Latest Efforts of an International Collaborative Team Studying Differences in Effects for Gamma, E-beam and X-ray Irradiation Applications

April 14, 2026

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Aerial-CRT, FRANCE

TEAM NABLO @PNNL

Aerial

U.S. DEPARTMENT OF ENERGY BATTELLE

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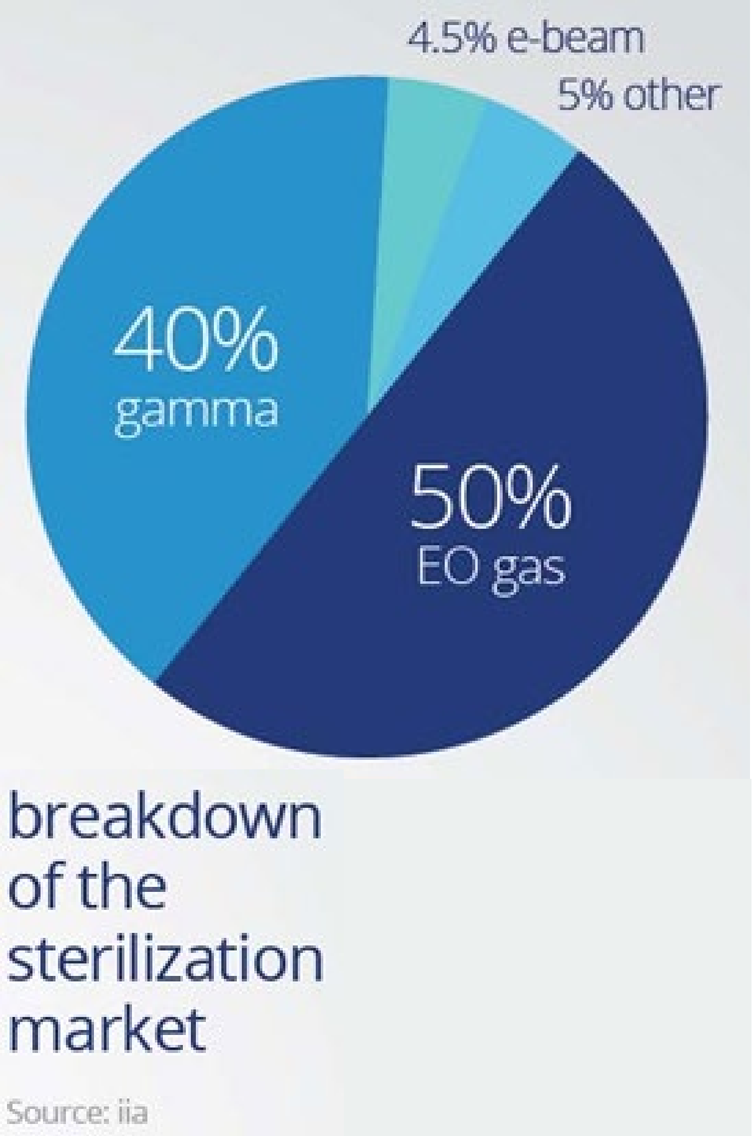
PNNL-SA-222021

CIRMS 2026 — Need

Annual Meeting · April 13–15, 2026

Progress in **Dosimetry for Low Energy (<1MeV), Low Dose (~1kGy) Electron Beam processing** is currently blocked because we lack **tools to quantify dose, account for energy, and ensure consistent treatment**, which prevents **efficient 1) sterilization of food/medical devices, 2) phytosanitary fruit/veg irradiation**.

Sterilization of Single Use Medical Devices



- Ethylene oxide (EO) gas (~50%)
  - Challenged by concerns over toxicity, throughput
- Cobalt-60 gamma radiation (~40%)
  - Challenged by regulatory requirements, supply chain
- Electron beam (eBeam) and X-ray (~5%)
  - Appealing machine-based options for sterilization
  - Challenged by availability, knowledge gaps

https://gipalliance.net/wp-content/uploads/2013/01/GIPA-WP-GIPA-iia-Sterilization-Modalities-FINAL-Version-2017-October-308772.pdf

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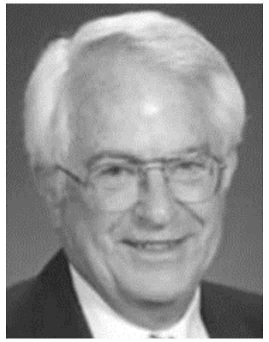


Samuel V. Nablo 1931-2018

2018

G vs EB vs XR for blood collection tubes/sets (2 products, 6 polymers)

BD SteriTek Pacific Northwest TEXAS A&M UNIVERSITY

2019

stryker

G vs EB vs XR for orthopedic surgery (3 products, 9 polymers)

2020

Aix*Marseille université Socialement engagée

SARTORIUS

G vs EB vs XR for multilayer biopharma film

Dose rate studies

iba Aerial

2021

BAYER Boston Scientific MilliporeSigma PALL

Repackaging studies G vs EB vs XR for relevant doses

2022

EB energy studies low/medium/high energy

PUFFin Dev1.0 E-BEAM TERUMO

2023

FDA

Extractable and leachables

2024

Chemical filled syringes

Stanford HEALTH CARE

Kroc, et al., Fermilab. “Accelerator-driven Medical Sterilization to Replace Co-60 Sources”. (2017). https://lss.fnal.gov/archive/2017/pub/fermilab-pub-17-314-di.pdf

IAEA (2019) Consultancy Meeting on ““Radiation effects on polymer materials”, 25-28 Nov. 2019, Vienna. http://www-naweb.iaea.org/napc/iachem/working_materials/IAEA Consultancy Meeting Final Report.pdf

Examples of Products Tested and Key Properties to Consider

L. Fifield, et al. *RPC* 2021 109505
L. Fifield, et al. *RPC* 2021 109282

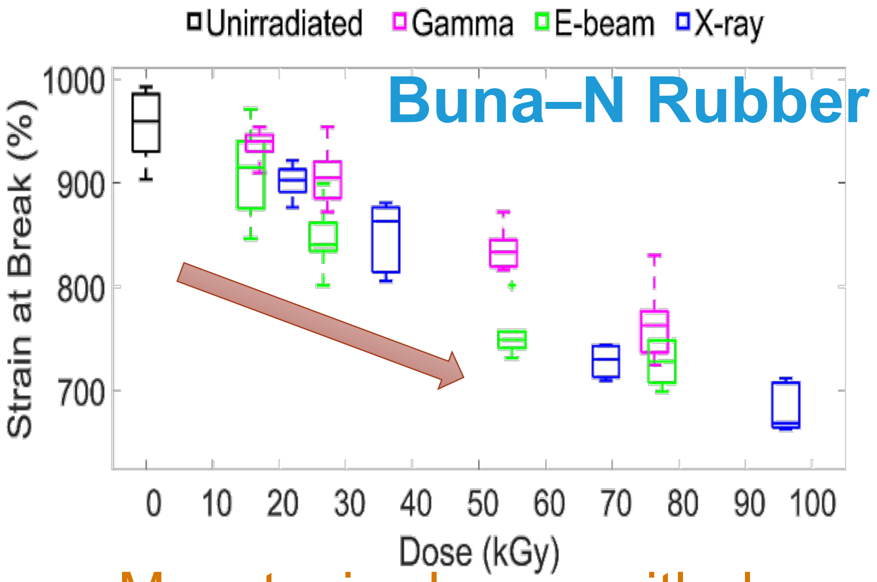
D. Li, et al. *RPC* 2024 111971 D. Li, et al. *RPC* 2025 112188

M.K. Hasan, et al. *PDS* 2024 110677

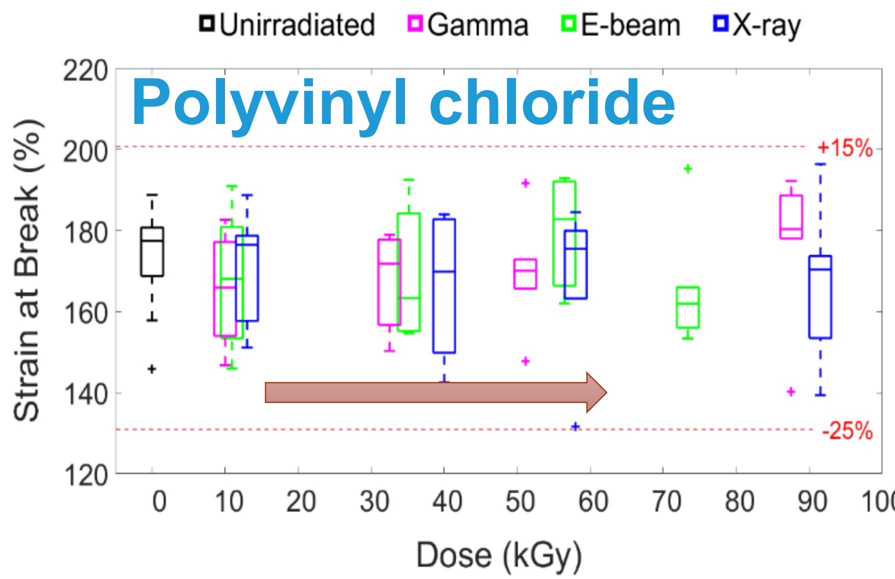
B. Krieguer, et al. *RPC* 2024 111607
Y. Ni, et al. *npj Mater Degrad* 2023

- **Functional**
  Device/component operation
- **Visual**
  Color, Clarity
- **Physical**
  Mechanical, Thermal, Surface
- **Chemical**
  Extractables & Leachables
- **Biological**
  Biocompatibility & Toxicology

