Destructive and Nondestructive Analyses of Low-Burnup Low-Enriched Uranium for the Validation of MCNP

MTV Workshop, 2022

March 23, 2022

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Introduction and Motivation

- Test the accuracy of MCNP6.2 for isotopic accumulation. And experimentally measure proliferation signatures of foreign nuclear fuel cycles.
  - Burnup, reactor-type, TSI.
- Classic validation of MCNP burnup simulations using radiochemical measurements. The MURR was used for experimental and simulation work. Two LEUO_2 pellet (17 and 18 mg) were irradiated to ~1 GWd/MTU.
- This study also builds on the previous nuclear forensic studies completed at Texas A&M University.

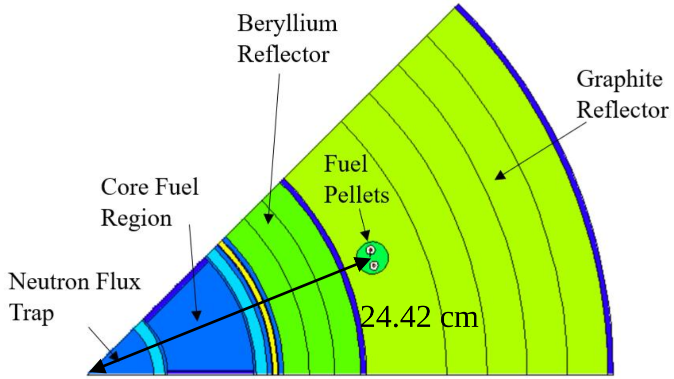
Mission Relevance

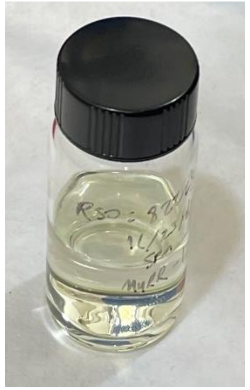
- This work falls under Thrust Area 2: Signals and Source Terms for Nuclear Nonproliferation.

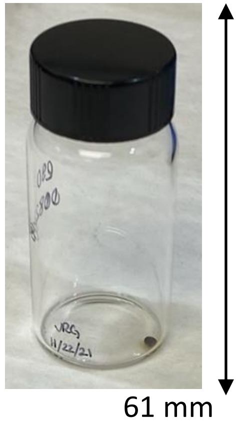
  "Preventing nuclear weapons proliferation and reducing the threat of nuclear and radiological terrorism around the world are key U.S national security strategic objectives that require constant vigilance."

Technical Approach

- Two ~17-mg $LEUO_2$ pellets were irradiated in the MURR to a burnup of 1 GWd/MTU.
- The study can be broken into two sections: Simulation and experimental.
- Simulation: MCNP6.2
- Experimental: HPGe, PIE, and ICP-MS

