INL-LTD-21-63462

Sampling of Contamination - Lessons for Nuclear Forensics Collections and Analysis

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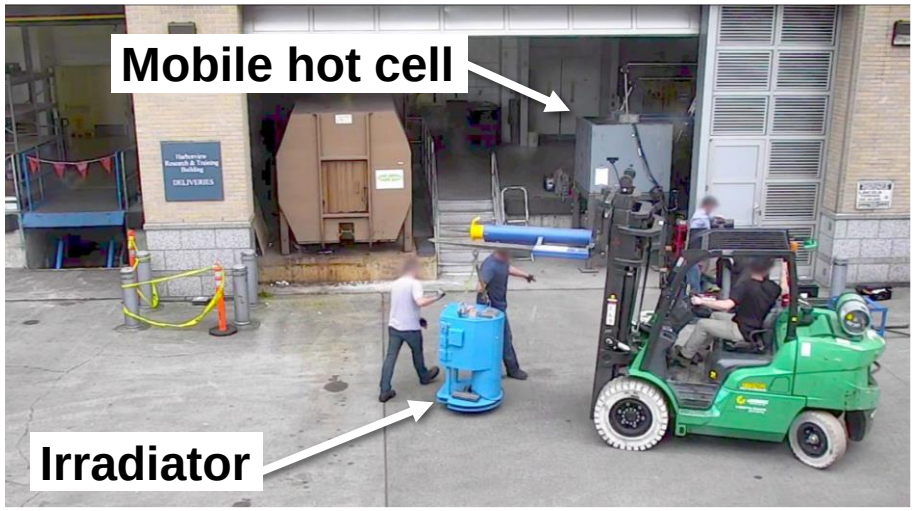
Overview

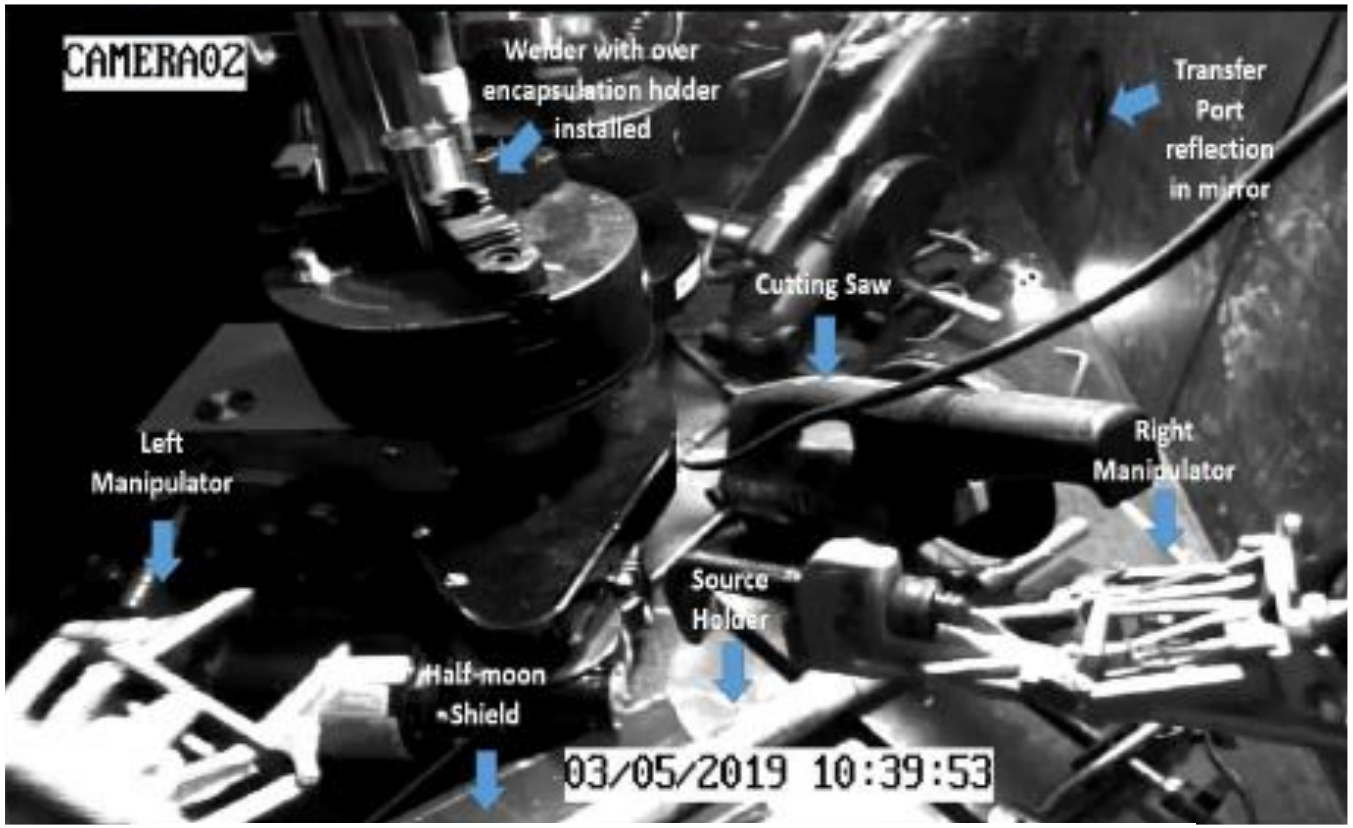
- On 2 MAY 2019 a radiological accident occurred at the University of Washington Harborview Research and Training Facility (HRT) in Seattle, Washington
  - Work was underway to dismantle a ^{137}Cs-based blood irradiator
  - During dismantlement a circular saw inadvertently cut into the source container, ~1.25 Ci (~4.62 x 10^{10} Bq; ~0.14 g) of ^{137}Cs was released into the local environment, contamination spread within HRT
- The mobility and spread of ^{137}Cs was greater than expected
- Total cost of remediation exceeded $200M
- INL have been working to analyze particulate from HRT, with a goal to determine if/how the material was different than expected under such conditions
  - INL received a variety of samples taken at HRT, and then later directly from the dismantlement equipment
  - This sampling and analysis work has similarities to how samples might be collected and analyzed for nuclear forensics
- This presentation summarizes this effort and presents some lessons learned to guide future NF collections and analyses

The Accident - A Brief Summary



Transport of the irradiator to the loading dock

Cesium source holder being moved into position

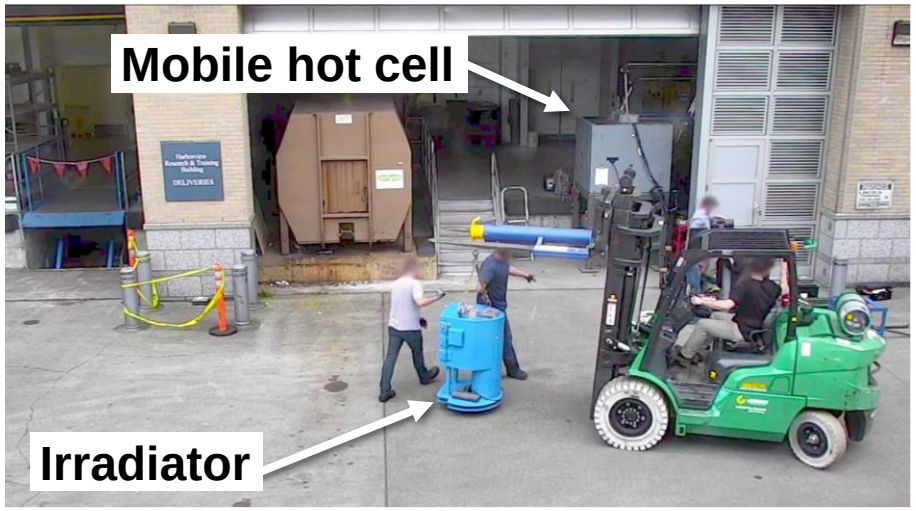


View of the rod, under the cutting saw, in the hot cell

www.energy.gov/sites/prod/files/2020/04/f73/JIT-Seattle-Cesium-Event-2019-05-02.pdfJIT-Seattle-Cesium-Event-2019-05-02.pdf

https://youtu.be/O9kkE1FPCSw

The Accident - A Summary in Pictures



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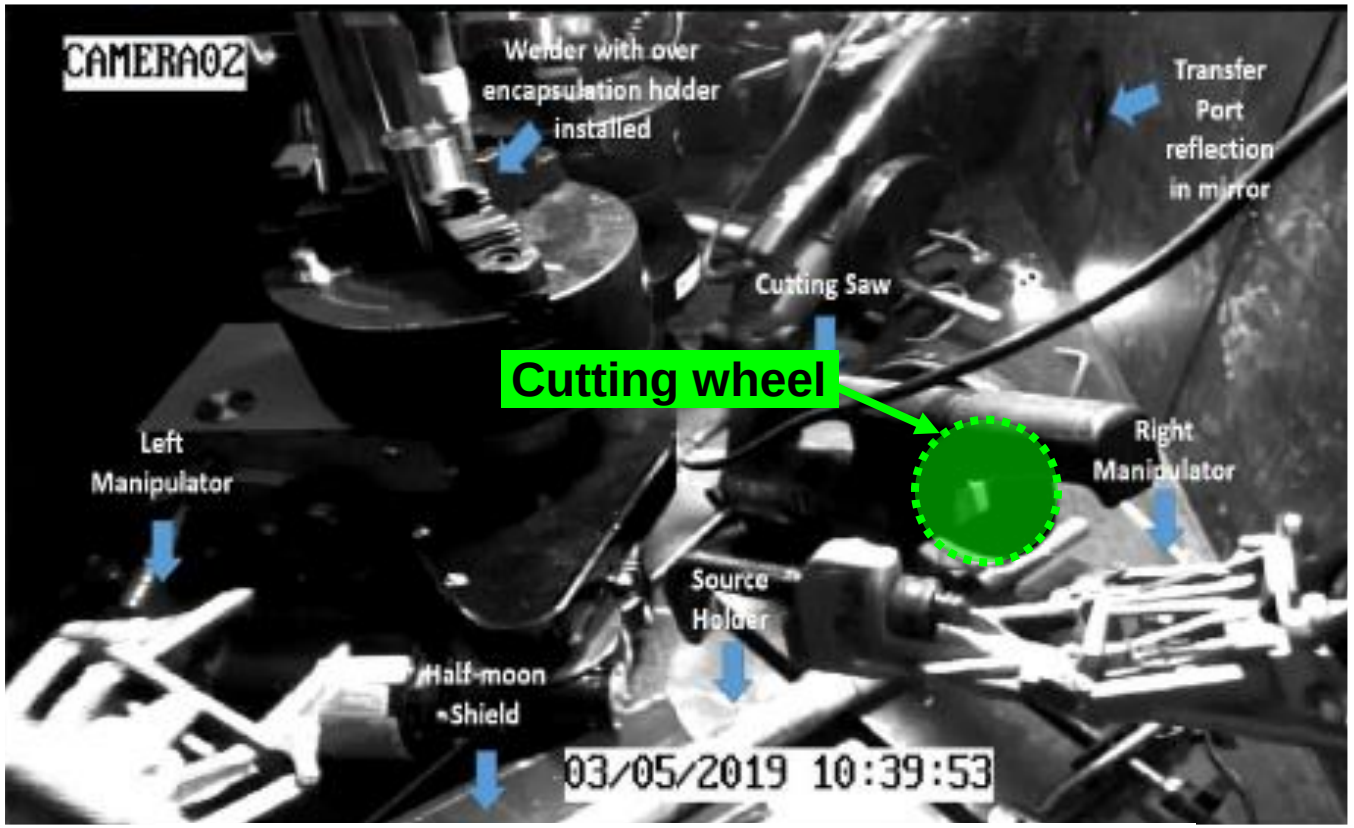