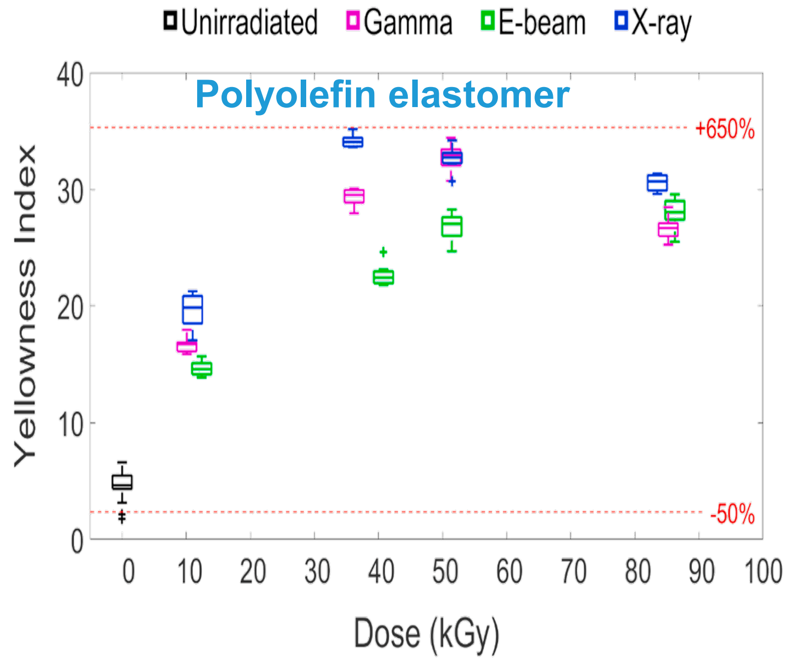
Types of Dose Effects Observed



Monotonic change with dose



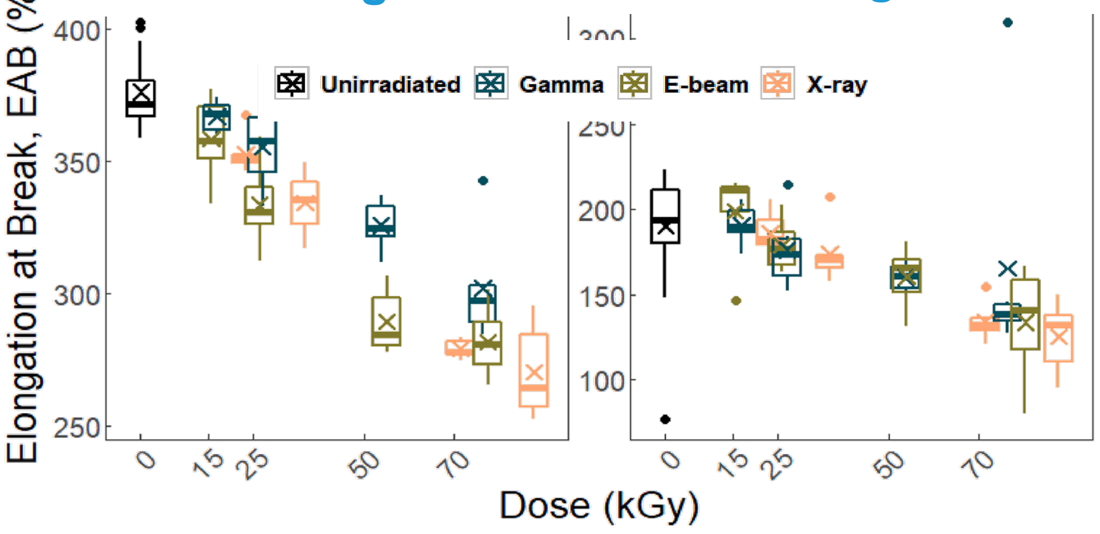
Little effect with dose



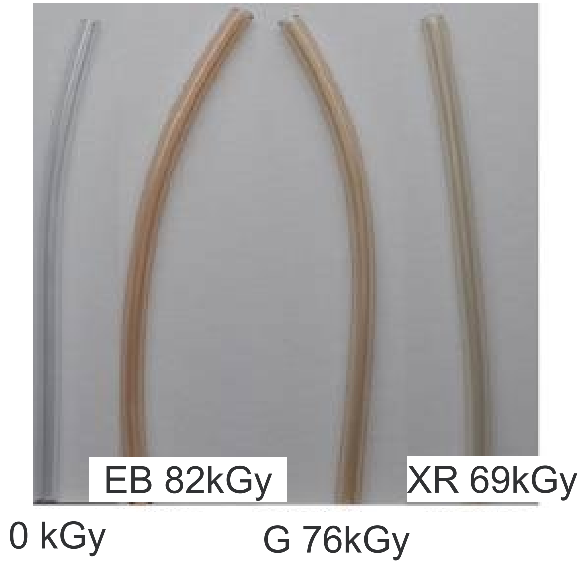
Variation with radiation source

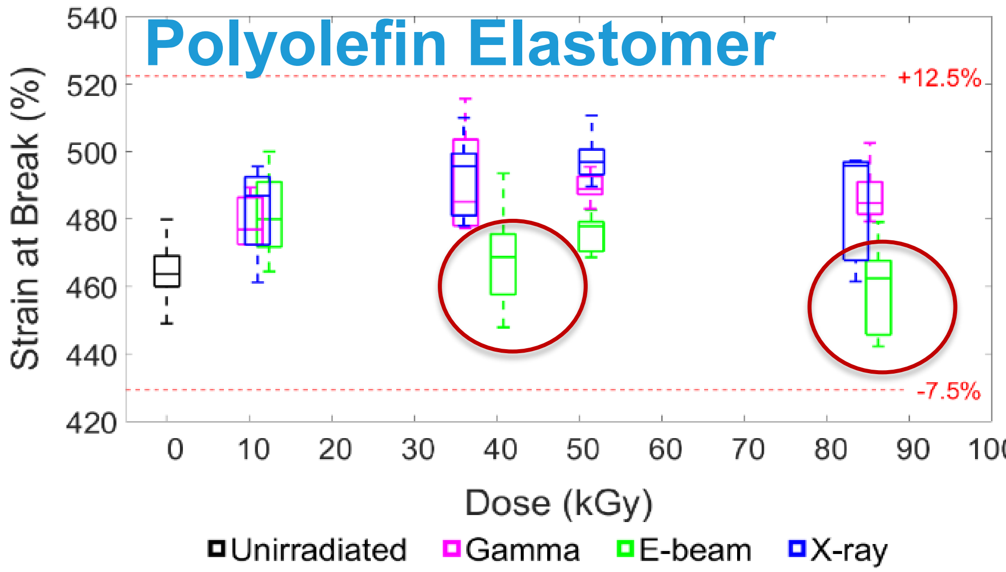
Black O Ring Buna N

Red O Ring Silicone



D. Li, et al. *RPC* 2025 112188





L. Fifield, et al. *RPC* 2021 109505

stryker BD

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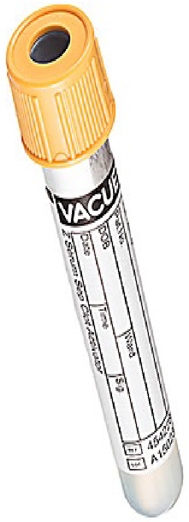
Influence of Radiation Source on Clinical Results of Filled Tubes

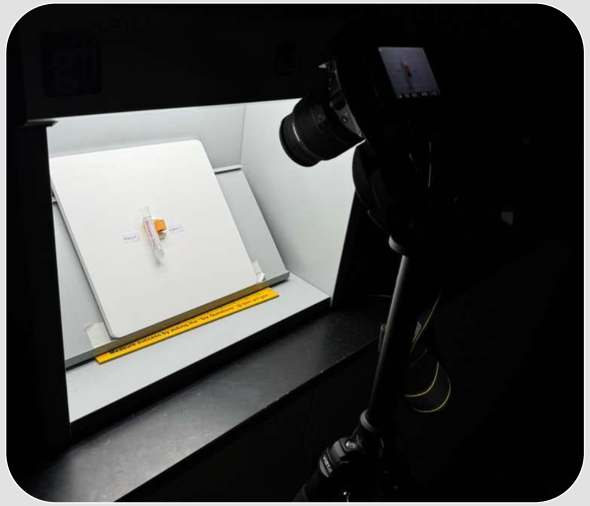
Material and blood analyte analysis of effects of Gamma, eBeam, X-ray on evacuated blood collection tubes with functional additives (Serum Separator, Clot Activator)



Irradiation

- Gamma, eBeam, X-ray
- 0, 25, 50, 75 kGy
- Gold Cap tubes

Greiner Bio-One Filled Tubes Characterization



Color

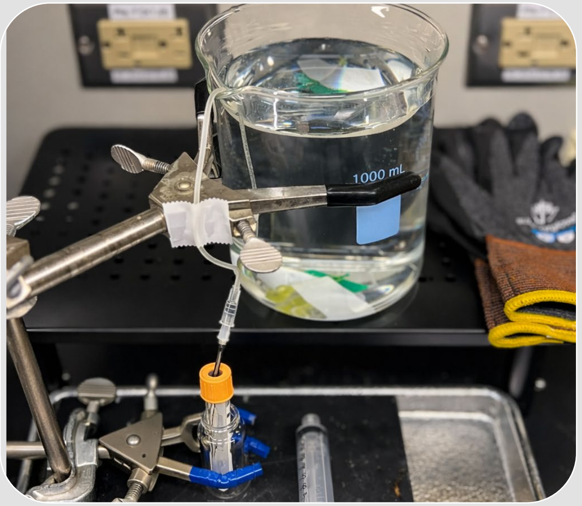
ASTM E313
ASTM D2244

- Yellowness
- Total Color

Hardness

ASTM D2240

- Shore D
- Shore M



Functional

ISO 6710

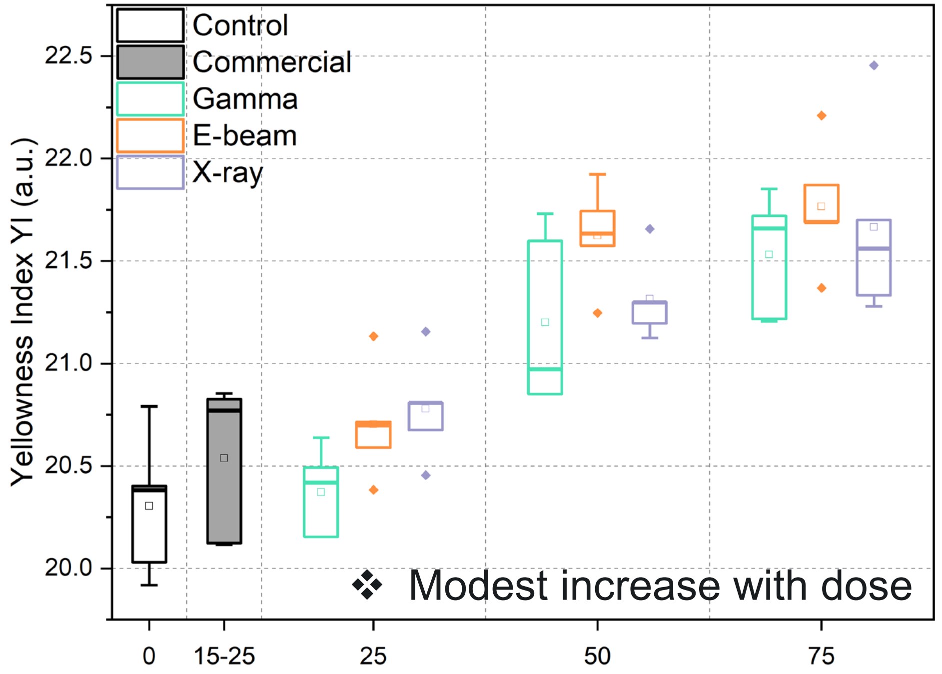
- Draw Volume
- Leak Test
- Centrifuge Integrity

