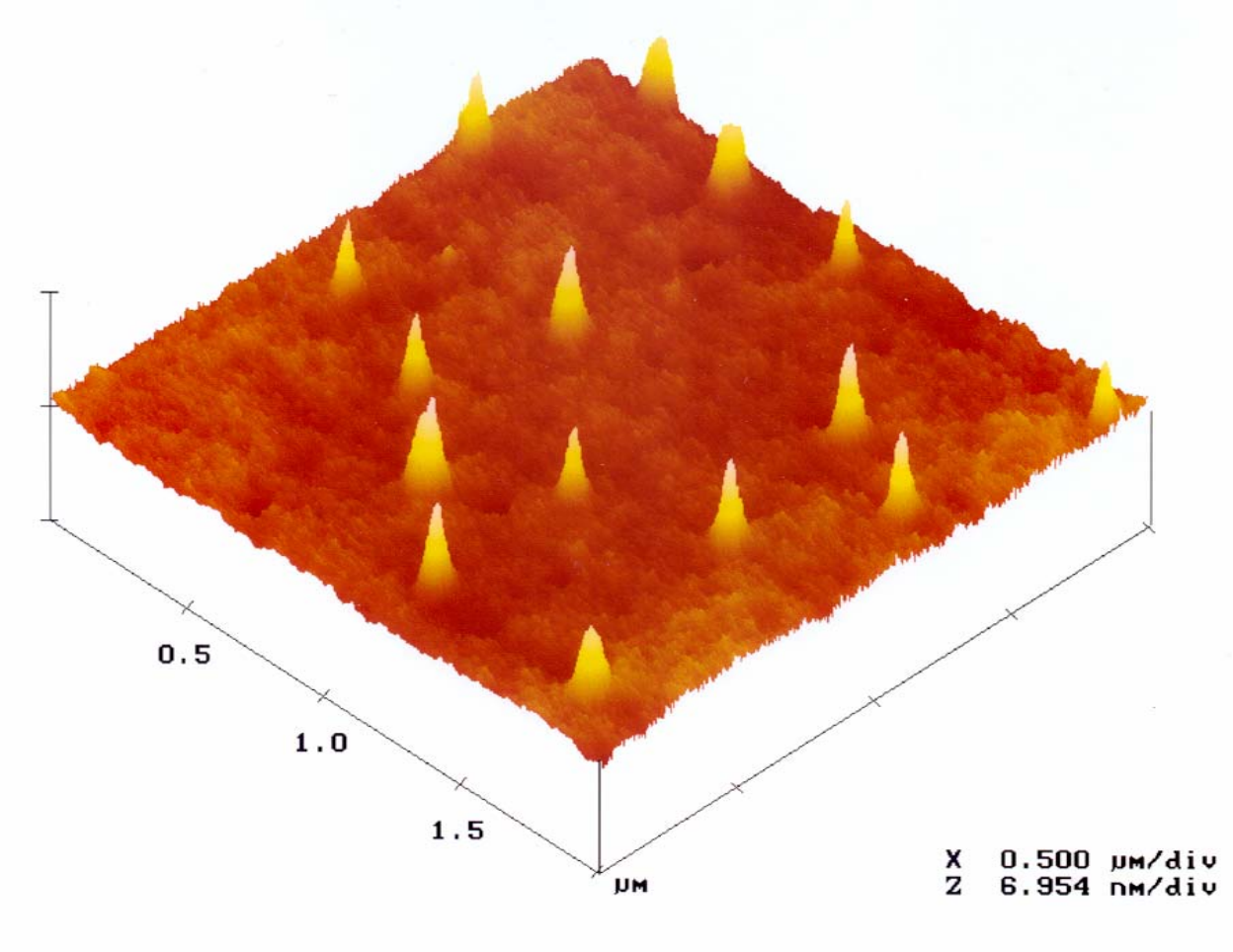
- Doped Optical Random Access Memory (ORAM) material responds quantitatively to energetic neutrons & charged particles.
- Mean and variance of penetration depth are functions of particle type & energy.
- The number of state changes (fluorescence intensity) is a function of dose unique for each radiation type.
- Response is reversible – the sensor can be re-used.
- Locations of state changes in the sensor can be determined by 2-photon excitation for track reconstruction and spectrometry.
- Surface features in CR39 exposed to neutrons also appear to be energy-dependent.
- We are now investigating the utility of high-resolution surface imaging of ORAM for determining the neutron spectrum and identifying the source material.

Anthracene/Anthracene Dimer





Anthracene is a stable dopant that responds reversibly to neutrons. The 'unwritten' form is the fluorescent monomer. Exposure to neutrons converts anthracene to the non-fluorescent dimer form.

