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Active Dosimeter Development for Pulse Reactor Applications

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Abstract

Customer requirements for reducing the uncertainty associated with real-time dose information during reactor pulses and the absence of dosimeters that could provide that information motivated Sandia's Radiation Effects Sciences (RES) program to develop active radiation detectors. The limitations and constraints of the test environments required the development of several different active dosimeters to meet customer needs. The development and testing of Diamond Photo-Conducting Detectors (PCDs) and multiple types of calorimeters are presented. In addition, the methodology that the RES uses to characterize the time- and energy-dependent reactor n/γ environments is presented.

My Background

- B.A. (Physics) – Hendrix College (1996)
  - Senior Topic: “Large Ring Laser Interferometers As a Potential Detector of Seismic Waves”
- M.S. (Health Physics) – Texas A&M (1998)
  - Thesis: “Energy Deposition Spectra of Simultaneous Electron Emissions from Low Energy Protons”
- Ph.D. (Nuclear Engineering) – Texas A&M (2001)
  - Dissertation: “Radiation Effects on the Cell-Cell Communication of Mammalian Cells”
- Staff Member of SNL since 2001
  - Main responsibilities are in the Radiation Effects Sciences (RES) Program

Outline

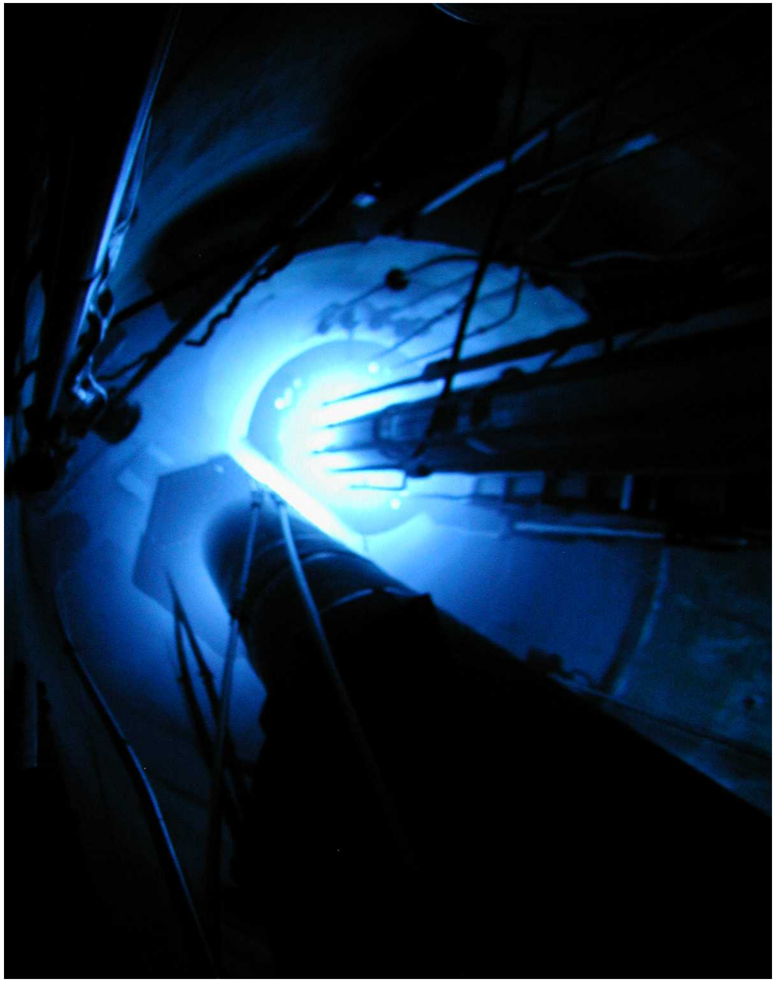
- Active dosimetry – Why does Sandia cares?
- Reactor facilities available to Sandia
- Diamond Photo-Conducting Detectors (PCDs)
- Use of calorimeters for active dosimetry
- Methodology used for characterization of the time-dependent and energy-dependent reactor n/γ environments

Active Dosimetry

- Why does Sandia care about active dosimetry?
  - The n/γ rate environments are important for our customers
  - Reduced uncertainty allows the customer to reduce "over-test"
- Dosimeters currently being used by the Radiation Effects Sciences (RES) program
  - Self-Powered Neutron Detectors (SPND)
  - Multiple fission chambers (^{235}U, ^{238}U, and ^{237}Np)
  - Diamond Photo-Conducting Detectors (PCDs)
  - Multiple calorimeters (Boron, Silicon, and Tungsten)
- Major Issues
  - Dosimeter response changes (neutron or total dose effect)
  - Undefined mixed field (n/γ) responses
  - Quantification of the uncertainty in the detectors

Annular Core Research Reactor (ACRR)

- Pool-type Reactor
  - Unique $BeO-UO_2$ fuel allows fuel temperatures up to 1400^{o}C
  - Pulse, steady-state and tailored transient rod withdrawal operation
- Performance Characteristics
  - Pulses
    - ~$3.00 reactivity insertion
    - ~30,000 MW peak power
    - 6.5 ms pulse width
    - 300 MJ of energy released
  - Steady-State
    - 2.0 MW reactor power
    - 4.0 MW for intermittent operations



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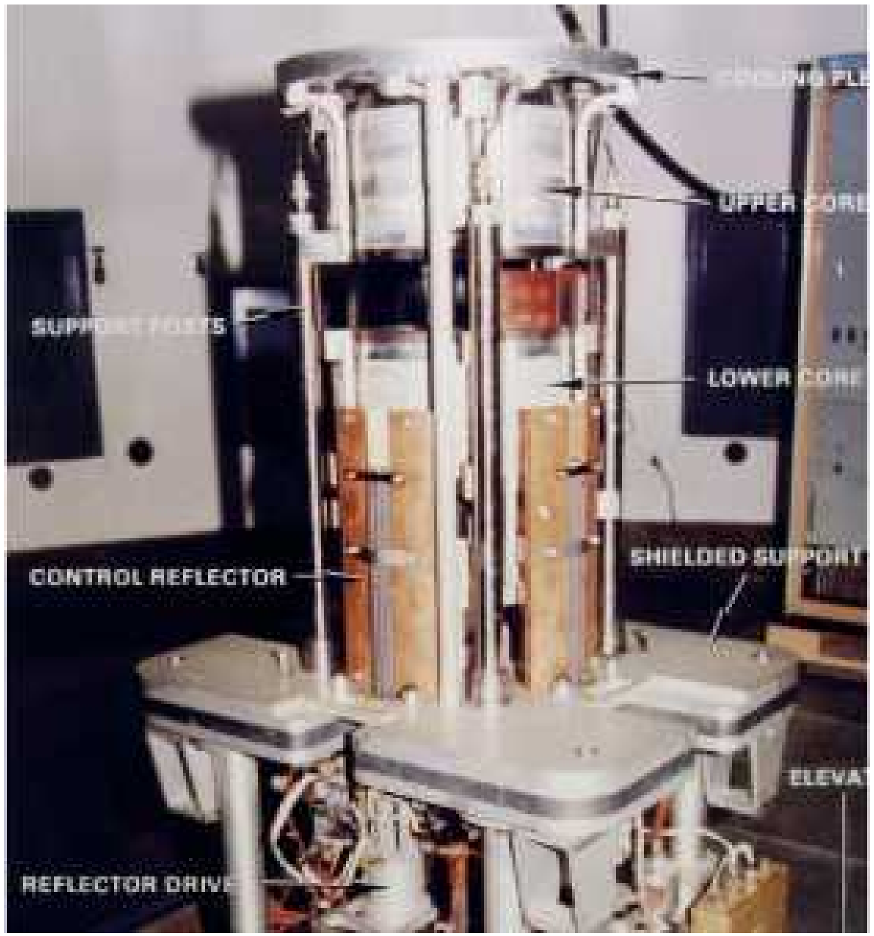
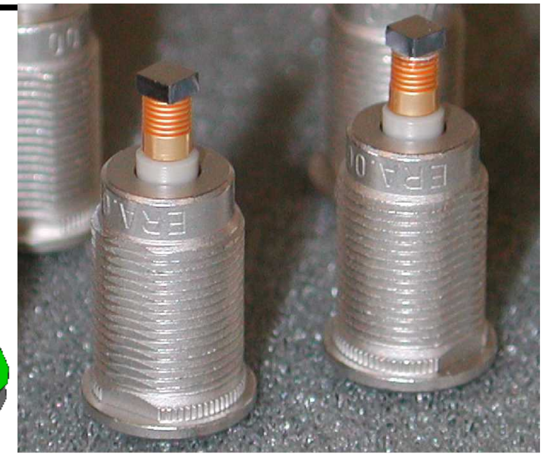
Sandia Pulse Reactor III (SPR-III)

