Investigation of an Alanine-Based EPR Dosimetry System at LLNL

CHRES CONSORTIUM OF HYBRID RESILIENT ENERGY SYSTEMS
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CHRES Technical Forum at NETL

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NUCLEAR SAFETY RESEARCH AND DEVELOPMENT Working Group

ENVIRONMENT SAFETY HEALTH

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Lawrence Livermore National Laboratory

Self - Introduction

Paige Witter

College/University: Colorado State University

Field of Study: Health Physics

Research Advisor: Dr. Brian Champine, LLNL
Dr. Alex Brandl, CSU

Research Location: LLNL/CSU

Project Title: Investigation of an Alanine Based Dosimetry System at LLNL

Jose A. Rosales Mata

College/University: University of Texas at El Paso

Field of Study: Synthetic Organic Chemistry

Research Advisor: Dr. Christopher Colla, LLNL

Research Location: LLNL/UTEP

Project Title: Investigation of an Alanine Based Dosimetry System at LLNL

What you like best about the summer research project/experience:
It has been exciting to be onsite and see first-hand the plethora of jobs, research, and interests that keep the Lab going. People are always happy to talk about what they are working on, and the collaborative environment has helped make connections between groups I would not have expected being able to work with.

What you like best about the summer research project/experience:
I like the several interactions we get to have with the people around us, learn new things as we encounter a scientific problem and help try to understand them. Generally, people here are eager to help you out with anything and encourage questions as they seem fit for incoming interns, and it is very helpful for us as well.

Description of your summer project:
As part of an ongoing project into the development of a prototype nuclear accident dosimeter, the use of alanine-based electron paramagnetic resonance (EPR) dosimetry is being investigated. Between LLNL's ES&H and Materials Science groups, we are working to stand up an EPR dosimetry system to try to expand the neutron spectra and energy ranges that could be used for alanine dosimetry. Research may expand to the impact of chemical treatments on fingernail samples that are used for EPR dosimetry.

Agenda

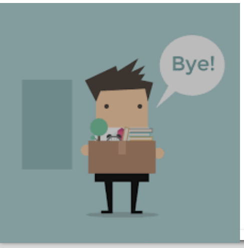
- Background
  - Brief description of need for Nuclear Accident Dosimetry
  - Problem definition and goals
  - Technical background
- Current Work
  - Alanine EPR Dosimetry
- Future Work
- Summary

Nuclear Accident Dosimetry

- A criticality event would stop work and shut down the facility – a potential significant impact to DOE programmatic missions and schedule.
- Radiation doses for workers nearby may far exceed the range of standard dosimeters.
- Nuclear Accident Dosimeters (NADs) fill the gap; can measure much higher doses.
- Dose determinations identify workers that need medical care.
- Data obtained may also be useful for the subsequent investigation of the event.

