TRTR 2024

Addressing Unclad and Unidentified Fissile Material in Highly Radioactive Debris

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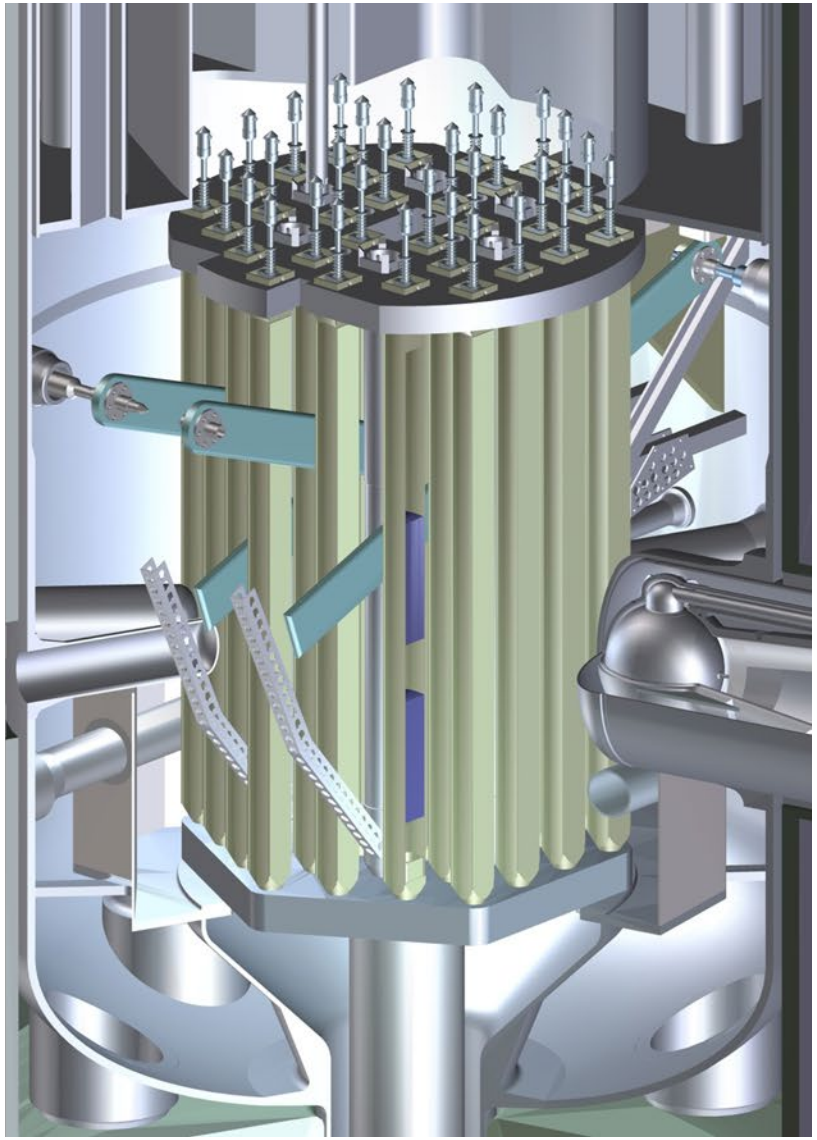
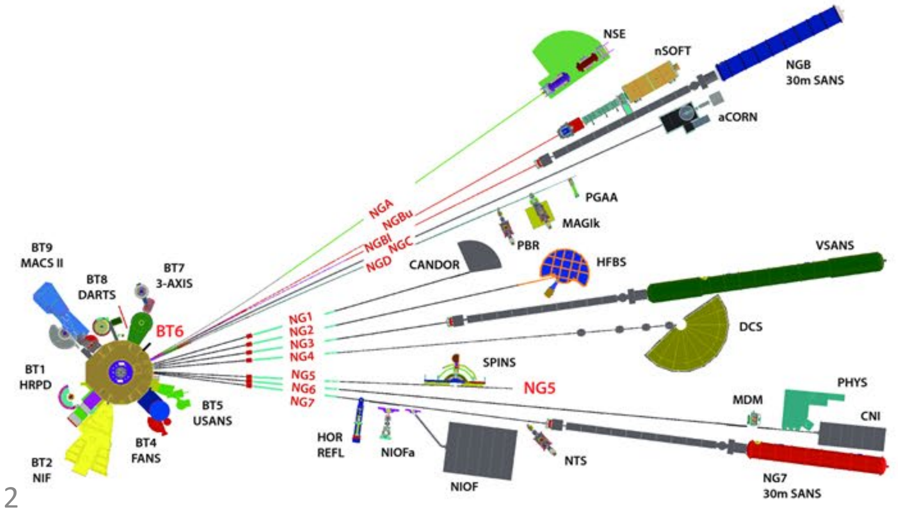
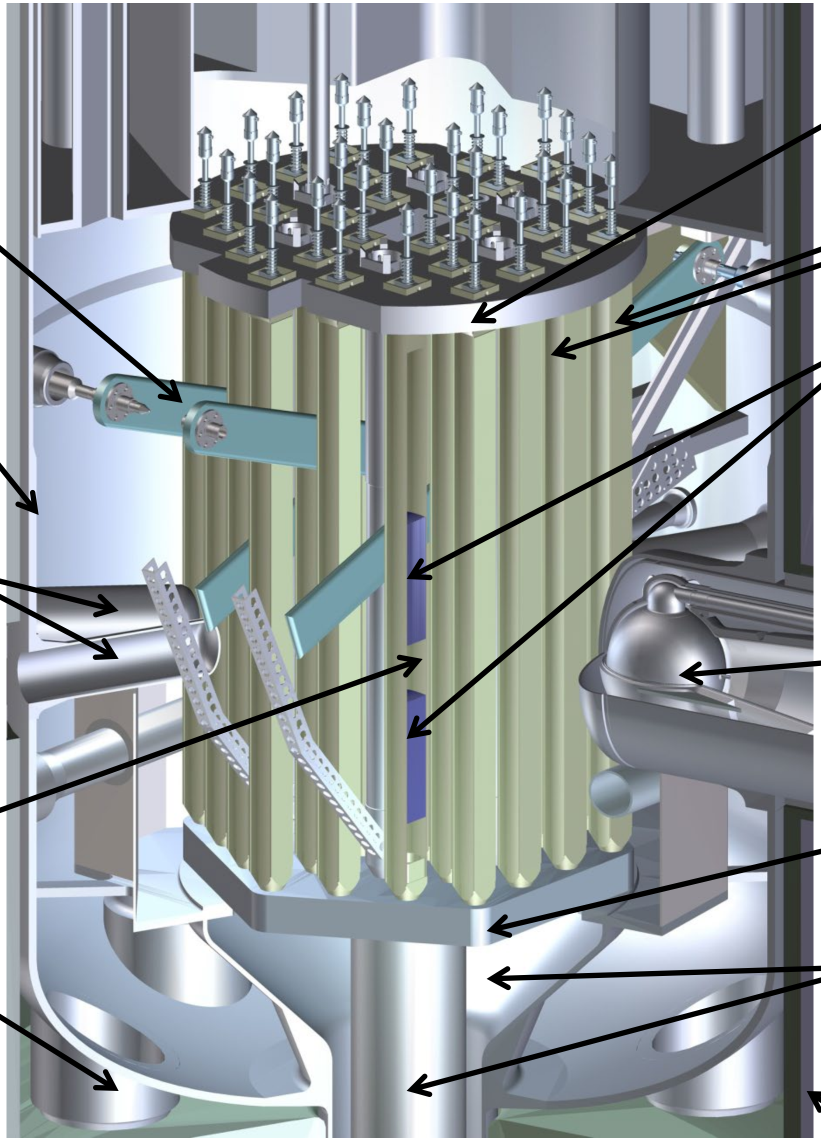
Introduction: NCNR & NBSR