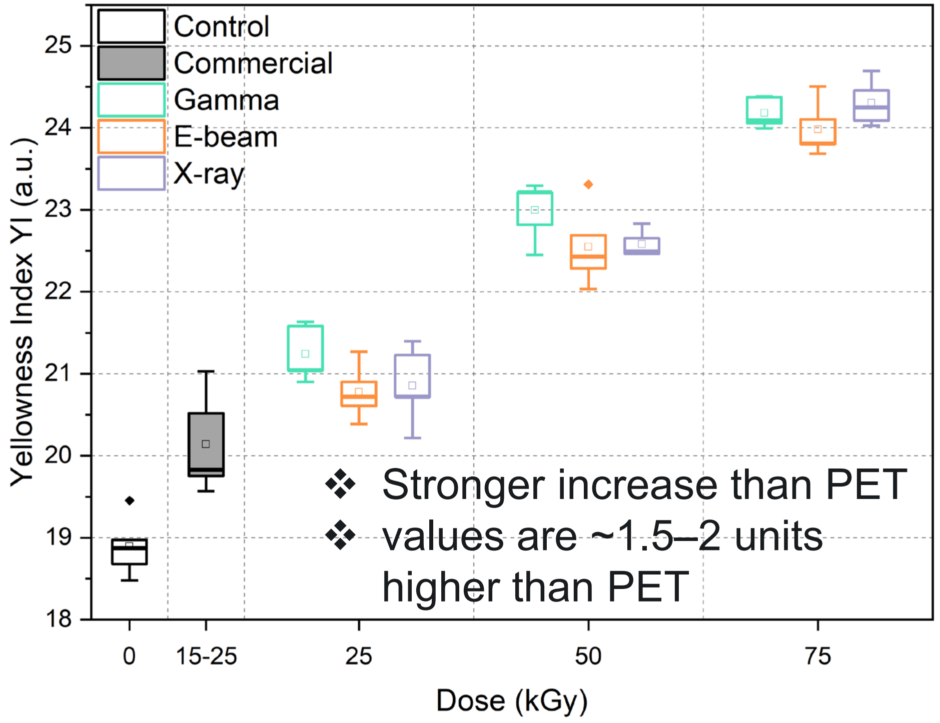
Thermal

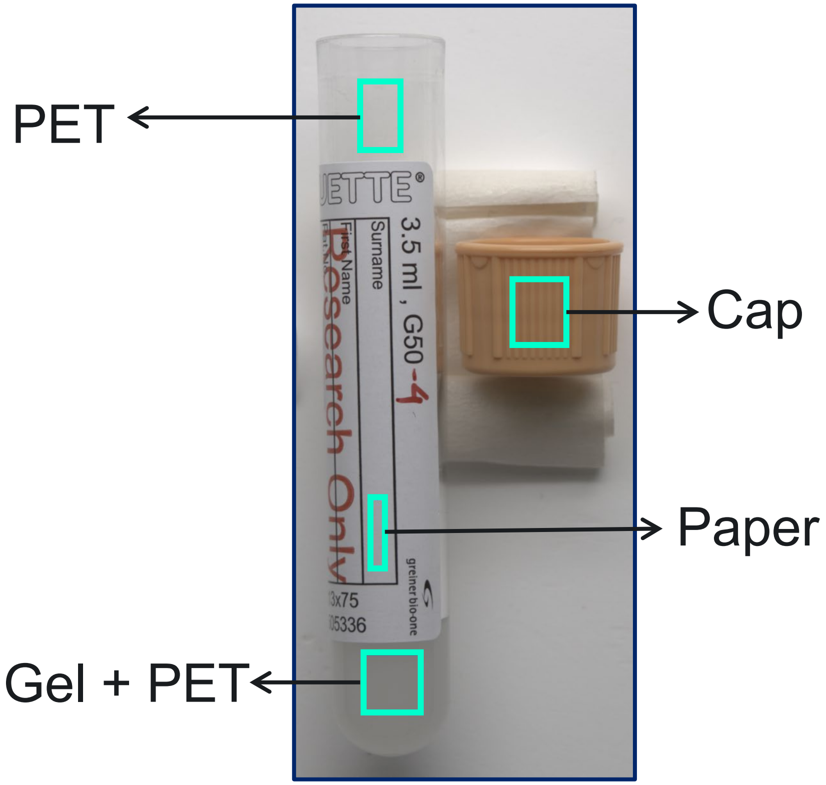
ASTM D3418

- Melting
- Crystallinity
- Glass Transition

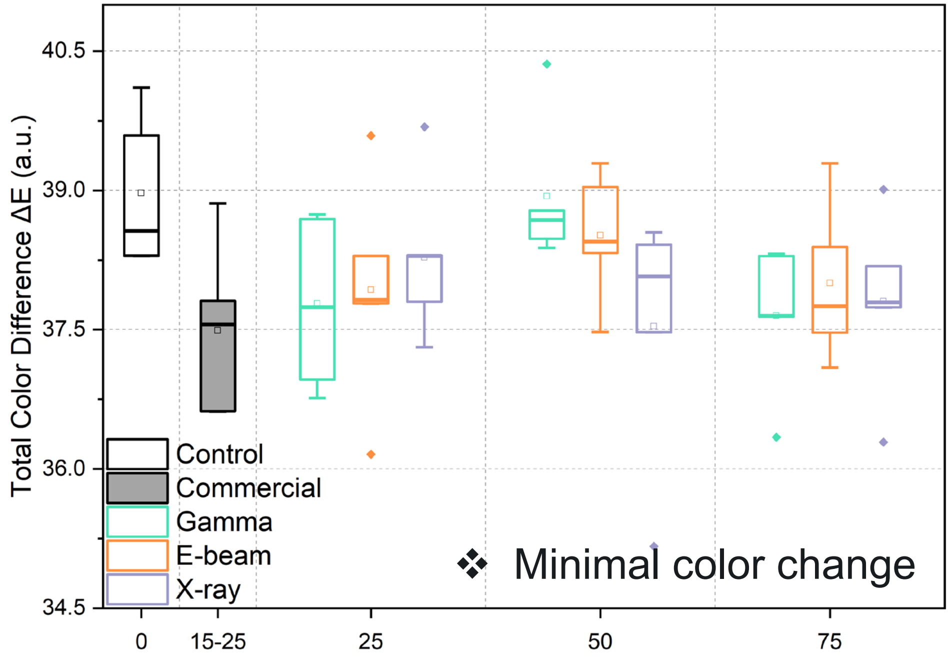
Color Measurement

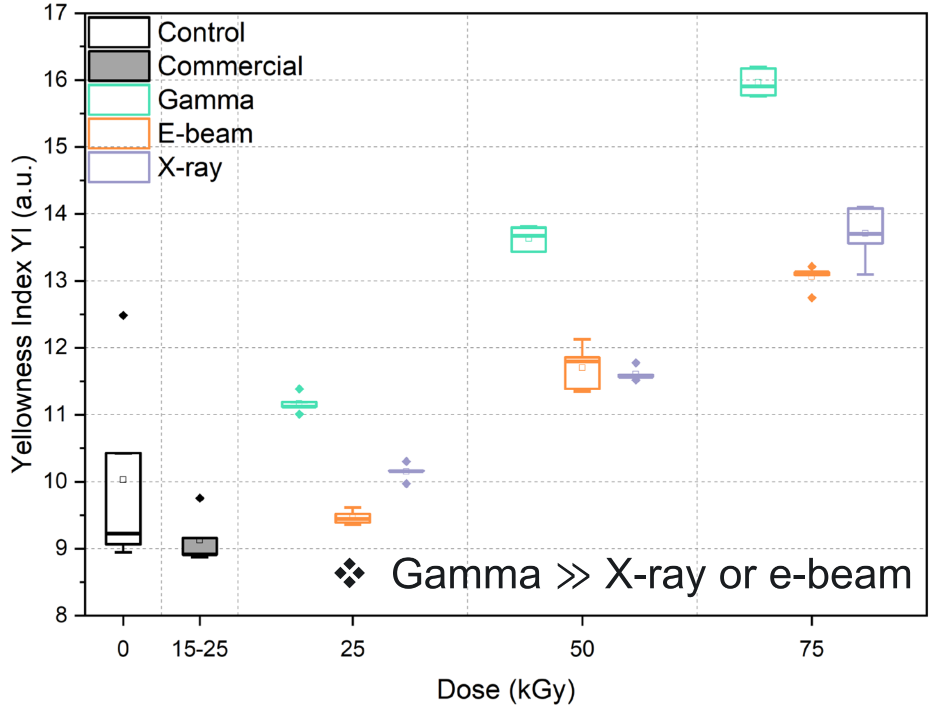




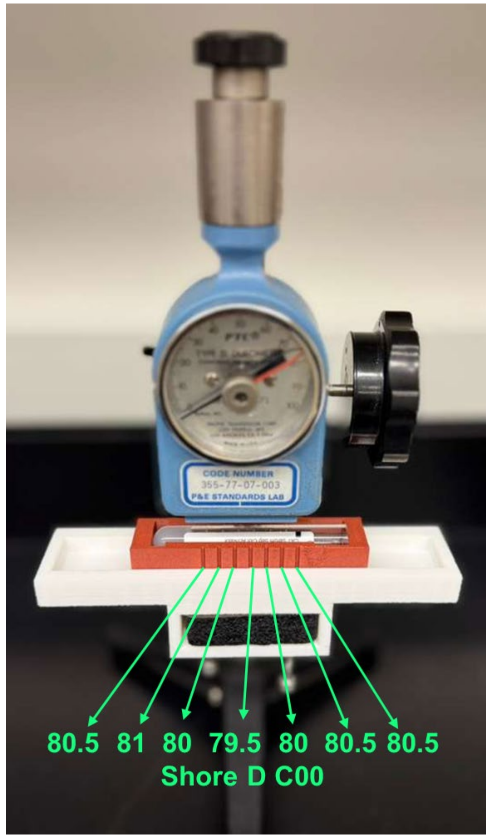


❖ Comparative Sensitivity to Yellowing
Paper ≫ Gel + PET > PET ≫ PE Cap



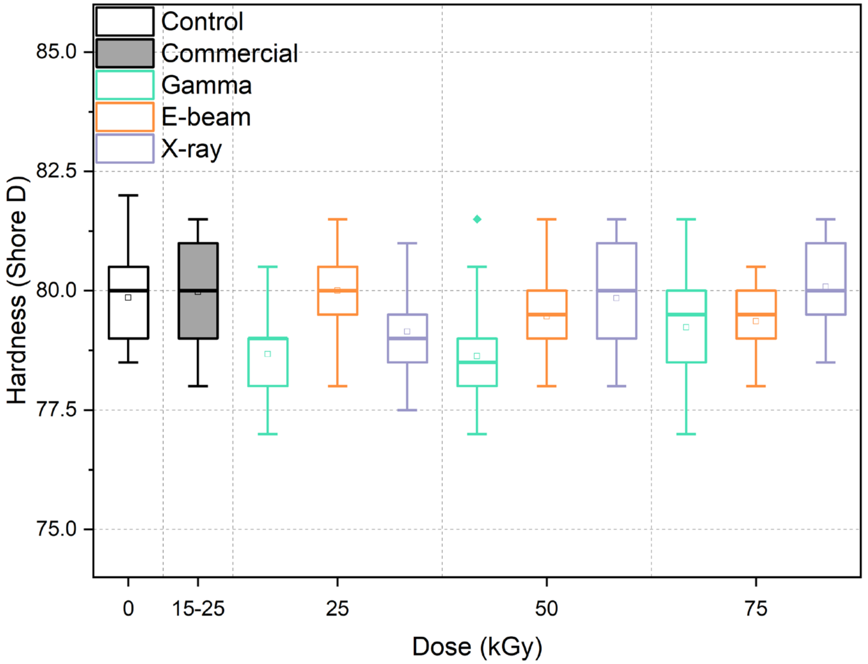


Hardness Measurement



PET Tube (Shore D):

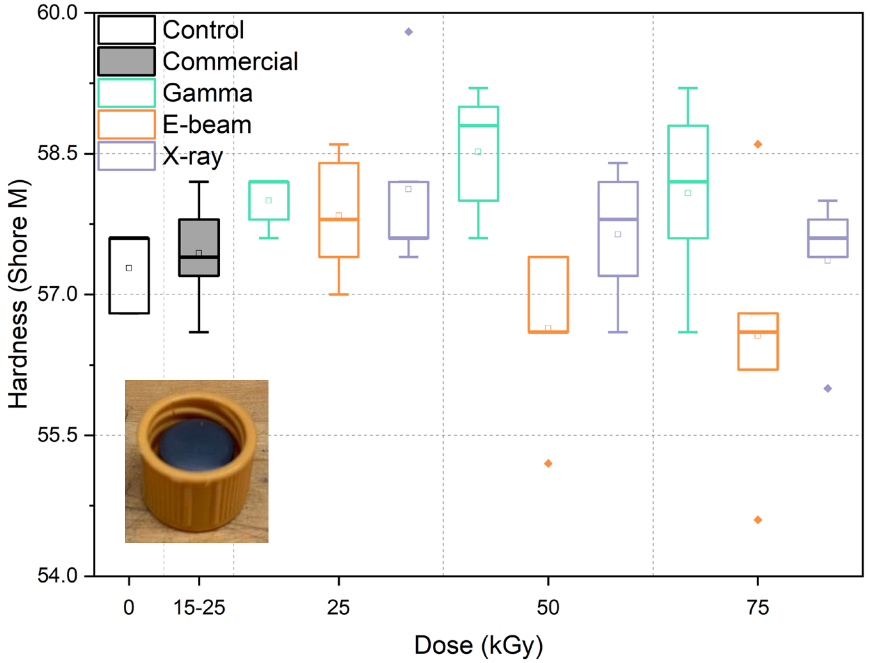
- ❖ 7 measurements per tube
- ❖ 5 replicate tubes



- PET maintains bulk stiffness across the sterilization dose range.
- Differences between sources are small and overlapping, indicating no strong source dependence.

