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Extension of Hybrid K-edge Densitometry for Pyroprocessing Material Accountancy Measurements

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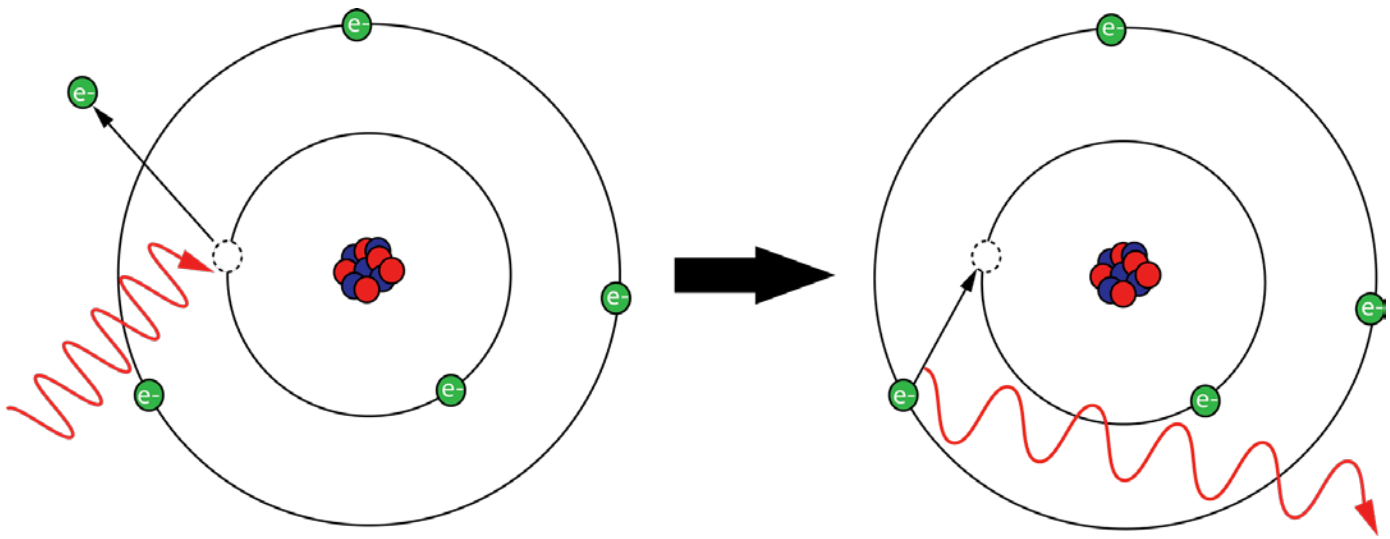
Agenda

- Hybrid K-edge densitometry (HKED) as a measurement method
- Application to pyroprocessing
- Discussion of physical processes involved in HKED
- Description of the Canberra HKED system
- MCNP6 model
- Aqueous validation cases
  - Uranium only
  - Uranium-plutonium
- Scoping studies
  - Molten pyro-salts from Mk. 4 electrorefiner
  - Voloxidation powders
  - Thorium surrogate
- Future work
- Conclusions

Hybrid K-edge Densitometry (HKED)

- HKED is a candidate for material accountancy measurements in pyroprocessing
  - Ability to measure through optically thick materials
  - Ability to make measurements in high radiation fields
- Modification and scoping studies with existing systems is sometimes not possible
  - High activity of pyrochemical solutions require a hot cell
  - Systems are rare and expensive

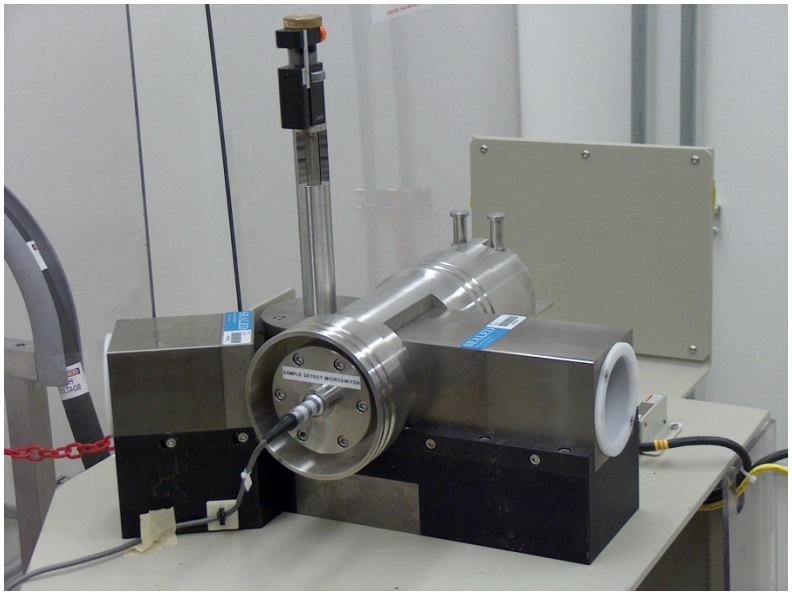
Physics Behind HKED



- K-edge densitometry
  - Tuned photon beam is directed at a sample of known geometry
  - Photons at the binding energy of an element's K-shell electrons are preferentially absorbed
  - Transmission difference at different concentrations of an element of interest are measured
- X-ray fluorescence
  - K-shell electrons excited by the photon beam leave a vacancy in the shell
  - Electrons from higher shells fill the vacancy emitting a characteristic x-ray
  - X-ray intensities indicate elemental concentration

Canberra HKED System

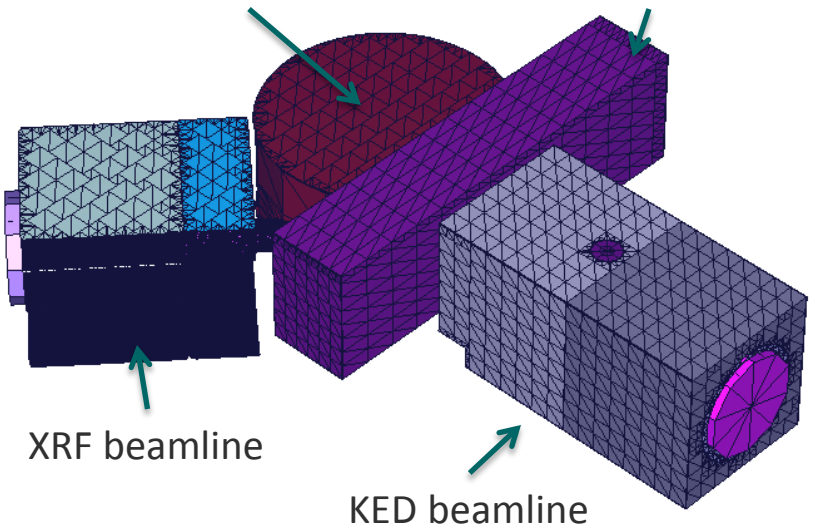
- Located at ORNL's REDC
- Components
  - X-ray source
  - Low energy HPGe detectors
  - Sample transfer system
  - Tungsten collimators
  - Beam filters
  - Shielding
- Currently used in commercial aqueous reprocessing operations



HKED system with exterior shielding removed

MCNP6 Model

- Model developed using measurements of HKED system in MCNP6
- Initial simulations used pulse height tally on two detector crystal volumes
  - Simulations required approximately 48 hours and 10^{12} starting particles to converge



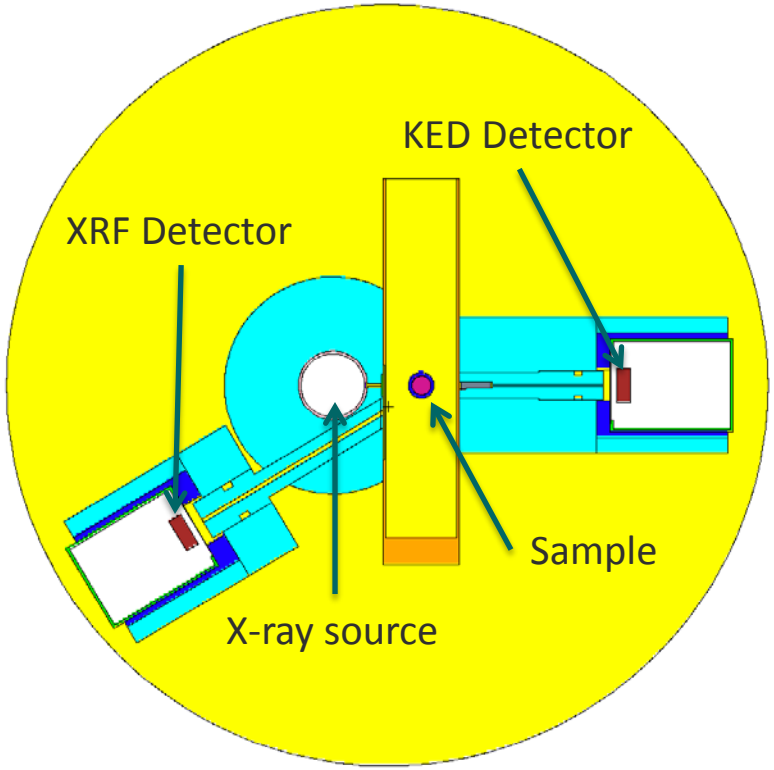
3D rendering of HKED external geometry using VisED

Mixed Solution Strategy

- Splitting problem into two separate simulations allowed variance reduction methods to be applied to each component as needed
  - XRF and KED portions required unique VR techniques
- Problem was further partitioned into stages
  - Transport using a F2 surface flux tally
    - Tally surface uses a small disc at the end of the beamline
  - Detector response using a F8 pulse height tally
    - Uses tally from first stage as source term for second