- NCNR is one of the USA’s primary resources for neutron research
- NBSR history of successful operation since 1967
- NBSR license to expire in 2029
- New NIST neutron source (NNS) is being conceptualized





NBSR Core Layout



Cd Shim Safety Arms (4)

Reactor Vessel

Radial Beam Tubes (9)

Split Core: 18-cm Unfueled Gap – Flux Trap

Primary Outlet (2)

Top Grid Plate

Fuel Elements (30)

Fuel Plates

Liquid Hydrogen Cold Neutron Source

Bottom Grid Plate

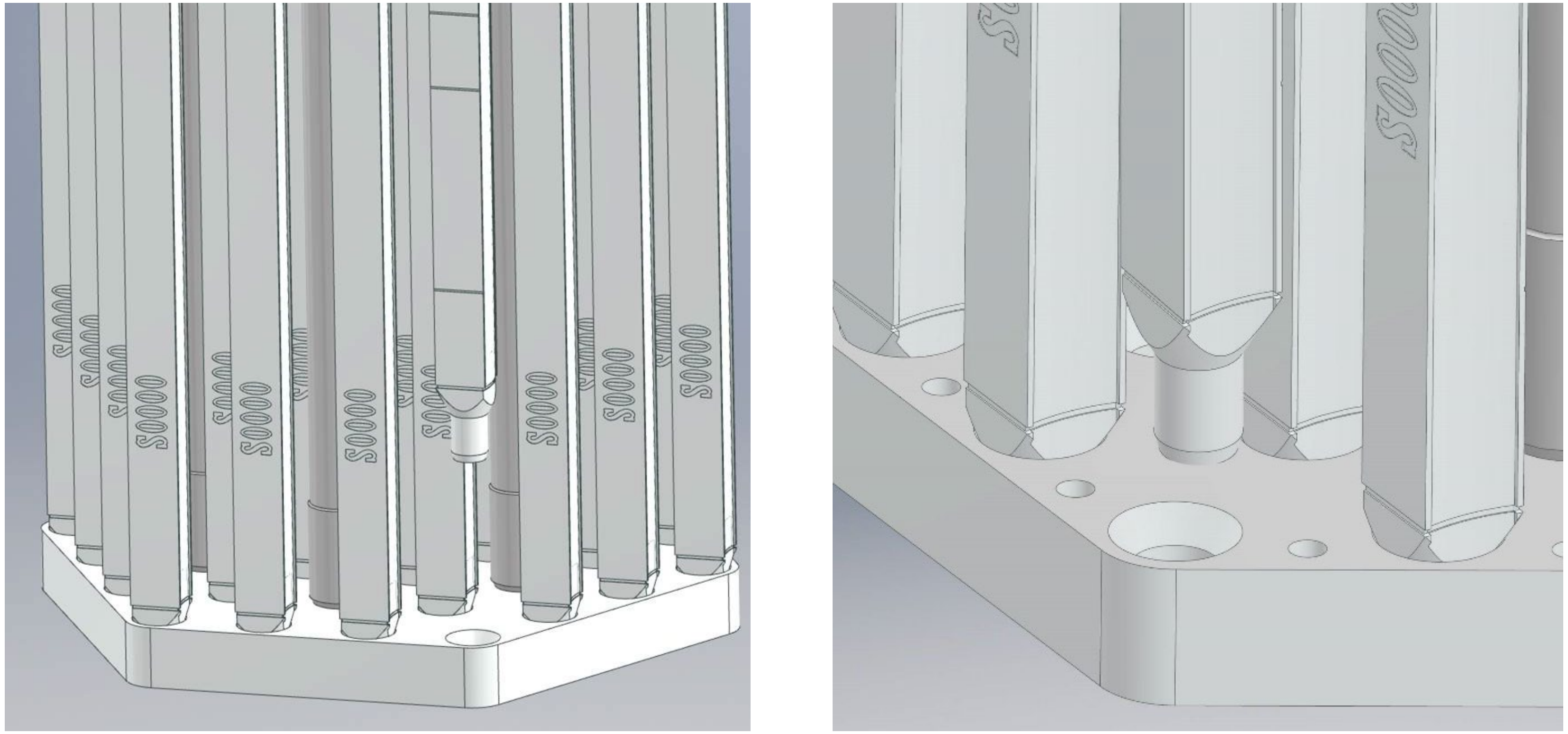
D_2O Primary Inlet Plenums

Thermal Shield

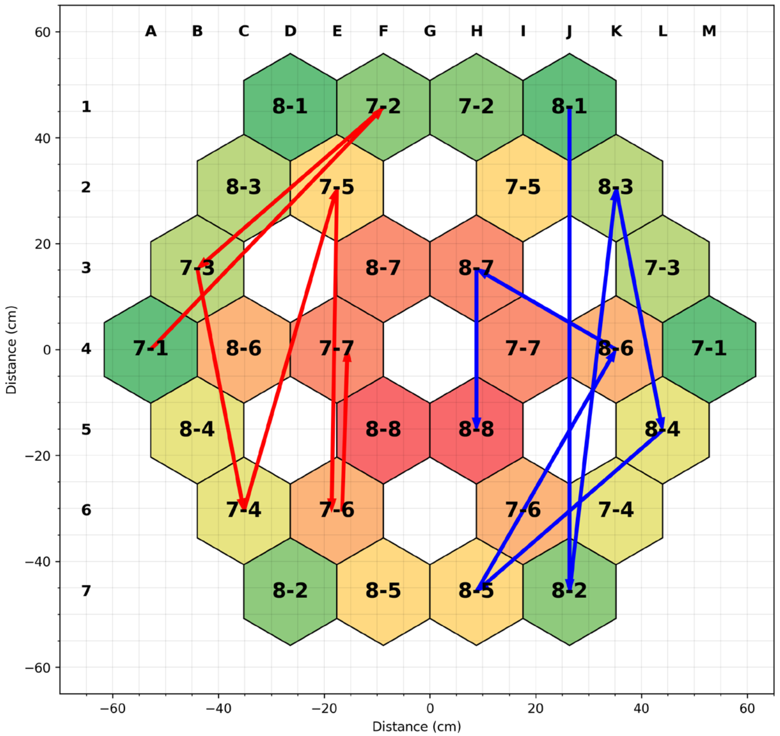
Introduction: NBSR

- HEU fuel (93% 235U) – U3O8+Al
  - 34 plates in a fuel element
  - Each fuel element has 350 g 235U
- 30 Fuel Elements
  - ~38-day fuel cycle at 20 MW
  - ~960 g of 235U consumed per cycle
  - 4 fresh elements per cycle (usually) with a management scheme to shuffle other elements in the core
  - 7 or 8 cycles per element depending on where it is loaded initially

Feb. 3, 2021 event: Unlatched Fuel Element



An unlatched element (J-7) floats on a jet of primary coolant emitted by the lower grid plate. (right) An unlatched element skews out of its conical seat due to multiple pump starts