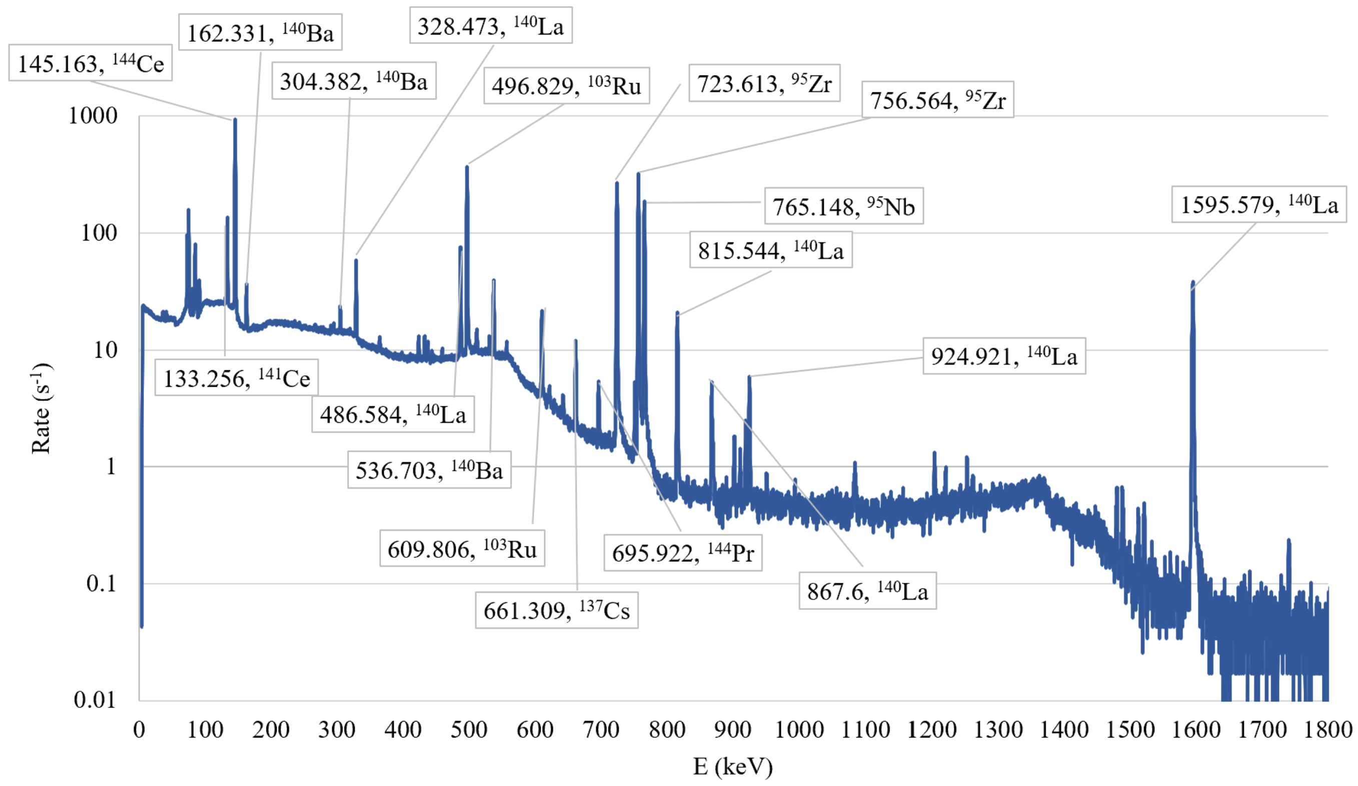






(Clockwise starting top-left) Visualization of pellet locations in MURR core. Opened irradiation can with two pellets. Undissolved pellet. Dissolved pellet