Application of variance reduction was dependent on beam line characteristics

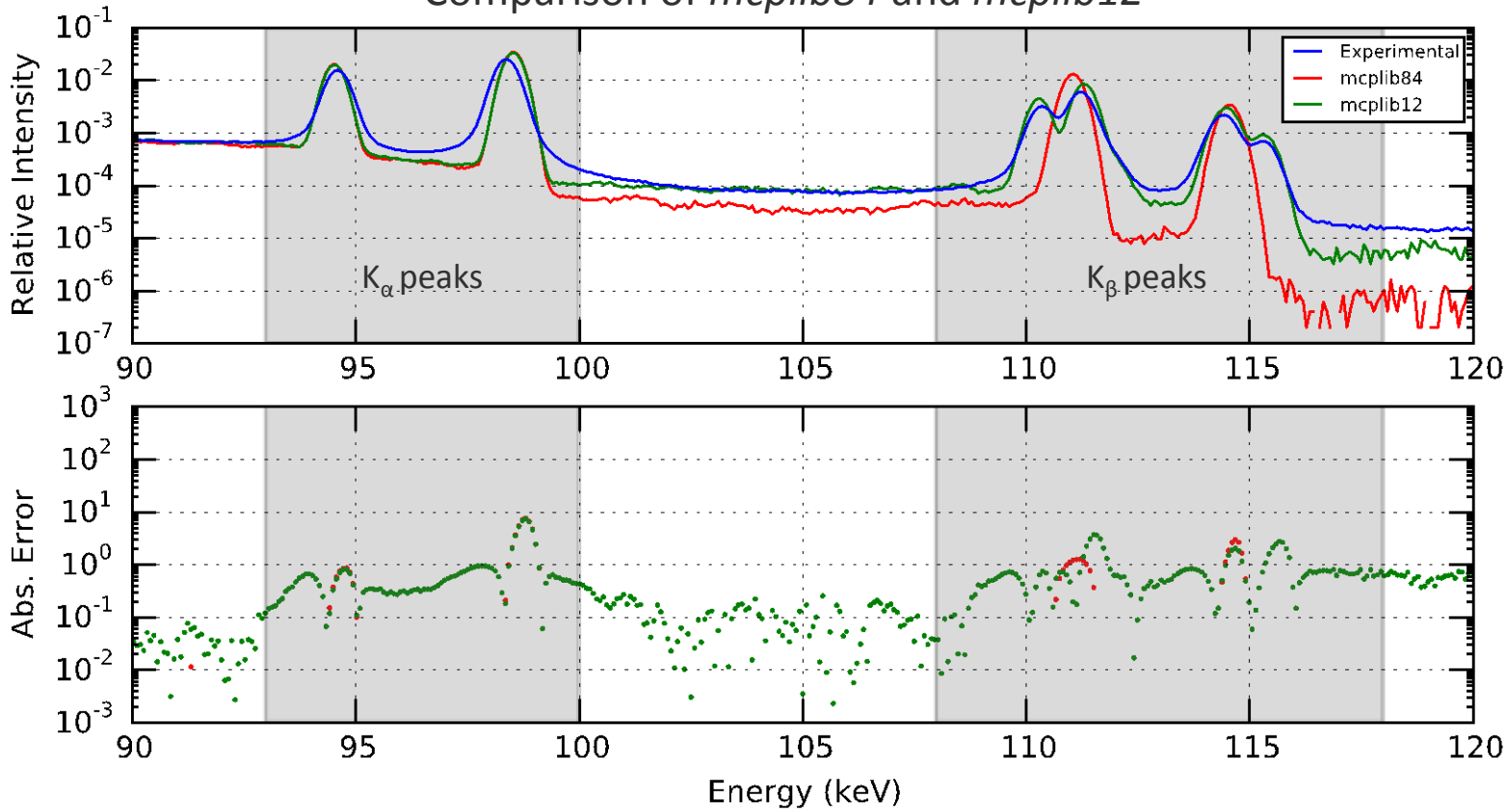
- XRF simulations
  - Forced collisions in the sample
  - Deterministic transport (DXTRAN) sphere
  - Energy & spatial weight window mesh
  - Source biasing
- KED simulations
  - DXTRAN sphere
  - Energy & spatial weight window mesh
  - Source biasing



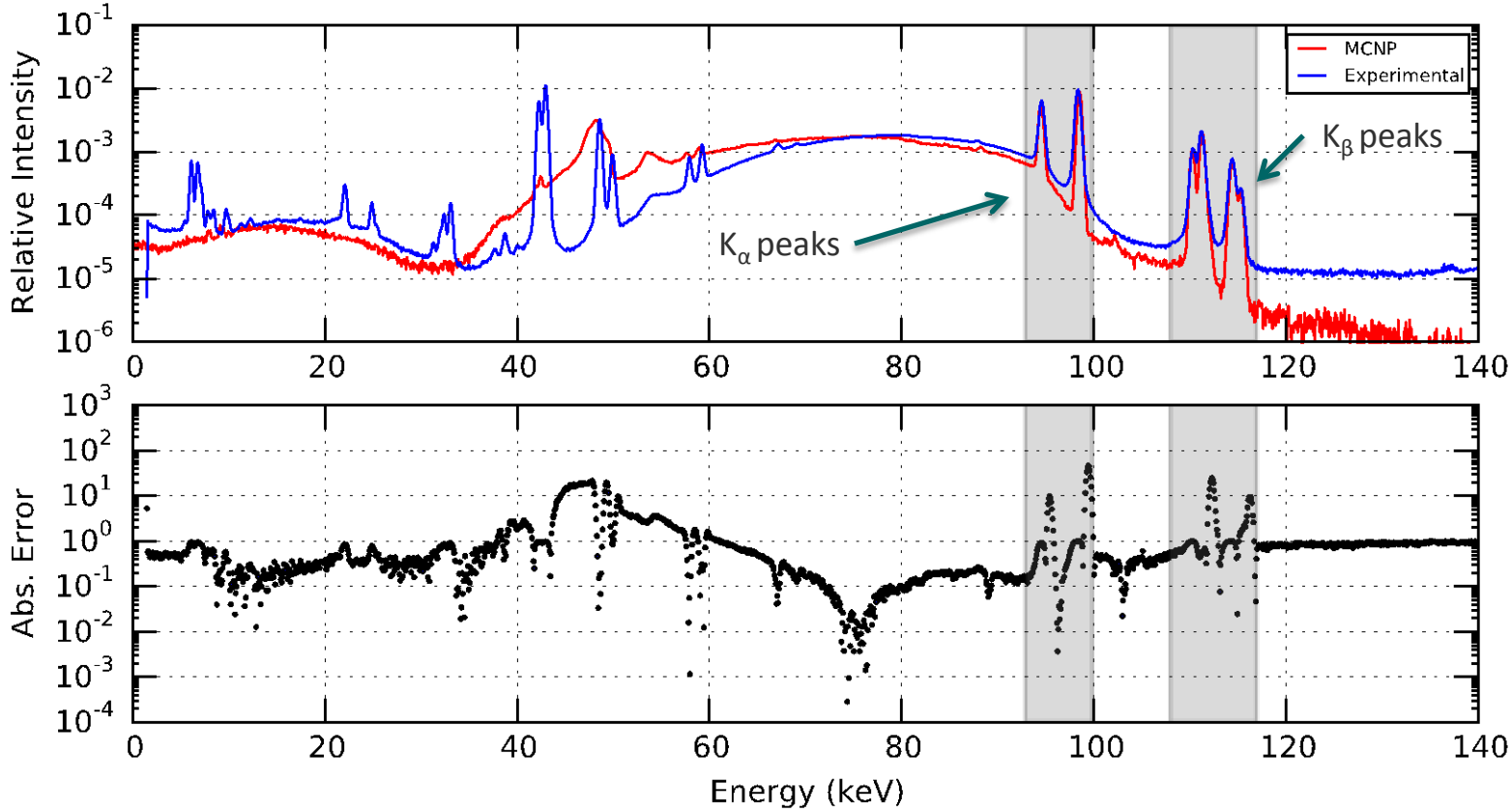
HKED Geometry
(X-Y Slice)

Previous work indicated an approximate 0.5 keV offset in energy and incorrect treatment of K_β lines. A correction factor to the spectrum was applied along with use of updated photon libraries.