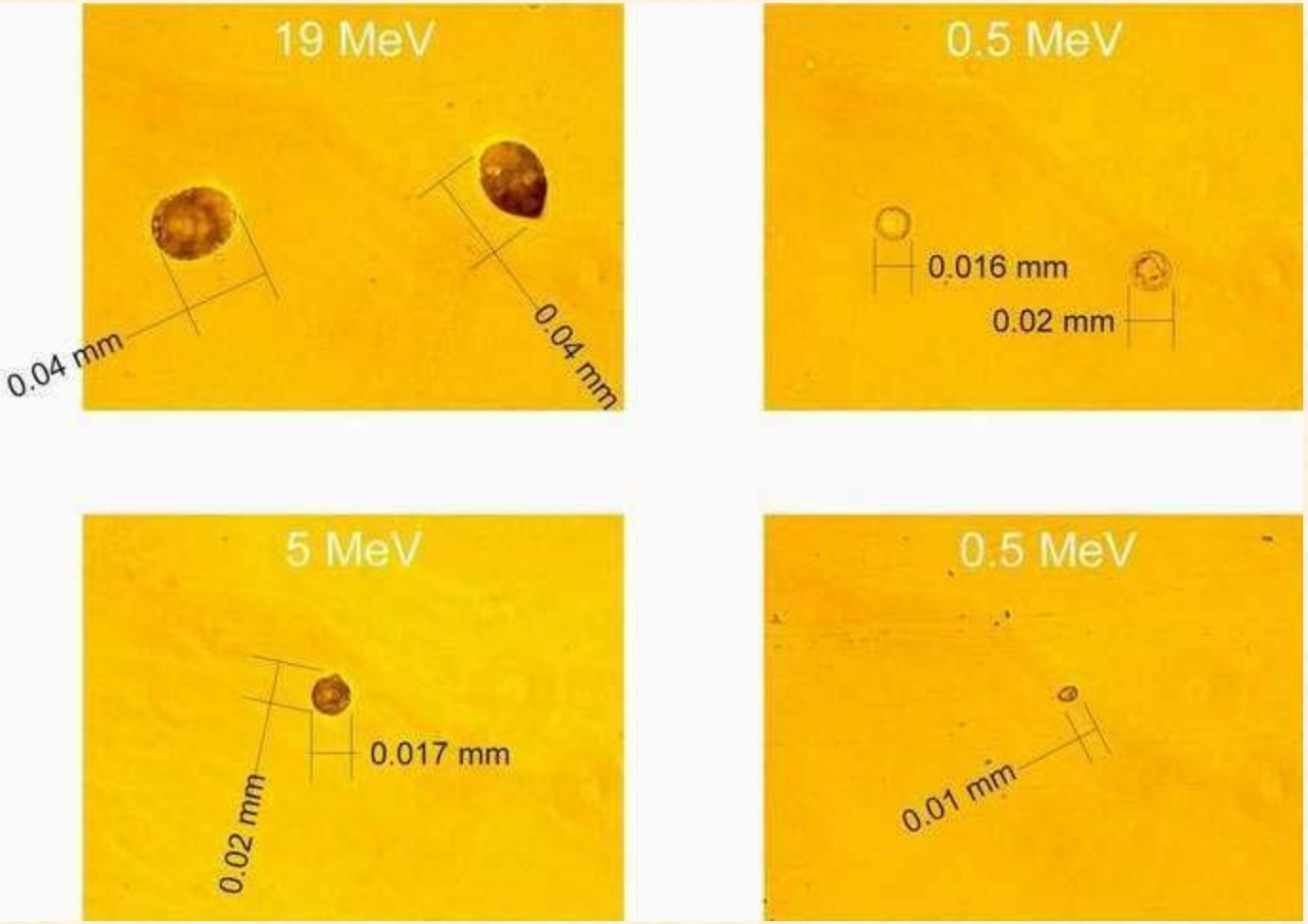


The average temperature rise from a 1-rad whole-body gamma exposure is about 2×10^{-6}°C (see Turner, Bogard, Hunt & Rhea, **Problems & Solutions in Radiation Protection**, p 256).