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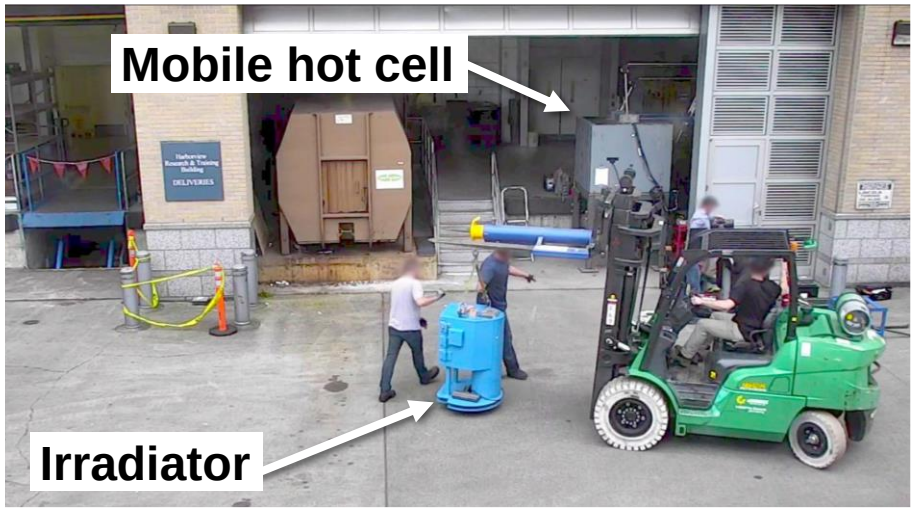

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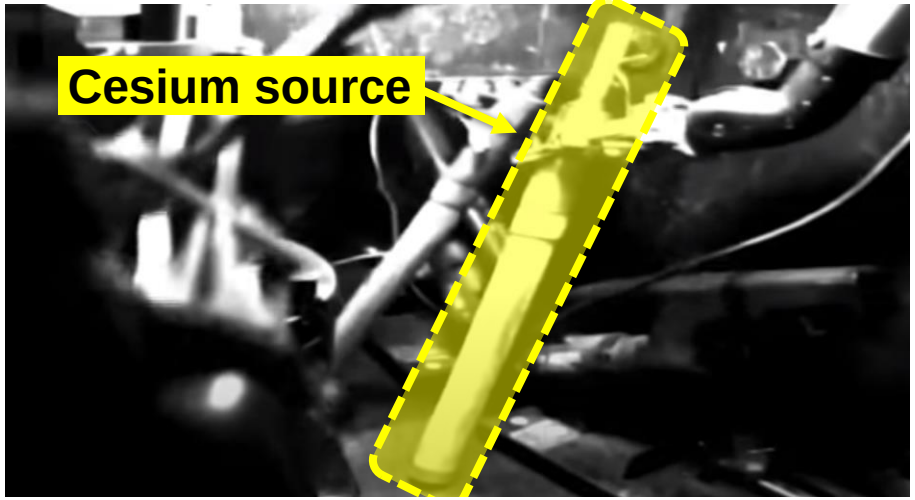
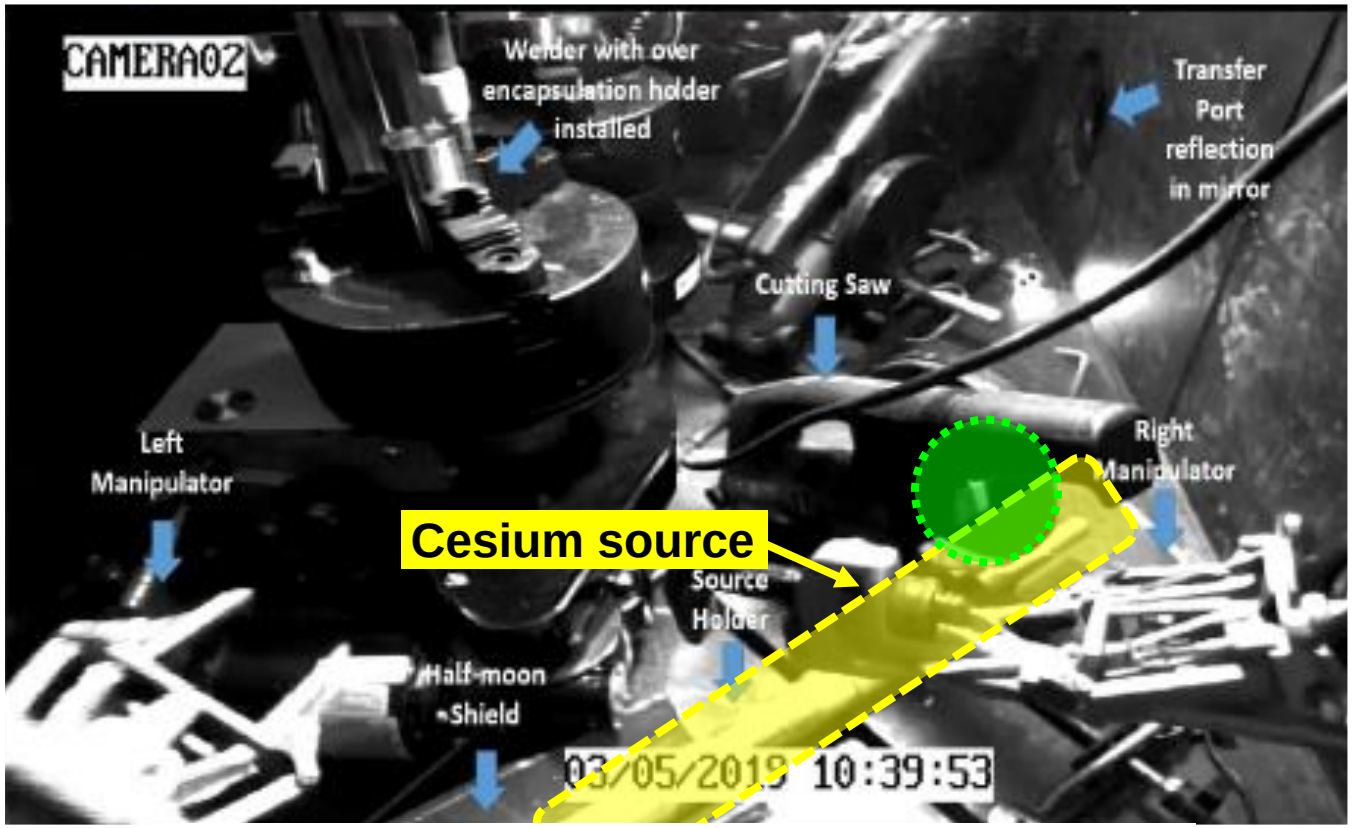


Cesium source holder being moved into position



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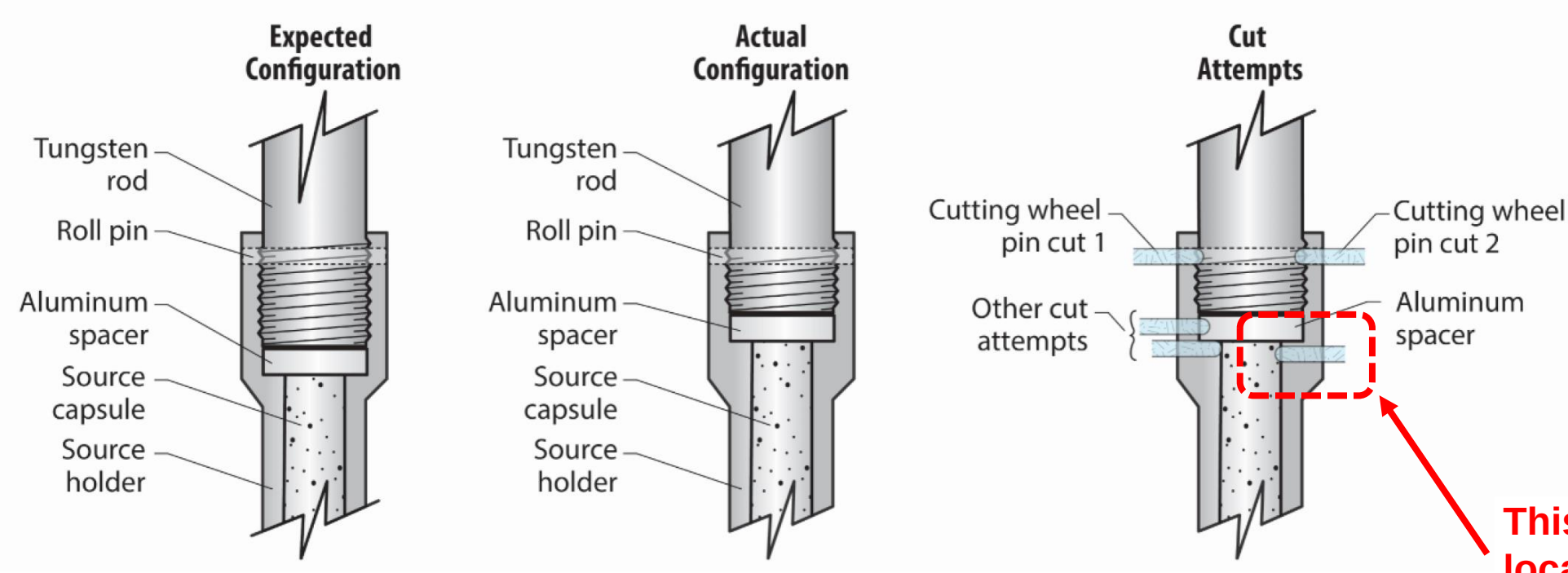