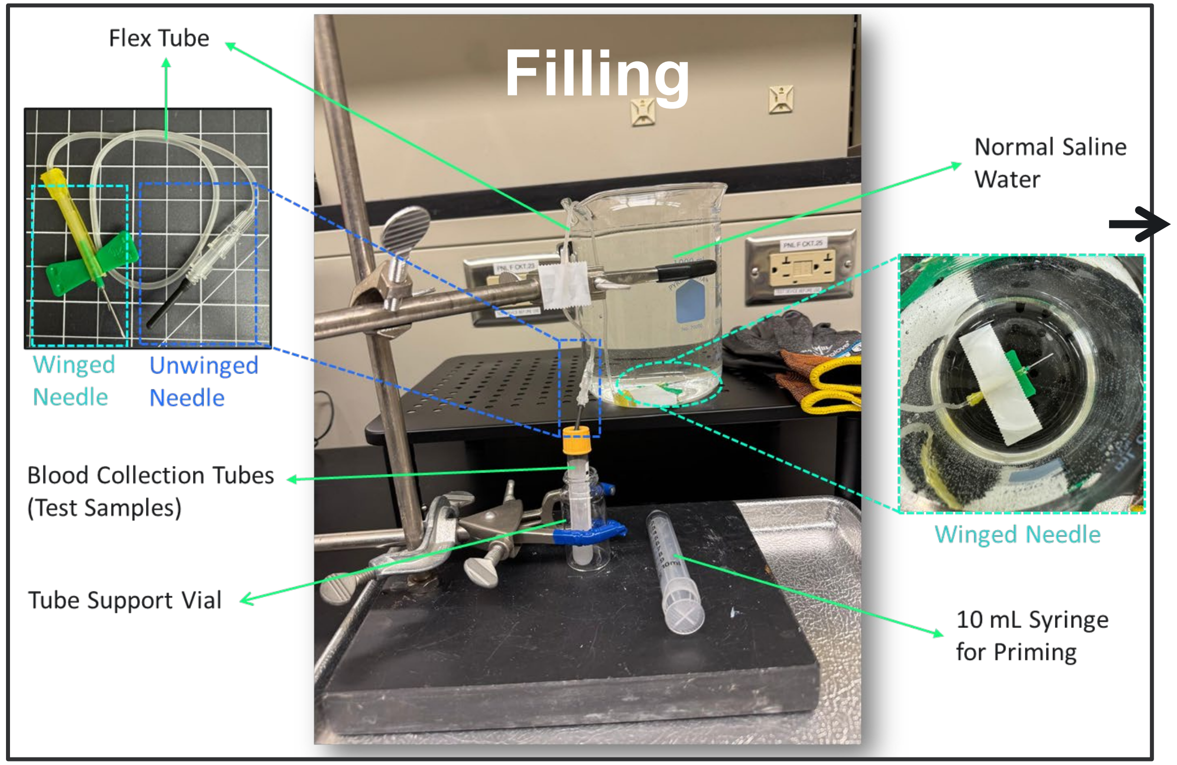
Rubber Stopper (Shore M):

- ❖ One hardness value per stopper
- ❖ 5 replicate stoppers per condition



- Radiation source influences rubber hardness slightly, with e-beam associated with modest softening, but the magnitude of change remains small.
- Occasional outliers (e.g., low E-beam values) likely reflect material or formulation variability, not systematic radiation damage.

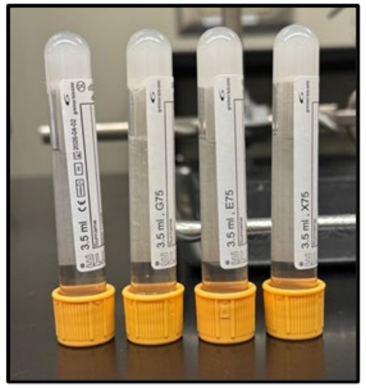
Draw Volume Test & Seal Integrity (Leak) Test



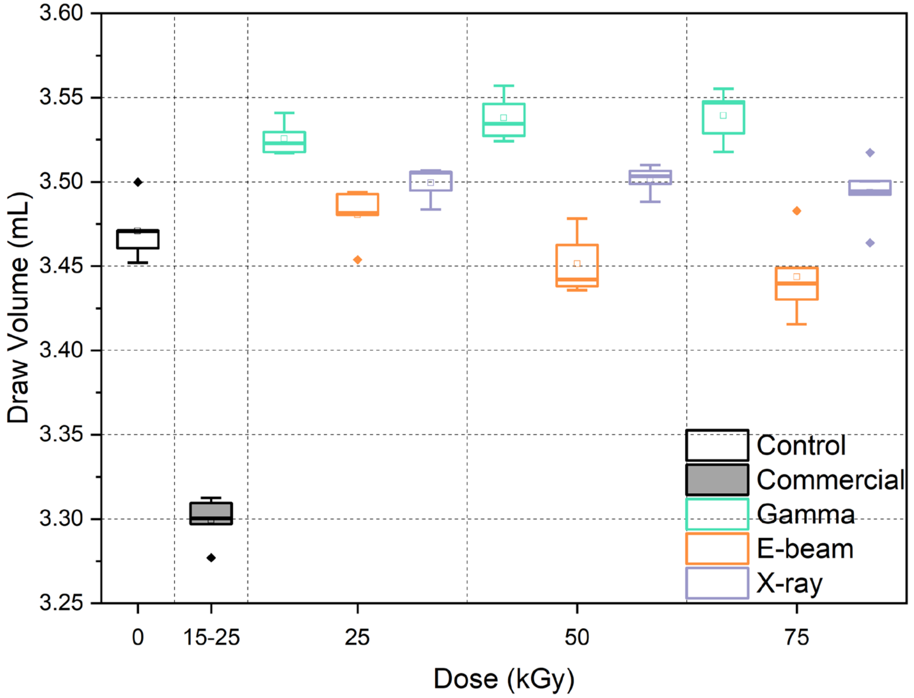
- Water fill
- Inversion test (5x + 10s hold)
- Post-Centrifuge Visual Inspection
- Post-Centrifuge Inversion