Results

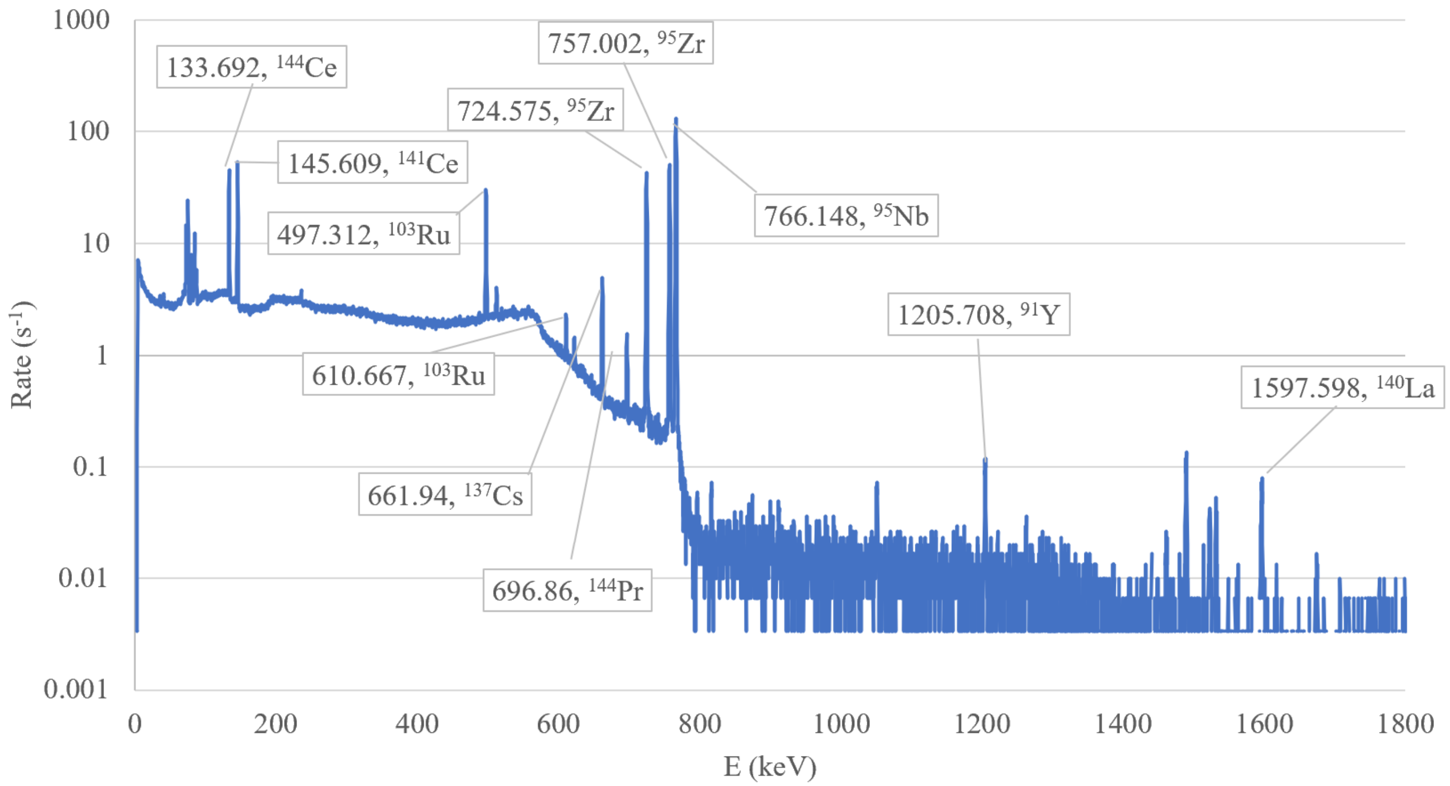


Nuclide	Calculated mass/U mass	MCNP mass/U Mass	S/E
^{133}Cs	(4.00±0.08) E-05	3.33E-05	0.83
^{137}Cs	(3.79±0.08) E-05	3.22E-05	0.85
^{148}Nd	(1.03±0.02) E-05	1.12E-05	1.09
^{149}Sm	(2.20±0.10) E-06	2.39E-06	1.08
^{150}Sm	(3.88±0.12) E-06	3.74E-06	0.96
^{152}Sm	(2.27±0.12) E-06	2.09E-06	0.92
^{153}Eu	(9.40±0.48) E-07	9.42E-07	1.00
^{154}Eu	(7.76±0.83) E-09	6.83E-09	0.88
^{239}Pu	(2.97±0.05) E-04	3.00E-04	1.01
Pu(all)	(2.99±0.06) E-04	3.02E-04	1.01

	Average Burnup (GWd/MTU)
S	0.873±0.011
E	0.940±0.025
S/E	0.929±0.027

Top-left: HPGe gamma spectrum of irradiated LEUO_2 material 70 days since discharge. Top-right: S and E values of ICP-MS measured nuclides. Bottom: S and E values of burnup.

Results (cont.)



Ratio	Calculated TSI (70 d)	Calculated TSI (173 d)
$^{95}Zr/^{137}Cs$	84.9	171.6
$^{103}Ru/^{137}Cs$	80.0	175.5
$^{141}Ce/^{137}Cs$	73.9	176.5
$^{144}Ce/^{137}Cs$	94.4	211.4
$^{95}Zr/^{144}Ce$	82.7	161.2
$^{103}Ru/^{144}Ce$	77.8	170.2
$^{141}Ce/^{144}Ce$	71.5	172.5
$^{103}Ru/^{95}Zr$	71.7	180.7
$^{141}Ce/^{95}Zr$	62.5	181.4
$^{141}Ce/^{103}Ru$	45.4	182.9
Average	74.5±4.0	178.4±4.0

Left: HPGe gamma spectrum of irradiated $LEUO_2$ material 173 days since discharge. Right: TSI estimations using nuclide ratios measured with HPGe.

Expected Impact

- Expand nuclear forensic database of experimental nuclear forensic signatures.
- Demonstrate strengths and weaknesses of MCNP burnup simulation of $LEUO_2$ in predicting certain proliferation signature isotopes.

MTV Impact

- Human capacity building in radiochemistry and reactor physics to support nuclear forensics and nonproliferation work.
  - Including internships: ANL and LANL.
- Collaboration with LANL to measure $^{239}Pu/^{240}Pu$ total-Q using a method, Decay Energy Spectroscopy (DES).

Conclusion

- This work builds upon and add new data to support nuclear forensics and proliferation signatures for $LEUO_2$.
- Good agreement (<15% difference):
  - HPGe: ^{95}Zr, ^{103}Ru, ^{140}Ba, ^{140}La, and ^{144}Ce.
  - ICP-MS: ^{137}Cs, ^{148}Nd, ^{149}Sm, ^{152}Sm, and ^{154}Eu.
- Excellent agreement (<5% difference):
  - HPGe: ^{137}Cs and ^{141}Ce.
  - ICP-MS: ^{150}Sm, ^{153}Eu, and ^{239}Pu
- Poor Agreement (>20% difference):
  - HPGe: ^{95}Nb.
  - ICP-MS: ^{134}Cs, ^{135}Cs, ^{136}Ba, and ^{138}Ba.
- The experimental and computational works support identification of plutonium (burnup, reactor-type, TSI) produced in foreign nuclear fuel cycles.

Next Steps

- Proliferation signatures measured in this work will support prior and ongoing tools from Texas A&M University.
  - Maximum Likelihood Methodology[Osborn et al.]
  - Machine Learning Algorithm[O'Neal et al.]

References

- J.M. Osborn et al., "Experimental Validation of a Nuclear Forensics Methodology for Source Reactor-Type Discrimination of Chemically Separated Plutonium", *Nuclear Engineering and Technology*, 51, 2 (2019).
- O'Neal, Patrick J., Sunil S. Chirayath, and Qi Cheng. "A Machine Learning Method for the Forensics Attribution of Separated Plutonium." *Nuclear Science and Engineering* (2022): 1-13.

Acknowledgements

The Consortium for Monitoring, Technology, and Verification would like to thank the NNSA and DOE for the continued support of these research activities.

This work was funded by the Consortium for Monitoring, Technology, and Verification under Department of Energy National Nuclear Security Administration award number DE-NA0003920