NAA at NC State

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NAA principle for U and Th

- Transmute long lived isotope into a short lived radioactive isotope through neutron activation
- Search for new isotope decay by identifying signature gammas through gamma ray spectroscopy
- Example:

$^{232}Th(t_{1/2}=1.4\times10^{10}) + n \rightarrow {}^{233}Th(t_{1/2}=22m)$

$\rightarrow e^- + {}^{233}Pa(t_{1/2}=27d)$

$\rightarrow e^- + {}^{233}U(t_{1/2}=1.6E5y) + \gamma$

17% prob. 312keV

NAA

- Activate sample with high flux of neutrons and search for signature gammas to identify contaminant

Element	Parent Isotope	Product Isotope and Decay chain	Gamma energy (keV)	Sensitivity
K	^{41}K	^{42}K ($t_{1/2}$=12.26 hr)	1524	Best limit 50 ppb Typical 50 ppb
U	^{238}U	^{239}U ($t_{1/2}$ = 24 min) → ^{239}Np ($t_{1/2}$ = 2.35d)	106, 222, 228, 277	Best limit 1ppt Typical 20 ppt
Th	^{232}Th	^{233}Th ($t_{1/2}$ = 22 min) → ^{239}Pa ($t_{1/2}$ = 27d)	312	Best limit 1 ppt Typical 20 ppt

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NAA data taken from P.Ila talk at LRT2004, Sudbury Canada, Dec 12-13, 2004

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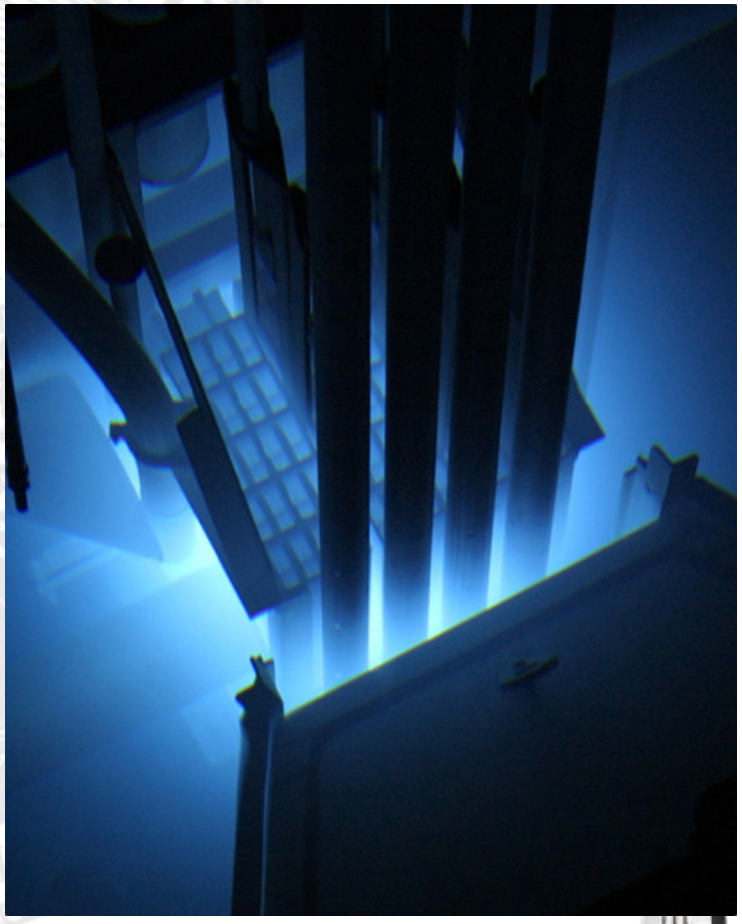
Pulstar reactor

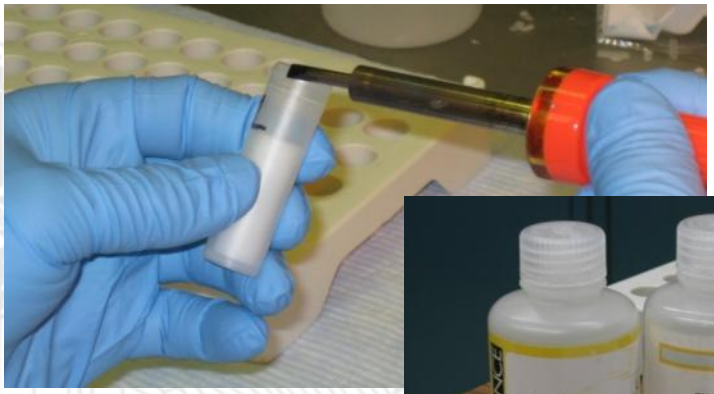
(http://www.ne.ncsu.edu/NRP/reactor_program.html)