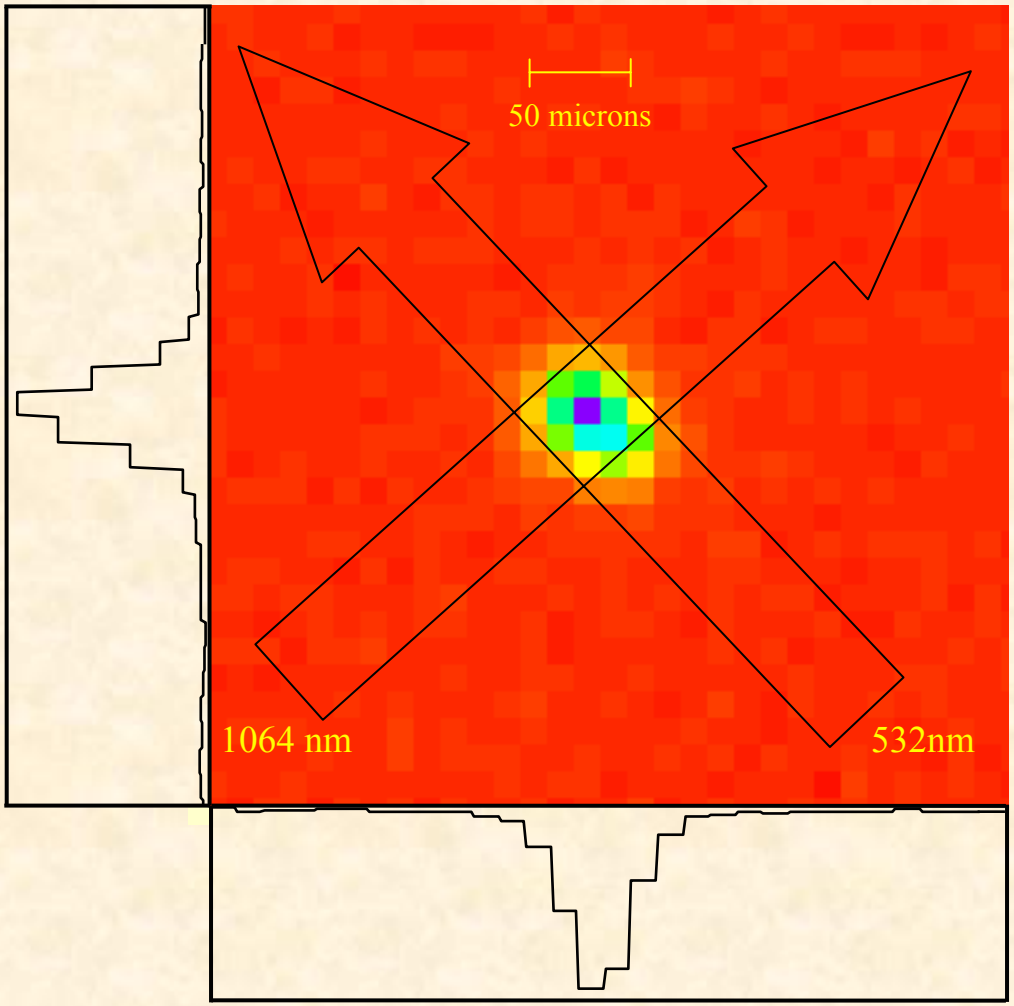
This shows that the temperature increase along a charged particle track is enough to melt and expand PMMA plastic!

Atomic-force microscopy image of SP/PMMA Film Exposed to 6.5-MeV $^{12}C^{3+}$

Photomicrograph of Etched CR39 Shows Feature Dependence on Neutron Energy and Incidence



Fluorescence Induced by 2-Photon 'Pumping' at Intersection of Crossed Laser Beams in Anthracene-Doped PMMA



ACORN 3

The Use of Natural TL Materials for Dating Archaeological Artifacts

Artifact Dating Collaborators

Jim Bogard	Oak Ridge National Laboratory
Mike Elam	University of Tennessee – Knoxville
Andrew Kramer	University of Tennessee – Knoxville
Bob Weeks	Oak Ridge National Laboratory (Ret.)
Dan Weinand	University of Tennessee – Knoxville