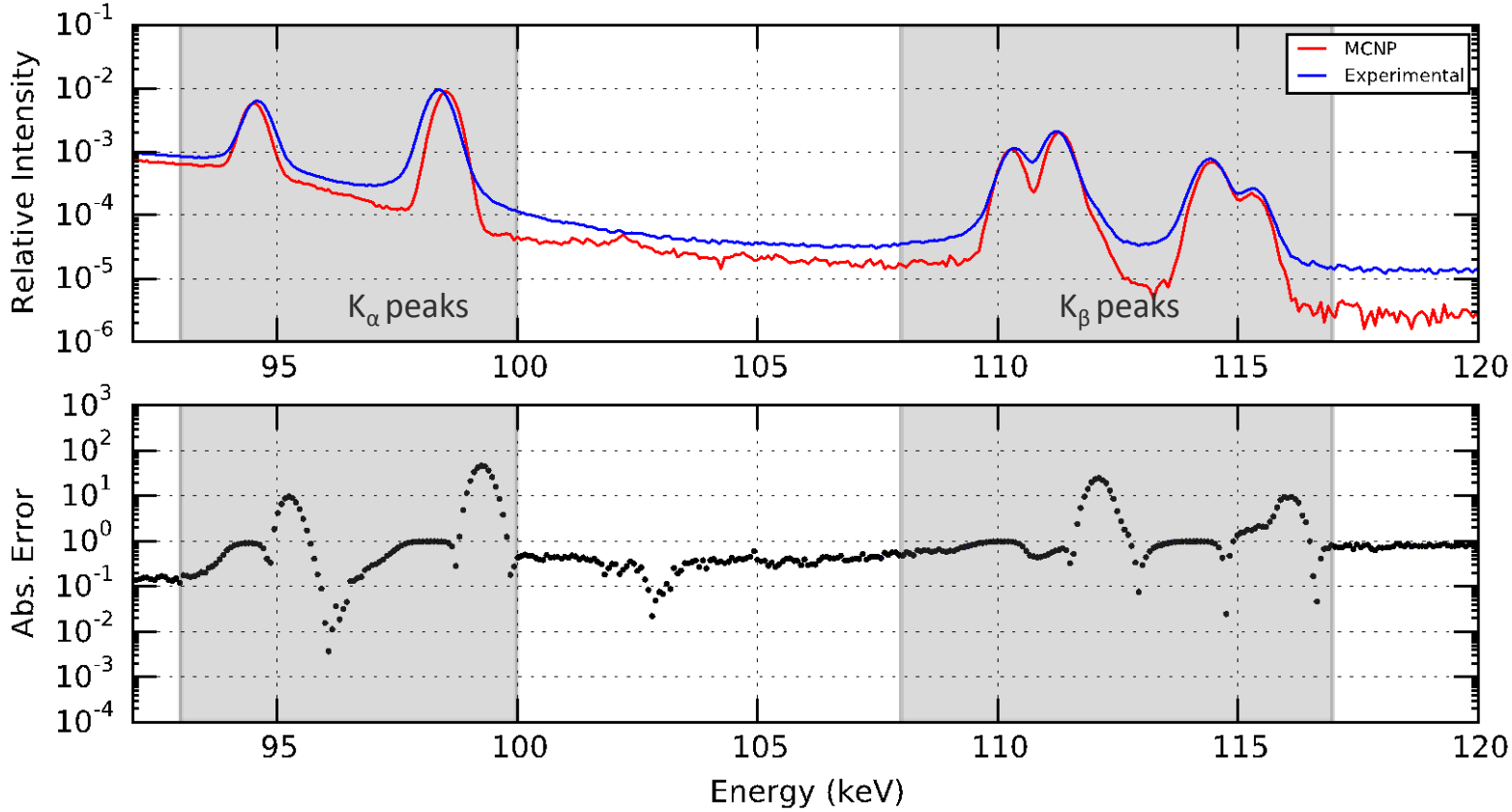
Comparison of *mcplib84* and *mcplib12*



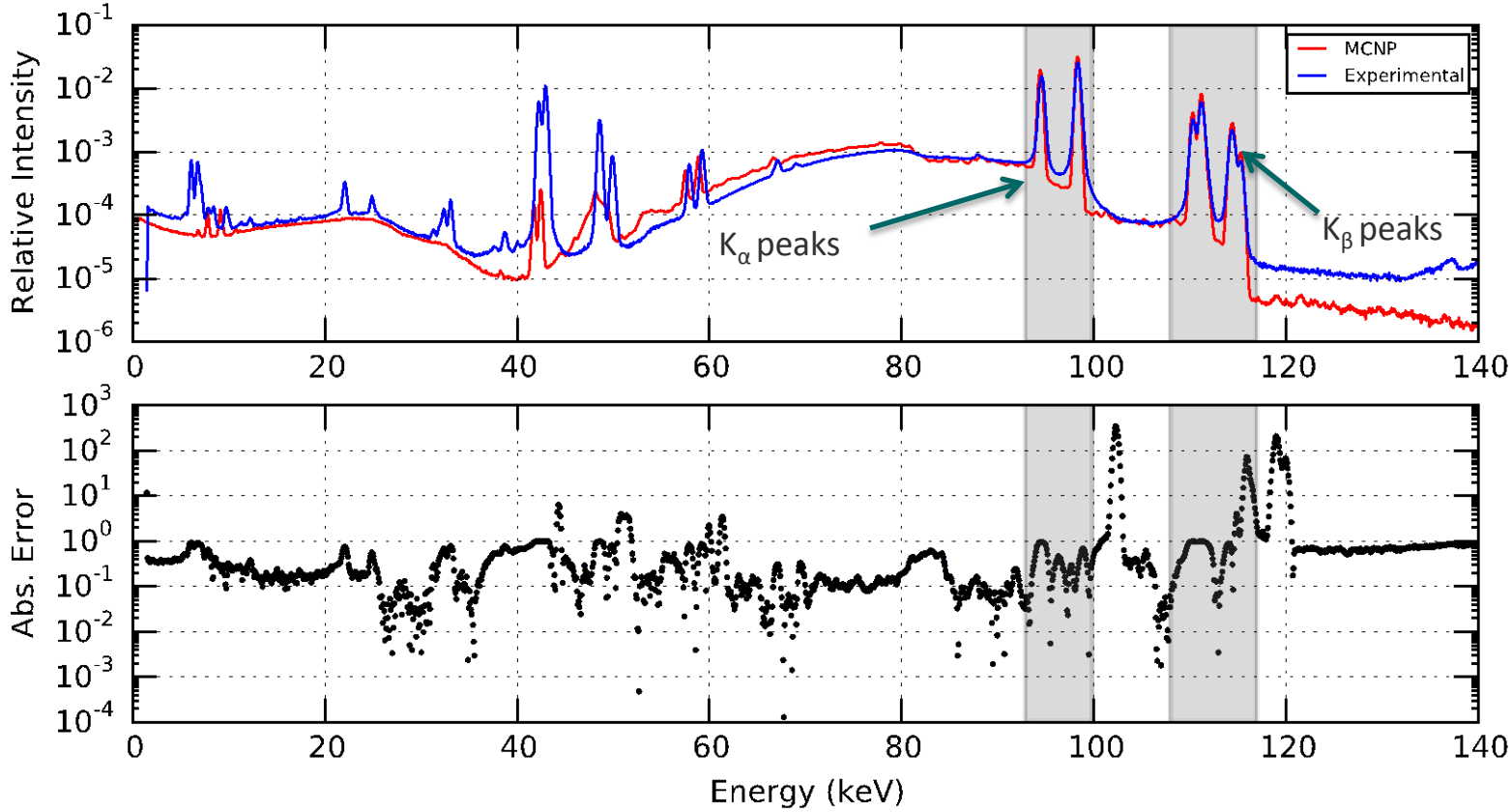
XRF Low Concentration
45 $^{g}/_{L}$ U in 3M HNO_3



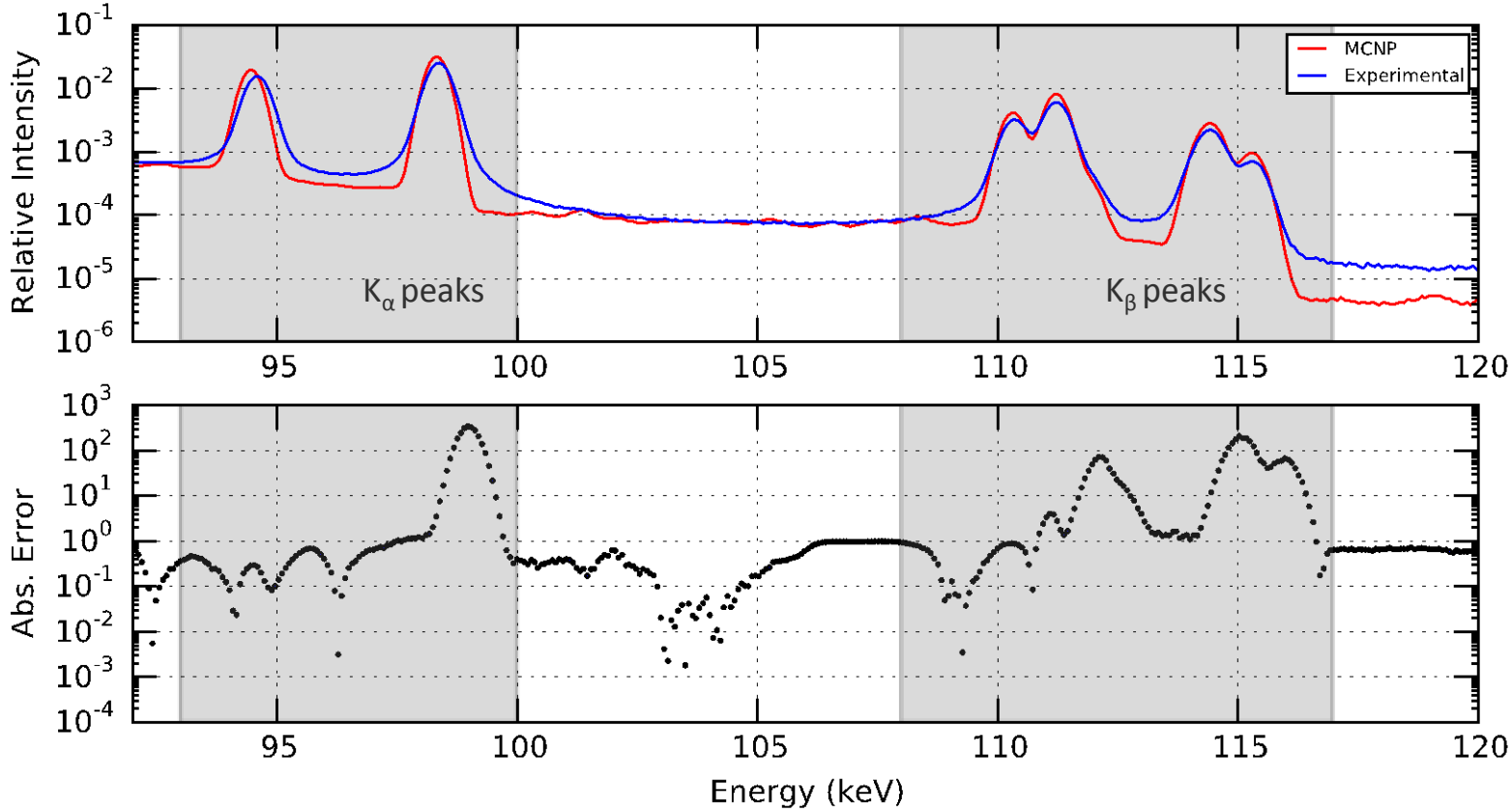
XRF Low Concentration 45 $^{g}/_{L}$ U in 3M HNO_3 (ROI Detail)