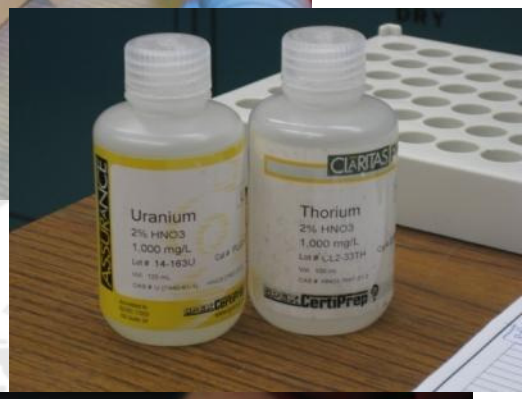
- Educational reactor
- Provides services for basic research
- 1 MW pool type reactor
- 4% enriched uranium dioxide fuel
- Thermal n-flux in 'Rotating Exposure Ports' = 4-8 x 10^{12} NV at 1 MW



NAA method at NC State

- Samples
  - Sealed in plastic vials
  - Vials are LDPE (tends to be cleaner)
- Standards
  - Aqueous solution sealed in plastic vials (vials are HDPE, which holds up better to irradiation)
  - Known amount of U and Th
  - Irradiated along with samples
  - Similar geometry as sample
- Flux monitor
  - Antimony solution sealed in plastic vials
  - Irradiated along with samples
  - Used to monitor n-flux if necessary
- All vials are packed and sealed in irradiation bottles (nalgene bottle)





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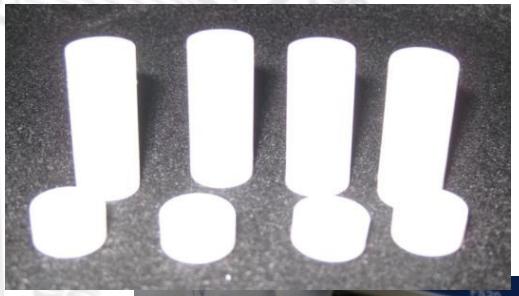
Sample and Standard prep

- Sample
  - Plastic = TE-6472
    - Identified by EXO collaboration
    - Used for MAJORANA crystal mounting
  - Machined at NC State to fill vial radially and 2/3 of the vial height
  - Mass – 38.8g
  - Plastic and vials etched and cleaned by Eric Hoppe –PNNL
  - Vial heat sealed at NC State
- Standard
  - U and Th in 2% HNO_3 aqueous solution
  - Height of standard liquid equals height of sample to match geometries
  - Starts as 1000mg/L, but further diluted to give required mass with correct geometry
    - Total U mass = 50µg
    - Total Th mass = 2.5 µg