NBSR Core Loading Pattern

Cleaning and Initial Operations

- Root Cause Evaluation Completed
- License Amendments (3),
- Startup Request, October 1, 2021
- 1st phase cleaning, throughout 2022
  - including manual pickup of large debris pieces, use of 20-micron-filtered dummy fuel elements to clean primary coolant, CO2 injection along with primary system filtration to encourage debris movement and vacuuming of the reactor pressure vessel and core internals.
  - ultrasonic cleaning was employed on primary piping hot spots to mobilize radioactive material.
- NRC Confirmatory Order, August 1, 2022
- March 16: initial criticality and startup to 50 kW

Cleaning 1st Iteration

- including manual pickup of large debris pieces
- use of 20-micron-filtered dummy fuel elements to clean primary coolant
- CO2 injection along with primary system filtration to encourage debris movement
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Helium Sweep Gas, Fission Products

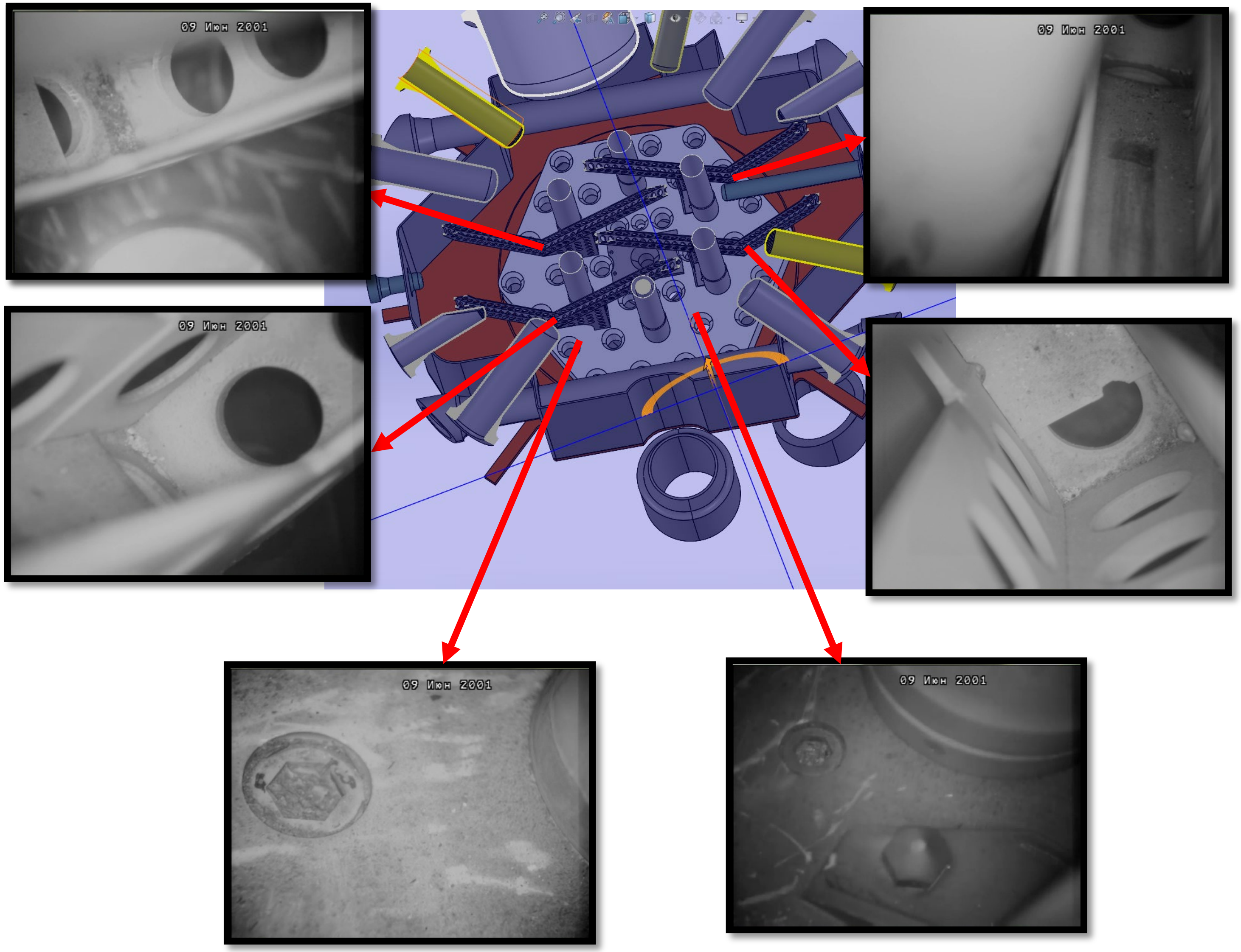
- Samples taken from the HSGS indicated the presence of Kr-85m, Kr-87, Kr-88, Kr-89, Xe-133, Xe-133m, Xe-135, Xe-135m, Xe-137, Xe-138.
- Using the gaseous FP activities, the mass of fuel/fuel-clad debris is estimated as 2.37 ± 0.66 g of fuel meat mixed with cladding (or 0.35 g of pure U3O8)
- Due to neutron flux variations within the vessel, this amount may vary up to 10 times based on the location.