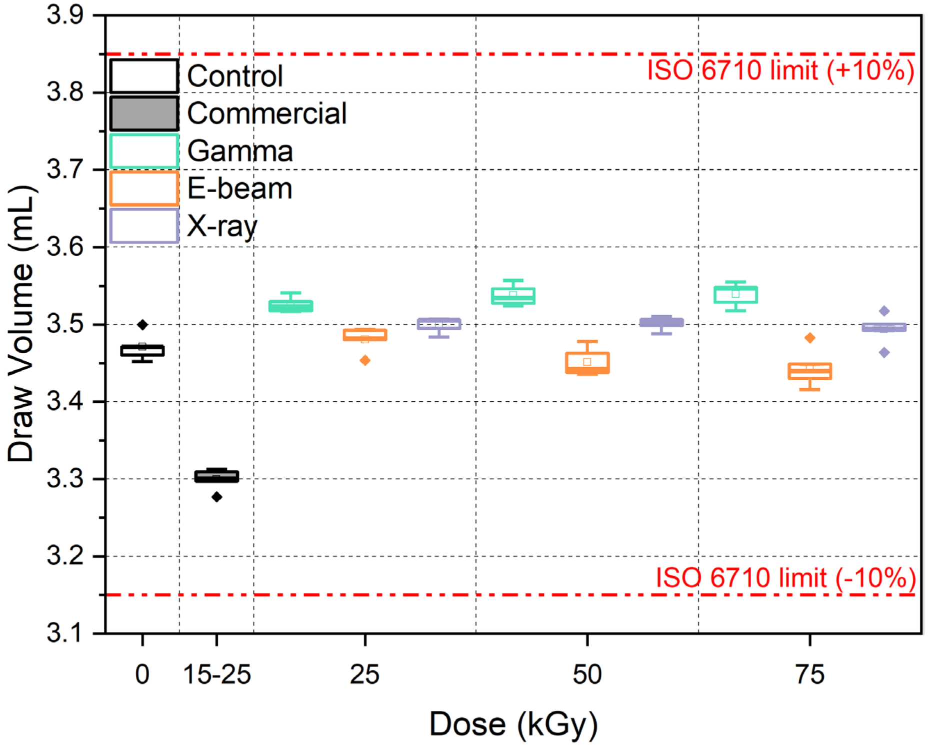
→ All tubes passed, no failures

Seal Integrity



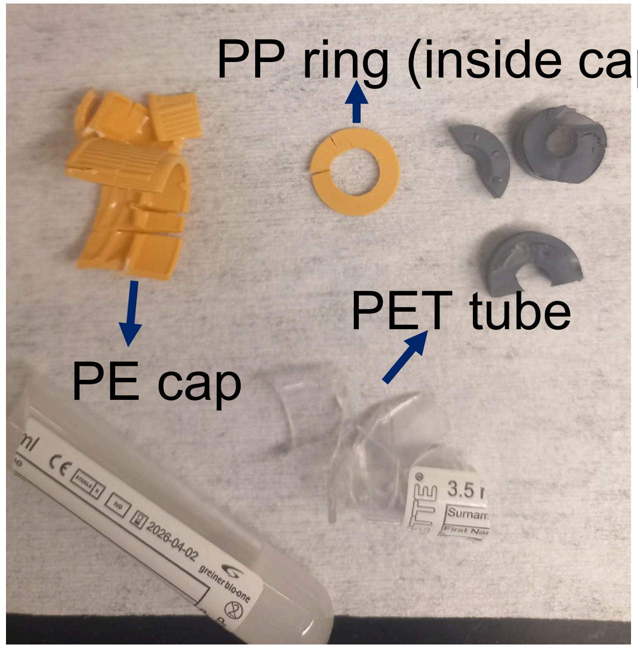
Draw Volume

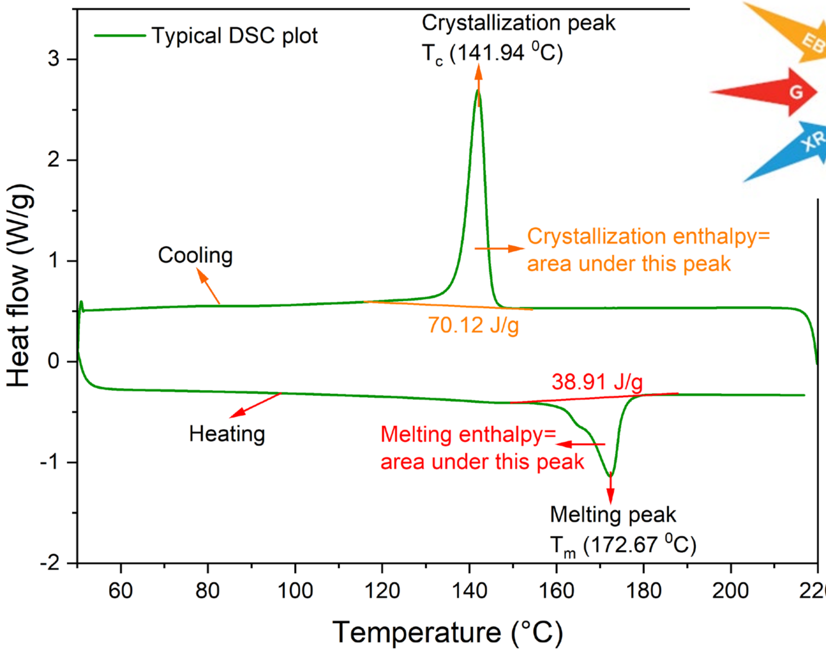




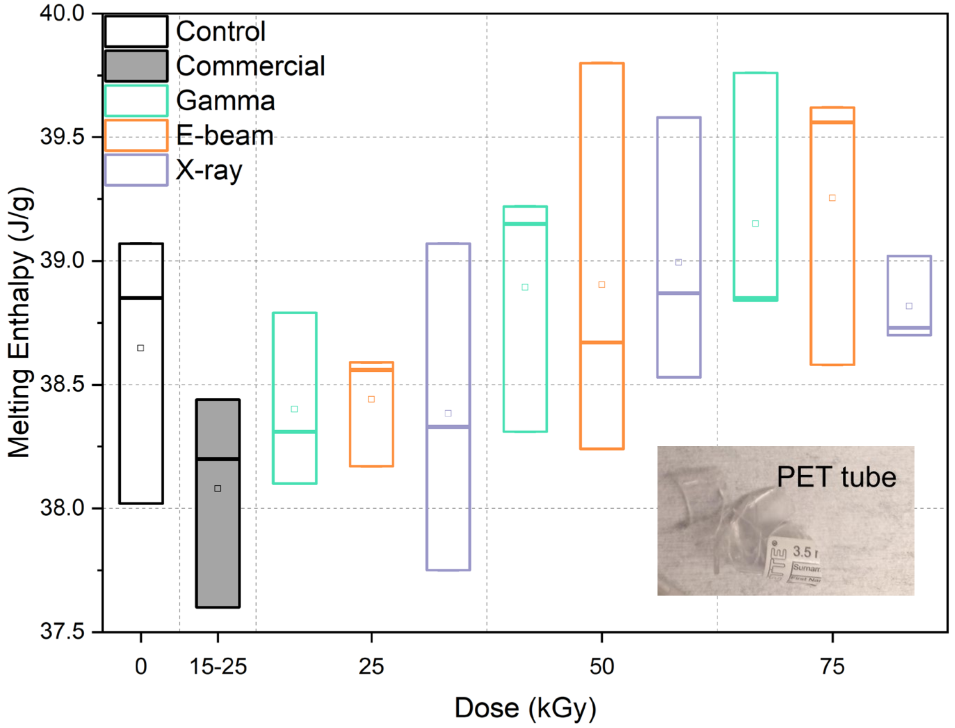
Thermal Measurement
Differential Scanning Calorimetry

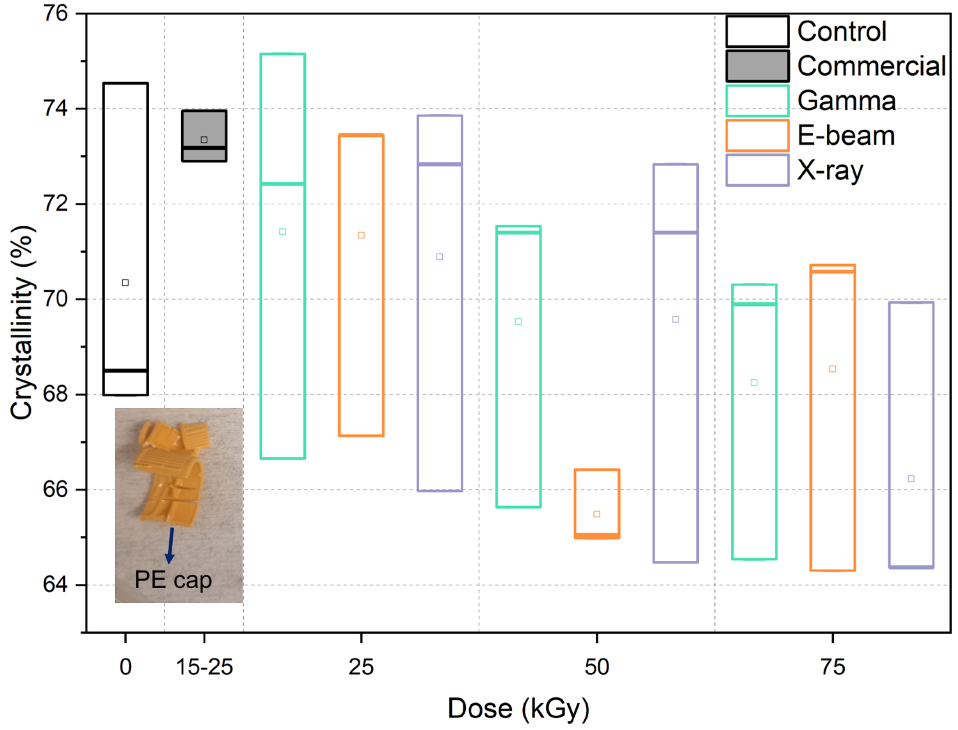
Component	Results extracted	Temp Range (℃)
PET tube	Tm, Tc, Xc, Tg	20 to 300
PE cap	Tm, Tc, Xc	40 to 180
PP ring	Tm, Tc, Xc	40 to 200





$$\text{Crystallinity, Xc} = \frac{\textit{Measured enthalpy of melting of polymer}}{\textit{Equilibrium enthalpy of fusion at 100\% crystallinity of same polymer}}$$