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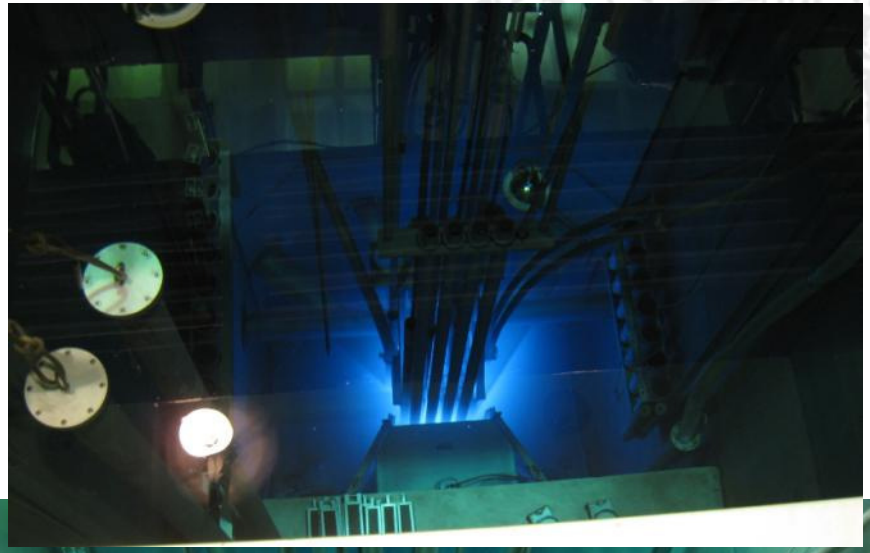
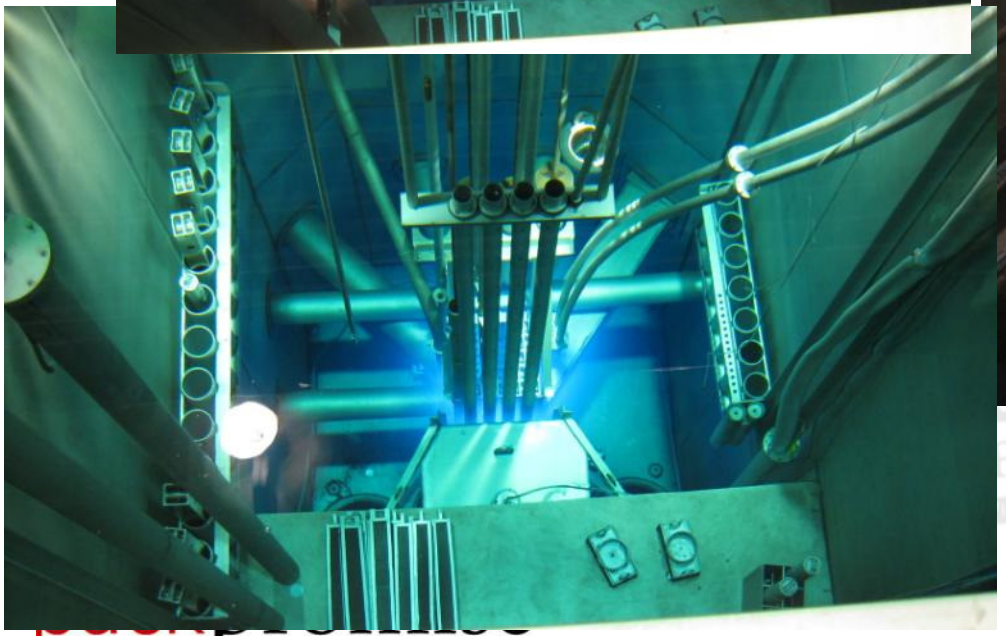
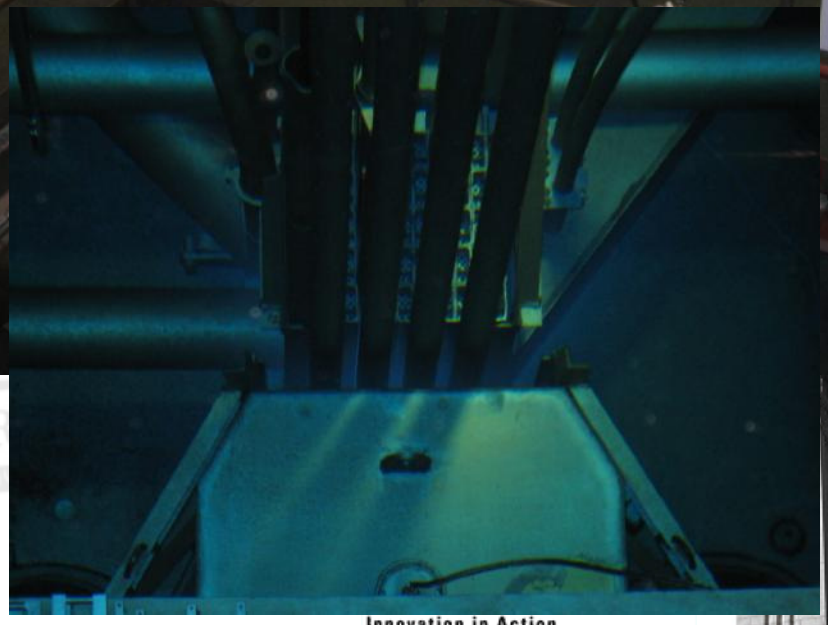
Reactor pictures