Selected Gaseous Fission Product Activates Measured from He-Sweep System

Measurement Date	**Kr-87 (µCi/mL)**	**Xe-138 (µCi/mL)**
3/30/2023	1.42E-02	4.00E-02
3/30/2023	1.85E-02	3.80E-02
4/6/2023	1.76E-02	5.68E-02
4/26/2023	1.53E-02	5.68E-02
Average (µCi/mL)	1.60E-02	4.60E-02

Potential Locations

Location of Interest
3.5” thimble off core axis 1 (NE-J3)
3.5” thimble off core axis 2 (N-G2)
3.5” thimble off core axis 3 (NW-D3)
3.5” thimble off core axis 4 (SW-D5)
3.5” thimble off core axis 5 (S-G6)
3.5” thimble off core axis 6 (SE-J5)
3.5” thimble on axis (G4)
2.5” thimble 1 (E-J4)
2.5” thimble 2 (N-G3)
2.5” thimble 3 (W-D4)
2.5” thimble 4 (S-G5)
Lower Grid plate / Lower Reserve Pan floor
Lower Grid Plate center plug crevice
Underside Upper Grid Plate
LOCA distribution manifold
Inner reserve tank floor (worst case)
Shim Arm Catchers
Shim Arms



2nd Cleaning -> Need for Assessment Method

- Various methods available, how to know which is more effective?
- these include but not limited to direct removal of visible pieces, power washing and vacuuming different areas of the core
- Some debris may be dust form, visual means of inspection may not reveal any conclusive results
- How do we know when to stop?
- How much Uranium did we recover at a given time?
- Measurement methodology is proposed to effectively assess recovered Uranium
  - perform differential measurements of the U-235 mass in the filters or trash cans

Literature Review

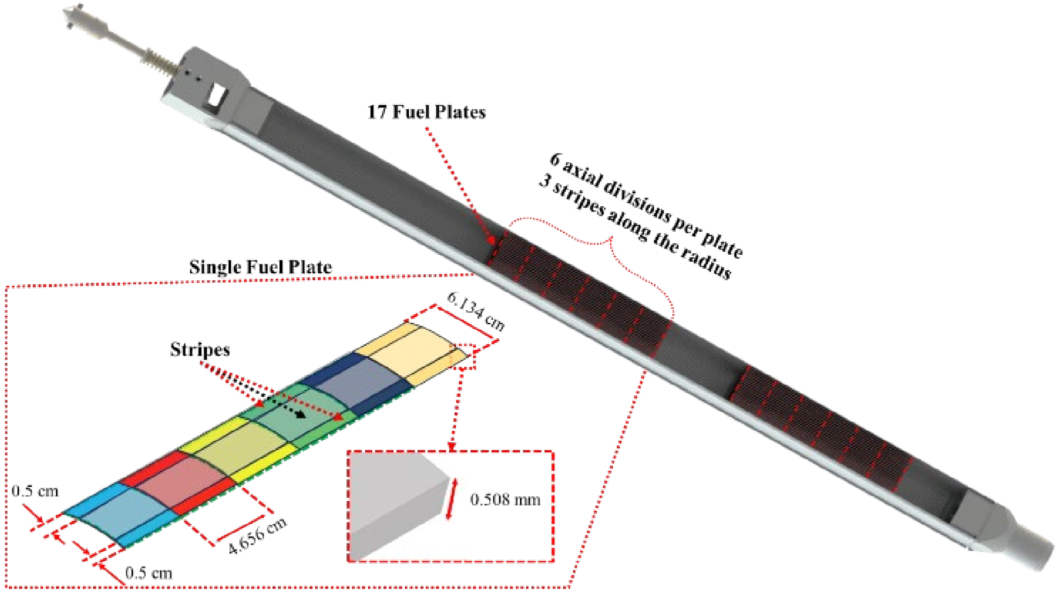
- burnup levels and uranium compositions of fuel elements have been under development since the early 1960s
  - B. E. Paige, “SIMPLIFIED METHOD FOR THE CALCULATION OF FISSION-PRODUCT ACTIVITIES AND CONCENTRATIONS,” Phillips Petroleum Co. Atomic Energy Div., Idaho Falls, Idaho, IDO-14542, Nov. 1960. doi: 10.2172/4031864.
  - K. Tasaka, “Estimation of irradiation history of a spent fuel by gamma-ray spectroscopy,” Nucl. Technol.; (United States), vol. 29:2, May 1976, Accessed: Jan. 30, 2024. [Online]. Available: https://www.osti.gov/biblio/7280940
- Recent studies have explored the use of short-lived fission product isotopes to measure the fissile compositions
  - J. Knowles, S. Skutnik, D. Glasgow, and R. Kapsimalis, “A generalized method for characterization of 235U and 239Pu content using short-lived fission product gamma spectroscopy,” Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment, vol. 833, pp. 38–44, Oct. 2016, doi: 10.1016/j.nima.2016.06.112.
  - Y. Ge et al., “Measurement of Cumulative fission product yields on 235U induced by 2.8 MeV neutrons,” Applied Radiation and Isotopes, vol. 200, p. 110907, Oct. 2023, doi: 10.1016/j.apradiso.2023.110907.
- these approaches have not addressed scenarios involving unclad and unidentified debris containing fissile material