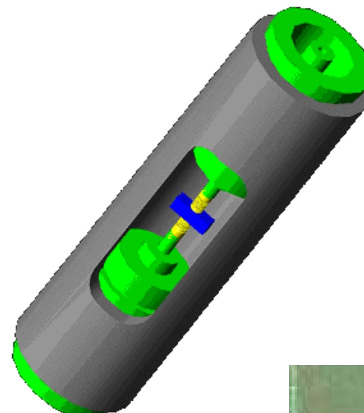


- Fast burst reactor
  - Unmoderated cylindrical assembly of HEU alloyed with molybdenum
  - Capable of pulse and steady-state operations
- Performance Characteristics
  - Pulses
    - ~1.13 reactivity insertion
    - $450^{o}C\ \Delta T$
    - 76 μs pulse width
    - 8.0×10^{18} n/cm^2-s peak flux
  - Steady-State
    - 10 kW reactor power

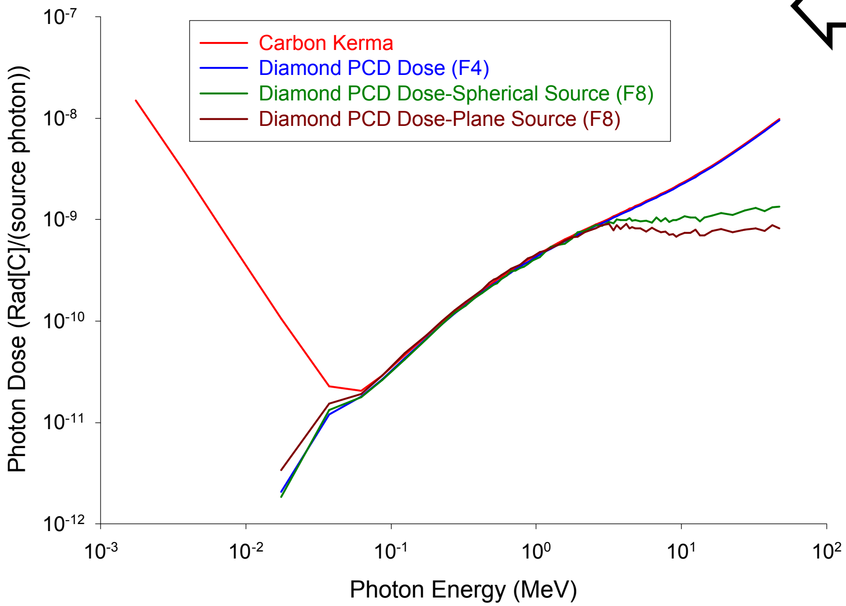
PCD Theory/Design/Fabrication



- **Material**
  - Requires high resistivity, large bandgap single crystal
  - 1x1x2 mm Type IIa natural {100} diamond with ohmic contacts
- **750 VDC Bias**
  - Just above 3 kV/cm critical saturation electric field
- **RTV potting material, Peek dielectric, Lemo connector**



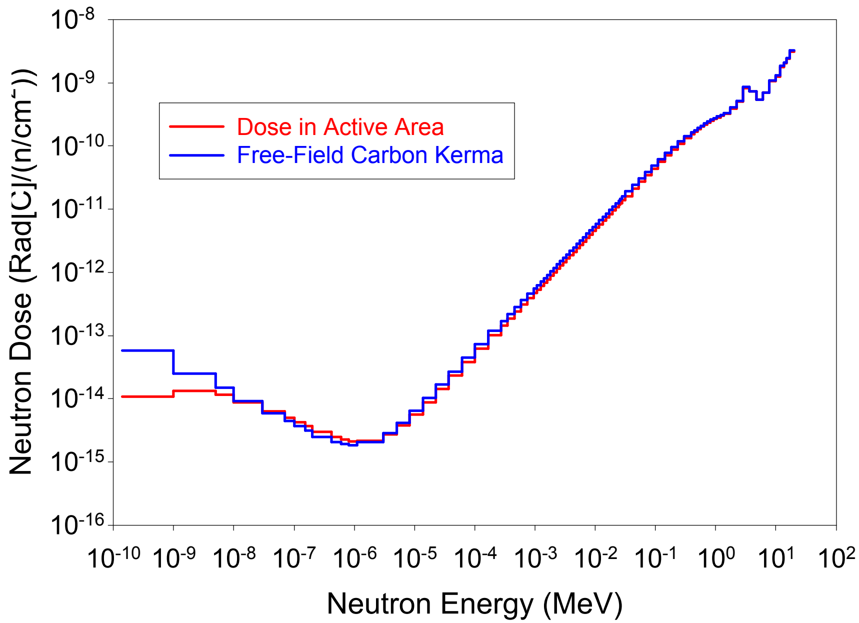
Modeled PCD Response



Photons:

- **Low energy attenuation**
- **High energy non-CPE**

Neutrons:



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PCD Maintenance

- Calibration performed on an individual basis
  - Take baseline data GIF and Hermes-III half power (photon sources)
  - Validate performance at ACRR for 20, 50, and 300 MJ ; comparison with SPND profile
- Calculate field-dependent conversion factors
- Always bias one side
  - Apparent trapped charge issue
- Pre-dose with 100 rad(C) within 8 hr of use
  - Automatic for ACRR due to pre-dose environment
  - Use first shot at linac
- Periodic stability check for neutron damage

PCD Issues

- Discrepancy in GIF and Hermes-III calibrations
  - Possible dose-rate effect or calibration issue
- Calibration of neutron response
  - Pure neutron calibration field not available
  - ACRR testing (with Hermes-III calibration) suggests reduced (~50%) response to neutron dose

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Calibration Discrepancy!