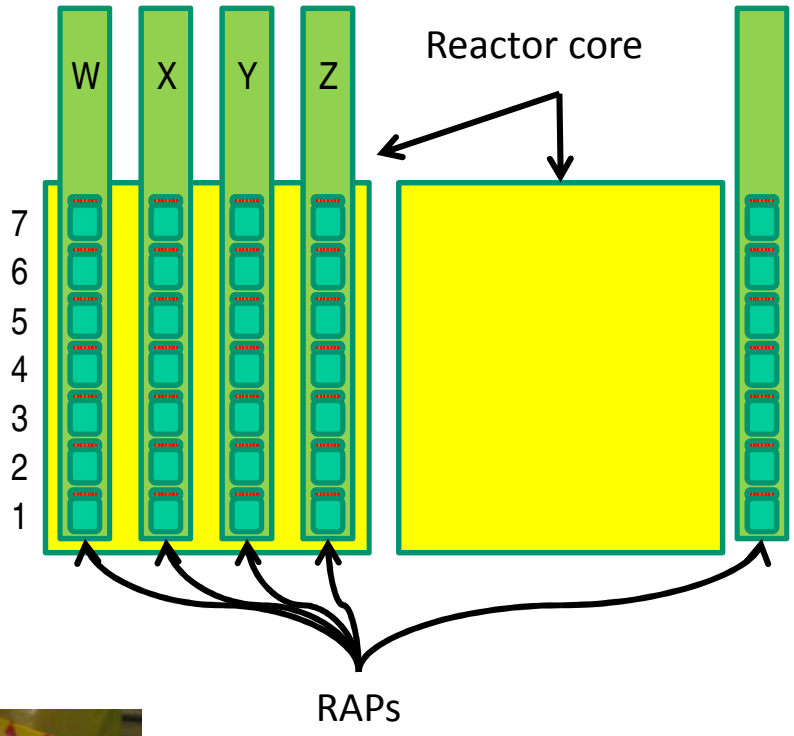


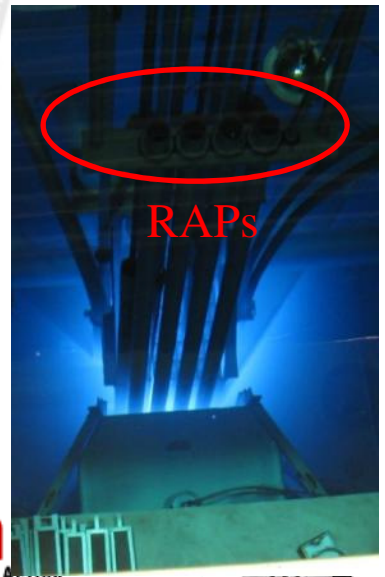




Irradiation

- Rotating Exposure Ports (RAPs)
  - Thermal n-flux in RAPs = 4-8 x 10^{12} NV at 1 MW (the flux varies by position)
  - Four RAPs each hold 7 irradiation bottles
  - Irradiation bottles suspended in aluminum stringers
  - Stringers must 'cool' in reactor pool for 4+ days before irradiation bottles can be removed
    - Eliminates ^{40}K determination (^{42}K $t_{1/2}$ ~12hrs)
    - Reduces ^{238}U determination efficiency (^{239}Np $t_{1/2}$ = 2.35d)
- Our sample irradiation
  - Irradiation positions
    - Sample – 2Y (highest flux region)
    - U standard – 1Y
    - Th standard – 3Y
  - 18 MW hours





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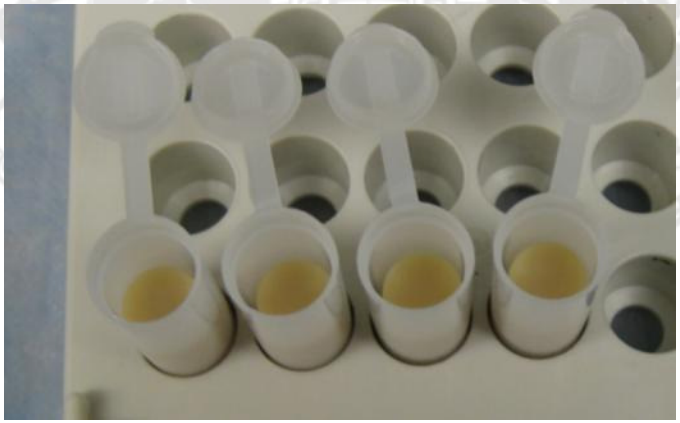
Post irradiation sample prep