Method

- Traditionally Cs-134 and Cs-137 isotopes are used for evaluating Uranium burnup
- Cs-134 and Cs-137 are soluble. At high temperatures, U3O8-Al fuel releases some of the Cs-137 content.
- When cladding is not intact, the amount of Cs-134 and Cs-137 releases significantly varied, ranging from 2% to 85%
  - R. E. Woodley, "Release of fission products from irradiated SRP fuels at elevated temperature Data report on the first stage of the SRP source term study," United States, 1986.
  - T. Shibata, K. Kanda, and K. Mishima, "Release of fission products from irradiated aluminide fuel at high temperature," Kyoto Univ., Kumatori, Osaka (Japan). Research Reactor Inst.; Argonne National Lab., IL (USA); Oak Ridge National Lab., TN (USA), CONF-821155-7, Jan. 1982. Accessed: Jan. 12, 2024. [Online]. Available: https://www.osti.gov/biblio/6487878
- isotopes Zr-95, Ru-106, Sb-125, Ce-144, and Eu-154 shown to be mostly within fuel matrix

Method - Activities

- Use Neutronic Model/Calculations to estimate relative activities of selected isotopes per initial gram of U-235
- Use licensed core model, and particular burnup for the failed element
- Calculate average values for the top and bottom section of the fuel element (along with uncertainty to account for variations)



Relative activities of select fission products per gram of initial U-235 loading, decay corrected to the date February 3, 2021

Activity (Ci/1g ^{235}U) ± Std Error (1σ)		
Isotope	Upper Section	Lower Section
Zr-95	2.50E+01 ± 4.63E+00	2.26E+01 ± 4.75E+00
Ru-106	4.67E-01 ± 8.70E-02	4.25E-01 ± 8.93E-02
Sb-125	1.58E-02 ± 2.94E-03	1.44E-02 ± 3.02E-03
Cs-134	3.39E-02 ± 1.03E-02	3.02E-02 ± 1.00E-02
Cs-137	2.72E-01 ± 5.08E-02	2.48E-01 ± 5.21E-02
Ce-144	8.03E+00 ± 1.49E+00	7.30E+00 ± 1.53E+00
Eu-154	8.30E-04 ± 2.58E-04	7.02E-04 ± 2.55E-04

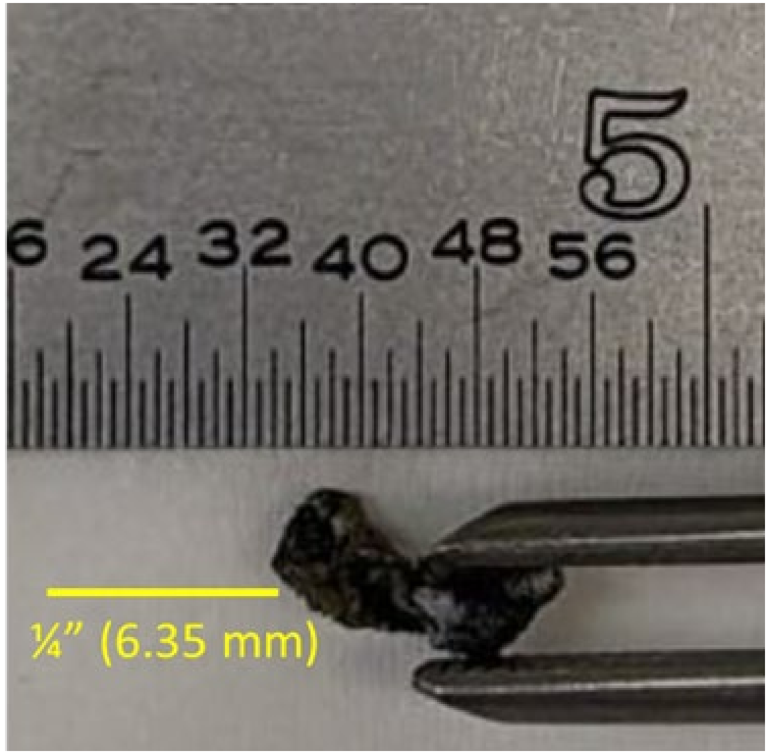
Method – Gamma Spectroscopy

- Apex-Gamma analysis package (Mirion)*
  - Efficiency based on LabSocs simulation of the measurement geometry
- A second analysis software, VRF (Snakedance Inc)*
  - VRF performs the least square fitting of the whole gamma spectrum that incorporates the Compton scattering (similar to Sandia Lab GADRAS) with internally simulated detector response
  - Absolute activity normalized to Cs-137 activity from Apex-Gamma analysis
- Estimate of amount of fuel in the recovered debris
  - The measured activity of each fission product is converted to mass based on the MCNP modeled specific activity (activity/g-U)

* Trade names and commercial products are identified in this paper to specify the experimental procedures in adequate detail. This identification does not imply recommendation or endorsement by the authors or by the National Institute of Standards and Technology, nor does it imply that the products identified are necessarily the best available for the purpose.