Common Natural TL Materials

- Quartz
- Feldspar
- Tooth Enamel

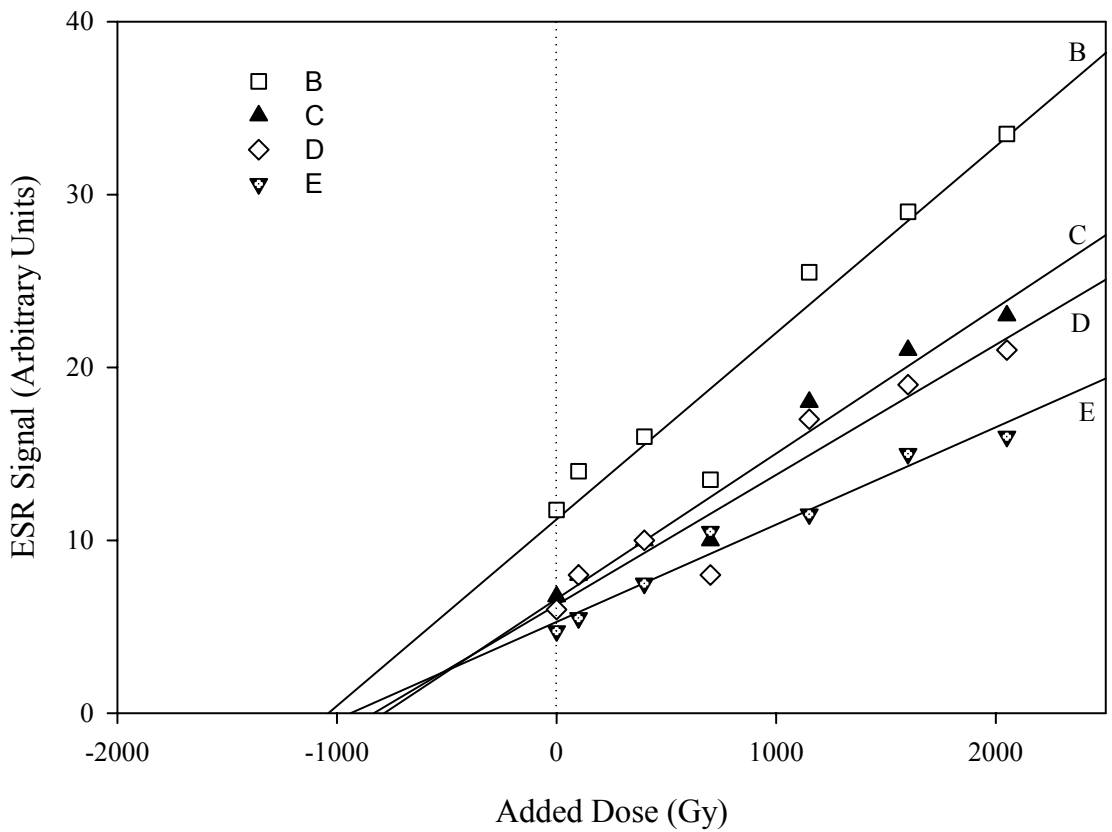
Steps for Determining Artifact Age

- Estimate contextual dose rate
  - Terrestrial, from radionuclide concentrations
  - Cosmogenic
- Estimate total delivered dose to artifact
  - ‘As-found’ TL signal
  - Dose Response
- Divide dose by dose rate
- Publish results, impress friends

Use of Electron Paramagnetic Resonance Instead of TL

- EPR measures the same promoted electrons that provide the TL signal
- Method is non-destructive
- EPR is a spectroscopic method – provides lots of information

Example of Dose Response Curves from Fossil Tooth Enamel Using Four EP Resonances



This sample appears to be about a million years old.
(Contextual dose rate ~0.8 mGy/a.)

ACORN 4

National Standards

HPS/ANSI N13.27

Performance Requirements for Pocket-sized Alarm Dosimeters and Alarm Ratemeters

Writing Group Background

Jim Bogard (Chairman)	National Laboratory
Steve Browne	Commercial Nuclear Power
Bill Casson, Sr.	National Laboratory
Larry Donovan	Military
Joe Leon	Dosimeter Manufacturer
Charlene Raddatz	Regulatory Agency
Mark Simpson	Commercial Nuclear Power
Bob Ulman	Dosimeter Manufacturer
Joe Zlotnicki	Dosimeter Manufacturer

Status of HPS/ANSI N13.27

- Initial draft contained both type and performance requirements
- Type requirements removed – performance requirements remain
  - Type requirements are provided in ANSI Standard N42.20-1995
  - Performance requirements are generally the same as in ANSI N13.11-1993, except
    - No test in photon mixtures (Category VI)
    - Beta-Photon Mixtures (Category VII) delivered simultaneously
    - Neutron-Photon Mixture doesn't require ^{137}Cs (uses tramp photons from the neutron source)

SUMMARY
Line them acorns up…

RADIATION DOSIMETRY IS A GREAT FIELD

- Personnel Dosimetry
- Microdosimetry
- Sensor Development
- Archaeology
- Standards

… all in just 20 short years.

My Wish for You

- Enjoy the Harshaw TLD User's Group Symposium
- Don't lose sight of the broader picture
- Stretch – Have as much fun in your career as I've had in mine