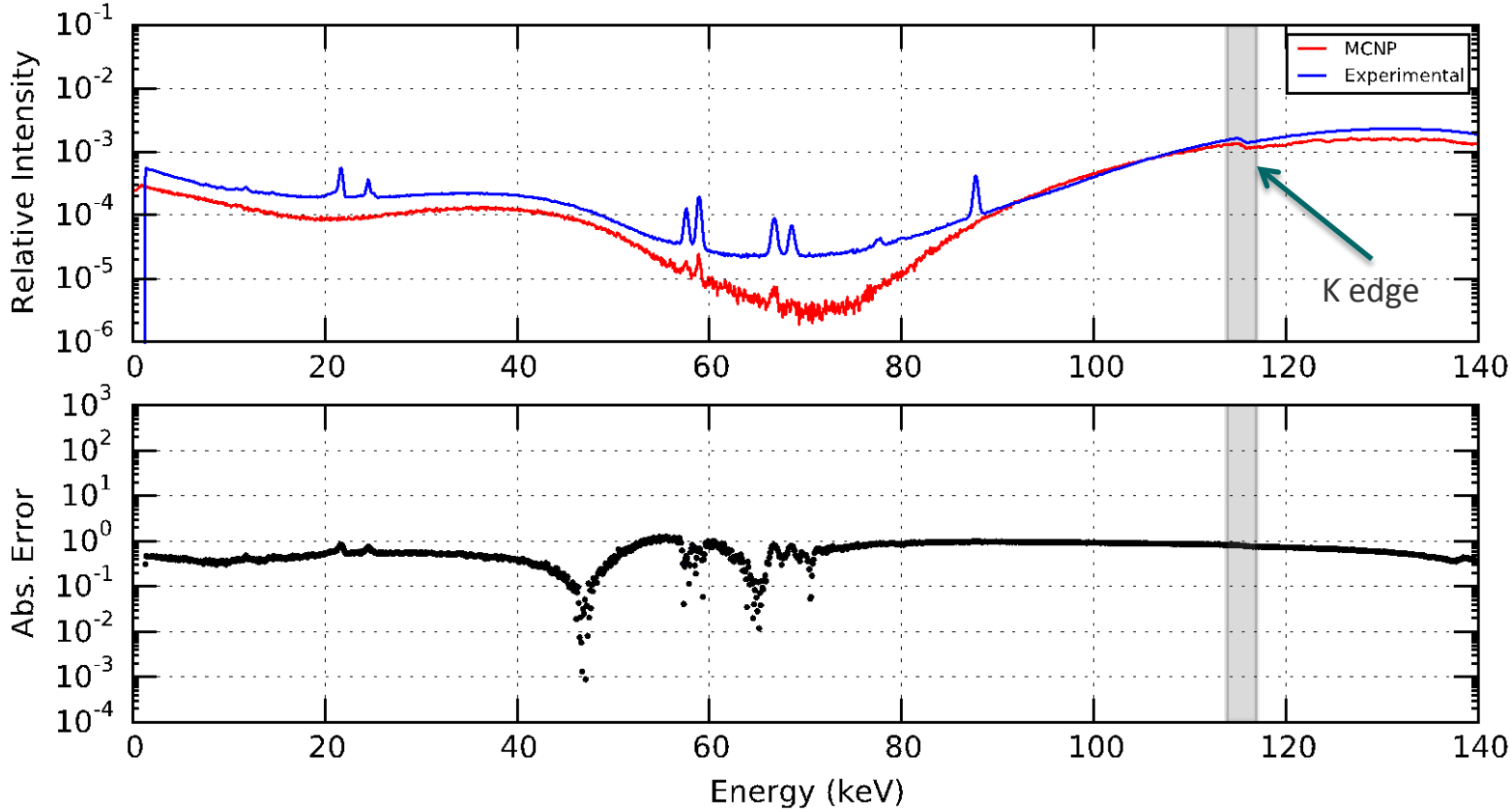
XRF High Concentration Validation Case
300 $^{g}/_{L}$ U in 3M HNO_3



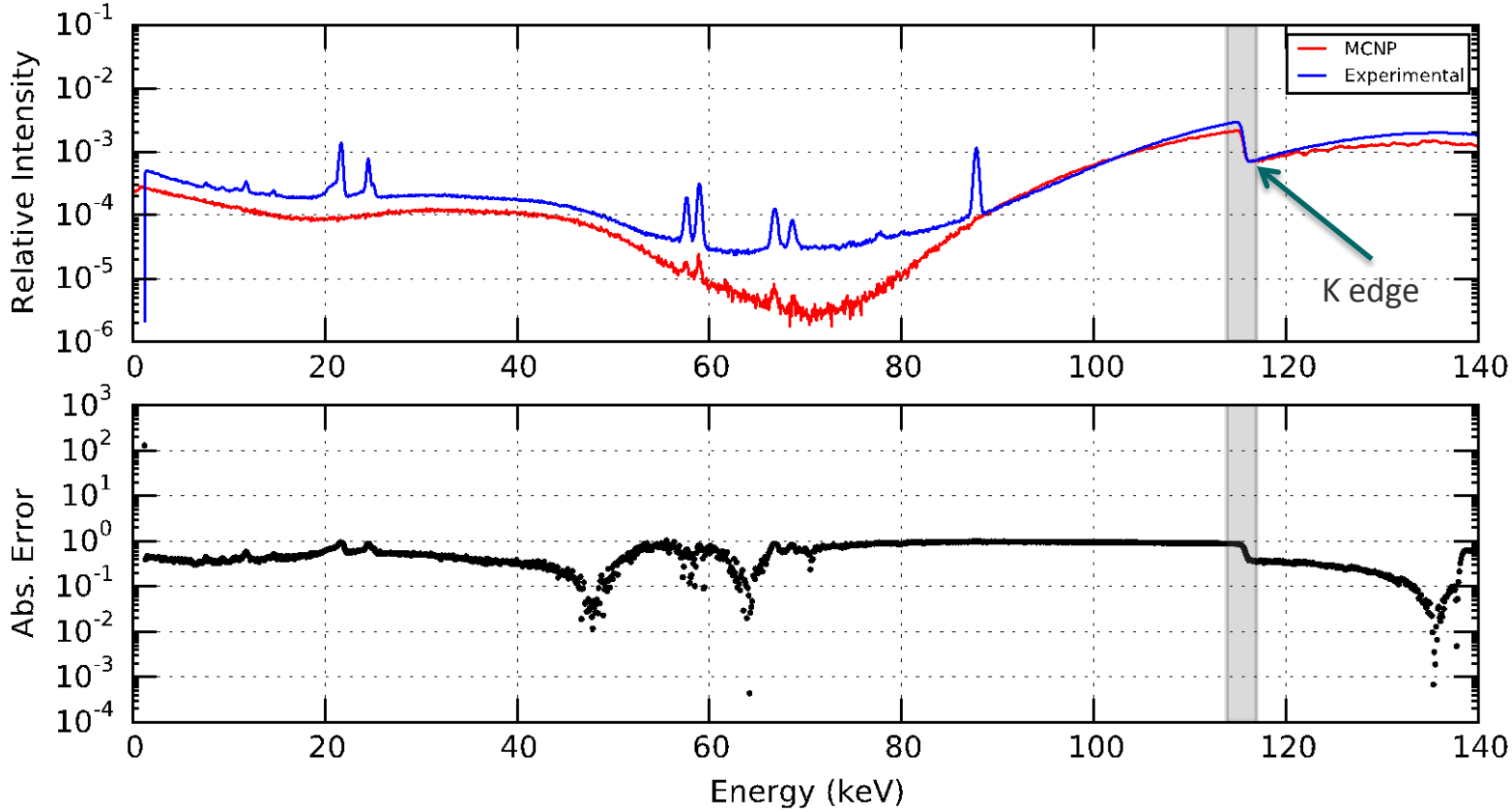
XRF High Concentration 300 g/L U in 3M HNO_3 (ROI Detail)