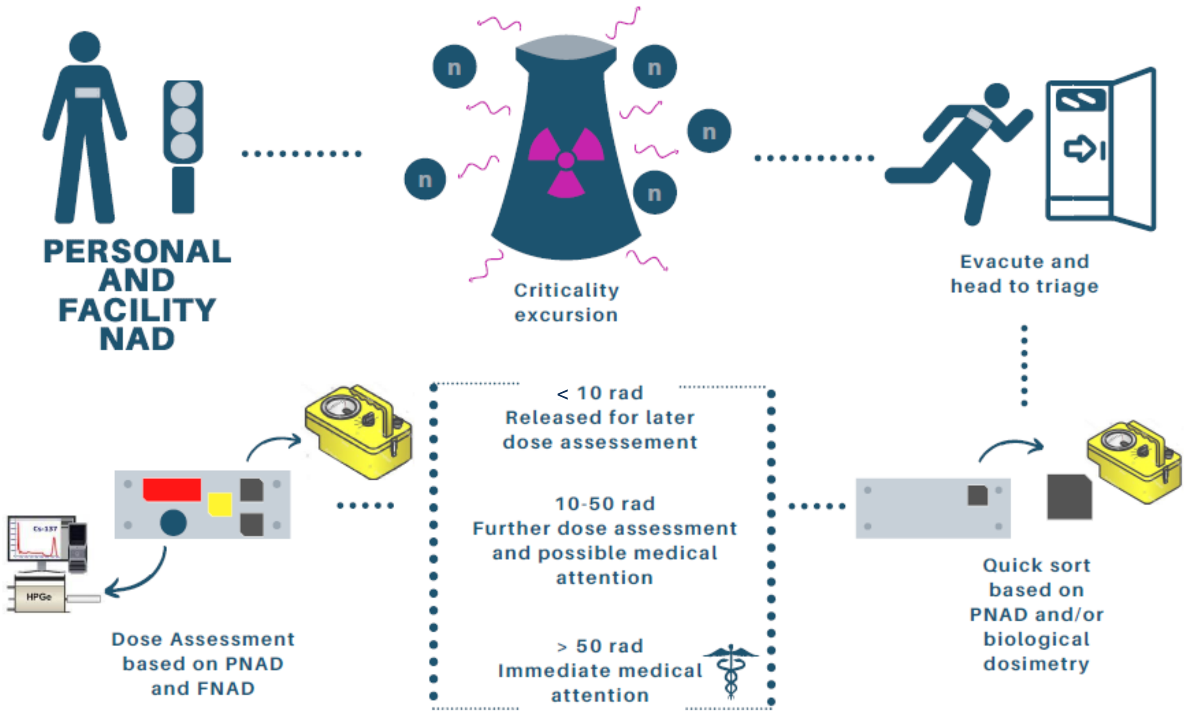
Nuclear Accident Dosimetry

- Intent: We are developing and testing new NADs to improve safety and enable standardization across the DOE/NNSA complex.
- Problem Definition:
  - ~30% of NADs did not meet DOE neutron dose standards during recent blind testing of a simulated accident using reactor bursts.
  - Most NADs were developed in the 1960s and 1970s and do not take advantage of advancements in detection technology.
  - Each DOE facility has its own custom device with limited resources to maintain proficiency or make improvements to the design.
  - Small criticality dosimetry groups mean large reliance on skill and expertise from workers vacating the position and/or close to retirement.