Influence of Radiation Source on Clinical Results of Filled Tubes

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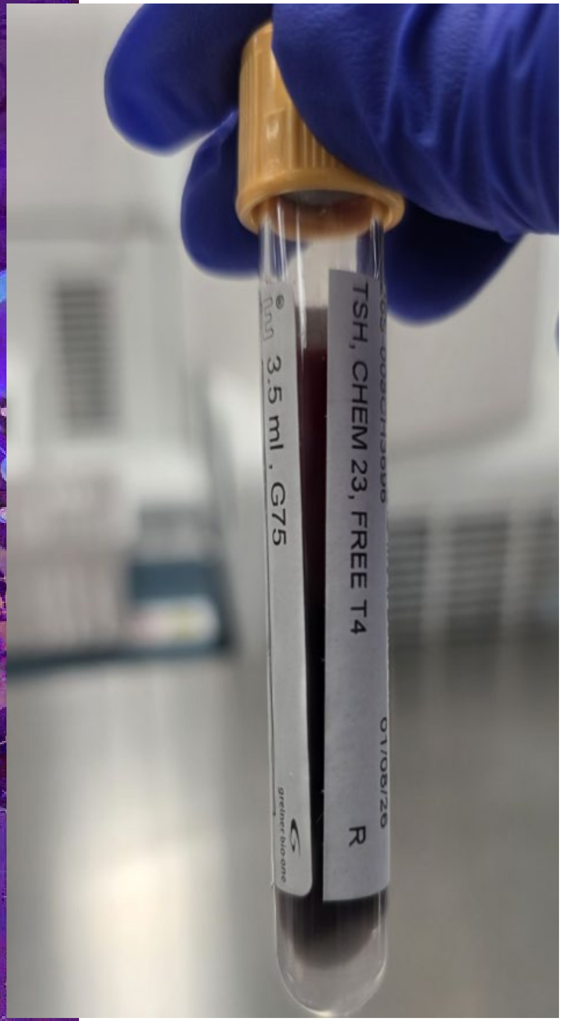

- 40 volunteers (20 male/ 20 female)
- Thyroid Stimulating Hormone (TSH)
- Free Thyroxine (FT4)
- Routine Chemistry Analytes (23)
- Statistical Analysis of results

Irradiation

- Gamma, eBeam, X-ray
- 0, 25, 50, 75 kGy
- Gold Cap tubes

Ongoing…

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greiner BIO-ONE

Influence of Radiation Source on Extractables and Leachables

- Gamma, eBeam, X-ray
- 20, 50, 80 kGy
- 2 formulations of polypropylene
- 1 polyisoprene rubber

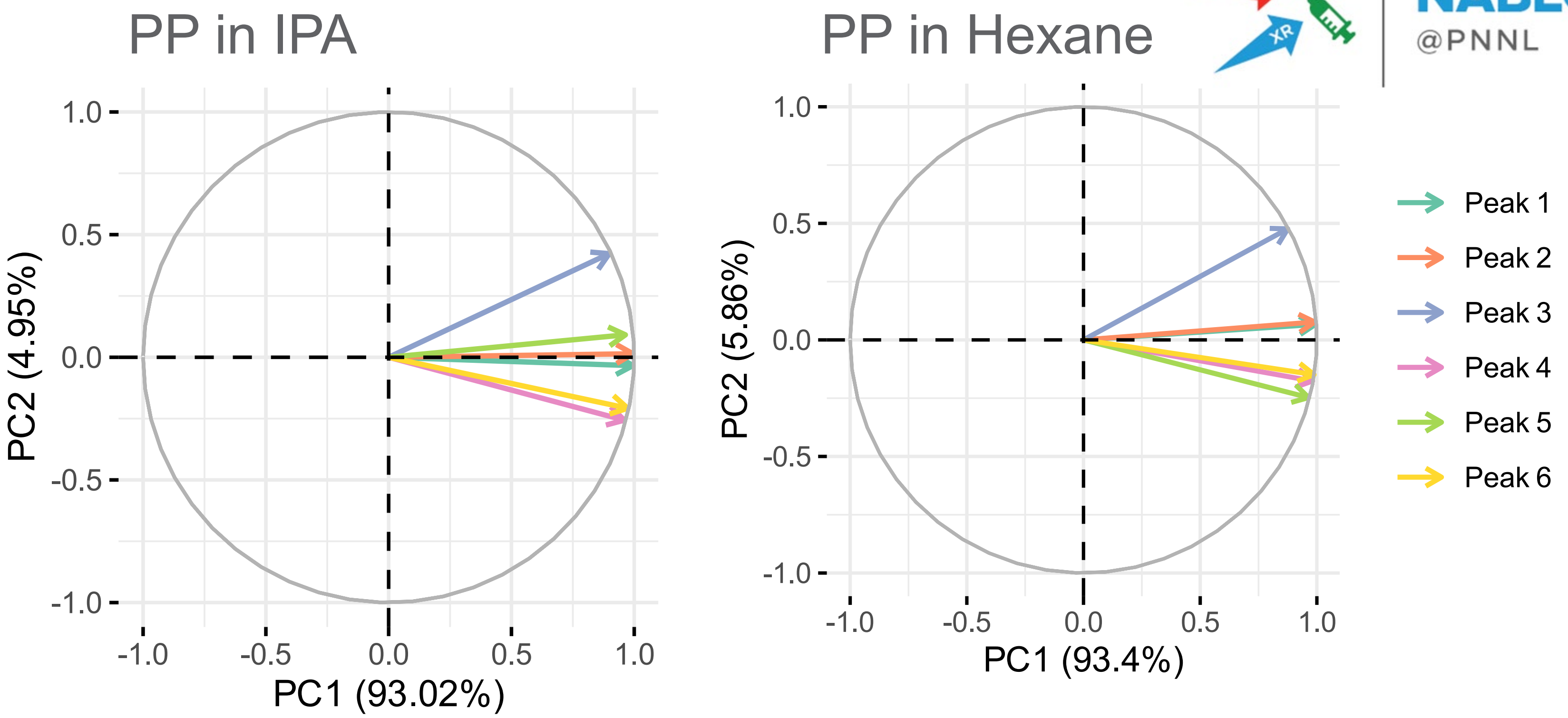
- Extractions in isopropanol (IPA) and hexane
- Use of Principle Component Analysis (PCA) to evaluate major GC-MS peaks

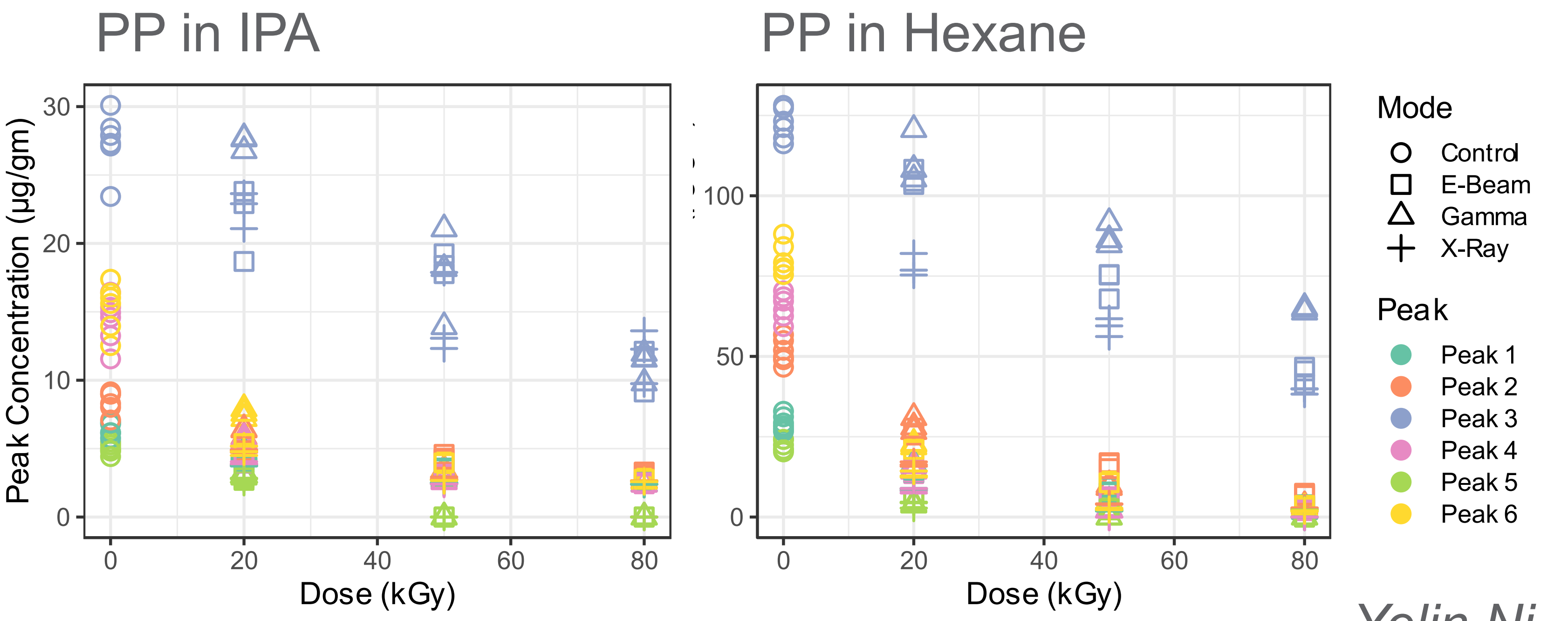
Polyisoprene Polypropylene

Polypropylene

Peak	Chemical
1	Cyclooctane, 1,4-dimethyl-, cis-
2	1-Nonene, 4,6,8-trimethyl-
3	Benzoic acid, 4-ethoxy-, ethyl ester
4	1-Octadecene
5	IPA: 9-Eicosene, (E)-
	Hexane: 5-Eicosene, (E)-
6	1-Docosene

- From both isopropanol (IPA) and Hexane extractions, all six peaks are positively correlated (they all decrease with dose)
  - Peaks 1&2, 4&6 are highly correlated
- PC1 explains 93% of the variance in data – PC1 can be a "single-value index" to reflect the effect of dose to PP2