Orientation of the CsCl and the cut locations

This cut location breached the cesium pellet, releasing powdered CsCl

www.energy.gov/sites/prod/files/2020/04/f73/JIT-Seattle-Cesium-Event-2019-05-02.pdfJIT-Seattle-Cesium-Event-2019-05-02.pdf

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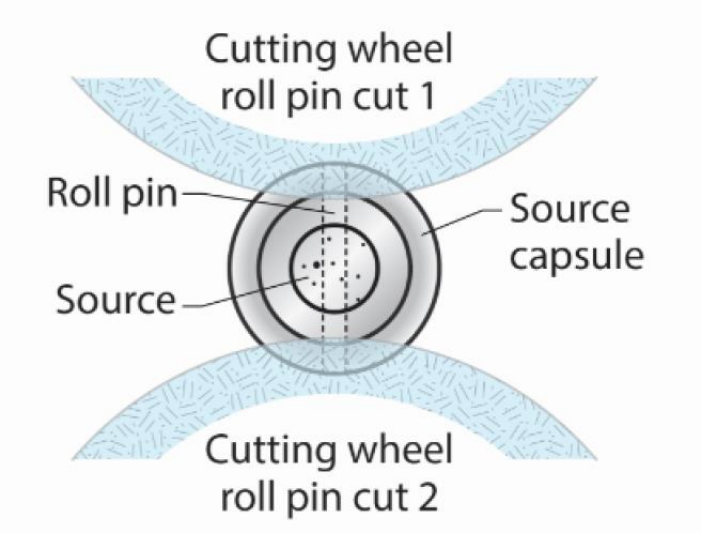
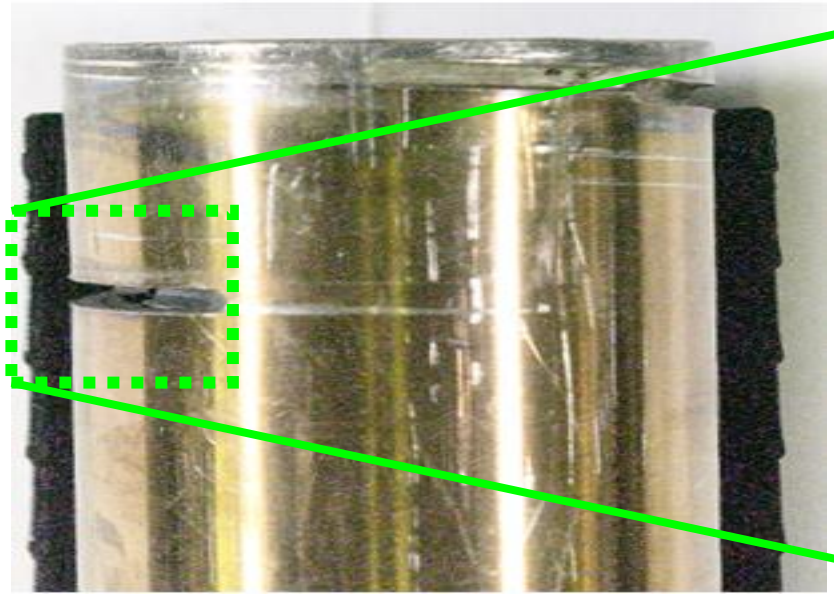


Geometry of cutting wheel entry into pellet



Damage to inner source capsule breaches steel wall and removes some of the CsCl

Close-up view of exposed pellet

The cutting wheel

www.energy.gov/sites/prod/files/2020/04/f73/JIT-Seattle-Cesium-Event-2019-05-02.pdfJIT-Seattle-Cesium-Event-2019-05-02.pdf

Schwantes, J., "Nuclear Forensics in Support of the Investigation into the Breach of a ~3000 Ci Sealed Radioactive Source in Downtown Seattle," PNNL 2021

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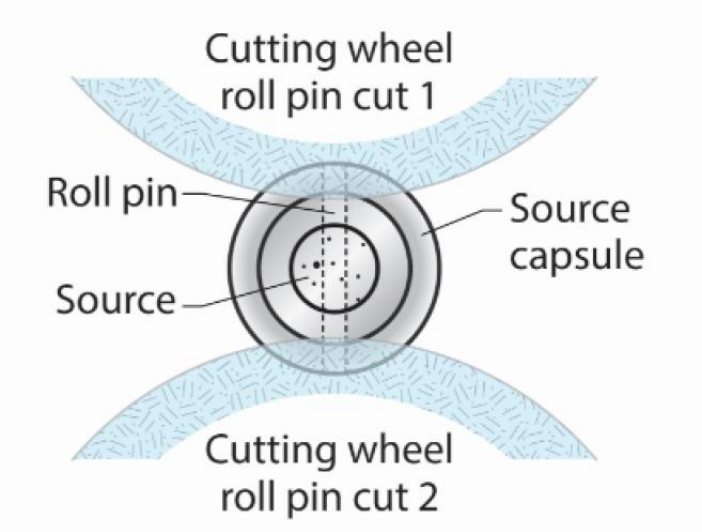

Geometry of cutting wheel entry into pellet

The cutting wheel

PNNL estimated the maximum potential ^{137}Cs activity released inside the building, based on the removed volume of CsCl

- ~1.25 Ci (4.62 x 10^{10} Bq)
- ~0.14 g
- Less than 0.04% of the total activity

- 13 people were contaminated
- All nine floors of the HRT were contaminated
- The total cost of remediation is >$200M

www.energy.gov/sites/prod/files/2020/04/f73/JIT-Seattle-Cesium-Event-2019-05-02.pdfJIT-Seattle-Cesium-Event-2019-05-02.pdf

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INL Forensic Analysis

- Brookhaven National Laboratory leading a research effort to understand how/why the CsCl dispersal was so extensive (Steve Musolino and Fred Harper)
- During the response activities, multiple "health physics" survey smears were taken at the Harborview Training and Research Facility
  - Taken for contamination control and monitoring
  - Taken as a part of the overall "consequence management" response
  - Not taken with the express purpose for nuclear forensic analysis
- Previous analyses were unable to locate and analyze particulates - INL asked to try

Ten HP smears, individually held within plastic film bags, shipped to INL for analysis

INL Plan:

1. Gamma-ray spectrometry to estimate total ^{137}Cs activity on each smear; correlation with contact dose rates
2. Autoradiography to map activity on each smear
   - Photography to visualize smears and co-locate activity
   - Image analysis to map locations, quantify hotspot activity, direct SEM
3. Scanning electron microscopy (SEM) analysis to search for particles
   - Particle morphology
   - Energy dispersive x-ray analysis (EDAX) for elemental mapping

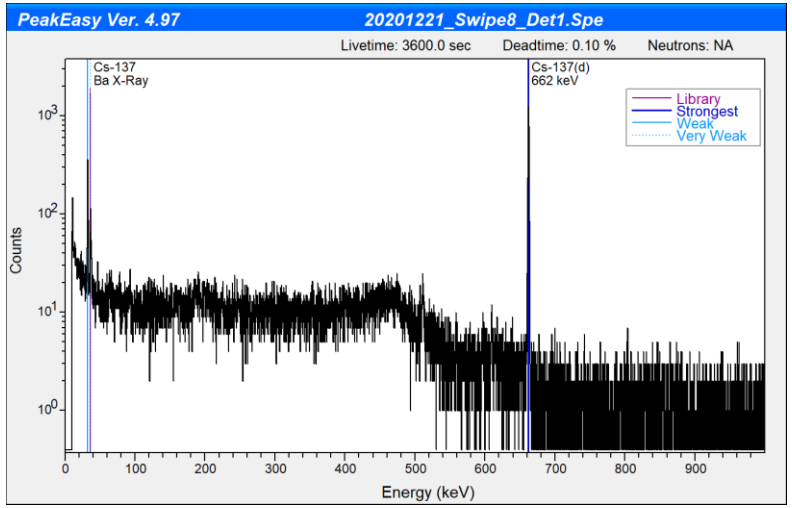
Example of Samples

Ten Harborview cesium "HP" smears

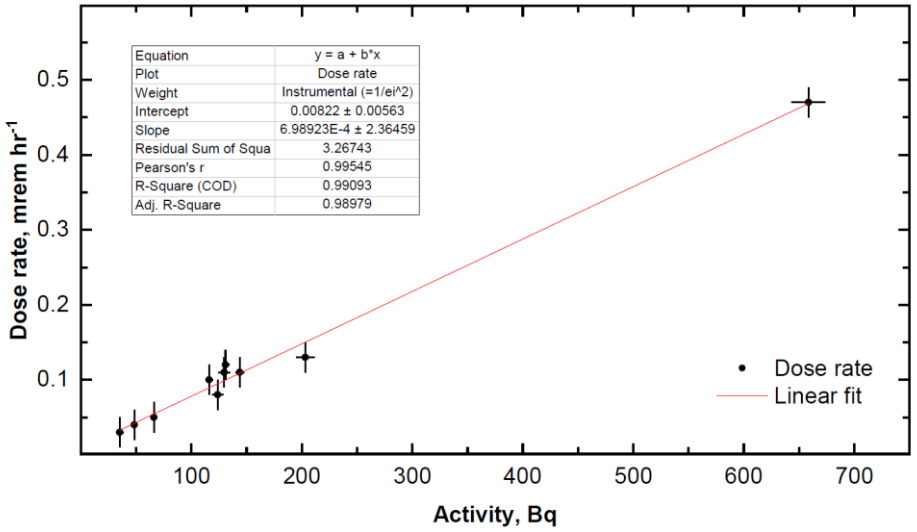
Close-up view of cotton-fiber HP smear, still attached to wax paper backing

Gamma-Ray Spectrometry and Dose Rate

- Performed gamma-ray spectrometry to confirm understanding of activity, exclude presence of other radioisotopes
  - ^{137}Cs observed, along with Ba x-rays (not seen in sealed sources)
  - No sign of ^{134}Cs or other significant radioisotopes
- Reasonably good correlation between measured "on-contact" $\beta + \gamma$ dose rates and estimated ^{137}Cs activity (γ dose rates were all 0 on contact)