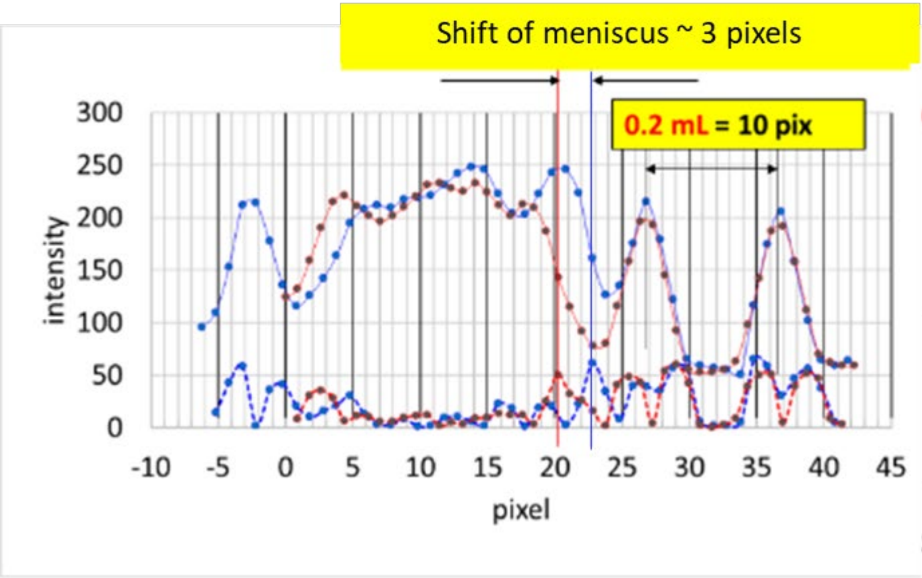
Method – Demonstration -Sample

- A sample piece recovered during the 1st cleaning, mass was 85.9 mg and 85.1 mg, density of 1.47 g/cm^3 +/- 0.3 g/cm^3