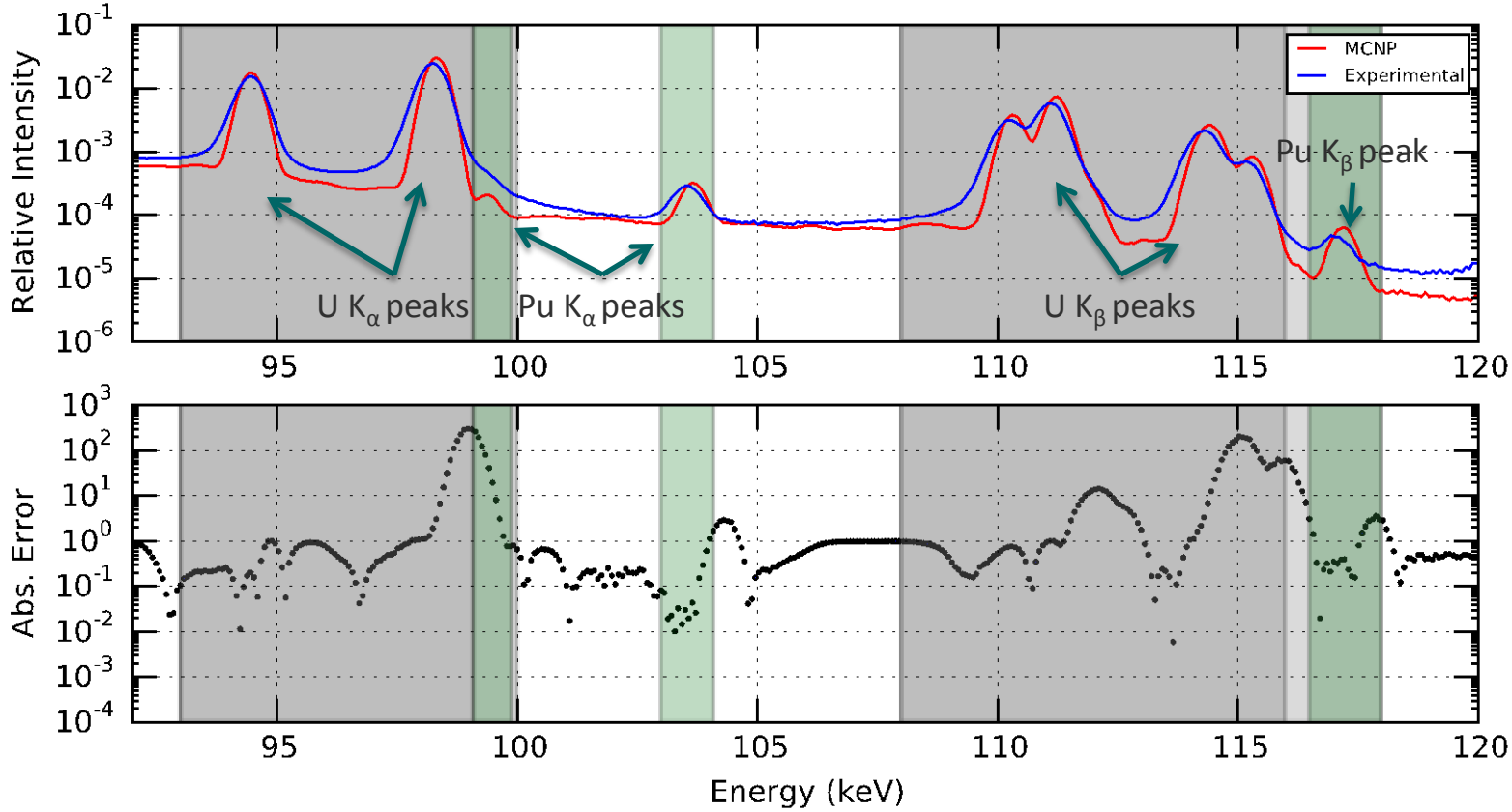
KED Low Concentration
45 $^{g}/_{L}$ U in 3M HNO_3



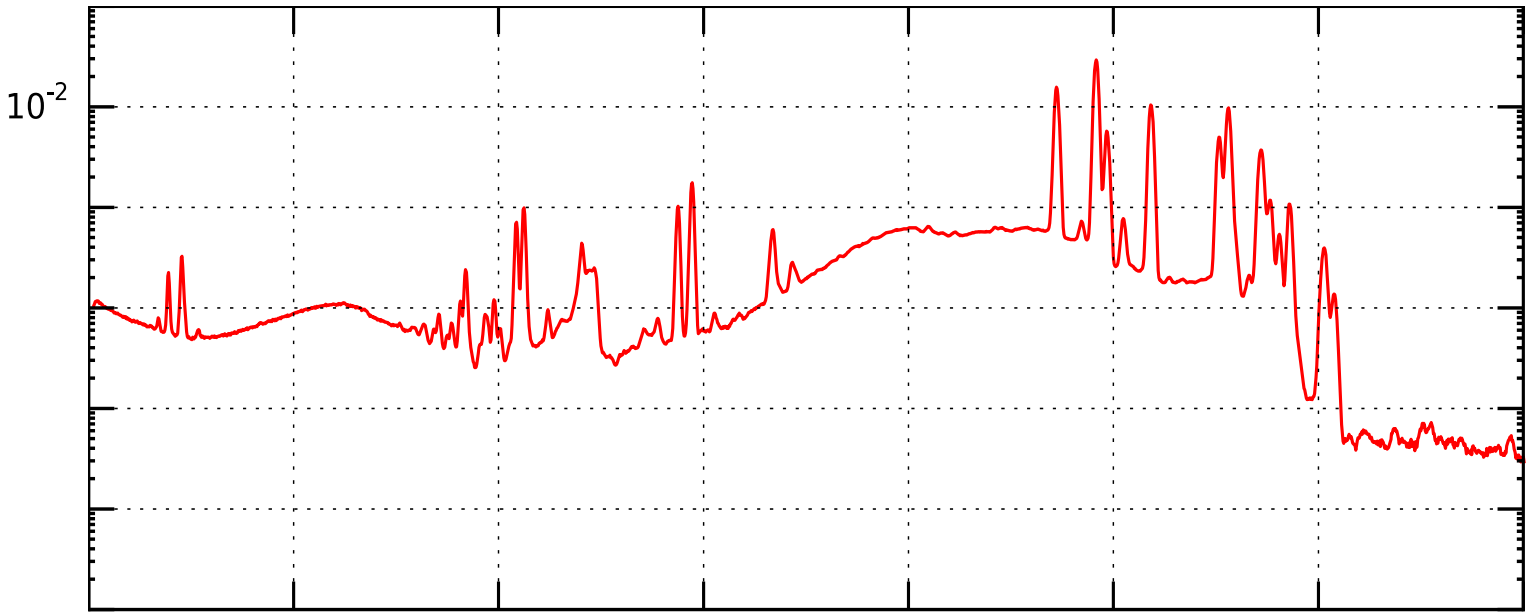
KED High Concentration 300 g/L U in 3M HNO_3



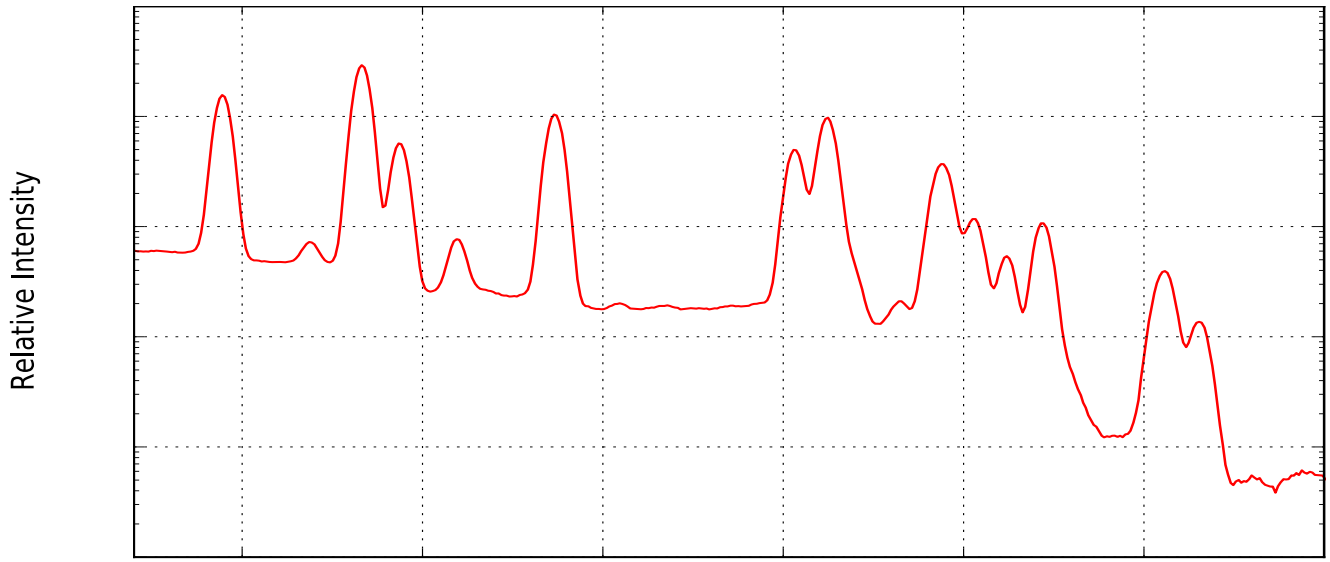
XRF High Concentration
250 $^{g}/_{L}$ U and Pu at 100:1 in 3M HNO_3
(ROI Detail)



INL Mk. 4 Electrorefiner (XRF)