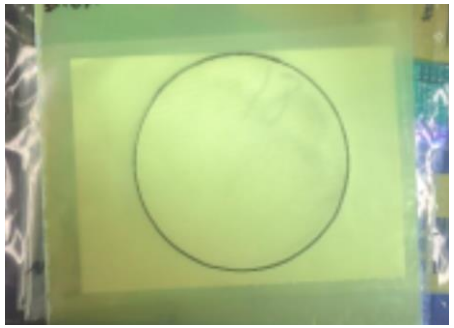
Representative gamma-ray spectrum



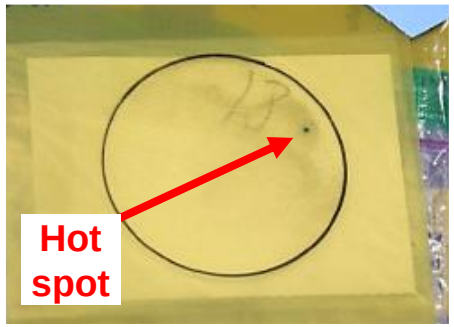
Correlation between observed on-contact dose rates and ^{137}Cs activity estimated from gamma-ray spectrometry

Autoradiography

Smear #18



ARAD film over #18

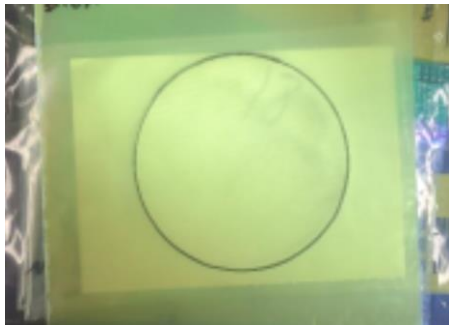


After 7 days exposure

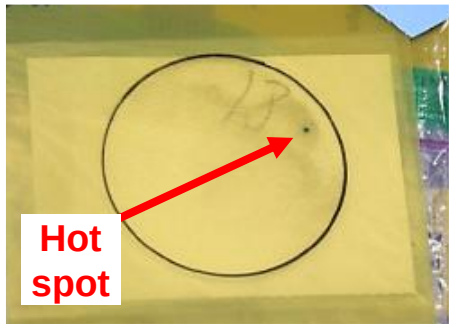
- HP smears placed on a lab bench
- GAFChromic EBT3 autoradiography (ARAD) film placed on top of smear
  - A black Sharpie marker was used to stencil a circle the same diameter as the cotton smear, intended to help registration
  - Tape was used to keep the film in place w.r.t. the smear
  - Photographs were taken before and after placement of the ARAD film
  - A heavy weight (tungsten block) was placed on top of the ARAD film
  - Exposure was for seven days
- A post-exposure photograph was taken before moving the film
- ARAD films were digitally scanned using an EPSON desktop scanner
- Automated image analysis tools were developed to locate spots, differentiate hot spots from dust/scratches, quantify spot intensity, correct for spots overlapping the stencil ring

Autoradiography #18

Smear #18



ARAD film over #18



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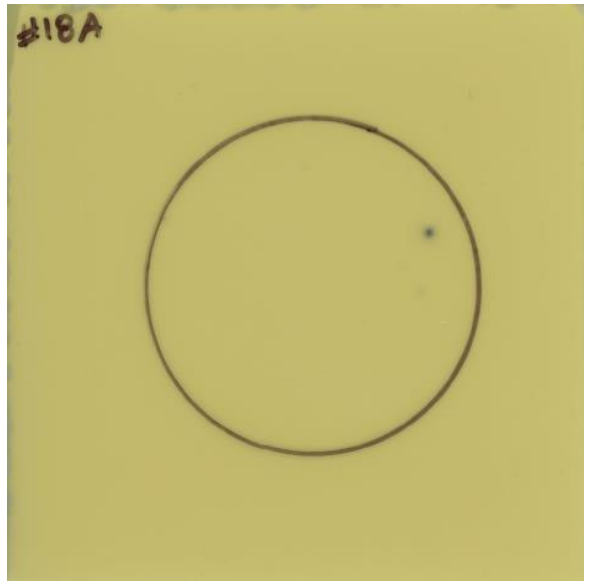
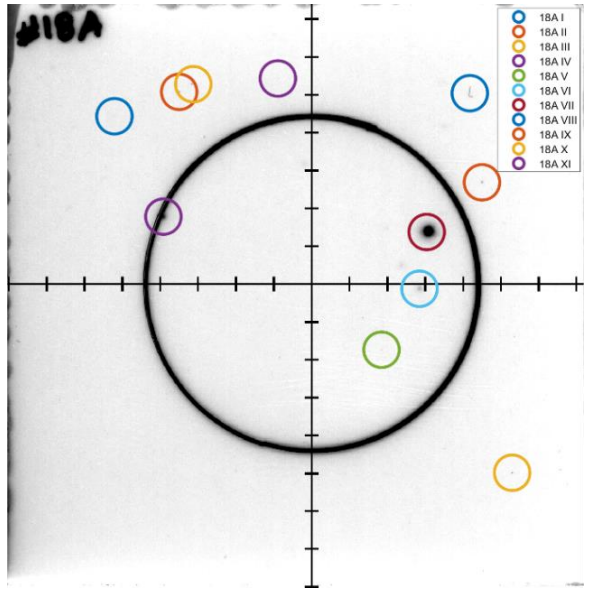


Digitized ARAD image

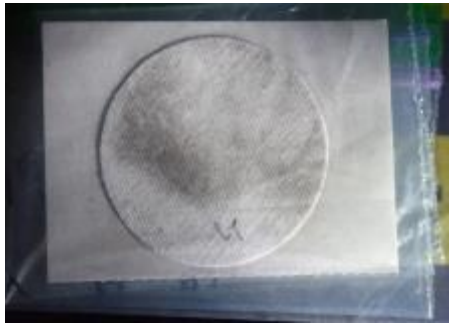


Analyzed ARAD image

Estimated ^{137}Cs activity per localized ROI

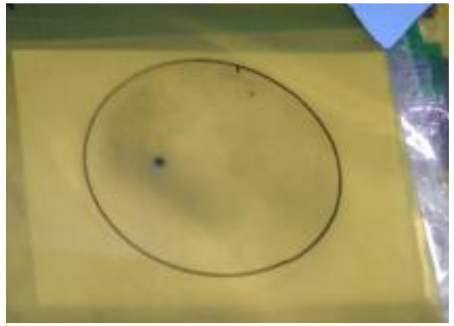
Region of Interest	Activity, Bq
VII	104
XI	5
VI	3
VIII	2
III	1
Balance	1
Total	116 ± 3

Autoradiography #27

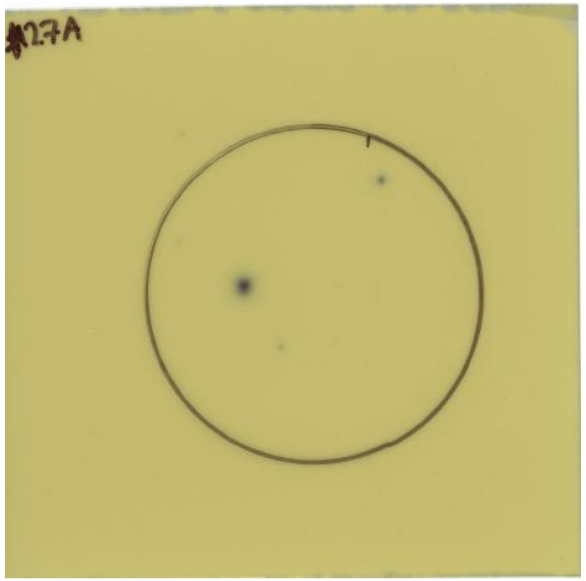


Smear #27

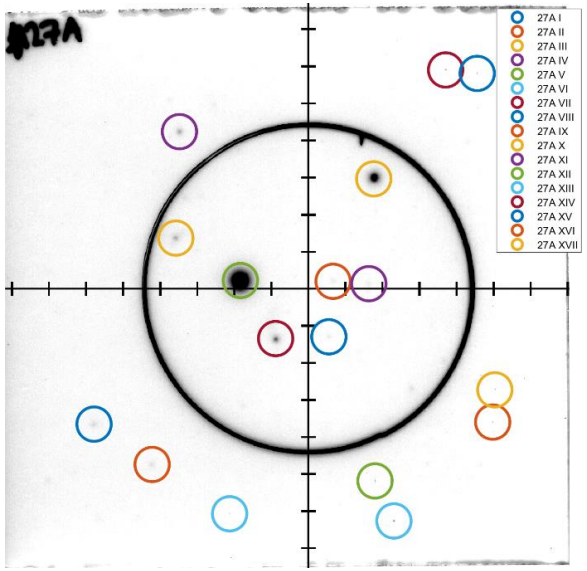
ARAD film over #27



After 7 days exposure



Digitized ARAD image

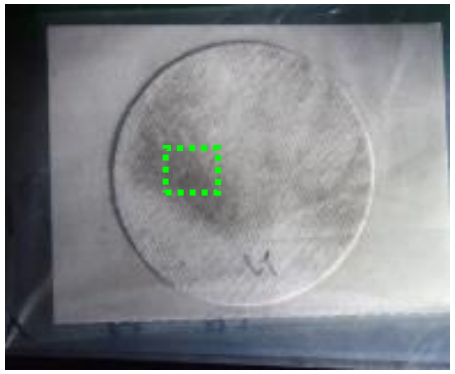


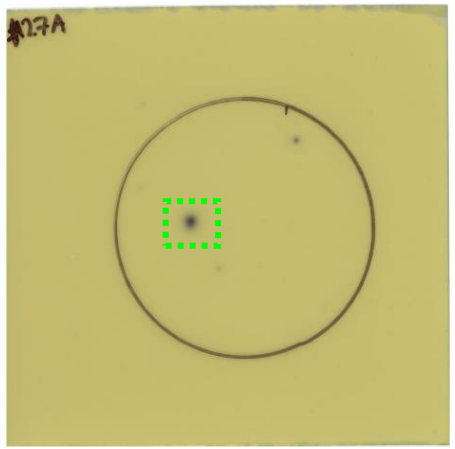
Analyzed ARAD image

Estimated ^{137}Cs activity per localized ROI

Region of Interest	Activity, Bq
V	528
X	89
VII	21
IV	7
I	3
Balance	11
Total	659 ± 15