PCD #	GIF		H-III Half Power		Var. of Cal.
	Rsp.	Cal.	Rsp.	Cal.	
	mV-s /rad(AlN)	mV-s /rad(C)	mV-s /rad(TLD)	mV-s /rad(C)	
AA22	1.43E-7	1.58E-7	1.87E-7	2.24E-7	41%
AA24	1.30E-7	1.43E-7	1.81E-7	2.17E-7	51%
AA26	1.46E-7	1.61E-7	1.91E-7	2.28E-7	42%
AA28	1.44E-7	1.59E-7	1.89E-7	3.26E-7	43%
AA29	1.64E-7	1.82E-7	2.00E-7	2.40E-7	32%
AA30	1.30E-7	1.43E-7	1.62E-7	1.94E-7	36%
AA33	1.51E-7	1.67E-7	2.03E-7	2.43E-7	46%
AA46	1.34E-7	1.48E-7	1.73E-7	2.07E-7	40%
	Avg. Var. = 41%; abs. std = 6%				

n/γ Response of PCD in ACRR Irradiation

Matl.	Neutron Kerma	Gamma Kerma	% Dose from neutrons
Alanine	469.2	208.2	69%
Diamond	74.3	192.9	28%
Silicon	16.0	203.2	7%
CaF_2:Mn TLD	31.45	202.4	13%

n/γ dose components?

- Photon kerma varies with Z
- Neutron kerma varies with material composition

Energy Loss Mechanism	% Energy Loss	
	Primary Ion	Recoil Atoms
Ionization	67.1	9.34
Vacancies	0.22	0.71
Phonons	1.39	21.23

Only some of the neutron energy is deposited as ionization.

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PCD Summary

- We have developed and calibrated a radiation-hard diamond PCD dosimeter
  - Useful for relative time profiles in reactor testing
  - Good precision (<15%) within peak + 3 FWHM
- Response normalization
  - Neutron calibration is still an open issue
  - Dose rate sensitivity can not be ruled out
  - Current protocol is to normalize integral to silicon calorimeter at peak + 3 FWHM

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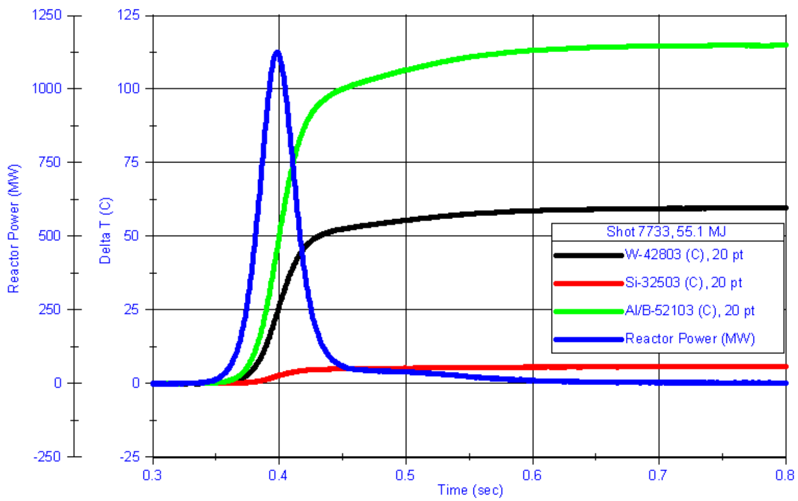
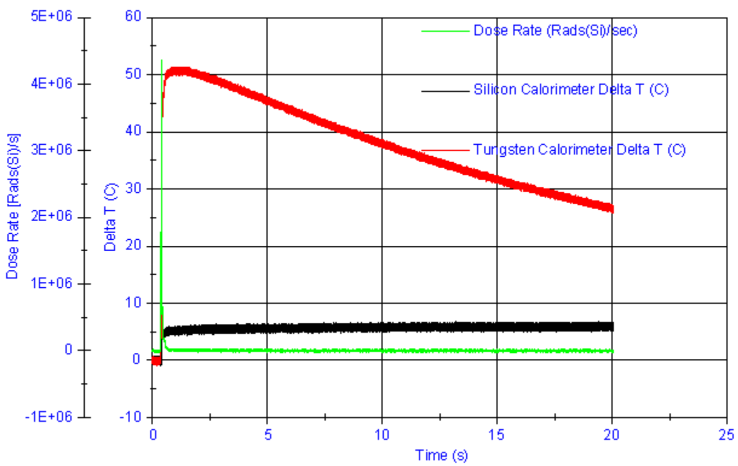
Calorimeters

- Si, B, and W used (other materials possible)
- Conversion to rad(Si) difficult for other materials
- Small signal (5° in ACRR 20-MJ, 0.2° in max. FBR)