Major XRF peaks from Mk.4 ER salt

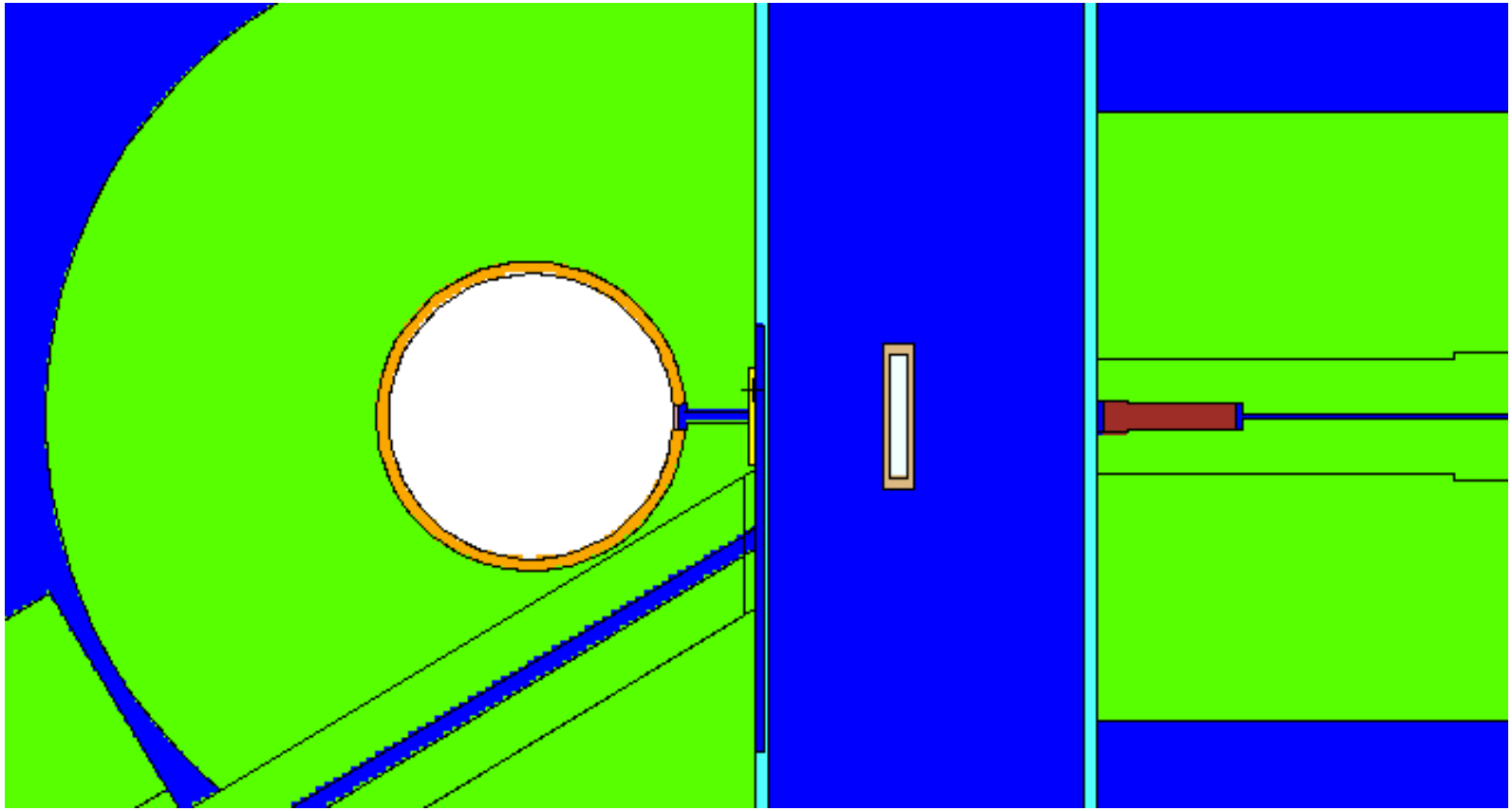


Element	$K_{\alpha 1}$	$K_{\alpha 2}$	$K_{\beta 1}$	$K_{\beta 2}$	$K_{\beta 3}$
U	98.4	94.6	111.3	114.5	110.4
Pu	103.7	99.5	117.2	120.7	116.2
Np	101.6	97.1	*114.2**	*117.5**	113.3

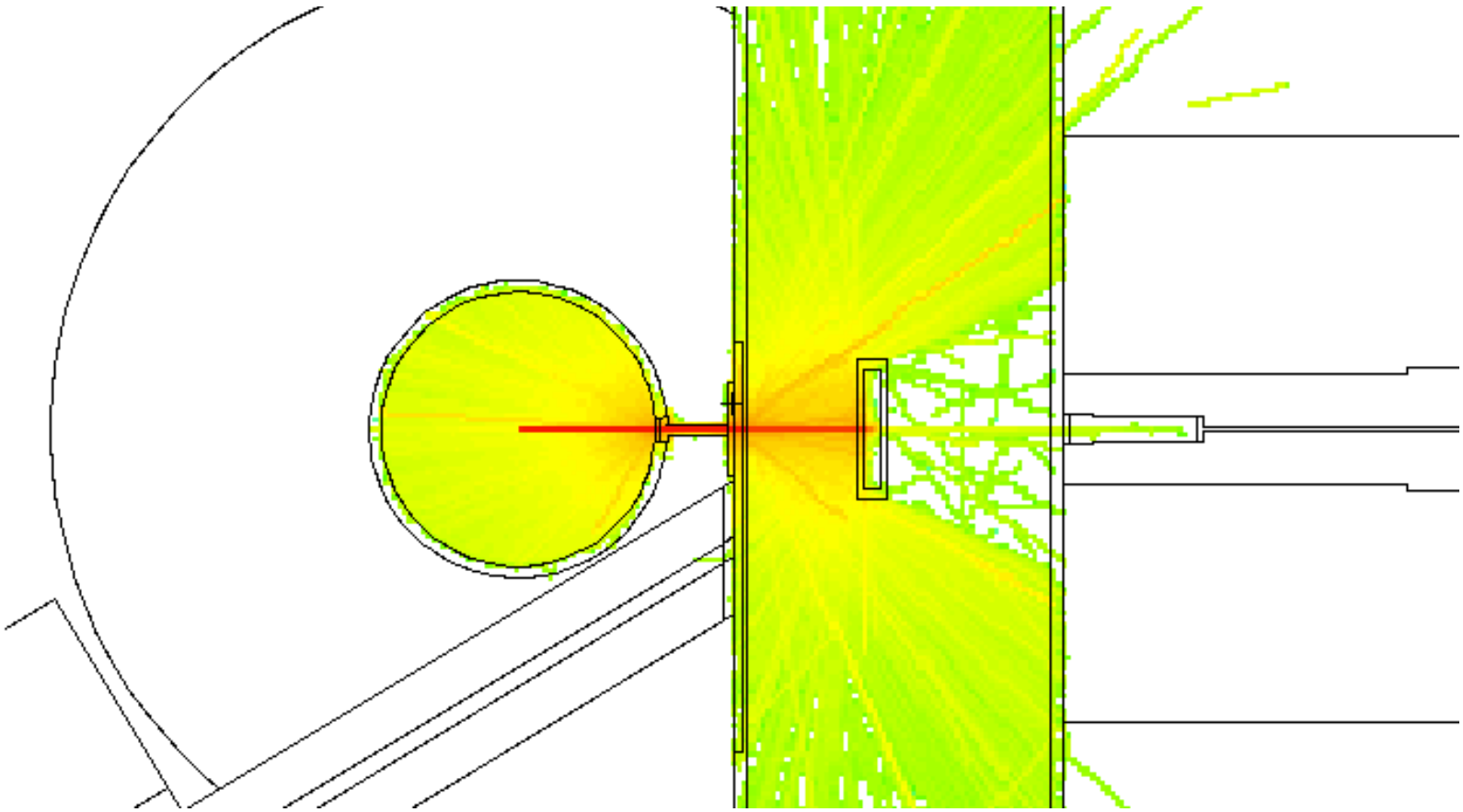
All energies in keV

** Np $K_{\beta 1}$ and $K_{\beta 2}$ peaks are obscured.*

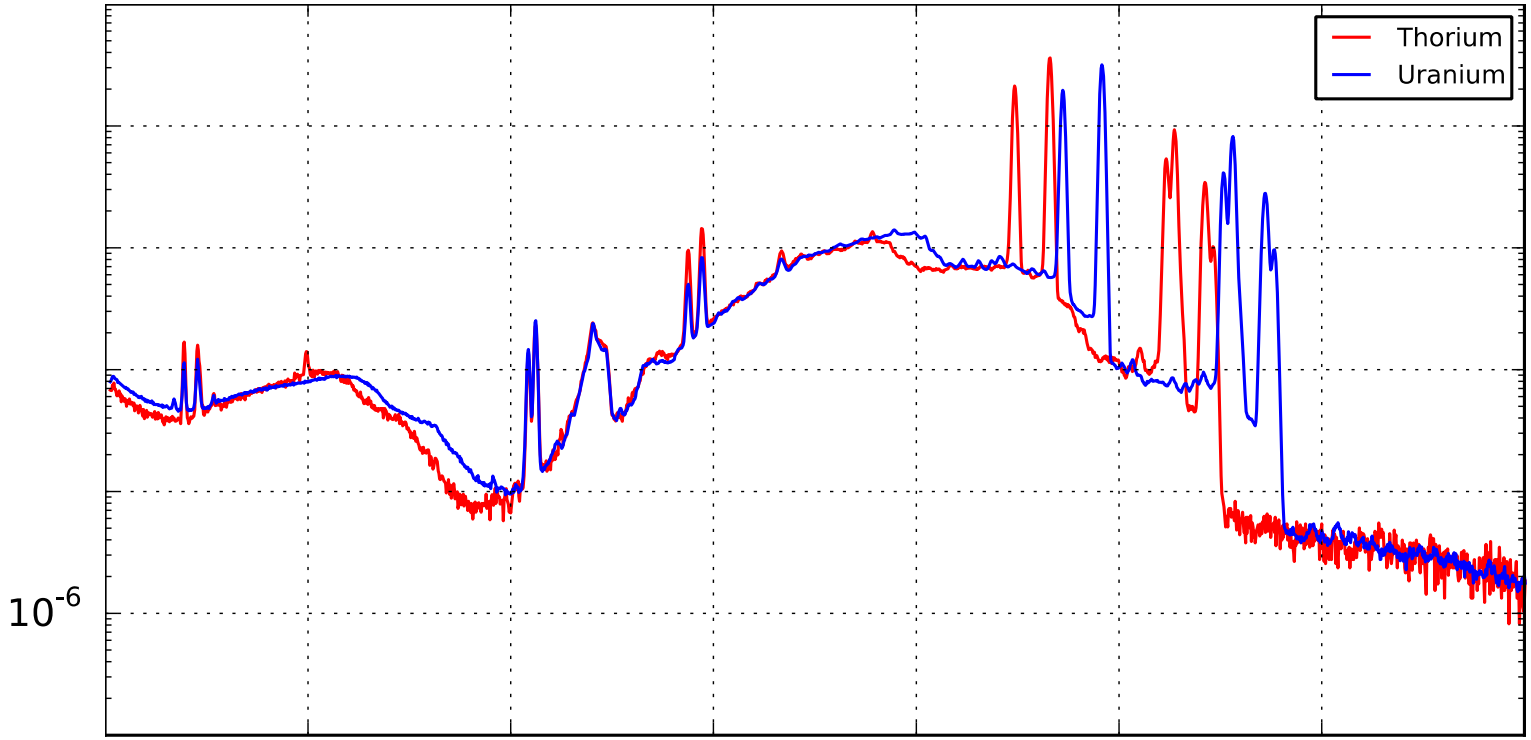
Voloxidation powders require a new sample geometry. Standard sample geometries were not optically transparent.



