Preliminary Findings from RIMS Analysis of Spent Fuel

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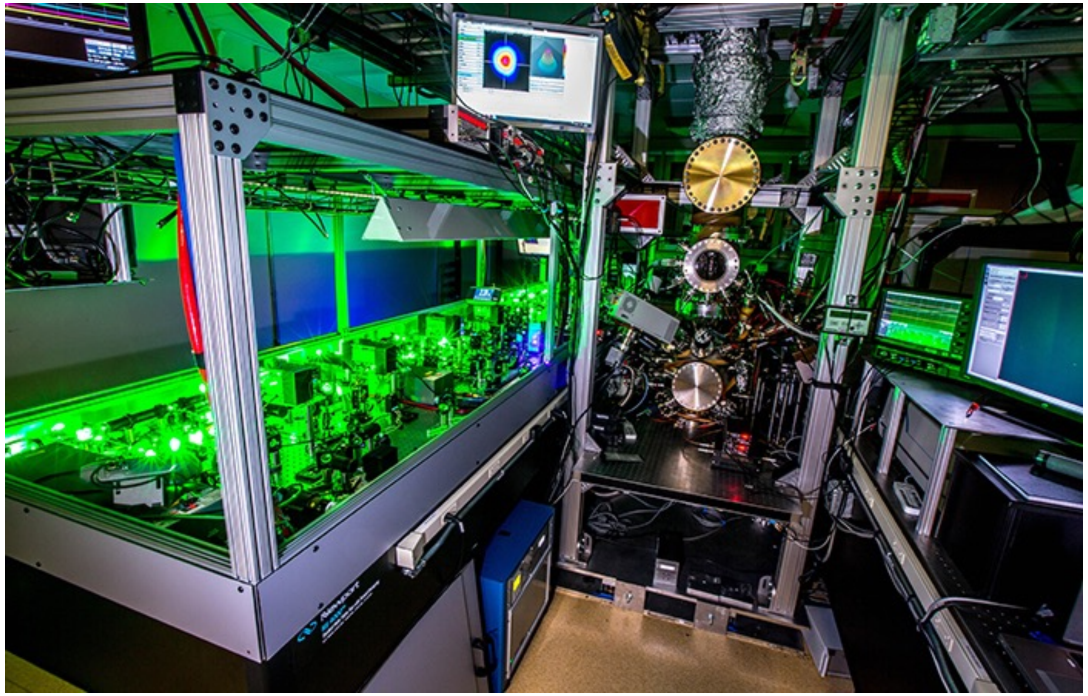
Introduction and Motivation

- Novel forms of isotopic analysis are generally useful for increasing the scope and precision of nuclear forensics.
- Ideally, a method of isotopic analysis should be accurate, useful, easily applicable, and suited to the isotopes of interest.
- Isotopes with small concentrations (ppb or ppt) may be forensically useful but require sophisticated mass spectrometry methods to measure.
- Resonance Ionization Mass Spectrometry is one such method.

Resonance Ionization Mass Spectrometry

- Resonance Ionization Mass Spectrometry (RIMS) uses tuned lasers to selectively ionize the element of interest.
- The ions are accelerated through an electric field.
- Isotopes can be distinguished from each other by time-of-flight (ToF) measurements.
- RIMS is highly accurate for many isotopes, distinguishing concentrations on the order of ppb or ppt.