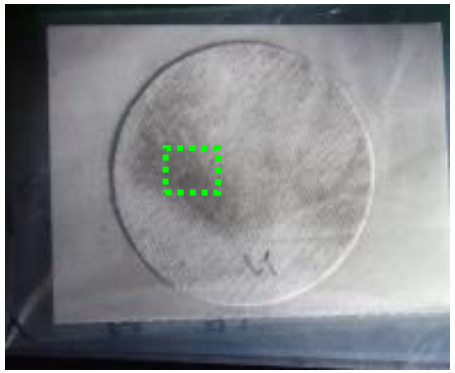
SEM Search for Particles

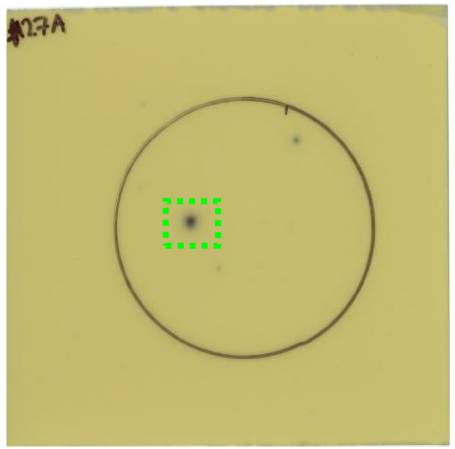


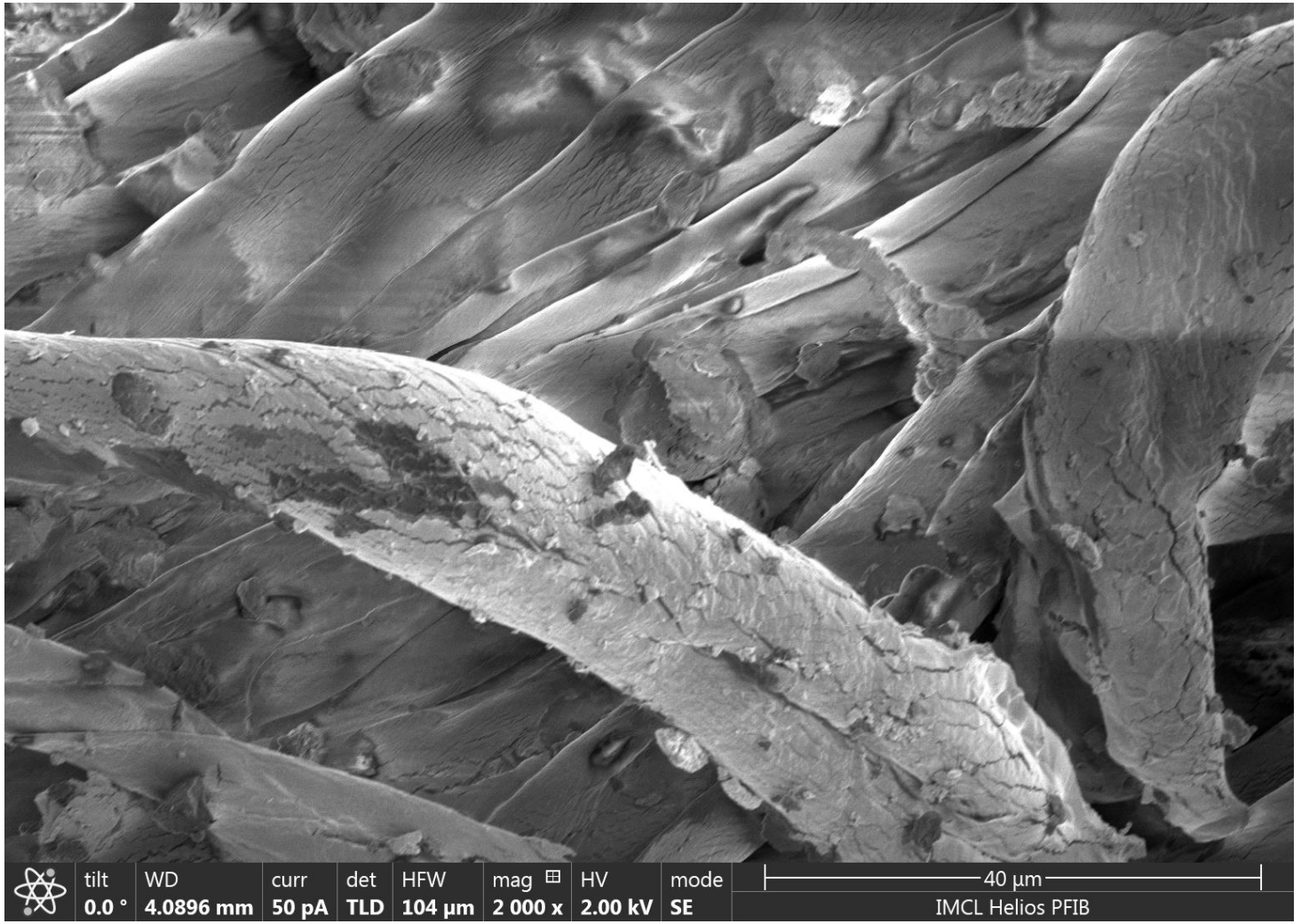


- With the ARAD data as a guide, smears were transferred to INL's Irradiated Materials Characterization Laboratory for SEM analysis
  - Hot cell, glove box, and fume hood infrastructure to support nuclear materials R&D
  - Diverse suite of integrated analytical tools
- SEM
  - Normally used to study grain structures in irradiated materials and nuclear fuels
  - Instrument set for automated scanning to locate particle
- EADX
  - Normally used to study chemistry and composition of irradiated materials, fission product migration in fuels

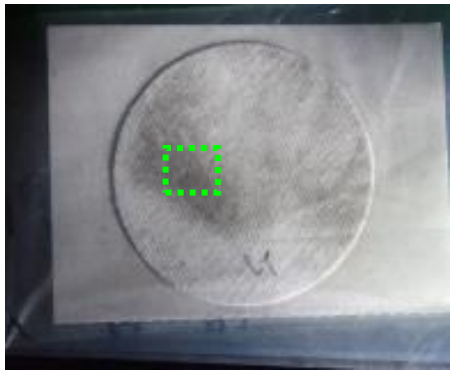
SEM Images: Smear 27, Hot Particle V (S27/HP V)

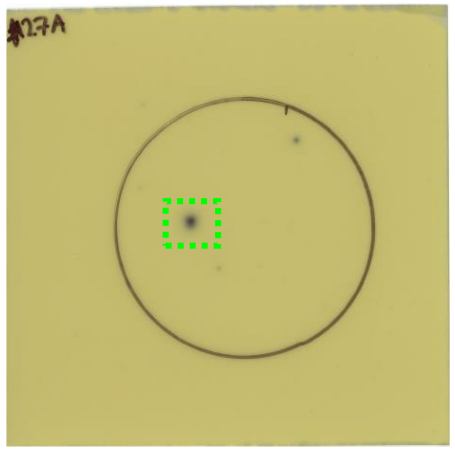


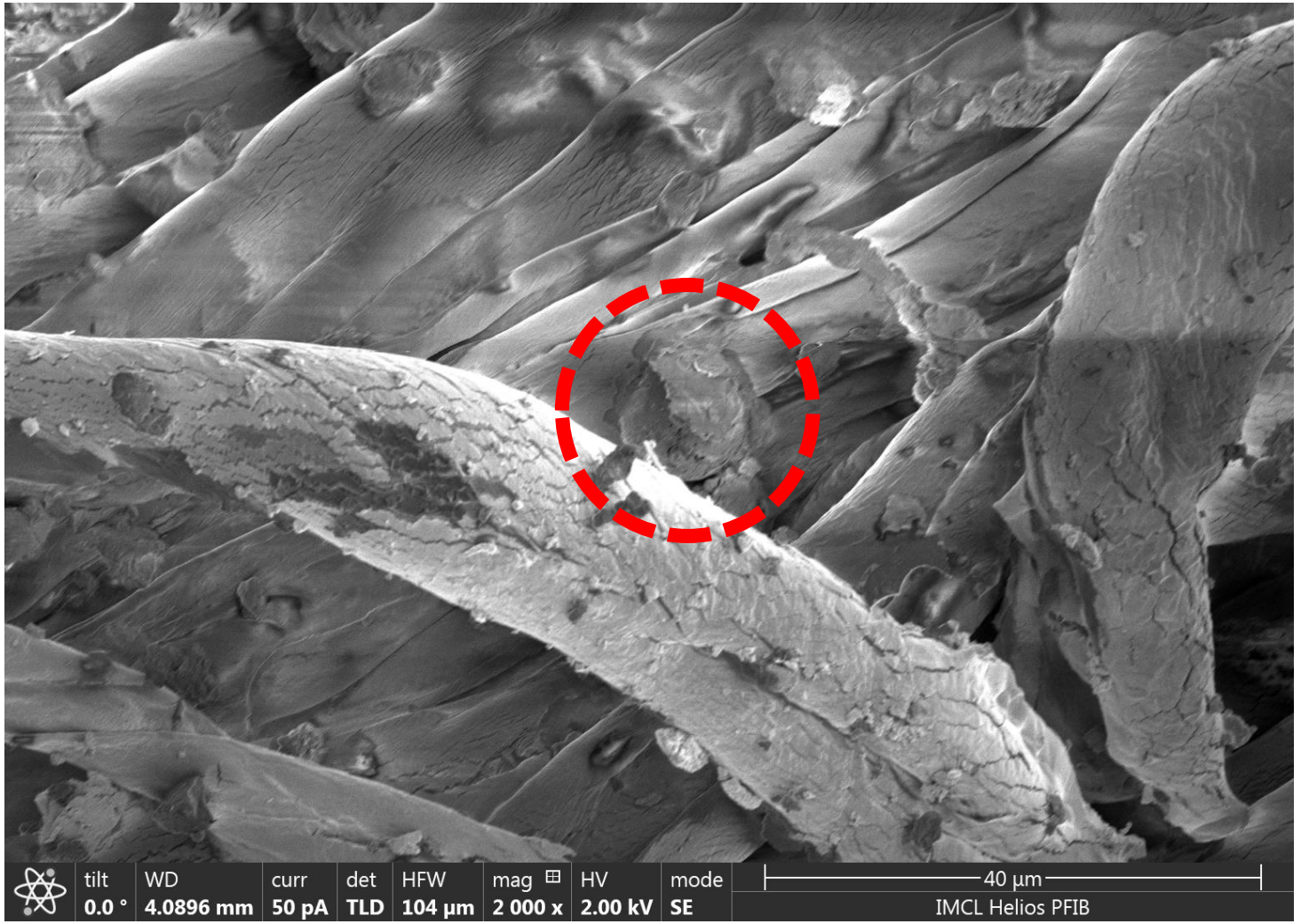




SEM Images: Smear 27, Hot Particle V (S27/HP V)