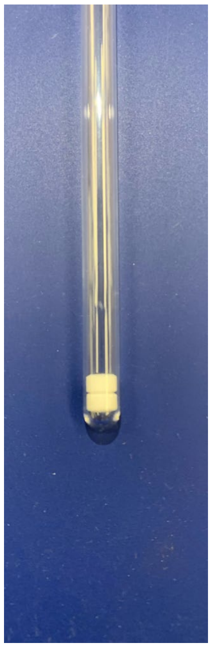


Ref. 1, 2, 3, 4

NAD Procedure



Project Goals

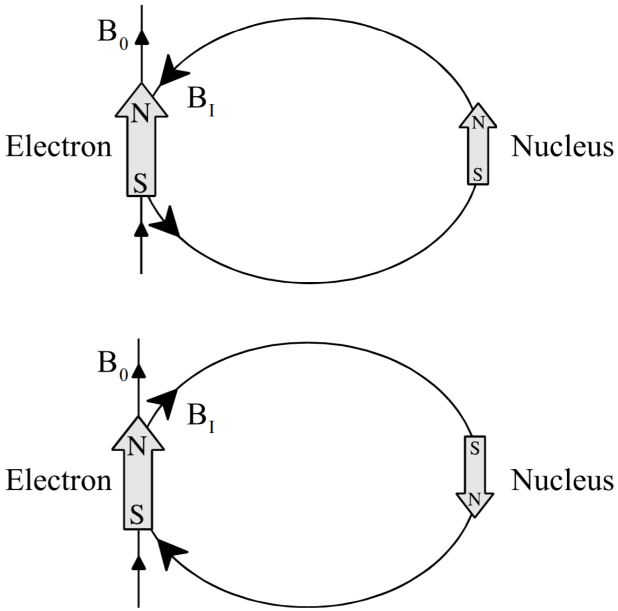


- Investigate alanine-based Electron Paramagnetic Resonance (EPR) as a potential method of neutron dosimetry.
  - Obtain alanine EPR reference spectra and dose-response curves in multiple radiation sources.
    - Gamma sources:
      - Co-60, Cs-137
    - Neutron sources:
      - Cf-252, Deuterium-Tritium neutron generator
  - Irradiate alanine in “unknown” neutron sources.
    - Nuclear accident dosimetry intercomparison exercises (Armed Forces Radiobiology Research Institute [AFRRI]).
  - Enhance neutron sensitivity and expand energy response using response-enhancing converters and/or dopants for alanine.

Universal Nuclear Accident Dosimeter

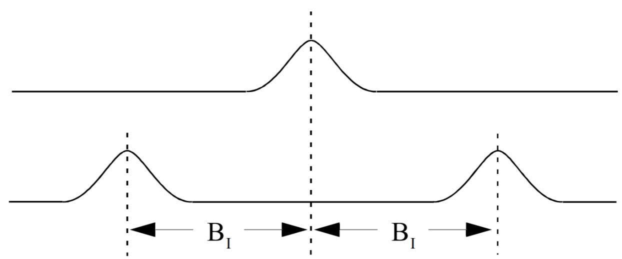
- Benefit: Improved support and response to a criticality accident.
  - Provide better triage to those exposed in a criticality accident and allow for quicker and better medical treatment.
  - Expedite responses in a criticality accident.
  - Expand collective knowledge and proficiency.
  - Improve performance to meet DOE requirements.
  - Standardize use of a single NAD across the DOE complex.
  - Reduce cost by streamlining fabrication.

Electron Paramagnetic Resonance (EPR) Spectroscopy



- Nuclei of atoms in a molecule often have a magnetic moment producing a local magnetic field at the electron.
- Interaction between electron and nuclei is called hyperfine interaction.
- Hyperfine interactions give identity, number of atoms and their distances from unpaired electrons.
- As number of nuclei gets larger, number of signals increases exponentially (2^N).

For a spin (I = ½) nucleus such as a hydrogen nucleus, we observe that a single EPR absorption signal splits into two signals which are each B_I away from original signal.