- Samples removed from irradiated vials
- Sonicated in dilute HNO_3 solution for 1 hour
- Rinsed with DI H_2O in ultrasonic bath for 1 hour
- Sampled placed in new, non-irradiated plastic vials to maintain geometry



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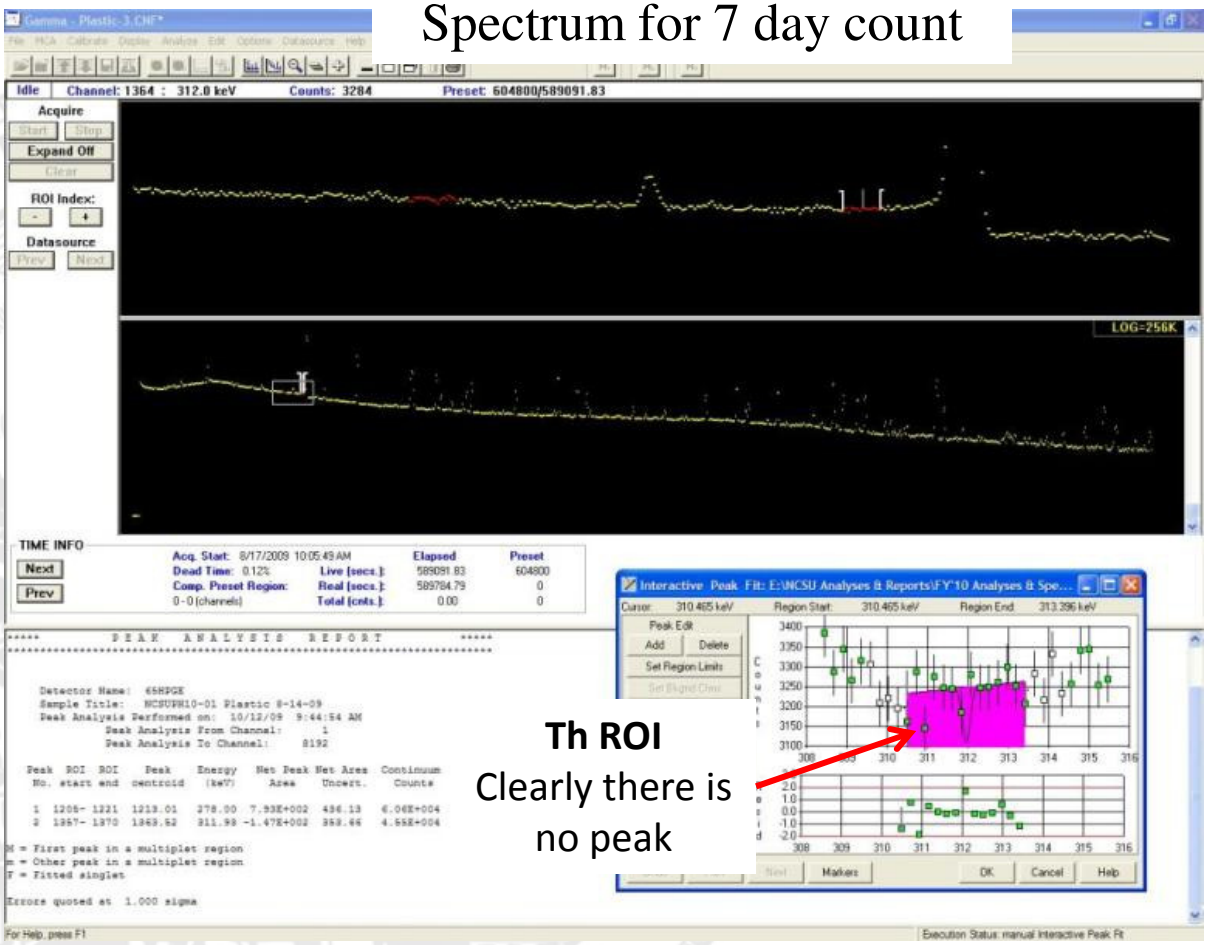
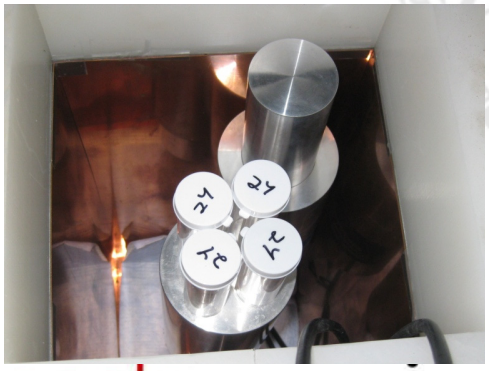
Counting