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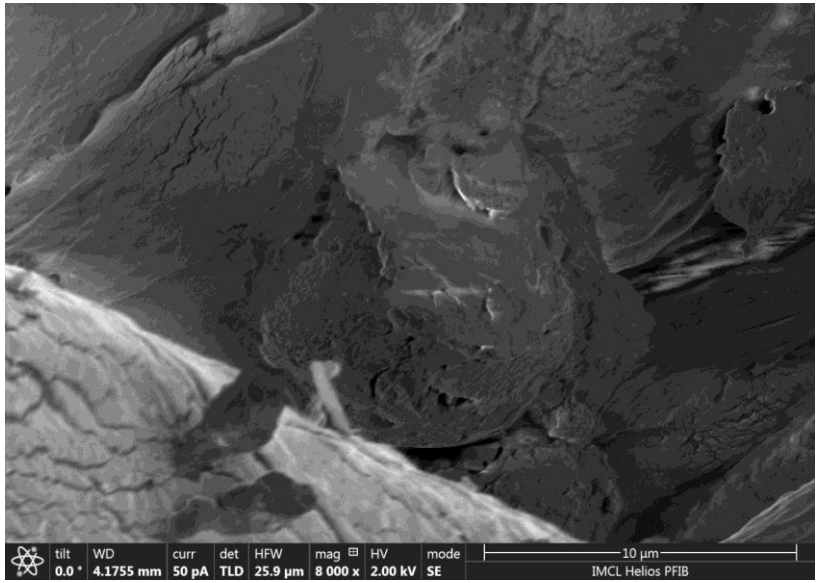
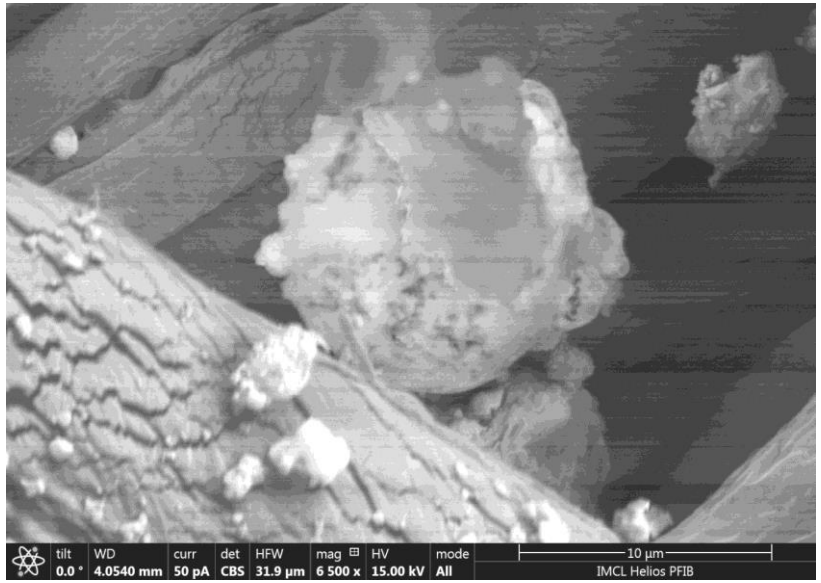
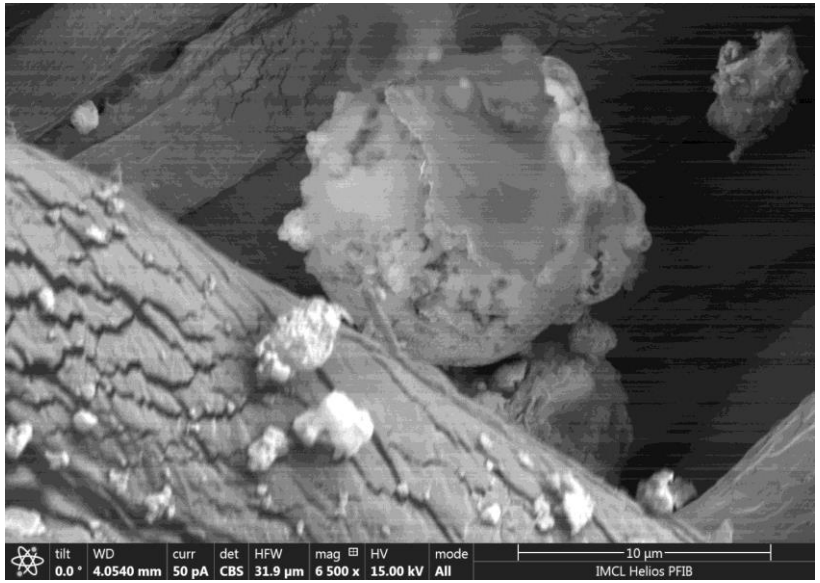
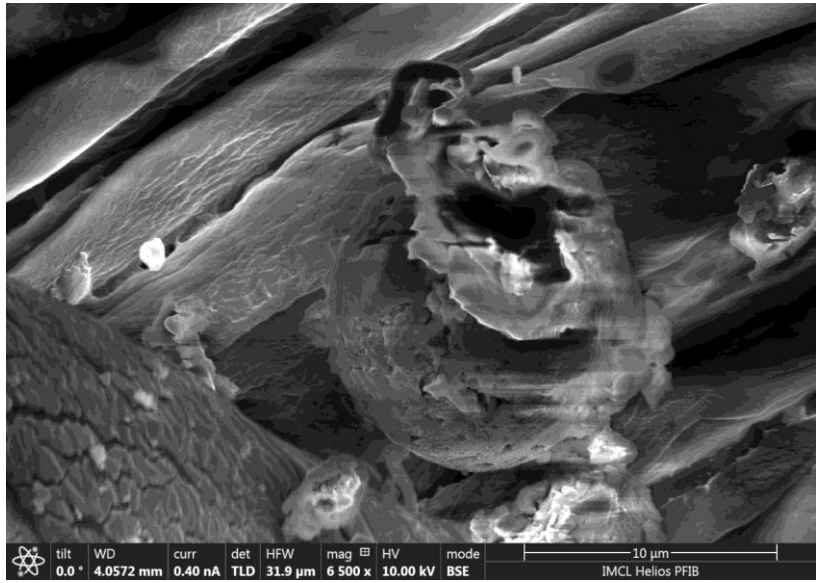
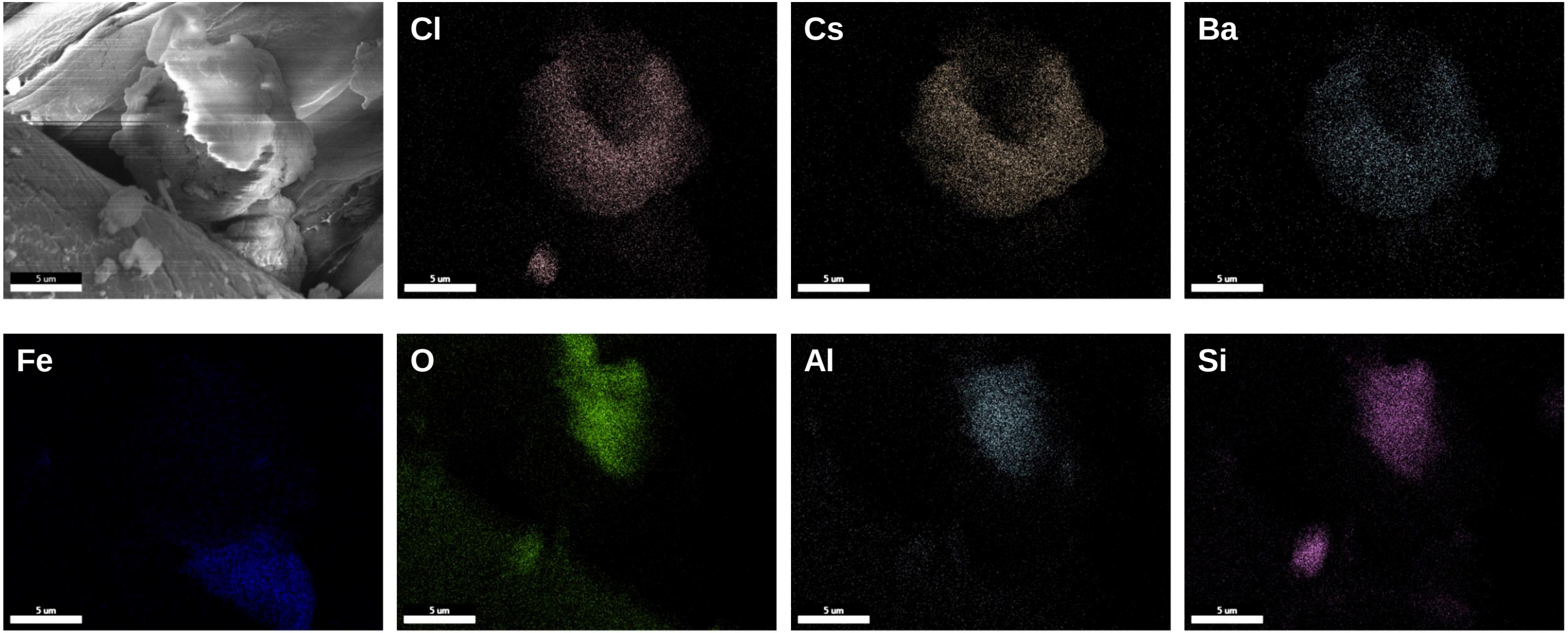