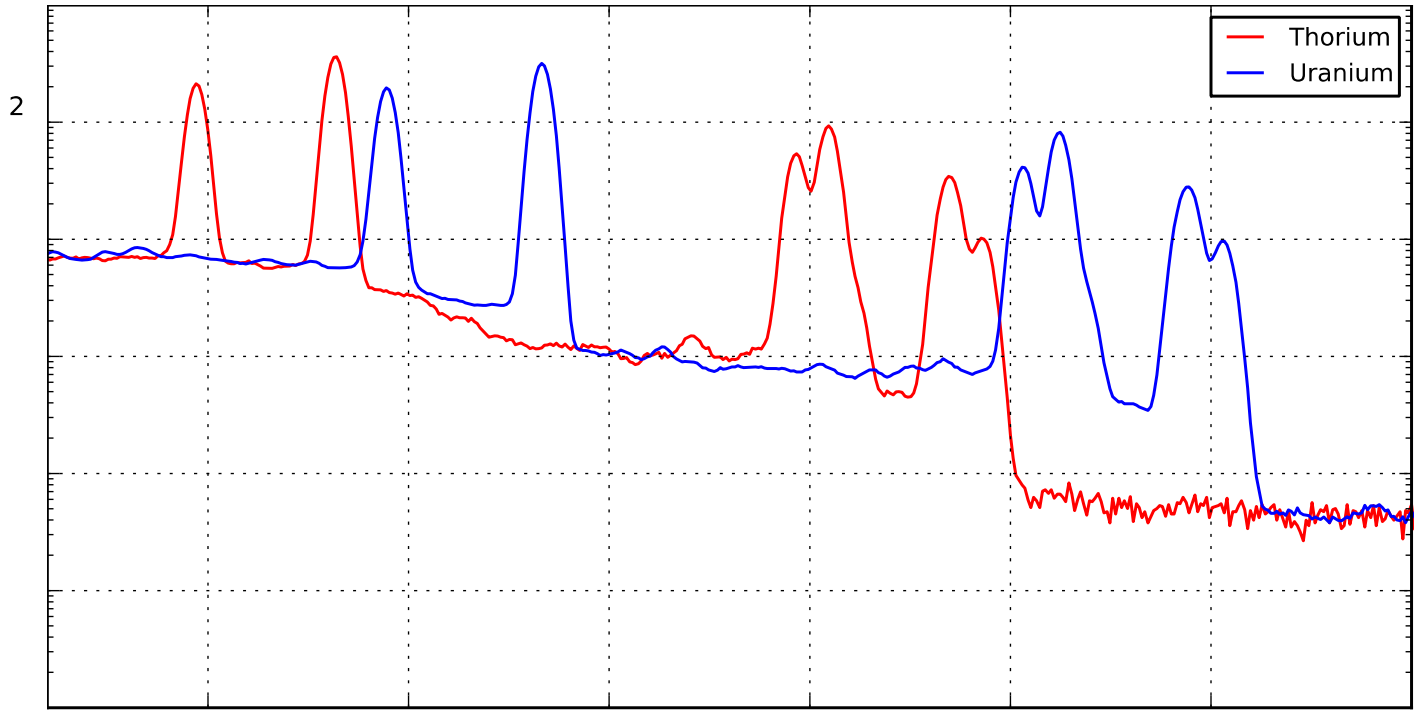
The voloxidation powder sample must be sufficiently thin to allow K-edge measurements to be made. XRF photons must also transport through the whole sample volume.



Using surrogate materials such as thorium may lower costs for mixed element samples.



Thorium appears to be an acceptable surrogate for uranium in planned scoping studies.



Future Work

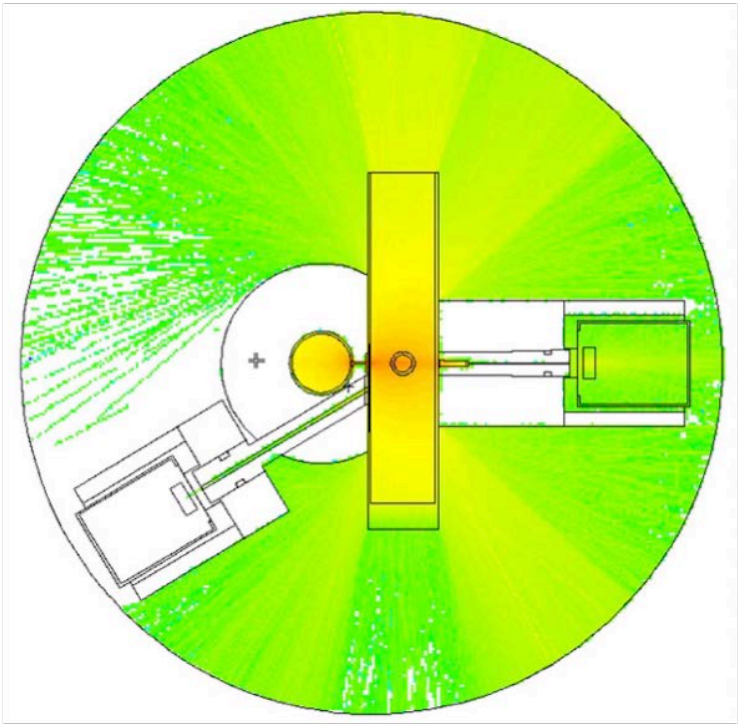
- More realistic pyrochemical studies
  - Incorporate ORIGEN models for developing more realistic and different types of samples
  - Determine optimal geometry for voloxidation powders
  - Determine method for assaying liquid cathode
- Explore possibility of online in-situ measurements for real time assay and process monitoring
- Exploration of background source term
  - Use ORIGEN and MCNP to predict the gamma-ray background present in real world samples
  - Self-induced XRF source term
- Further exploration of detector response
  - Incorporate other detector modeling tools to simulate the detector electronic response

Conclusions

- HKED appears to be a viable method for materials accountancy measurements in pyroprocessing salts
  - Application to voloxidation powders will require further modification method
- MCNP model of the HKED system will be useful for scoping studies
  - Photon library update essential for accurate simulations
  - Average correction factor adequate for current studies but further refinement of method is required
  - Model facilitates scoping studies to further evaluate HKED modifications
- Further refinement of the model is needed
  - More cases are needed to improve model's accuracy
  - More realistic treatment of detector response
  - Modification of system geometry to match what is needed for pyroprocessing (goal of in situ real-time measurements)

Thank you for your time.

Questions or Comments?