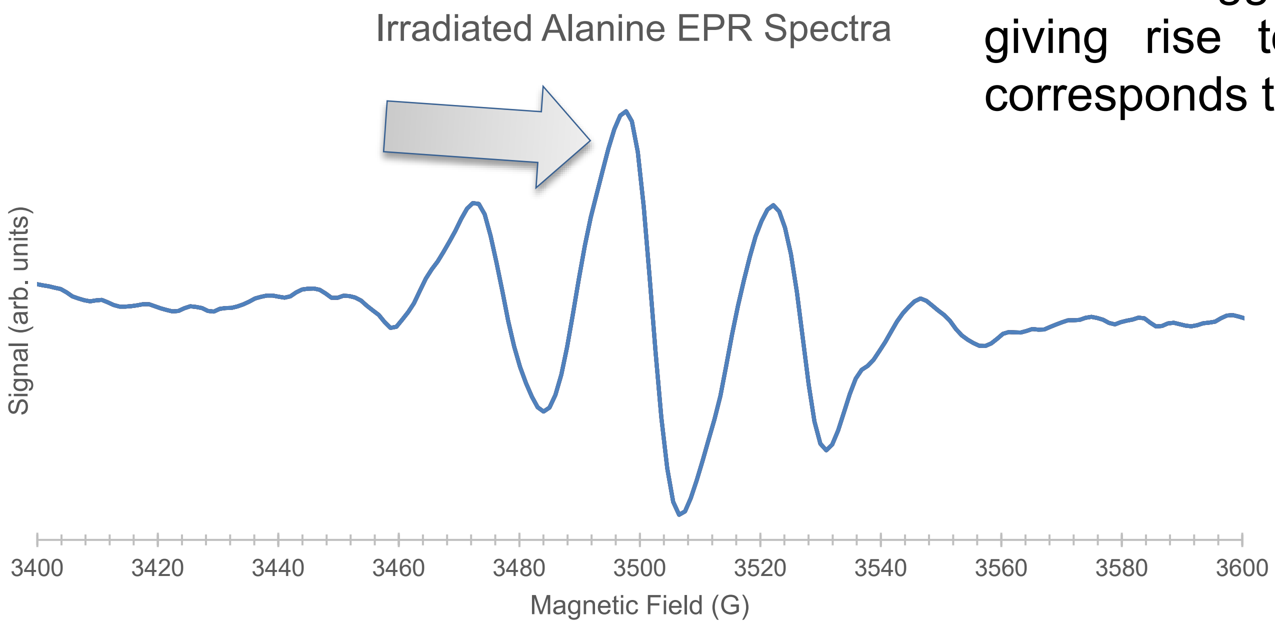


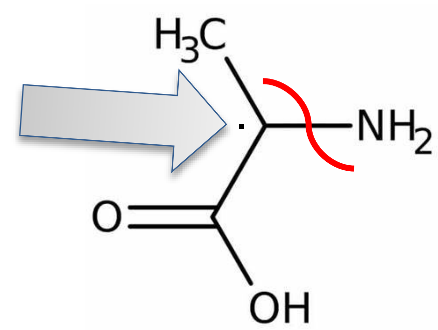
EPR Measurement of alanine pellets

Ref. 5,6,7

Alanine Irradiations in Co-60

Interaction of ionizing radiation with alanine triggers series of reactions giving rise to radicals. Most stable corresponds to breaking of NH_2 group.





- Presence of an unpaired electron with central carbon assigns paramagnetic properties and is responsible for central line of spectrum.
- Adjacent lines are due to hyperfine interactions of unpaired electron with four hydrogen atoms present in the radical CH_3–C•H–COO of alanine (2nI + 1) where n = 4.

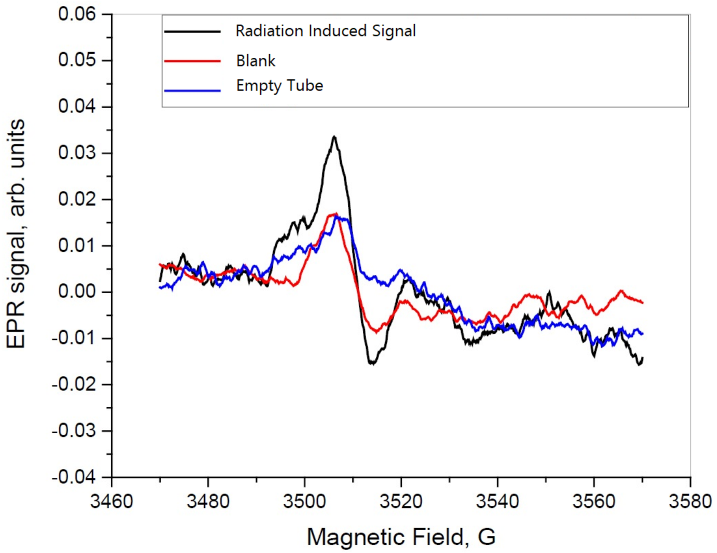
Ref. 8

Alanine Electron Paramagnetic Resonance

- Number of paramagnetic centers produced by ionizing radiation is related to the absorbed dose in the target material.
  - Measured as amplitude of primary signal peak.

- Alanine: IAEA-recognized secondary reference for high dose gamma dosimetry.

- Resources required:
  - EPR instrument (lab-scale or portable).
  - Specialized personnel.