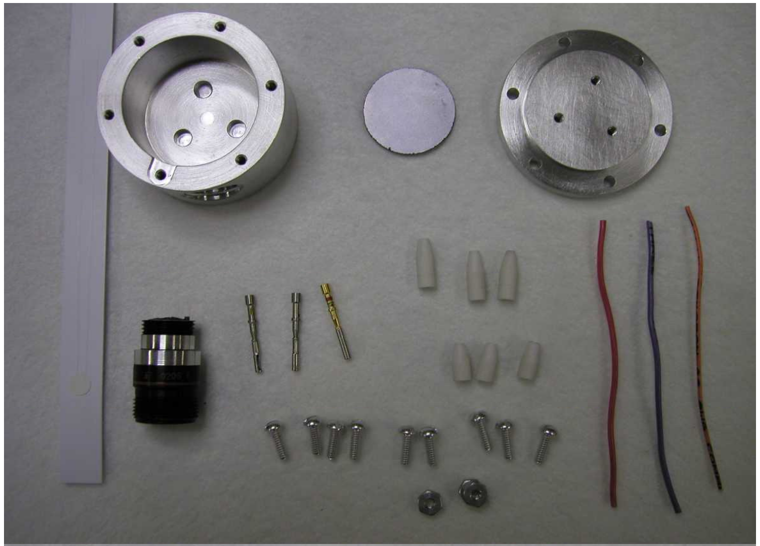
Temperature rise

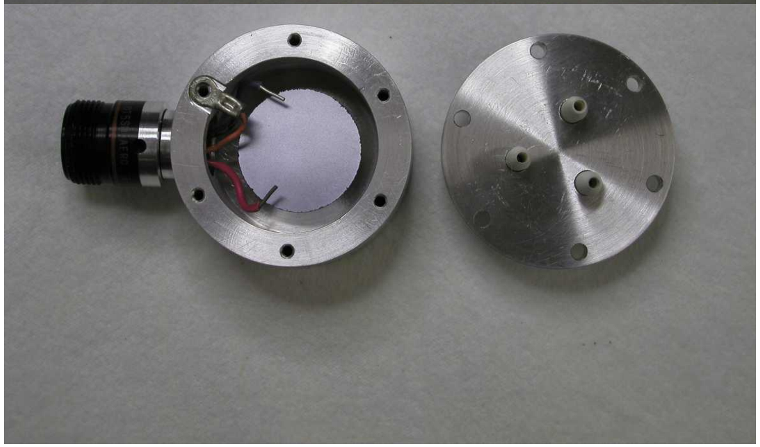


Temperature loss

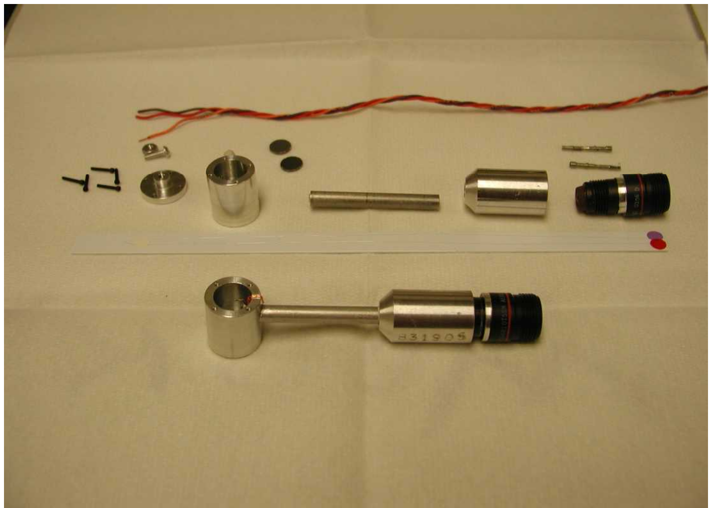
Early Calorimeter Designs

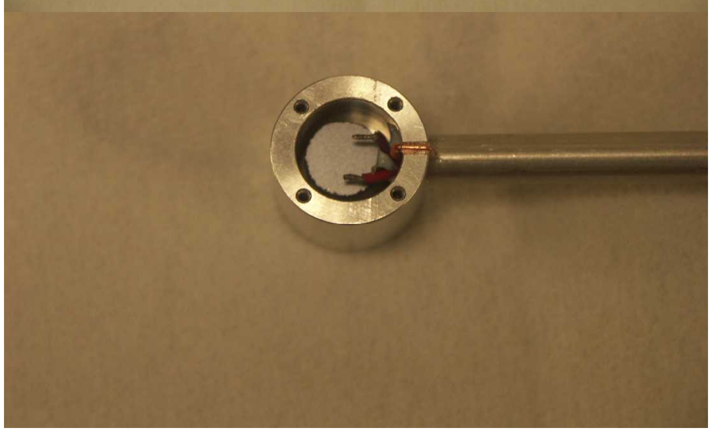
- **Two 1-inch (2.54 cm) silicon (or other material) wafers**
- **Type K thermocouple – (.001 inch, 0.0025 cm)**
- **Small aluminum housing**
- **Wires were not fully shielded**





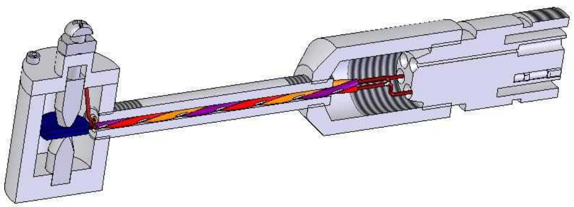
Current Silicon Calorimeter

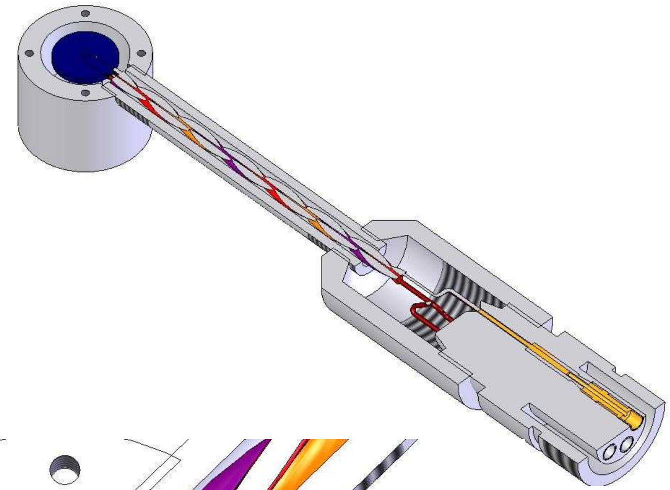


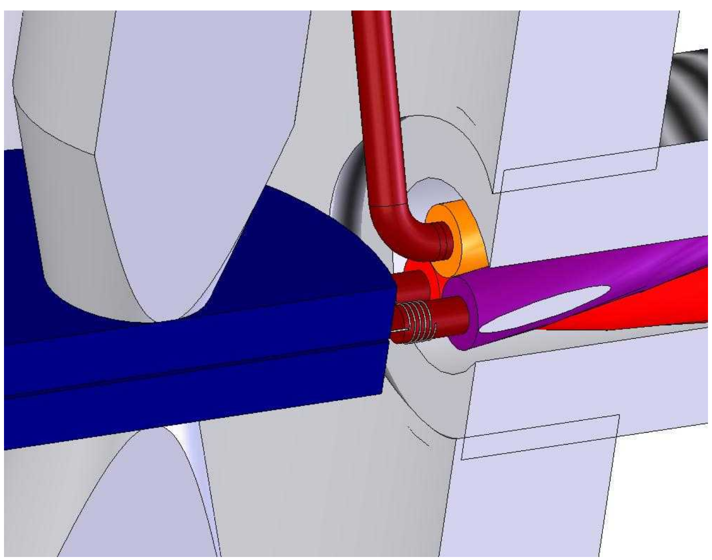


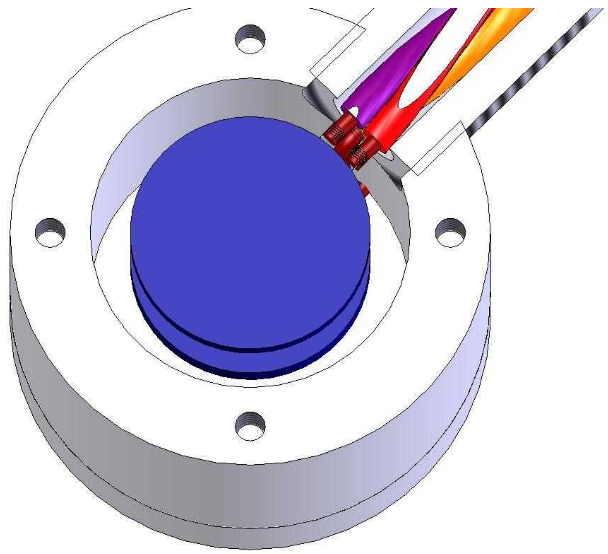
- **Totally EM shielded design**
- **Two float-zone disks of calorimeter material**
- **Low mass 1-mil Type- E thermocouple**
- **Twisted-shielded pair thermocouple cable to reduce noise.**