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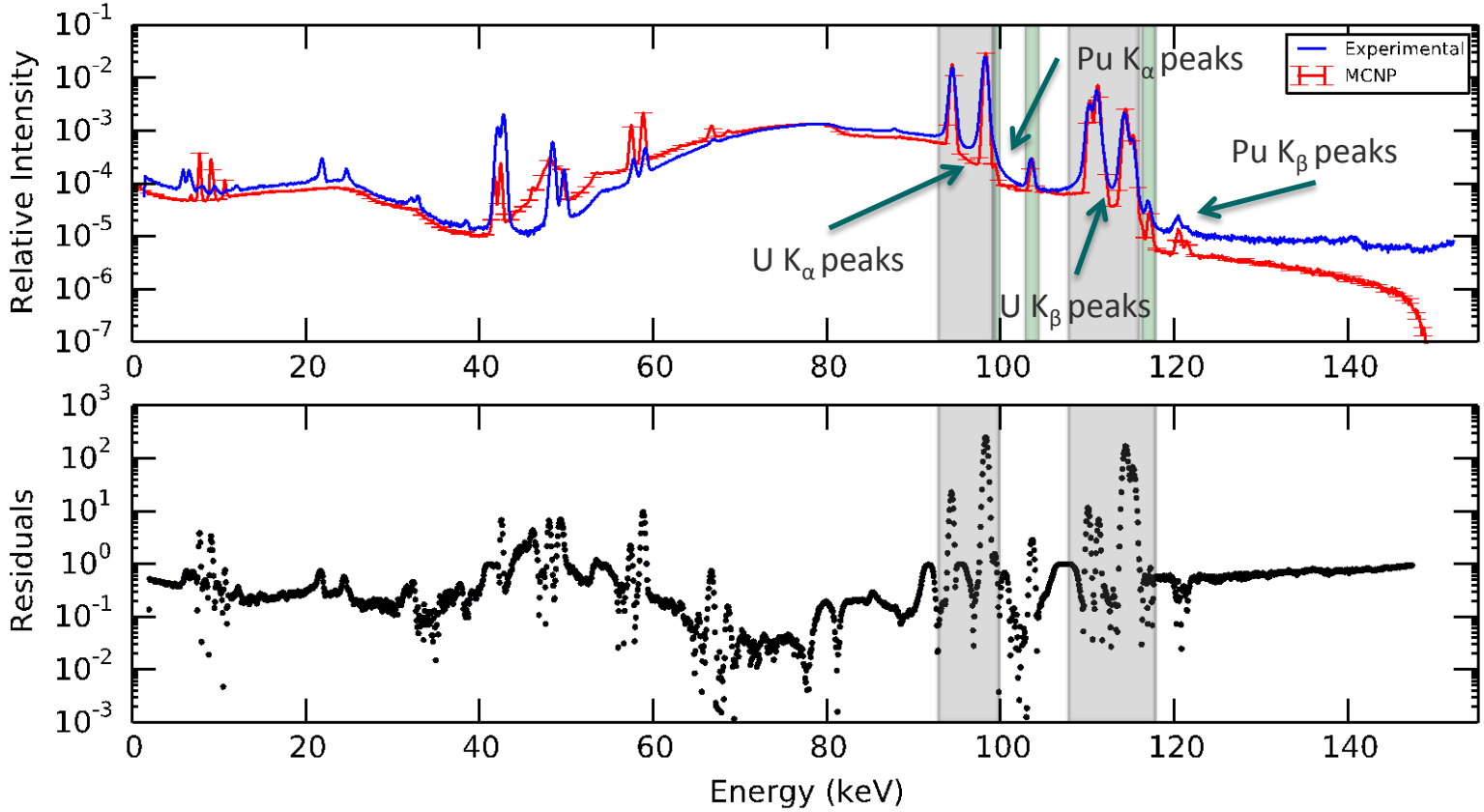
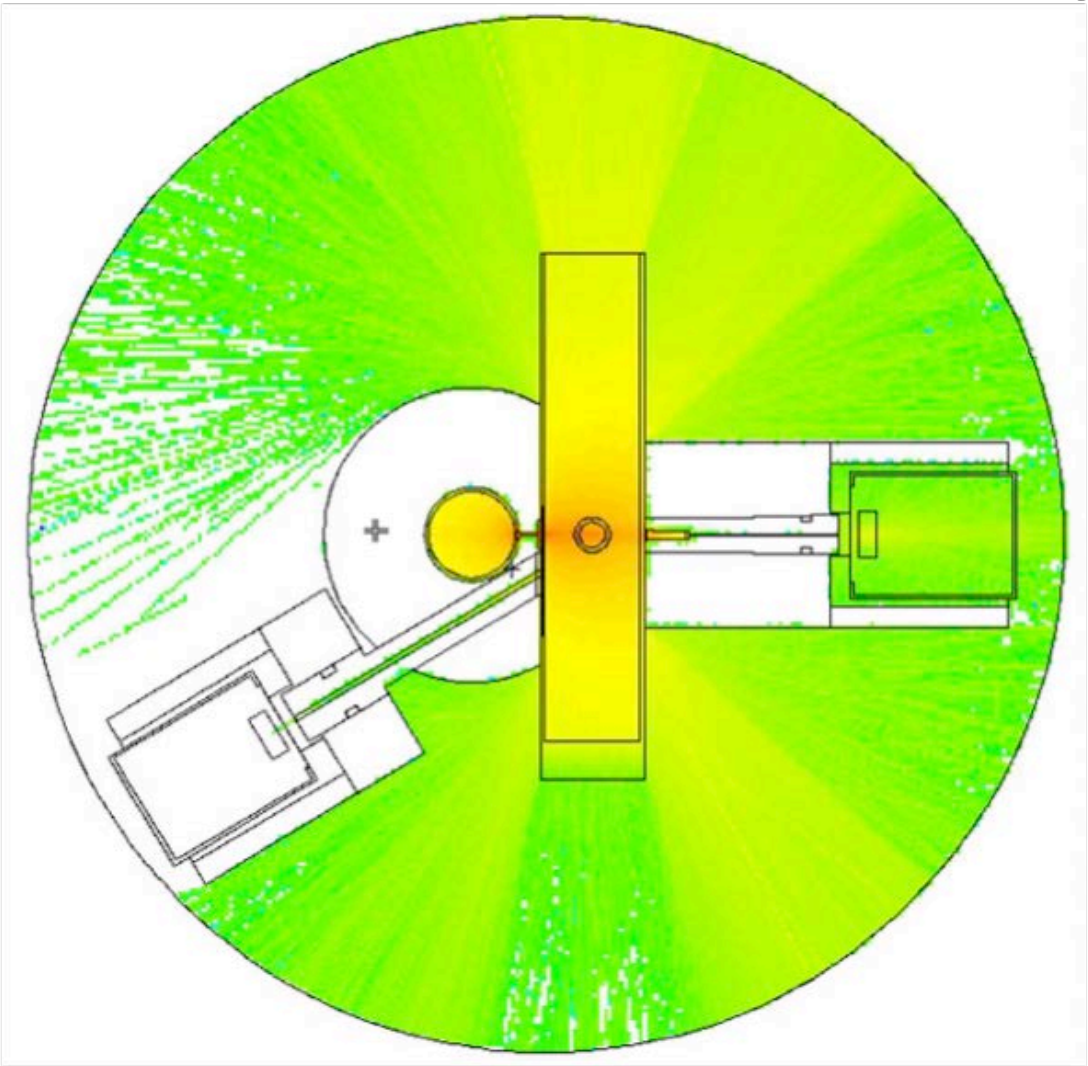
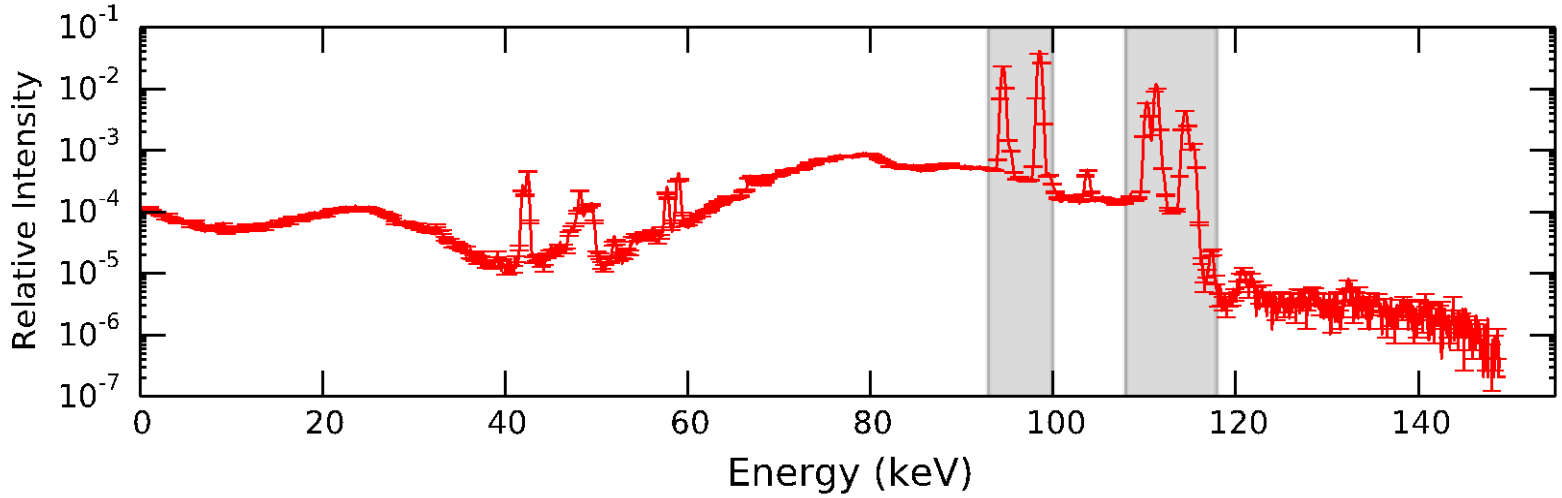
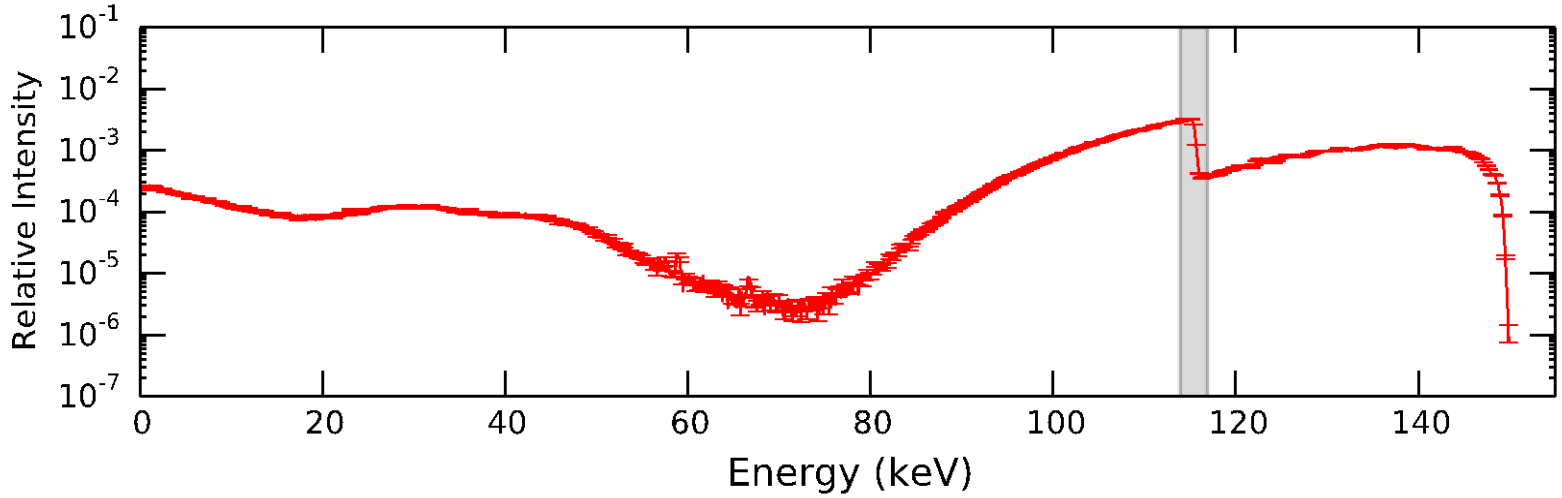


Volume Flux Mesh Tally



XRF Actinide Case (300 g/L U with Pu at 100:1)



KED Actinide Case (300g/L U with Pu at 100:1)