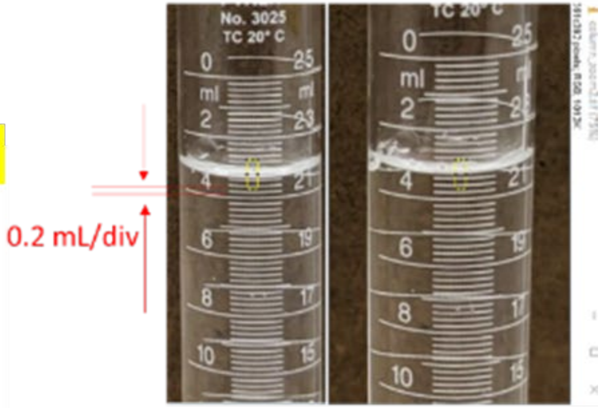


displaced water volume	0.058	cm^3
mass	0.0851	g
density	1.467	g/cm^3

Displaced water volume change = 0.058 mL = 0.058 cm^3



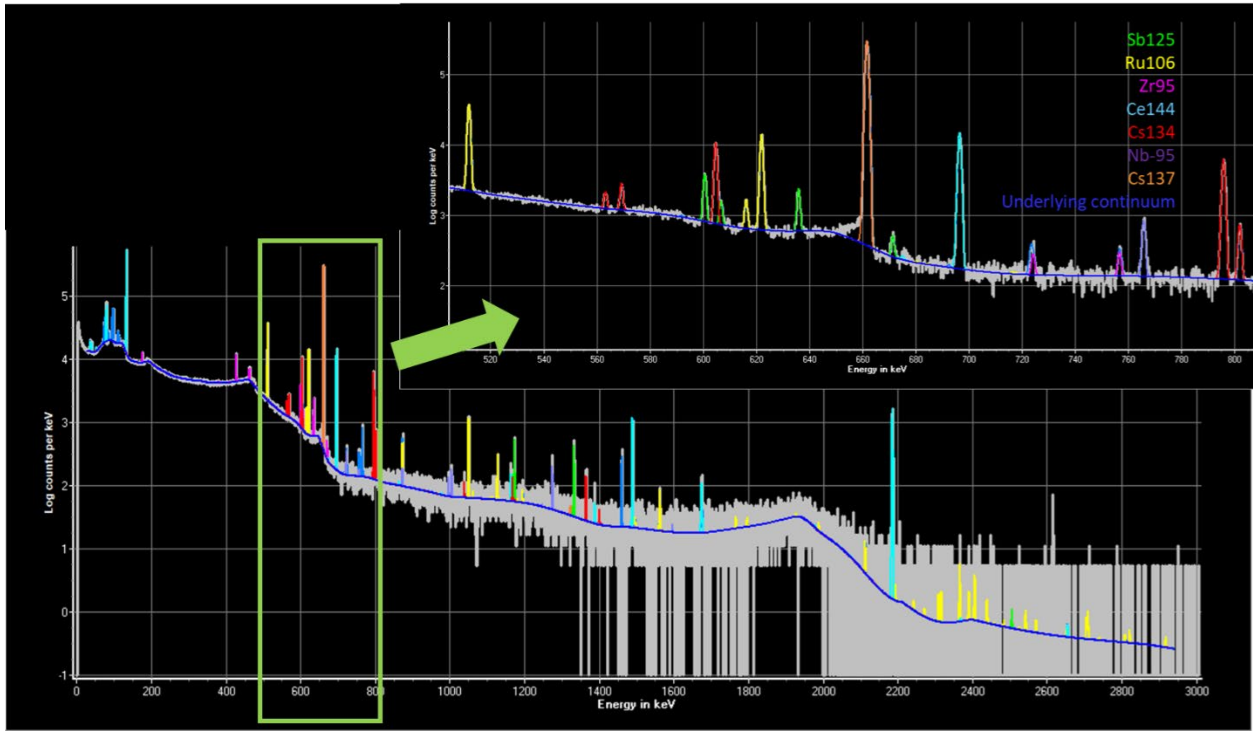
Submersion test, Tuesday March 5, 2024



Submerged piece

Method – Gamma Spectroscopy

- debris piece was placed at 16' 2.5" (4.95 m) from an HPGe detector for a 1-h live time gamma spectrum acquisition.
- determine the activities (Ci) of Zr-95, Ru-106, Sb-125, Cs-134, Cs-137, Ce-144, and Eu-154.



Method – Estimated U-235

- Derived U-235 mass based on MCNP calculated reference values for the upper and lower section, and the weighted mean and standard error of all 5 isotopes.

Isotope (VRF activity)	Upper Section (mg)	Lower Section (mg)
Cs-134	4.0 ± 1.3	4.6 ± 1.5
Cs-137	7.8 ± 1.5	8.5 ± 1.8

	Isotope	Apex	VRF
Upper Section	Zr-95	18.6 ± 5.5	11.0 ± 2.2
	Ru-106	10.5 ± 2.0	11.6 ± 2.2
	Sb-125	12.8 ± 2.4	11.6 ± 2.2
	Ce-144	9.1 ± 1.8	11.7 ± 2.2
	Eu-154	14.5 ± 7.9	9.7 ± 3.0
	Weighted Mean	10.9 ± 1.1	11.2 ± 1.0
Lower Section	Zr-95	20.6 ± 6.4	12.1 ± 2.8
	Ru-106	11.6 ± 2.5	12.7 ± 2.7
	Sb-125	14.1 ± 3.0	12.7 ± 2.7
	Ce-144	9.9 ± 2.2	12.9 ± 2.7
	Eu-154	17.1 ± 9.9	11.4 ± 4.2
	Weighted Mean	12.0 ± 1.4	12.5 ± 1.3

Method – Results

- Maximum and minimum weighted mean U-235 content was found as (12.5 ± 1.3) mg and (10.9 ± 1.1) mg, respectively (equivalent to around 40 mg U_3O_8-Al fuel meat).
- Zr-95 resulted in the highest estimate as (20.6 ± 6.4) mg U-235.
- The mean value for the upper and lower section, excluding Cs isotopes was (11.2 ± 1.0) mg and (12.5 ± 1.3) mg of U-235, respectively

Method – Results

- Zr-95, Ru-106, Sb-135 and Ce-144 provided most consistent results when analyzed with VRF and would be used in future assessments.
- About 60% of Cs-134 and 30% of Cs-137 is released from the piece.
- MCNP calculated Cs-134/Cs-137 ratio for the upper and lower sections were 0.125 and 0.122
- Measured Cs-134/Cs-137 ratio with Apex and VRF was 0.065 and 0.066
- Cs ratio would indicate inaccurate burnup/uranium amount

Conclusions and Future Work

- described our initial efforts to develop a traceable and quantitative method for assessing the U-235 content in highly radioactive debris
- Demonstration of the method is shown by assessing a piece of debris containing U-235.
- Acquire more information through repeated measurements of more debris pieces as recovered from the core.
- Derive isotope activity ratio’s as a function of burnup, seek linear relations such as Cs ratio (Cs-134/Cs-137 (or Ru-106)) but instead use Zr-95, Ru-106, Sb-125 and Ce-144
- activity ratios of Ru-106/Sb-125, Cs-134/Cs-137, Cs-134/Ru-106, Cs-144/Sb-125, and Ru-106/Ce-144 as a function of MWD/kgU

Thank you for listening!

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Questions??

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