Current Calorimeter Construction



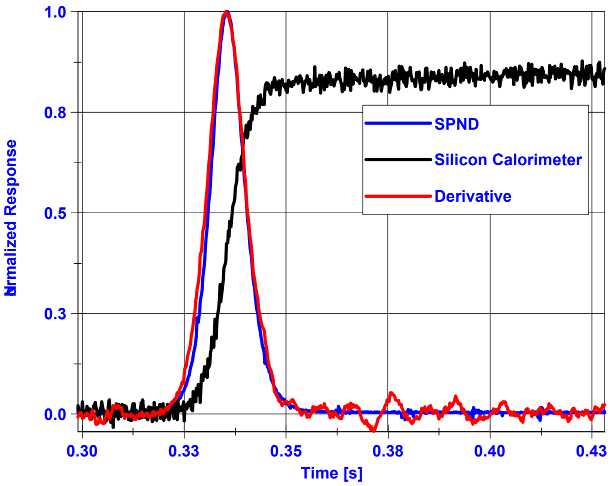


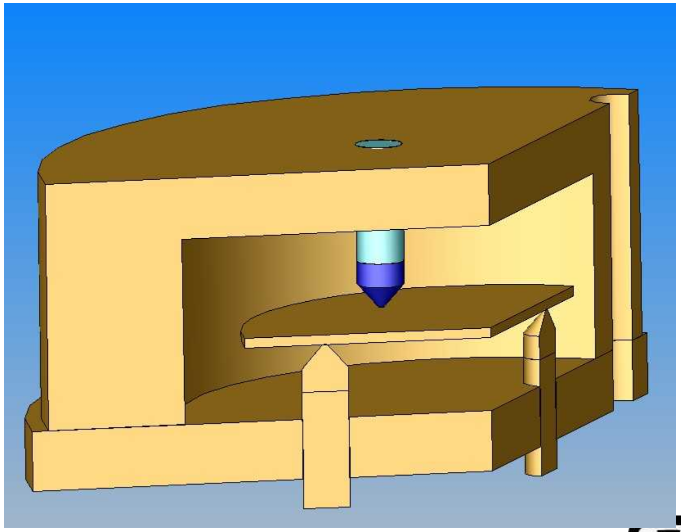




Modeled Calorimeter (Si) Response

- MCNP Calculation in ACRR
  - 7.5% of dose due to neutrons
  - 92.5% of dose due to gammas
  - 8.2 [krad(Si)/MJ] – Total
- Thermal model predictions are within 5% of measured values
- For ACRR tests, doses are within
  - 6% @ 23.3 MJ Pulse
  - 13% @ 53.6 MJ Pulse
  - 12% @ 200 MJ Pulse





Calorimeter Summary

- We have designed a real-time silicon calorimeter for use in reactors that matches calculated dose values during a reactor pulse.
- The calorimeter signal can be differentiated and is proportional to the SPND.
- A PCD can be used with the calorimeter for dose rate determination.
- The calorimeter is currently being used for routine customer support.

Methodology for Characterizing Reactor Environments

- **What is so different about these reactor characterization?**
  - Emphasis at Sandia is on the dynamic (rather than static) characterization
  - Requires the interpretation of active dosimeters during a radiation operation
- **Radiation Components**
  - Prompt fission neutron and gamma
  - Delayed fission neutron and gamma
  - Activation gammas
  - Late-time photoneutron environment in some reactors
    - $BeO-UO_2$ research reactor fuel (ACRR)

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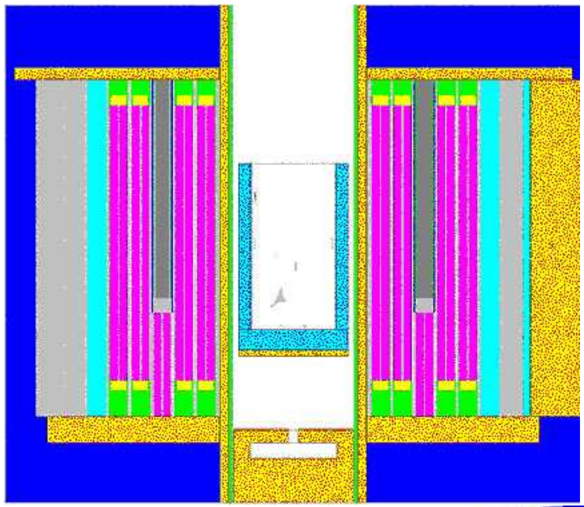
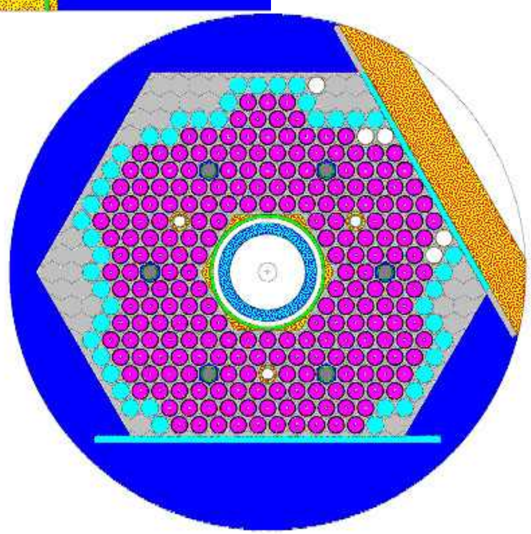
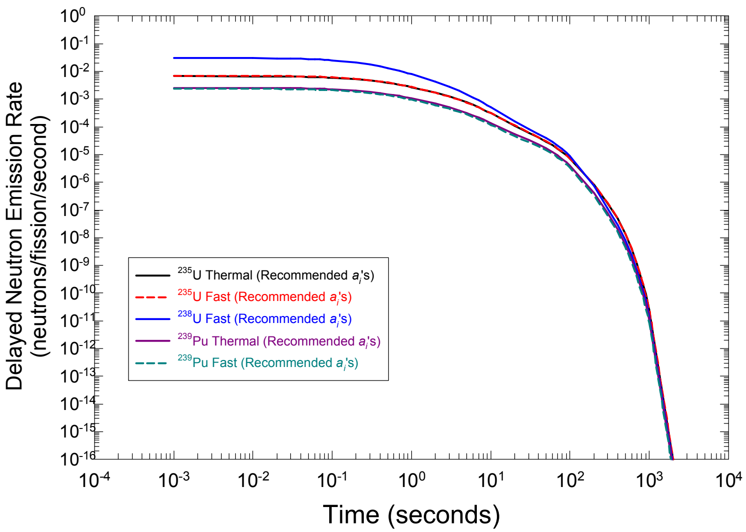
Calculation Approach (Prompt Radiation)