Mission Relevance

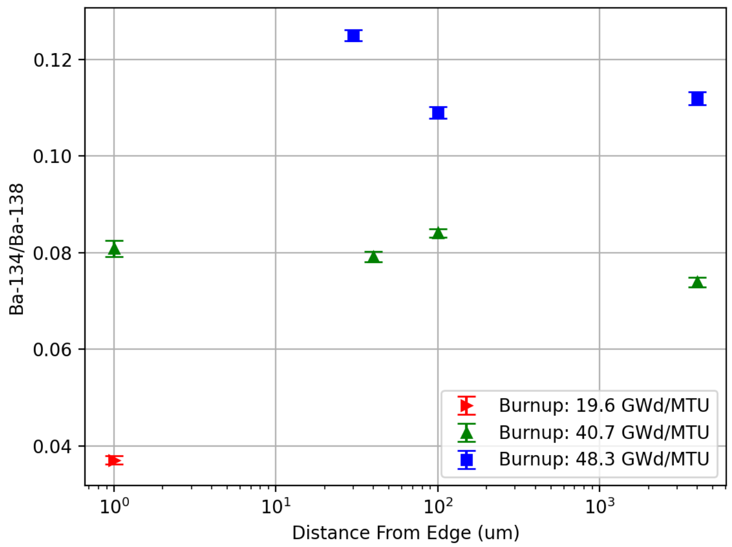
- This work relates to the NNSA mission by identifying and verifying applications of RIMS to nuclear forensics.
- As part of NNSA efforts to prevent weapon proliferation, it is necessary to have robust analysis methods for spent nuclear fuel to determine whether any nuclear material has been diverted from the fuel cycle.
- An understanding of the effects of reactor type and irradiation time on isotope ratios and the ability to measure those isotopes increases the ability to analyze spent fuel samples for nonproliferation studies.
- Focusing on isotopes that have not previously been used for nuclear forensics will expand the possibilities of spent fuel analysis and may help identify attempts to spoof isotopic signatures.

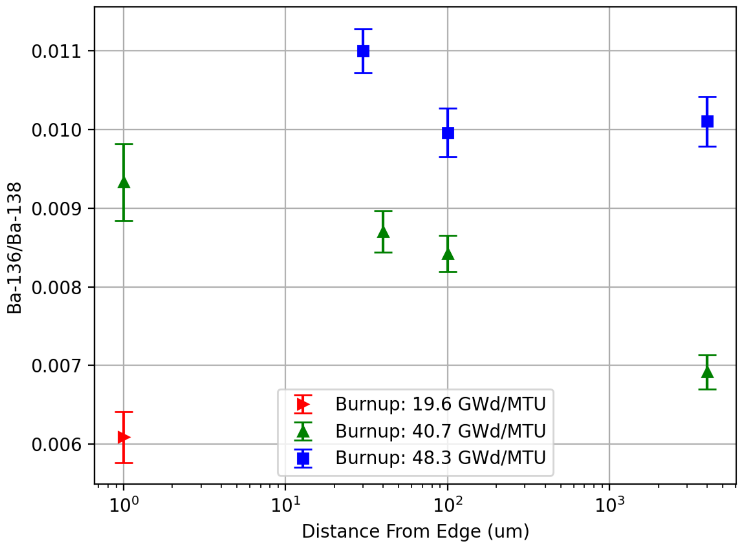
Technical Approach

- During summer 2023, RIMS analysis of spent fuel cubes was conducted at LLNL.
- Measurements focused on barium and molybdenum isotope ratios in cubes taken from different locations within a Belgian PWR.
- Samples of pure barium and molybdenum were measured to establish ionization rates and serve as a point of comparison for subtracting background contamination from the spent fuel measurements.

Results

- Experimental work focused on barium isotopes, present in small quantities (parts per 100 million), but stable (useful for analysis of older fuel samples).
- Ba-134/Ba-138 and Ba-136/Ba-138 were observed to vary substantially with burnup (dependent on axial position) but less with radial position within fuel pellets (Ba-134 especially stable over radial position).