Yelin Ni

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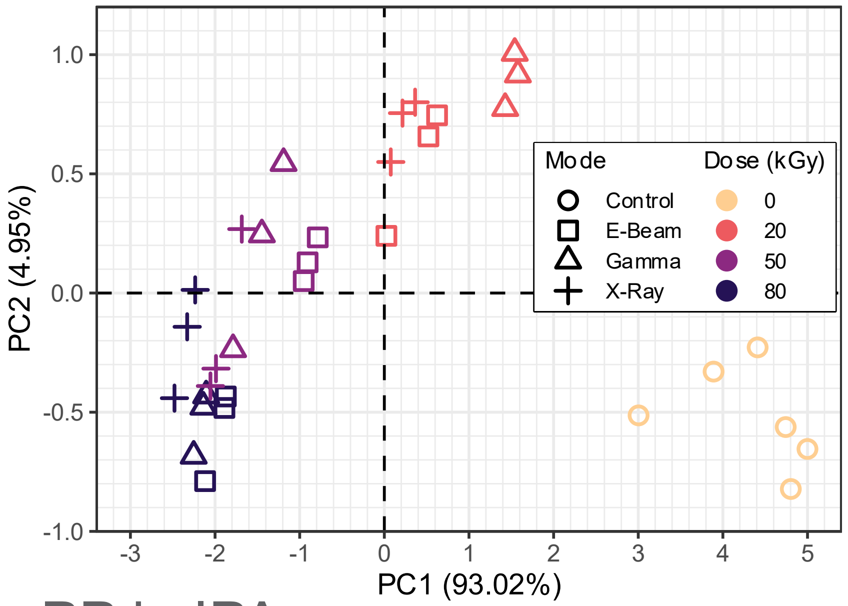
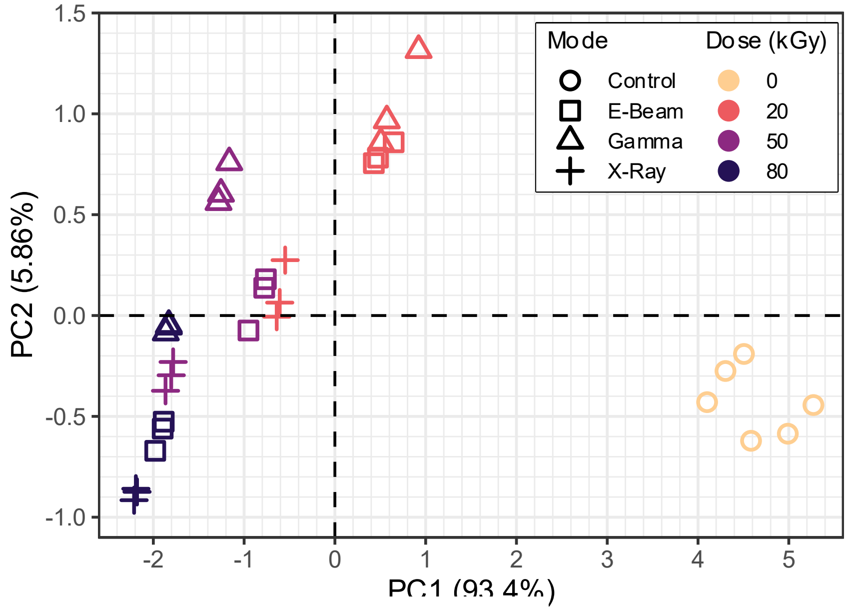
Polypropylene (PP)

- PC1 decreases with dose
- PC2 differentiates the effect of Mode:
  - Hexane: Gamma > E-Beam > X-ray
  - IPA: varies with dose

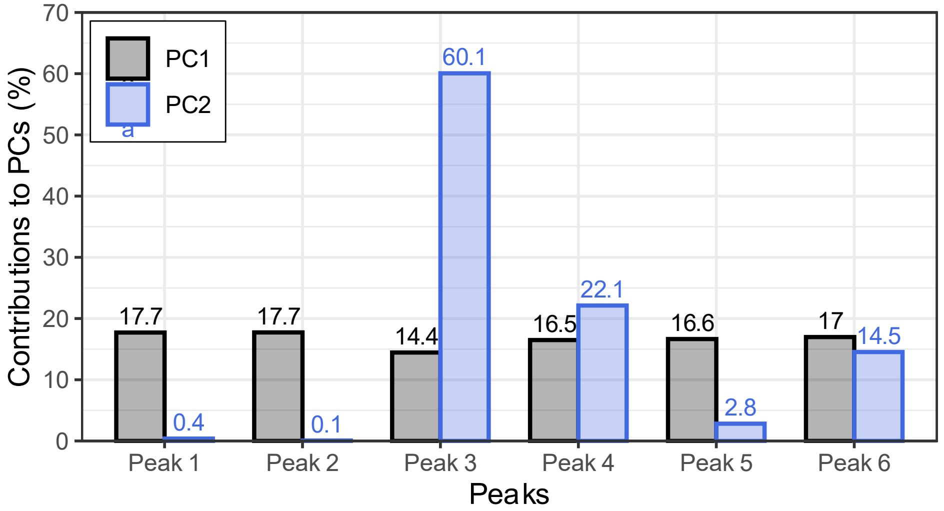
PP in IPA



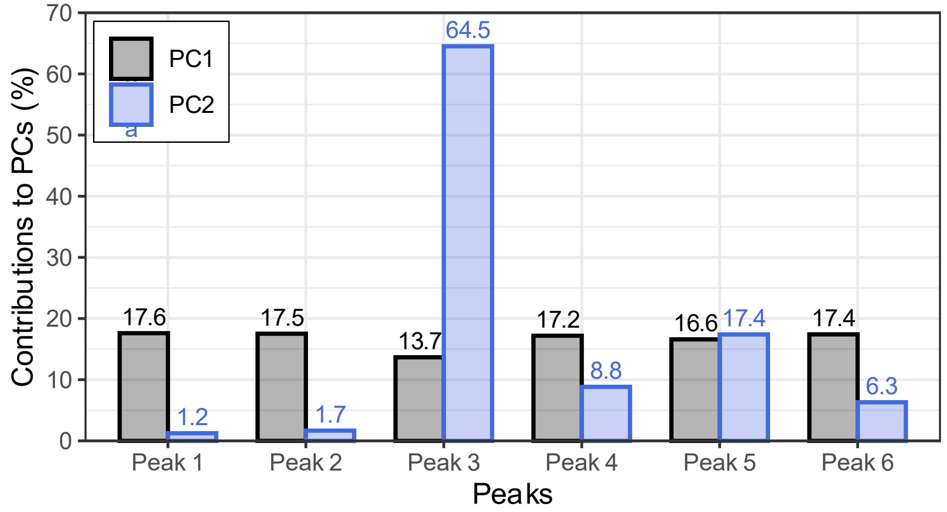
PP in Hexane



PP in IPA



PP in Hexane



- All peaks contributes to PC1 equally (~16.7%)
- PC2 is composed of Peak 3 primarily

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Dose Distribution Visualization

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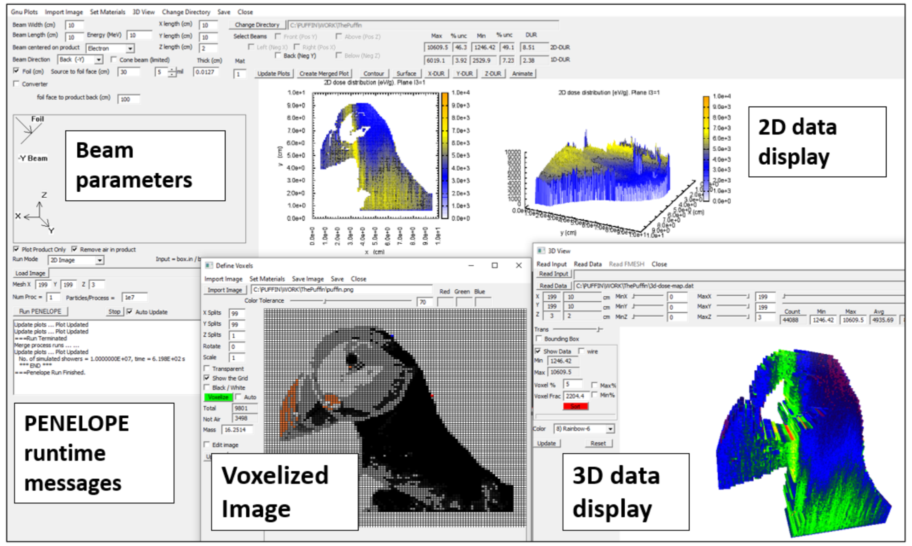
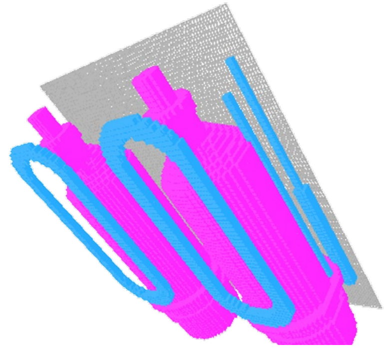
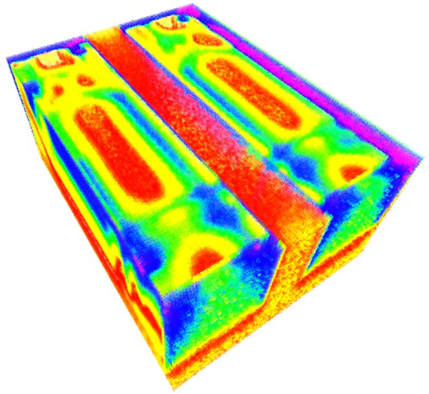
PUFFIn (PENELOPE User Friendly Fast Interface)[1]

- Utilizes PENELOPE[2] Monte Carlo code
- Visualizes dose distribution in simple product geometries using a personal computer
- Provides 2D & 3D visualization of dose distribution (dose uniformity ratio = DUR) in static products
- Accepts almost any input file – 2D images, CAD and DICOM files from a CT scan





Voxelized 3D Geometry



Energy Deposition for 10 MeV e^-

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1. *Schwarz, et al "PUFFIn – A user friendly fast interface for calculating and visualizing the dose distribution in materials," RPC, 2024, 111774*
2. *Salvat, F. 2019. PENELOPE, NEA/MBDAV/R(2019)1*

Dosimetry Needs for
- Low-Energy E-Beam (<1 MeV)
- Low Dose (<1 kGy) E-beam and X-ray

- Applications
  1. Surface sterilization for food packaging and medical devices
  2. Phytosanitary irradiation of fresh fruits and vegetables
- LEEB

  Commercial thin film dosimeters
  - Performance issues → significant errors
- Low-Dose E-Beam and X-ray

  Alanine pellets, optical waveguide, liquid dosimeters
  - Performance issues → large errors
  - Expensive readout equipment