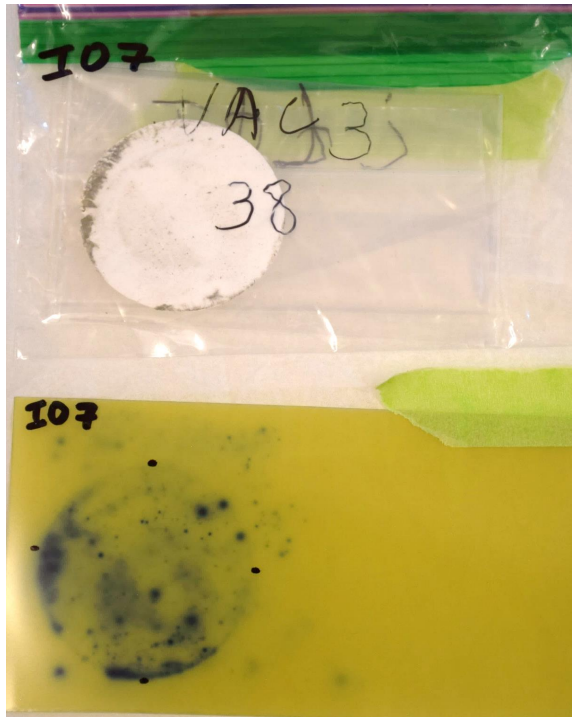


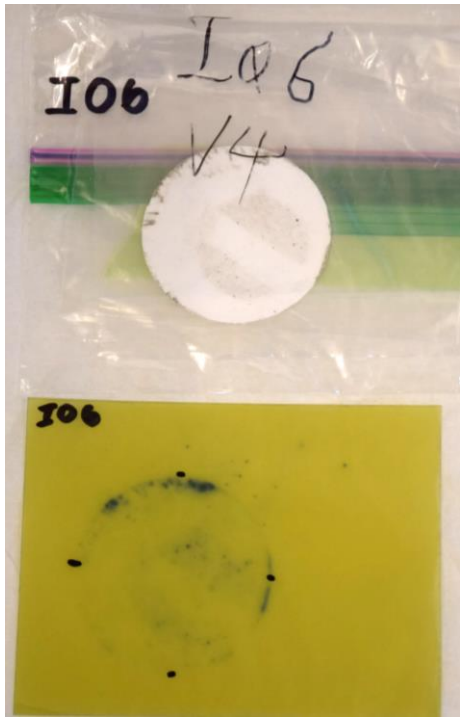
EDAX Elemental Analysis -- S27/HP V

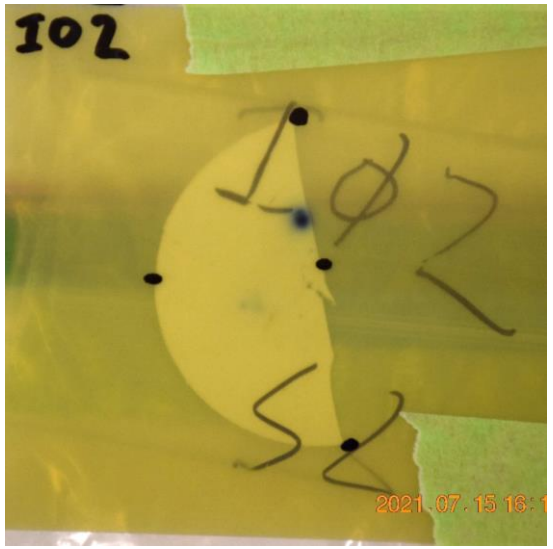


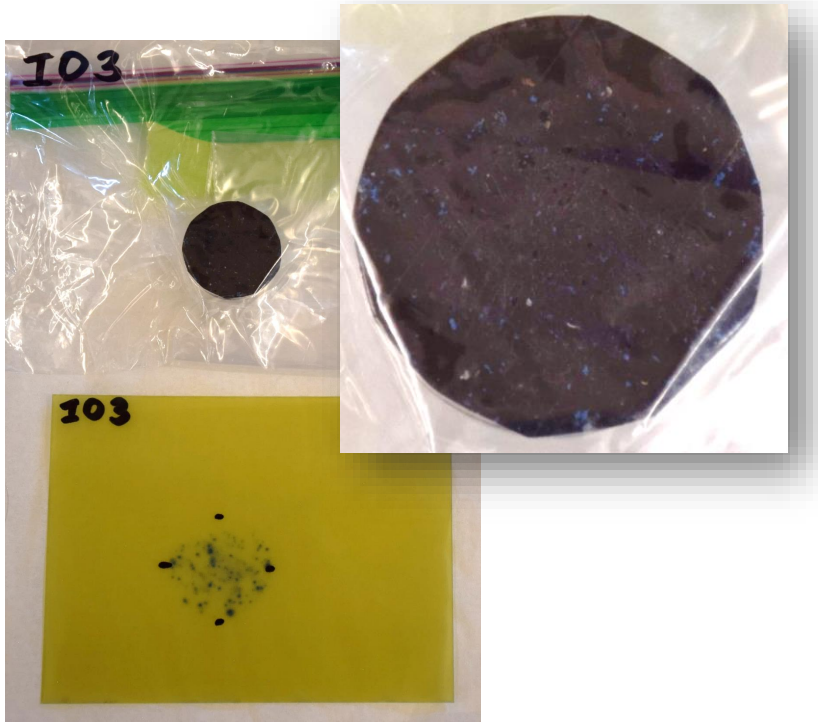
Follow On Analysis (Now Underway)

- After analysis of the ten HP smears, a second batch of samples was sent to INL for analysis
- Collected at PNNL from the equipment and materials recovered from Harborview
  - Much higher activity levels, highest at 200 mrem hr^{-1} (2 mGy hr^{-1}) on contact
  - Smears plus SEM stubs









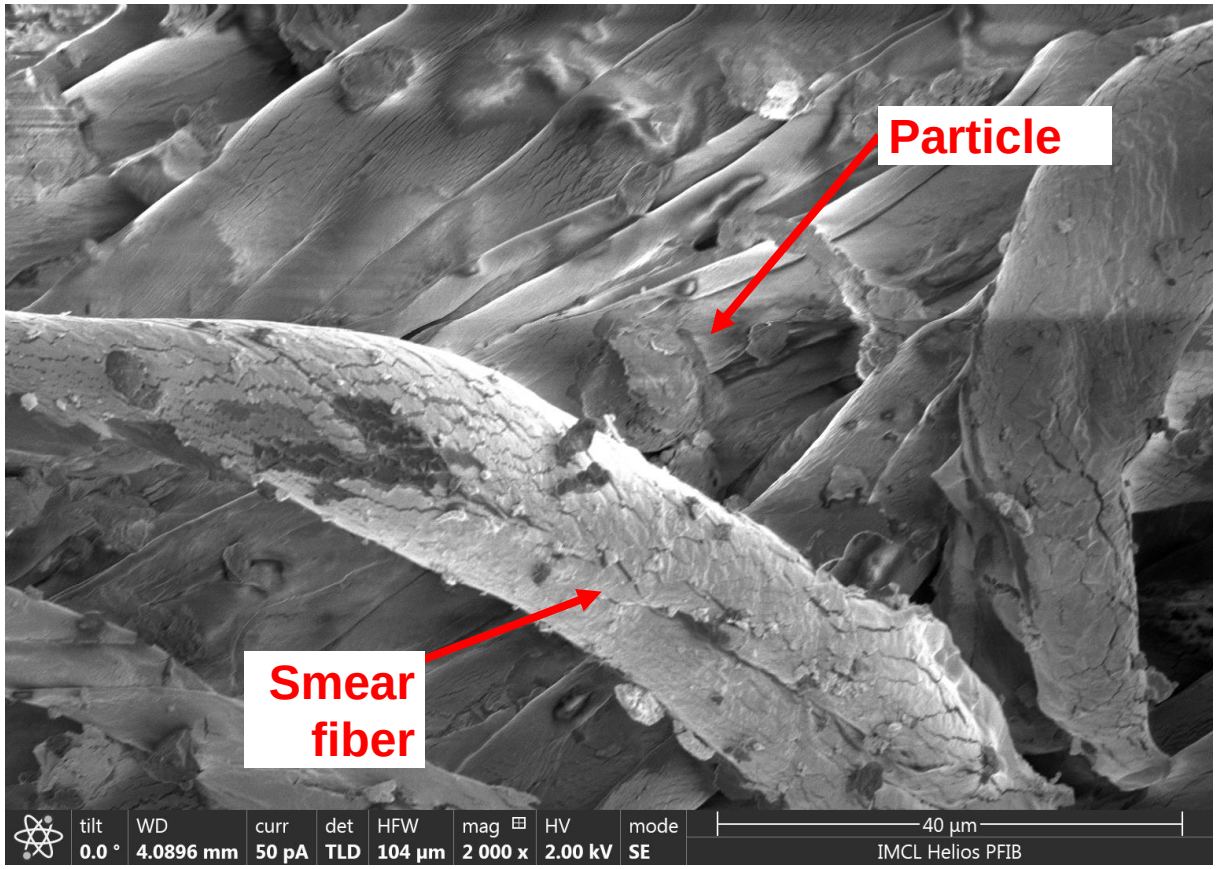
Take Away Lessons for Nuclear Forensic Sample Collection

Observation #1

Standard HP smears are too fibrous, very small particles can be caught inside the weave and make SEM detection difficult

Recommendations

Need to explore other options for materials for hand smearing when SEM is intended, such as natural and synthetic filter media



Take Away Lessons for Nuclear Forensic Sample Collection

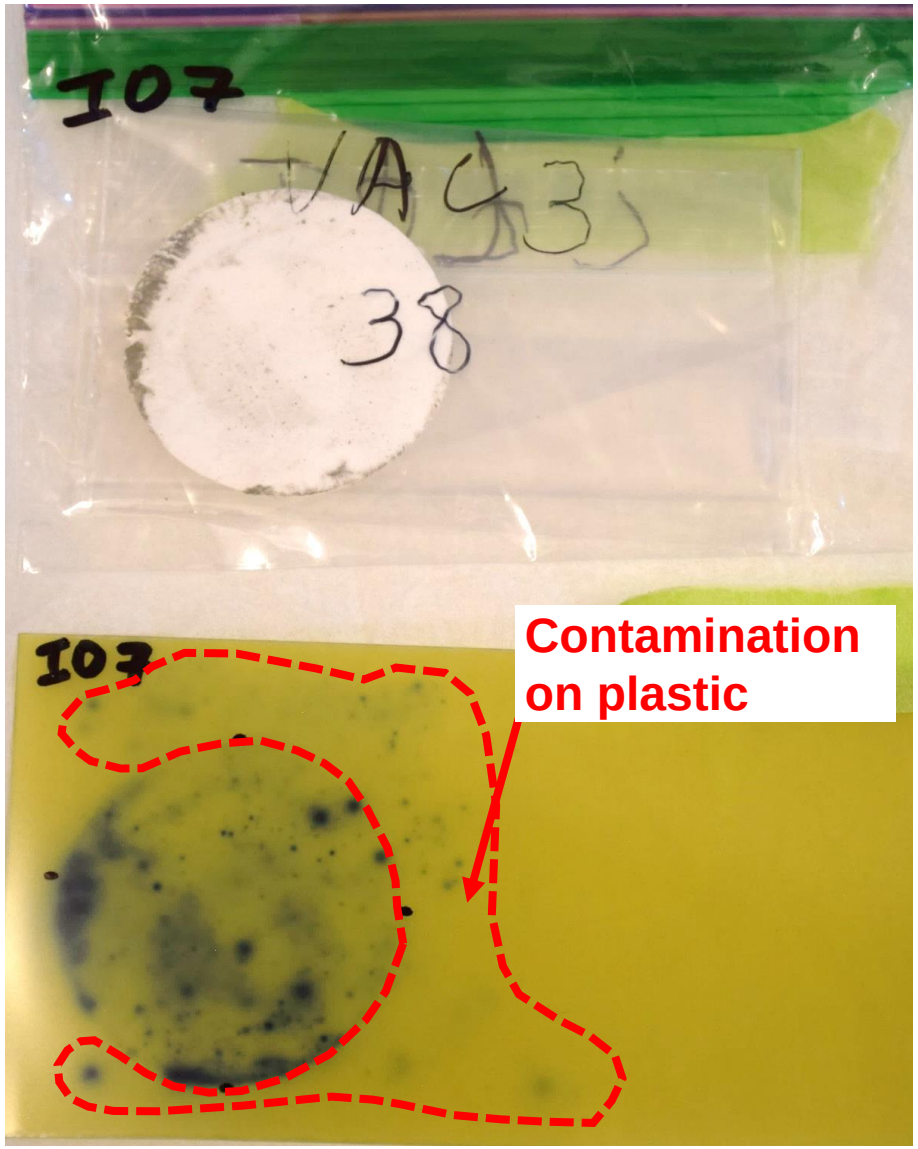
Observation #2

Rubbing of the smear against the inside surface of the poly bag appears to dislodge particulates and move them off the sample