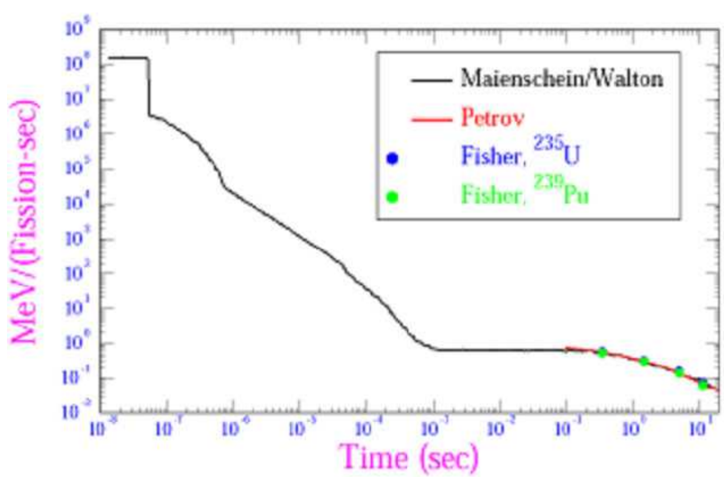




- Normal radiation transport
  - Do not turn on delayed neutron normalization
  - Addresses all prompt secondary gamma components – <u>if</u> – gamma production included in cross sections
    - Issue for some applications with Cd or Gd
  - Determine normal mode fission source distribution in fuel to support next step

Calculation Approach (Delayed Radiation)





- **Time-dependent spectrum from literature**
- **Radiation transport to capture spectrum modification from source to experimental location**
- **Convolute δ-function representation of transported source term with time-dependent emission rate model**
- **Emission rates from literature**
  - **Spriggs and Campbell for neutrons**
  - **Keepin/Maienshein for gammas**

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Calculation Approach (Activation Radiation)

- Radiation transport to get local neutron source spectrum at representative masses
  - Good resolution for local environment
  - Lumped mass-models for distant large masses
- Activation products with CINDER-90 or ANITA
- Activation gammas with TORI (Browne & Firestone)
- Time and energy-dependent gammas used as a source term for added radiation transport code

Survey of Active Dosimeters

- **Self-powered neutron detector (SPND)**
  - **Cd-based prompt response**
- **Photoconducting detector (PCD)**
- **Ionization chambers**
- **Fission chambers**
- **PIN diodes**
- **Calorimeters**
  - **Yes – in an active application but for "dose up to a time" not for a dose rate**

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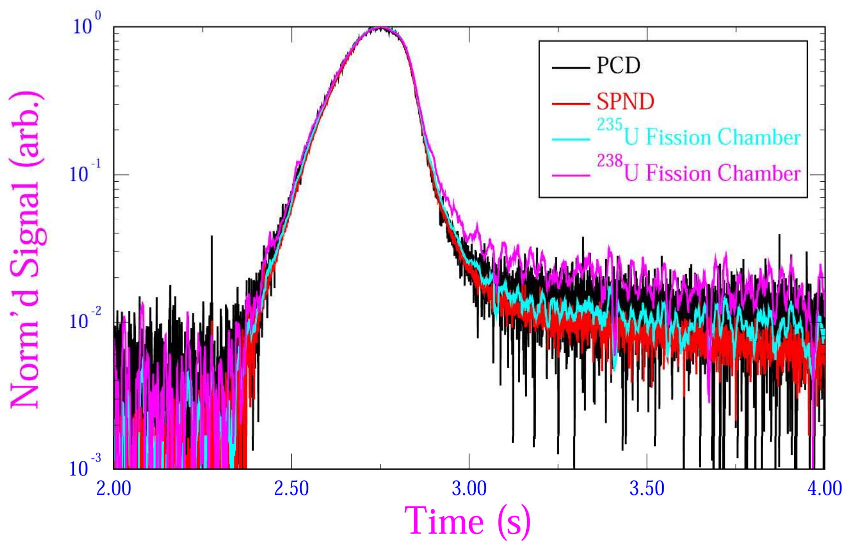
SPND

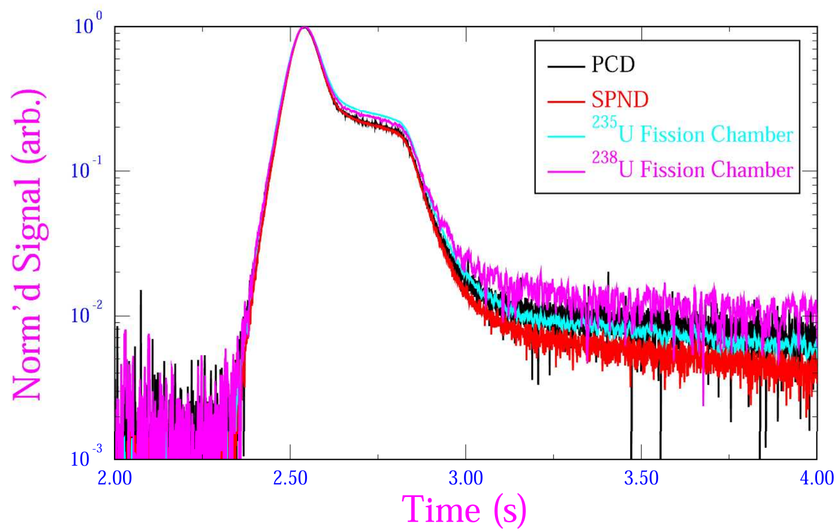
- **Dominant response is to thermal neutrons**
  - Cd-based prompt SPND
  - Delayed relative to fission source term by one generation
- **Some gamma response**
  - dominant at late times when neutron environment reduced
  - Gamma energy-dependent response
- **Issues:**
  - Size – typically long (0.4m)
  - Small signal
  - Location – next to fuel rather than at experiment

^{235}U, ^{238}U, ^{237}Np Fission Chambers

- Very sensitive neutron response
- Some gamma sensitivity in current mode
- Fission impurities (^{235}U in ^{238}U)



10-MJ ACRR Pulse



20-MJ ACRR Pulse

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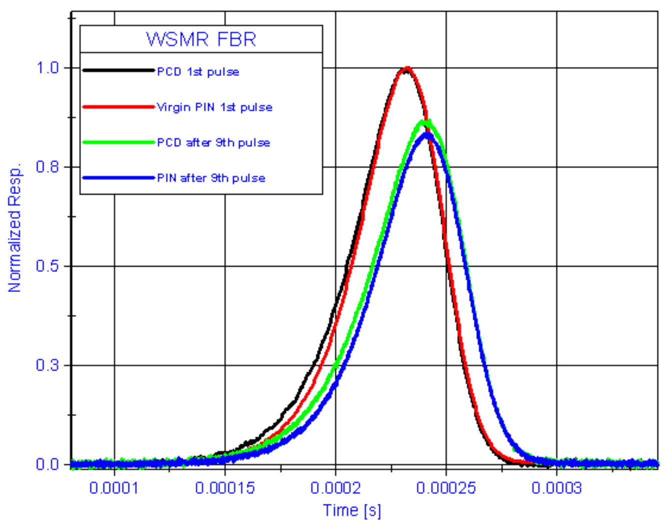
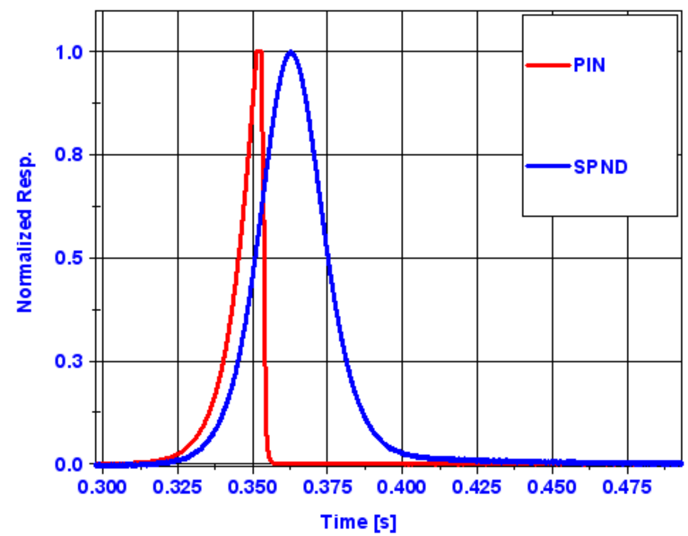
PIN Diodes