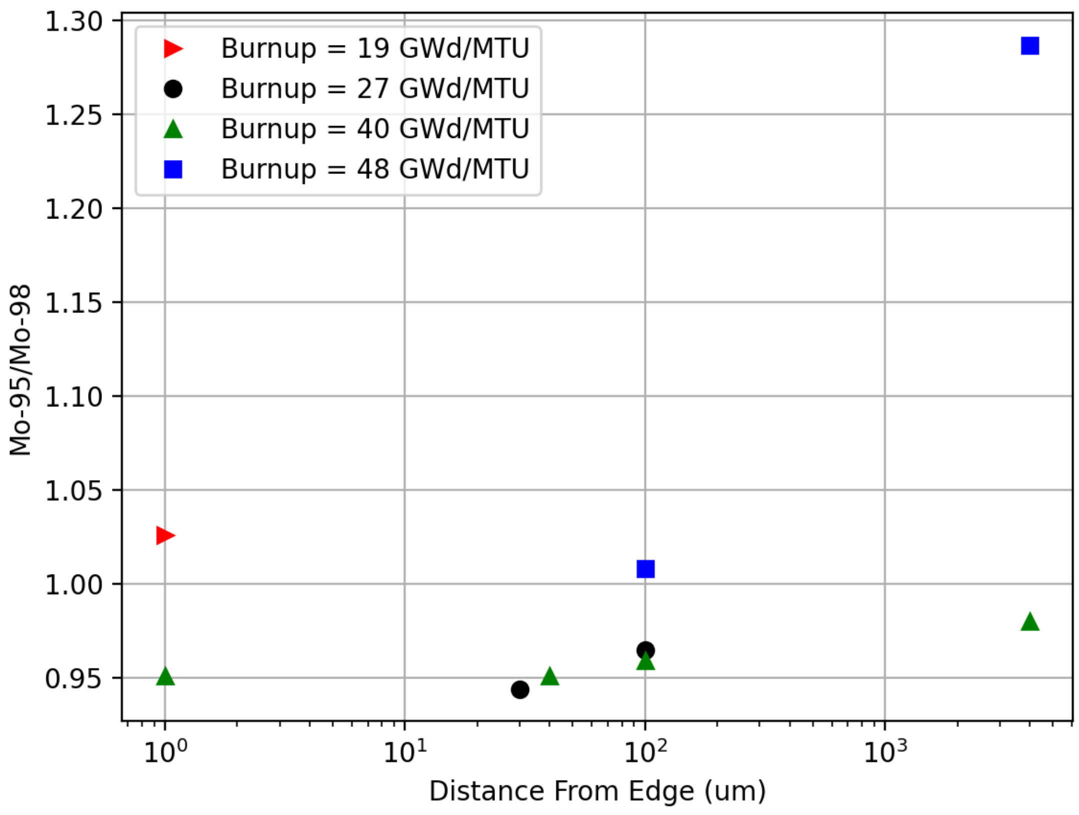




National Nuclear Security Administration

Results

- The molybdenum data is significantly more confusing, and it is unclear what role environmental contamination played in these results.
- The use of a gallium ion gun for sputtering may have produced gallium aluminide.



Expected Impact

- If successful, this work will provide additional forensic signatures to nuclear forensics teams and facilities inspectors.
- These new signatures could be used in tandem with existing signatures to provide more precise results.

MTV Impact

- This work was conducted at LLNL in collaboration with Michael Savina.
- It was made possible through connections fostered by MTV fellows.
- Future continuation of this work will similarly rely on collaboration with the RIMS team at LLNL, facilitated by MTV.

Conclusion

- Barium isotope ratios, particularly Ba-134/Ba-138, should serve as excellent position-independent burnup monitors. They exhibit low levels of environmental contamination and can be measured by RIMS even decades after removal from the reactor.
- Molybdenum may be able to serve a similar purpose, but further work is needed to determine whether the observed overlap in measurements between samples with different burnups is real or due to contamination.

Next Steps

- Conduct additional simulations in SCALE to determine likely isotopic ratios of barium and molybdenum under different conditions.
- Take additional spent fuel measurements, ideally from multiple reactors, to confirm results.
- Redo molybdenum measurements using laser sputtering to avoid gallium aluminide contamination.

Acknowledgements

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