- Detector
  - 65% HPGe
  - 8” Pb shield
  - 2” poly shield
- Counting
  - All four vials counted together
  - Geometry of vials kept the same in each counting
  - Total counting campaign > 10 days

Spectrum for 7 day count



Th ROI
Clearly there is no peak



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Date		Start time	Counting time (sec)
8/11/09	Sample	3:15:54 pm	78,767.3
8/12/09	Thorium standard	1:46:29 pm	636.1
8/12/09	Sample	2:08:05 pm	86,577.4
8/17/09	Sample	10:05:49 am	589,784.8
8/21/09	Uranium standard	3:35:59 pm	371.9

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Results

- Analysis
  - Standards give expected gamma rate for given U and Th mass
  - Matching counting geometries cancel any geometrical systematic errors

- **Results**
  - **^{232}Th ≤ 0.809x10^{-6} µg/g or 3.28 µBq/kg**
  - **^{238}U ≤ 2.77x10^{-6} µg/g or 34.5µBq/kg**
- Comparison – Best EXO results for TE-6472
  - ^{232}Th ≤ 0.26x10^{-6} µg/g
  - ^{238}U ≤ 0.78x10^{-6} µg/g

Systematic study of trace radioactive. **D. S. Leonard, et Al.** 2008, Nuclear Instruments and Method A 591, pp. 490-509.

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Summary

- NAA at NC State is a viable material assay method for DUSEL experiments
  - NC State Nuclear Engineering is very supportive (private conversation with director of reactor program)
  - Is done completely in-house at NC State (reliably)
    - Irradiation (others irradiate at other facilities)
    - Counting and analysis (Others send samples home and count in their own counters)
- The thorium limit is comparable to the best EXO measurement
- We would like to improve the Uranium limit
  - **Higher mass?** - Probably not much help. We would like to do ~5 times better, but 5 times more mass is 200 grams. Might be possible, but it will take more R&D
  - **Shorter cooling off period?** Probably our best bet. The ^{239}Np has a 2.4 day half-life. However, currently NC State uses aluminum stingers, which are very 'hot' when first removed. Requires more R&D.

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NAA at NC State

(conversation with S. Lassell in Nucl. Engineering)

- Currently 1200-2000 samples per year
- Large room for expansion
  - Room for more samples
  - Run reactor longer
- Sample size (3 different vials)
  - 0.13 ml – dia. 0.22" X 0.44"
  - 1.4 ml – dia. 0.38" X 0.87"
  - 8.06 ml – dia. 0.57" X 2.09"
  - www.lacontainer.com
  - Vials supplied by NC State
- Typical irradiation – 24 hr
- Neutron E and flux can be somewhat tailored
  - Further from core or shielding = lower flux & lower energy
- Detectors
  - 3 HPGe detector
    - Relative efficiency order 20-30%
    - One automated detector system for large number of samples
  - 1 low background HPGe detector with large shield
- Estimated costs for NC State physics
  - Irradiation - $100/hour
  - Analysis (typical including irradiation)
    - $55 for one element
    - $10-$20 for second element
    - $10/element after two
    - U and Th - $70/sample
- Sensitivity test underway (irradiation of polyethylene)
- More sensitive gamma at Kimballton (REF: P. Finnerty talk)

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Other facilities

- The University of Missouri-Columbia Research Reactor Center (MURR)
  - 10 MW research reactor
  - Offers NAA service and we can request quote online
  - http://www.murr.missouri.edu/
- Oak ridge – High Flux Isotope Reactor (HFIR)
  - 85 MW reactor
  - Uses both plastic and graphite vials (graphite = longer irradiation)
  - Quote from website *"The NAA systems support ORNL (DOE) programs, are used in work-for-others projects, and are available for use by students and faculty of universities through Oak Ridge Associated Universities and other programs."* – NC State, Duke, and USC are part of the Oak Ridge Associate Universities
- MIT Nuclear Reactor Laboratory (NRL)
  - 5 MW reactor
  - 4 HPGe detectors
  - More info at - http://web.mit.edu/nrl/www/research/neutron_activation.htm