- Very sensitive – easily debiased
- Mixed neutron/gamma response – boron dopant
- Sensitive to neutron damage – even rad-hard versions (Emerge Semiconductor)



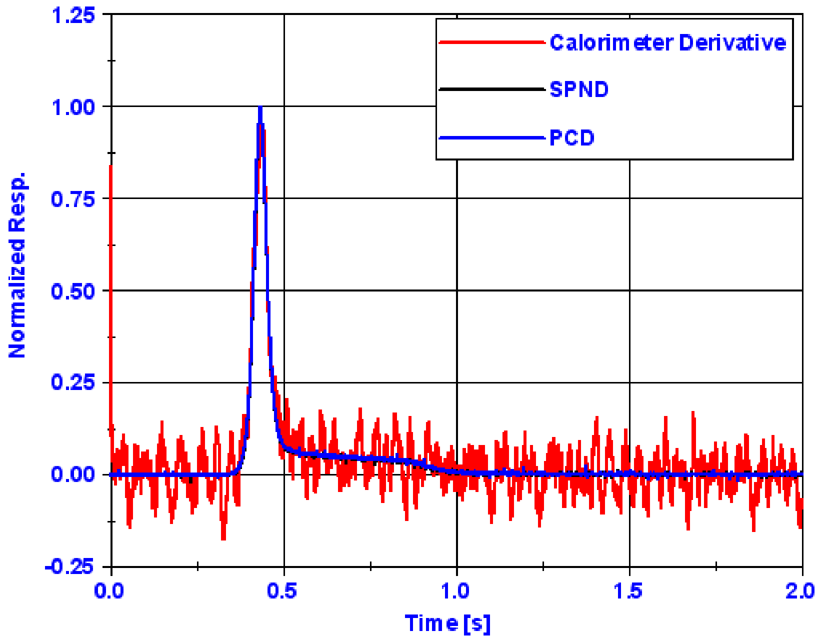
Calibration change

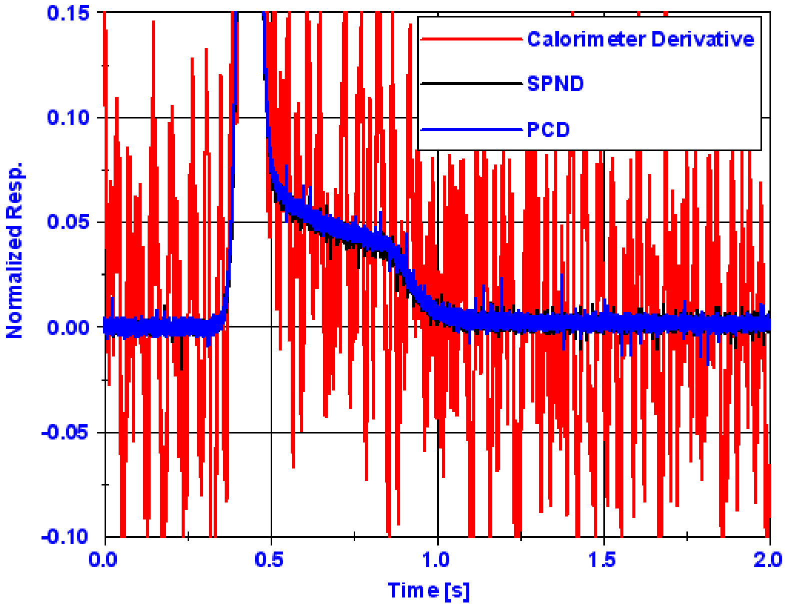


Failure

SPND, PCD, and Calorimeter Early-time Agreement

- PCD and SPND are indistinguishable
- Calorimeter derivative noisy but good





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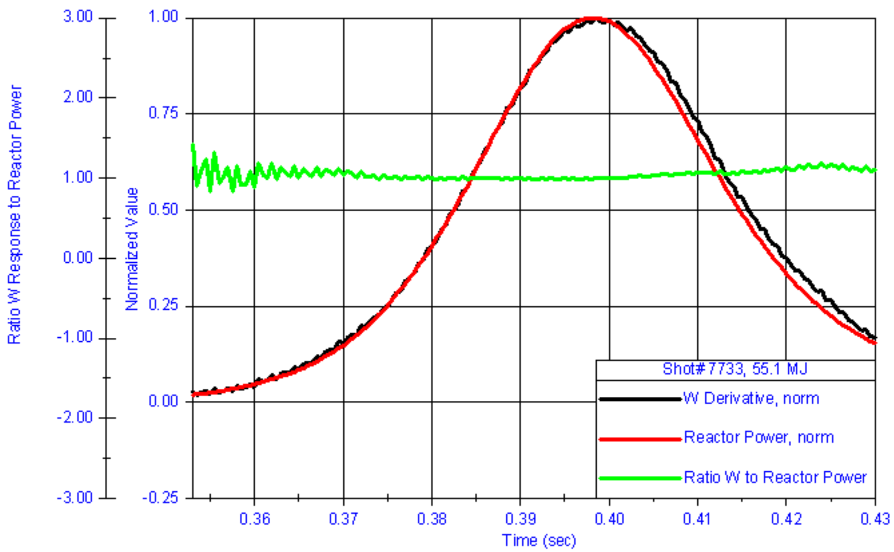
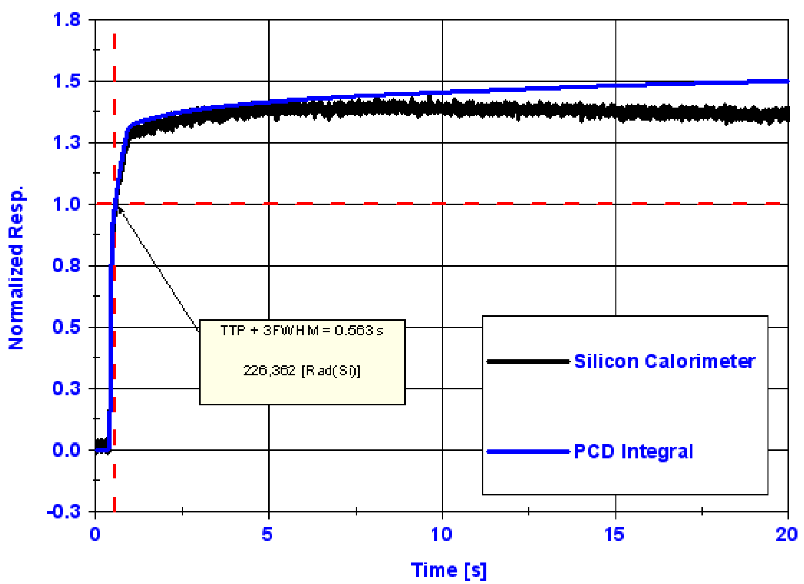
Calorimeter Late-time Agreement