Recommendations

Need to develop a better way to hold samples for storage and transport, consider mounting smear onto a SEM stub, and then use SEM sample holders

SEM stub in holding mount



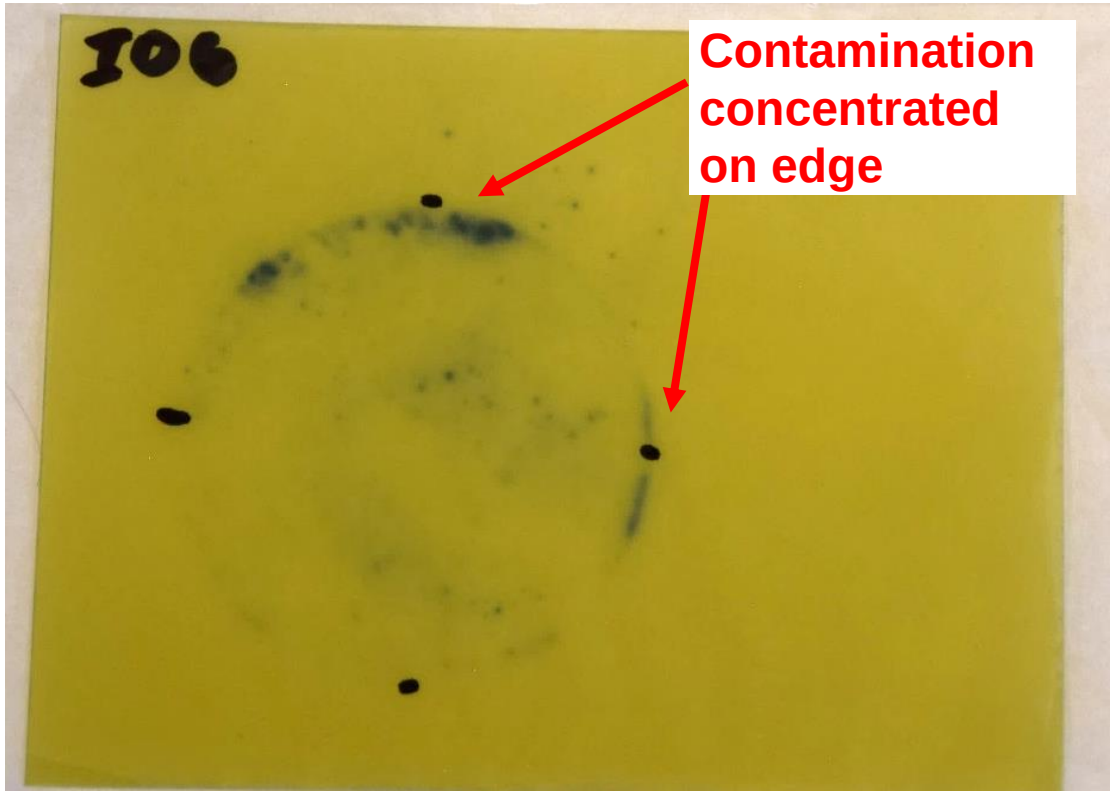
Take Away Lessons for Nuclear Forensic Sample Collection

Observation #3

Hand smearing can concentrate sample collection at the fingertips, and fingertip pressure can push particulate into the smear media

Recommendations

Should consider pushing straight down versus swiping, also consider using a foam backing to allow more even application of pressure



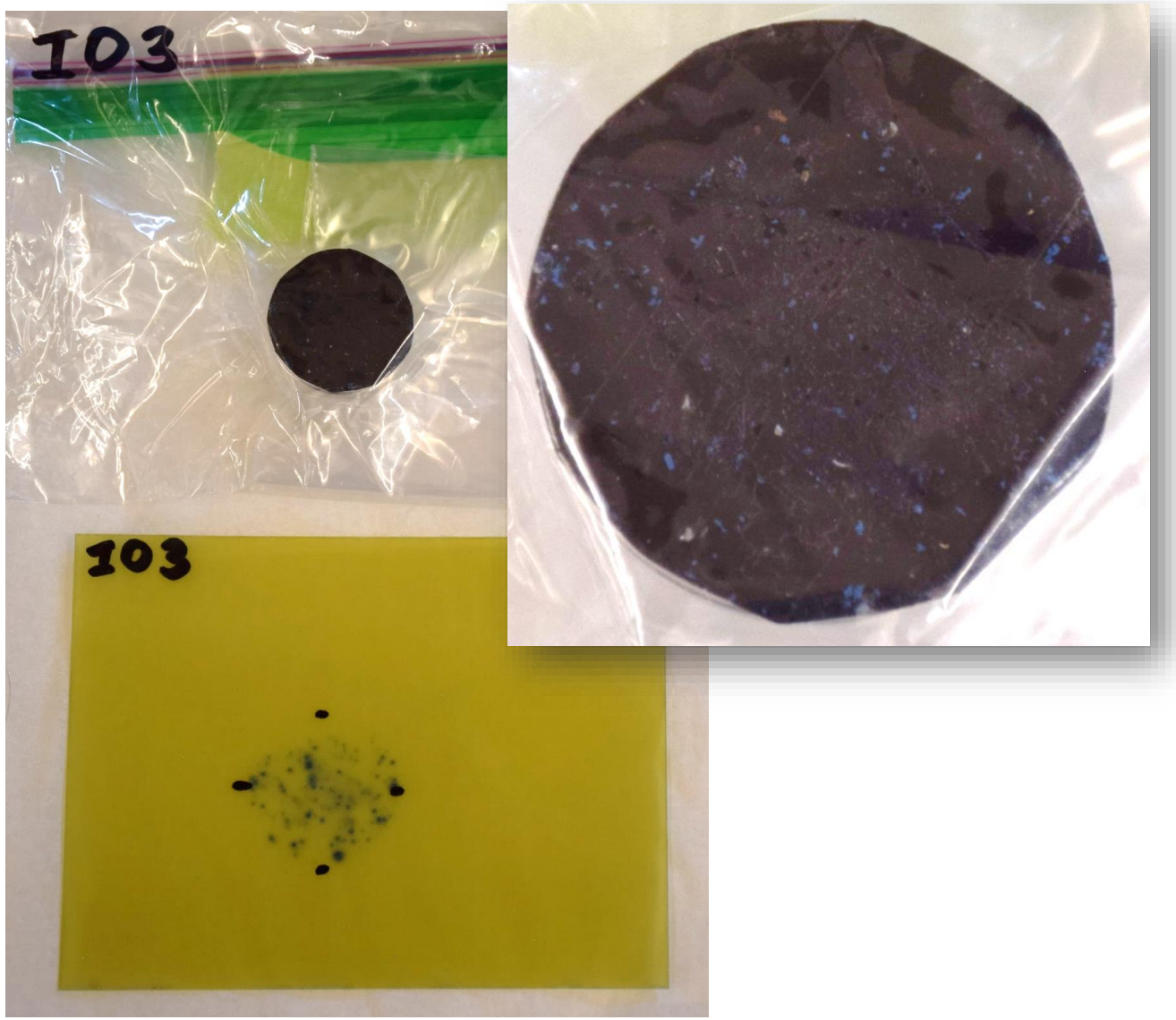
Take Away Lessons for Nuclear Forensic Sample Collection

Observation #4

SEM stub mounts with sticky, conductive tape may be ideal for sample collection, and allow direct transfer to SEM

Recommendations

Need to examine data collected from SEM stubs to assess quality



Take Away Lessons for Nuclear Forensic Sample Collection -- Other Observations

O#5: Direct gamma-ray scanning to assess dose works well

R:Need to improve detector models of efficiency, to reduce uncertainty

O#6: Use of stencil fiducial on ARAD film complicated image analysis

R: Need to improve ARAD fiducial, use color to allow separation (red vs. blue?)

O#7: Photography was performed ad hoc

R: Need better setup to perform photography, reduce glare from plastic bags, ensure focus, include reference scale, maintain magnification and orientation

O#8: SEM autoscan did poorly at finding hot particles

R: Need to optimize SEM autoscan spot size and speed

O#9: It was difficult to locate a fiducial in the SEM to match sample and ARAD

R: Consider pre-marking smear media (filters, SEM, etc.) with conductive ink/paint to allow position determination w.r.t. ARAD

O#10: Sample ID marking was inconsistent and irregular

R: Care needs to be taken to support chain of custody consistency

O#11: Health physics sampling techniques are not well matched for nuclear forensics

R: Consider development of specific NF evidence sampling kits, pre-deploy kits with first responders and train people in their use

Summary

- A recent real-world contamination release has opened our eyes to the challenges associated with the collection of nuclear forensics evidence
- It is clear that current best practices for collecting samples, in support of health physics surveys and contamination control, are not ideal for NF; methods can be improved with a focus on evidence quality and ease of use for later-stage analyses

https://www.energy.gov/nnsa/articles/nnsa-sponsoring-deep-dive-us-nuclear-forensics-capabilities

"Our ability to identify the origin of nuclear material, devices, or a detonation is a crucial pillar of deterrence."

JAY A. TILDEN
ASSOCIATE ADMINISTRATOR AND DEPUTY UNDER SECRETARY FOR COUNTERTERRORISM AND COUNTERPROLIFERATION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