Ref. 9,10

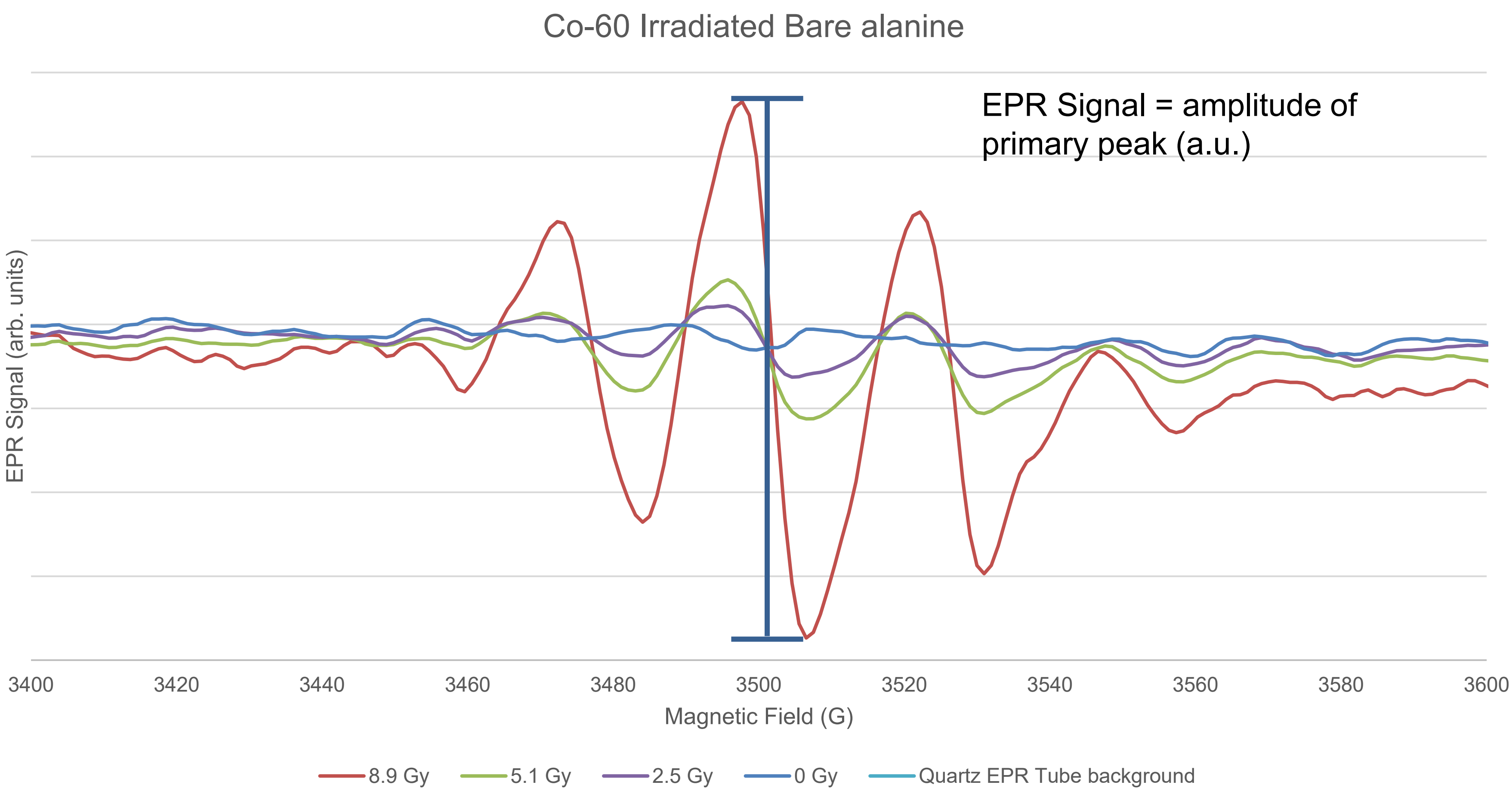
Alanine Electron Paramagnetic Resonance

- Advantages:
  - Tissue equivalence
  - Independent dose rate response to radiation energy
  - High stability of paramagnetic centers/ free radicals: small signal fading over time
  - Direct dose measurement using a dose-response calibration curve (known irradiation field) or can use multiple filters or dopants to cover neutron energy spectrum
  - Fast measurements with a single machine
  - Consolidated centers of expertise possible