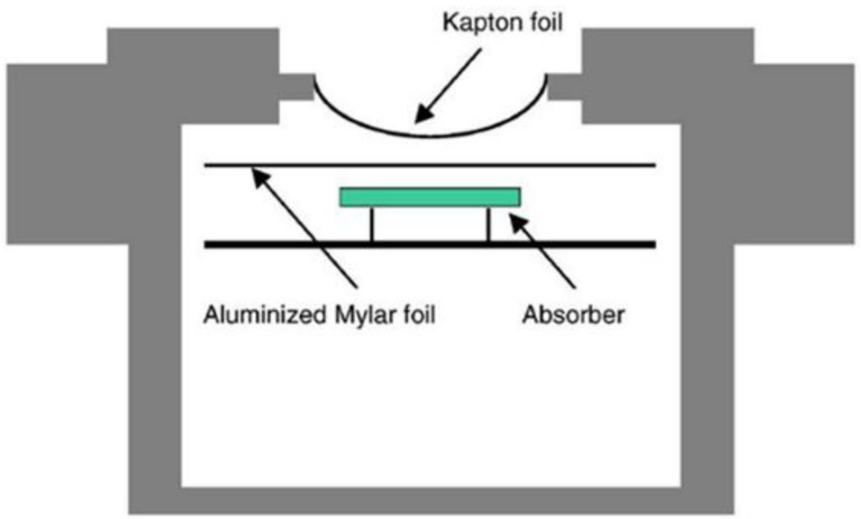
Mark Murphy

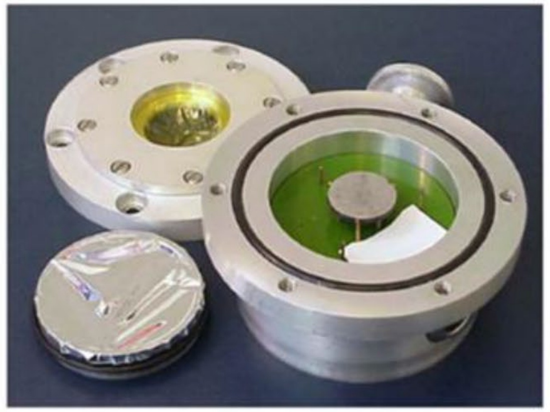
Desired Dosimeter Characteristics

- Dose response curve that covers range of the application, has a shape that is reproducible, and for which there is minimal influence of differences in the field energy spectrum.
- Thin enough that dose gradients within the dosimeter are minimal
- Overall uncertainty in measurement is within user requirements
- Influence quantities can be mitigated during in-plant calibration
- Shape in accordance with intended use and readout method
- Reasonable purchase and maintenance costs

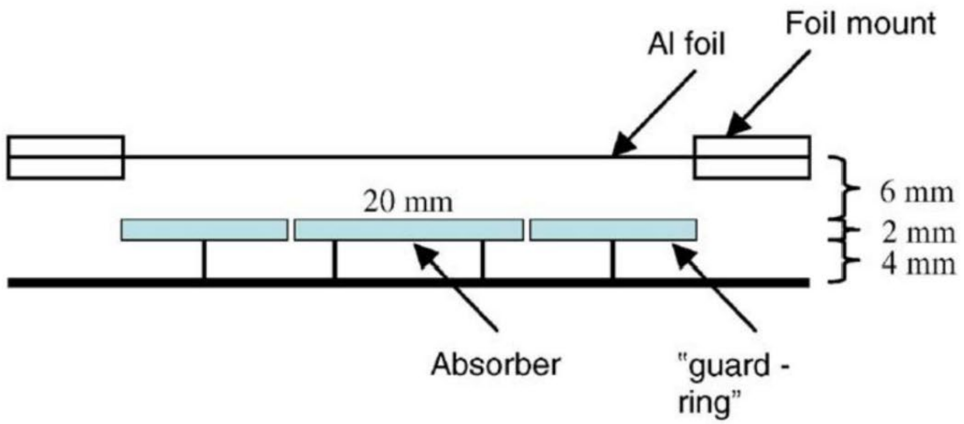
Low-Energy Electron Beam (LEEB) Dosimetry Development

- ORS is improving passive and calorimetric dosimetry technologies for beam energies below 300 keV
- Modeling and real-time monitoring are being integrated to enhance process control, efficiency, and reproducibility
- Calorimeters used for precise calibration of cellulose triacetate (CTA) and blue cellophane (BC) film dosimeters





Vacuum chamber calorimeter



Risø Developed calorimeter

Mark Murphy

Dosimetry Training Need

- **Purpose:** Address dosimetry knowledge gaps as institutions shift from Cs-137/Co-60 to X-ray irradiators.
- **Key Drivers:**
  – High dose-delivery errors (10–40%).
  – Significant irreproducibility in preclinical radiation research (~50%).
  – Shortage of qualified dosimetrists and radiation biologists.
- **Gap Analysis Findings:**
  – Lack of standardized dosimetry training and protocols.
  – Inconsistent measurement and reporting practices.
  – Users often unfamiliar with core dosimetry principles.
- **Recommendations:**
  – Develop low/no-cost training in dosimetry and irradiation protocols.
  – Engage users, RSOs, physicists, manufacturers, journals, and professional organizations.
  – Support facilities lacking dosimetry expertise.
- **Next Steps:**
  – Refine gap assessment and define learning objectives.
  – Compile standards and reference materials.
  – Develop, pilot, and evaluate training curriculum.

Heather Weidert

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Thank you

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Funded by the
U.S. Department of Energy National
Nuclear Security Administration
Office of Radiological Security

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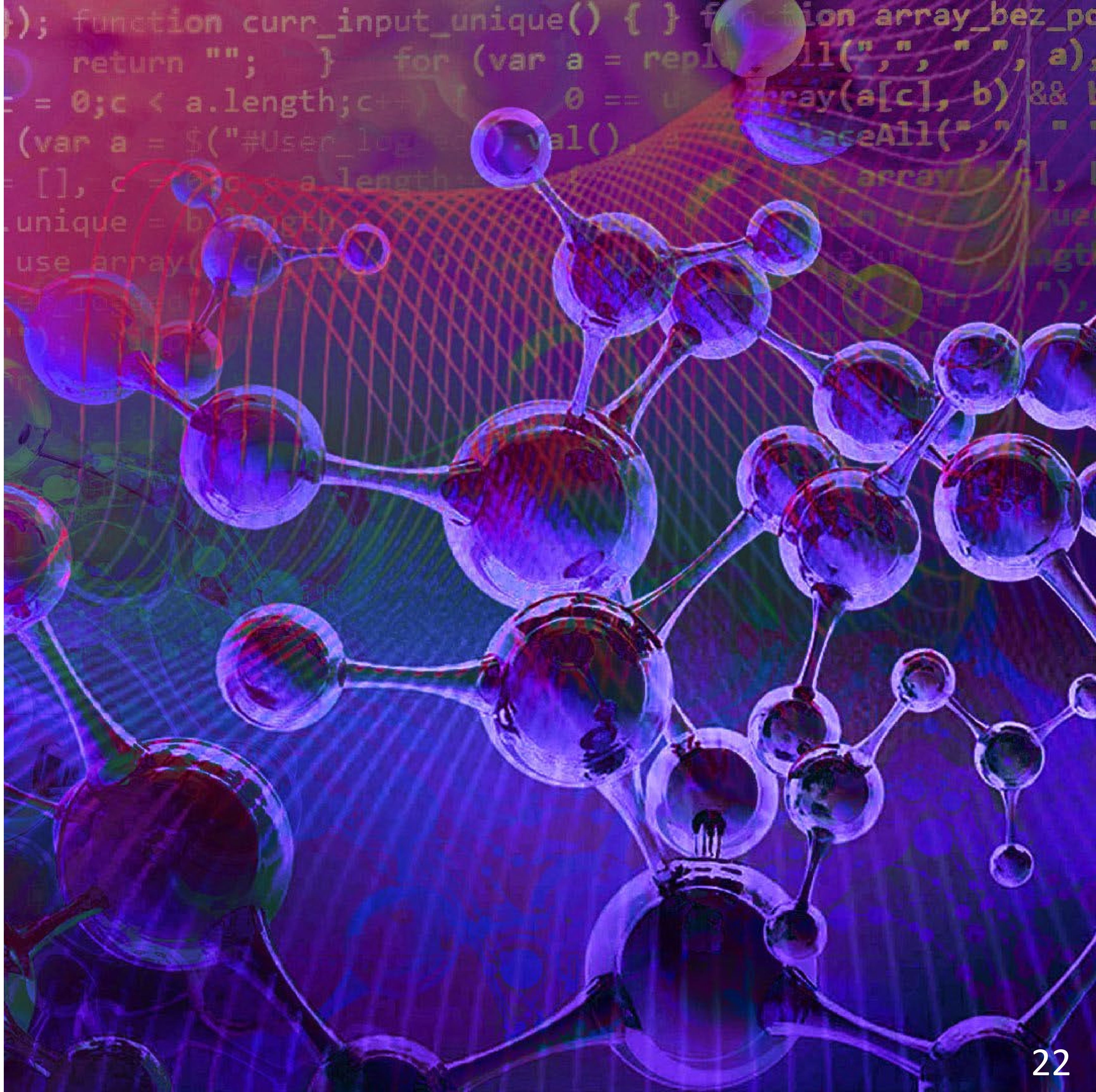

Topics for Emphasis in CIRMS Needs Report

- Regulator consideration of source equivalency
- Dosimeters for lower energy radiation detection (A.20.0, D.08.1)
- Reduced thickness/form factor dosimeters for better resolution (D.05.3)
- Accessible software for dose mapping as a function of source/energy (F.01.1)
- Better understanding of combined effects on polymer materials
- Improved “reproducibility” in radiation biology research (A.19.0)
  - X-ray fields at laboratories (provide “traceability”) that are more representative of the actual fields/spectra within the commercial cabinet X-ray irradiators (AAPM TG319)
- Education of calibrators and irradiator users on basic dosimetry and proper irradiation protocols, as well as minimum information to provide in published studies so others can accurately reproduce and/or validly compare

Communication of Team Nablo Results

- > 8 journal pubs (*RPC, PDS, npj Mat Deg*)
- > 60 presentations
  - Rad Confs
    (AccApp, CAARI-SNEAP, CIRMS, IMRP, ICARST)
  - Prof Society Confs
    (ANS, SEM, SPE MiniTec, ANTEC)
  - Industry Events
    (Kilmer, Q1, Marcus Evans, Fermilab)
  - Technical Exchanges
    (AAMI, IAEA, TUV-SUD, NAS)
- Webinars and > 5 Workshops (PUFFIn)
- ORS Portal
  - Datasets, publications, presentations

  https://orsportal.pnnl.gov/en/initiative/reduce-library

- *Radiation Phys & Chem* Special Issue

1. Li D., et al. "***Gamma, Electron Beam and X-ray Irradiation Effects on Polymers in an Advanced Bone Cement Mixer Device***" , Rad Phys Chem, Volume 226, January 2025, 112188 https://doi.org/10.1016/j.radphyschem.2024.112188
2. Li D., et al. "***Effects of gamma, electron beam, and X-ray irradiation on the plastic components of a universal bone cement mixer medical device***" Rad Phys Chem. Volume 223, October 2024, 111971 https://doi.org/10.1016/j.radphyschem.2024.111971
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