- For calorimeter response, excellent early-time agreement
- Late-time response shows thermal loss in competition with delayed gamma delivery



W-calorimeter derivative vs SPND



Si-calorimeter vs PCD integral

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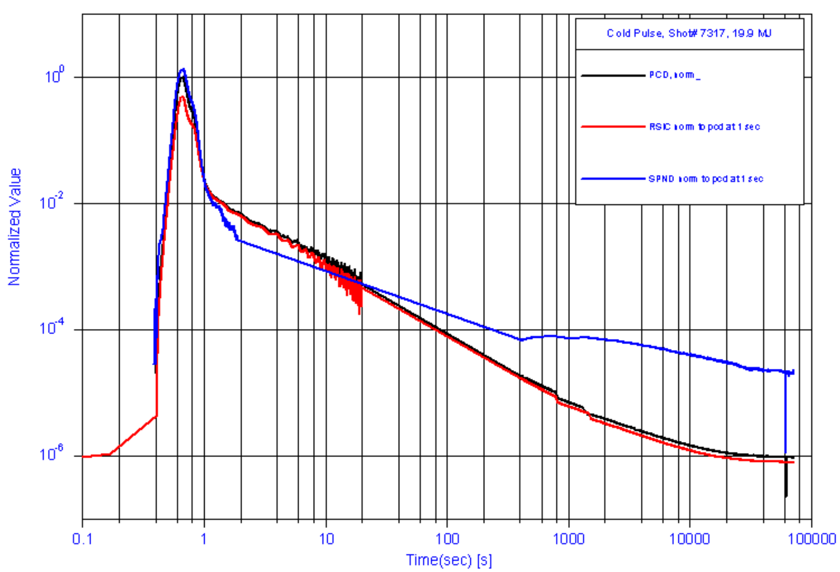
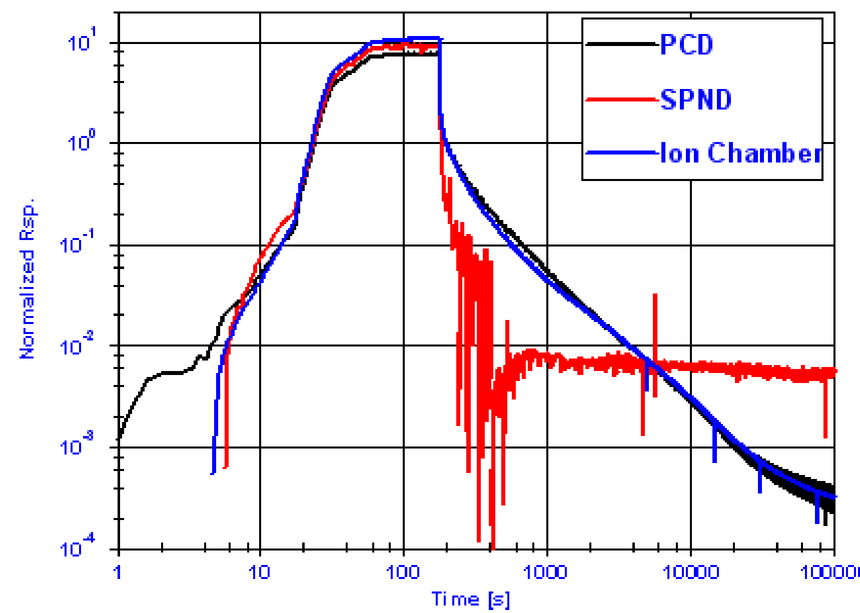
Late-time Dosimeter Comparison

- Late-time comparison shows effect of mixed field response and changing n/γ environment
- Dosimeter normalization effect seen
- Activation gammas may explain deviation from $t^{-1.2}$ decay



19-MJ Pulse



300-MJ 200-s SS

Conclusion

- Early-time dosimeter shapes in good agreement
- Normalization of detector response problematic due to mixed field response
- Detector relative response shifts (early/late) due to changing n/γ environment
- High fidelity calculations needed of time- and energy-dependent reactor field to help resolve issues
- Recommendation – use Si calorimeter for early-time normalization and PCD for temporal shape
- Caution – do not trust simple interpretation of late-time active-dosimeter response

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Recent Publications/Presentations

- P. J. Griffin, S. M. Luker, D. B. King, K. R. DePriest, R. J. Hohlfelder, and A. J. Suo-Anttila. "Diamond PCD for Reactor Active Dosimetry Applications." *IEEE Trans. on Nucl. Sci.*, vol. 51, no. 6, pp. 3631-3637, December 2004.
- K. R. DePriest and P. J. Griffin. "Neutron Contribution to CaF_2:Mn Thermoluminescent Dosimeter Response in Mixed (n/γ) Field Environments." *IEEE Trans. on Nucl. Sci.*, vol. 50, no. 6, pp. 2393-2398, December 2003.
- P. J. Griffin, S. M. Luker, P. J. Cooper, D. W. Vehar, K. R. DePriest, and C. V. Holm. "Characterization of ACRR Reference Benchmark Field." In *Reactor Dosimetry in the 21st Century: Proceedings of the 11th International Symposium on Reactor Dosimetry*. Jan Wagemans, Hamid Aït Abderrahim, Pierre D'hondt, and Charles De Raedt, editors, World Scientific, Belgium, June 2003 (pp.323 – 331).

- **Presented at the 12th International Symposium on Reactor Dosimetry**
  - P. J. Griffin, S. M. Luker, D. B. King, K. R. DePriest, and P. J. Cooper, "Characterizing the Time- and Energy-Dependent Reactor n/γ Environment."
  - S. M. Luker, K. R. DePriest, P. J. Griffin, D. B. King, G. E. Naranjo, and A. J. Suo-Anttila, "Application of a Silicon Calorimeter in Fast Burst Reactors."
  - S. M. Luker, P. J. Griffin, D. B. King, K. R. DePriest, G. E. Naranjo, "Development of a Silicon Calorimeter for Dosimetry Applications in a Water-Moderated Reactor."
  - K. R. DePriest, "Benchmark Experiments/Calculations of Neutron Environments in the Annular Core Research Reactor."
  - D. King, S. M. Luker, G. E. Naranjo, P. J. Griffin, R. H. Hohlfelder, "Development and Test Program for Radiation Hardened Diamond PCDs,"

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Question/Comments