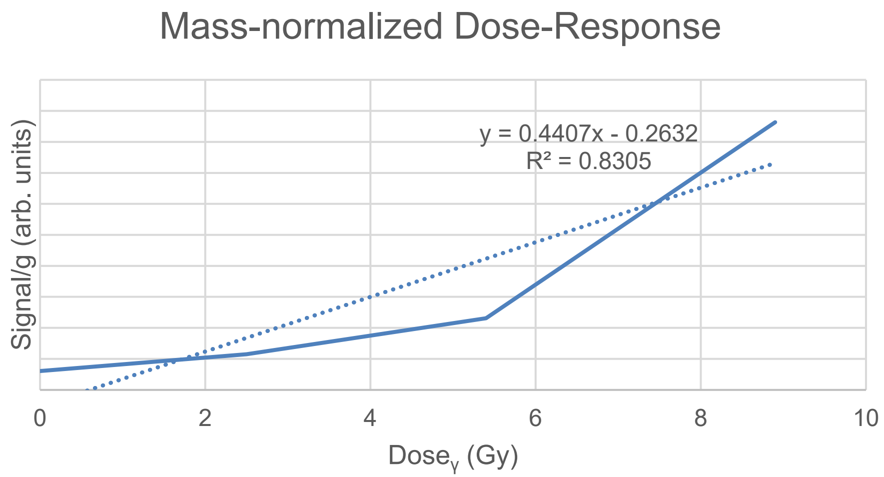
- Disadvantages:
  - More sensitive to gammas and high energy neutrons than thermal neutrons
    - Additional filtering or dopants required to cover expected neutron spectrum
  - Less common technique in nuclear accident dosimetry
    - Requires a new machine, procedures, additional expertise and training
  - Pure alanine Lower Limit of Detection (LLD) higher than 10 rad (0.1 Gy) criterion in DOE-STD-1098-2017, *Radiological Control*.

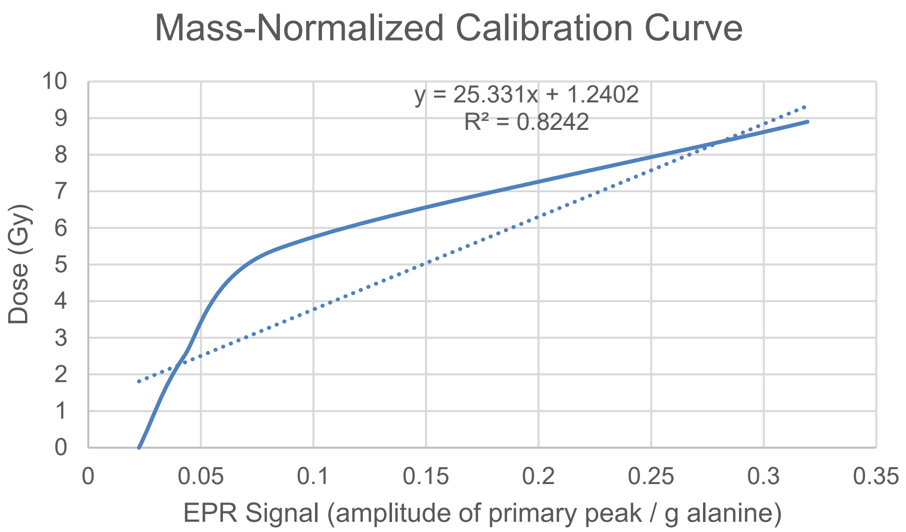
Ref. 11,12,13

^{60}Co Dose-Response Curve



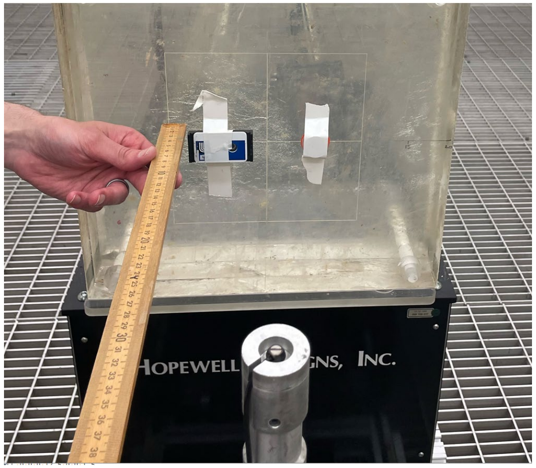
^{60}Co Unknown Dose Determination





Alanine Irradiations in additional fields

- Dose-response curve of bare alanine in difference reference spectra.
  - Gamma:
    - ^{60}Co
  - Neutrons:
    - ^{252}Cf
    - DT neutron generator



RCL ^{252}Cf irradiation of current NADs

- Steps:
  - Ongoing repair of LLNL Elexsys E500 EPR Spectrometer.
  - Obtaining benchtop E1000 MS500 ESR Spectrometer.


EPR Measurement of alanine pellets

Alanine Irradiations in additional fields

- Expand neutron spectrum information.
  - Increase thermal neutron sensitivity using materials that convert neutrons to particles that alanine is more sensitive to:
    - $^{6}Li + n \rightarrow \alpha + ^{3}H$
    - $^{113}Cd + n \rightarrow ^{114}Cd^{*} \rightarrow ^{114}Cd + \gamma$
    - $^{10}B + n \rightarrow \alpha + ^{7}Li$
    - plastic radiator
  - Collaboration with Naval Dosimetry Center, build on ongoing work there.

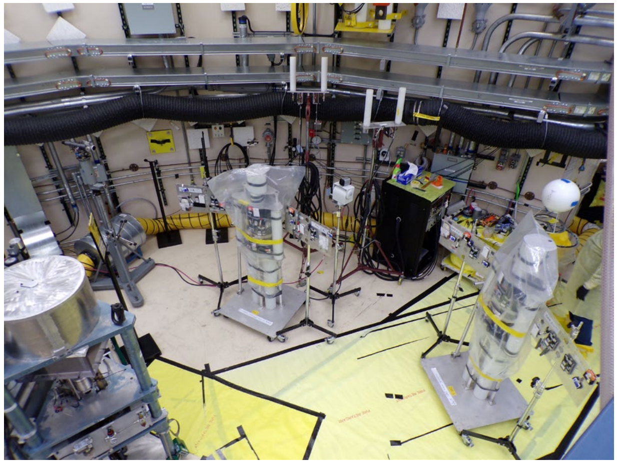
Alanine in quartz EPR tube for analysis

- Steps:
  - Prototype holder for filters:
    - Ten built, 3-D print or fabricate additional as needed.
  - Irradiations and dose-response curves in known fields:

Alanine Irradiations in Additional Fields

- Test converter NAD in "unknown" spectra.
  - Deploy filtered alanine in AFRRI characterization exercise, isolate neutron dose and parts of spectrum.
- Steps:
  - Calculate neutron dose and parts of spectrum using photon dosimeters to subtract photon contribution.
  - Use known response curves to estimate dose in unknown fields.

BOMAB phantoms with current NADs



NAD Irradiation at GODIVA-IV (2022)

Summary

- Collaboration with LLNL ES&H, Materials Science Division on EPR spectrometry of alanine.
  - Expansion of EPR spectrum analysis options.
- Investigation of converters to expand alanine's neutron-energy response range.
- Development of measurement and analysis technique at LLNL.

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Questions?

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