THE UNIVERSITY OF ALABAMA

NAA techniques and applications

Ryan MacLellan

LRT 2010

August 28, 2010

Techniques typically used to characterize natural radioactivity

Direct γ-ray counting

- Sensitive to the background creating isotopes of the decay series directly.
- Requires long counting time and large sample mass to achieve good sensitivity.

10^{-14}g/g U produces only 1 decay per day per 100kg of sample mass.

Mass spectroscopy

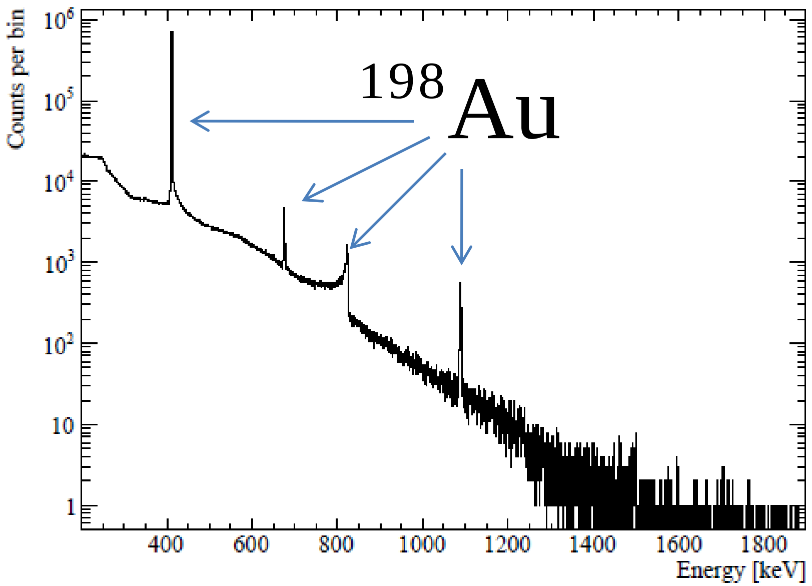
- Sensitivity limits of:
  - 10^{-9}g/g K
  - $10^{-12/-13}$g/g U and Th
- Requires sample to be digested by acids (ICPMS) or conductors (GDMS).

Neutron Activation Analysis

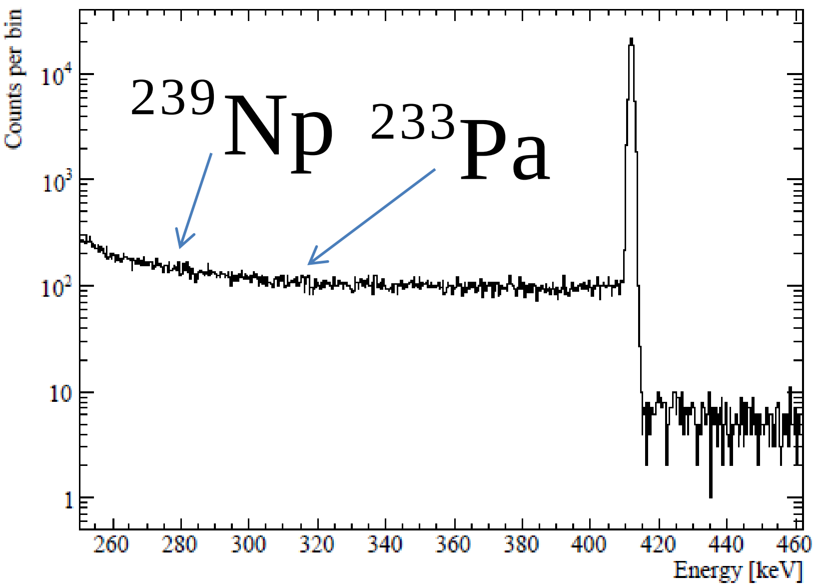
- Sensitivity limits of:
  - 10^{-9}g/g K
  - $10^{-12/-13}$g/g U and Th
- High sensitivity measurement can take up to one month due to long half life of Th activation product: ^{233}Pa.
- Only applicable to matrix materials with low neutron capture cross sections (most metals not suitable).

Gold plated silicon wafer (etched for ICP-MS)

2d after neutron activation



1 month after neutron activation



$$t_{1/2} = 2.7\text{d}$$

Neutron Activation Analysis

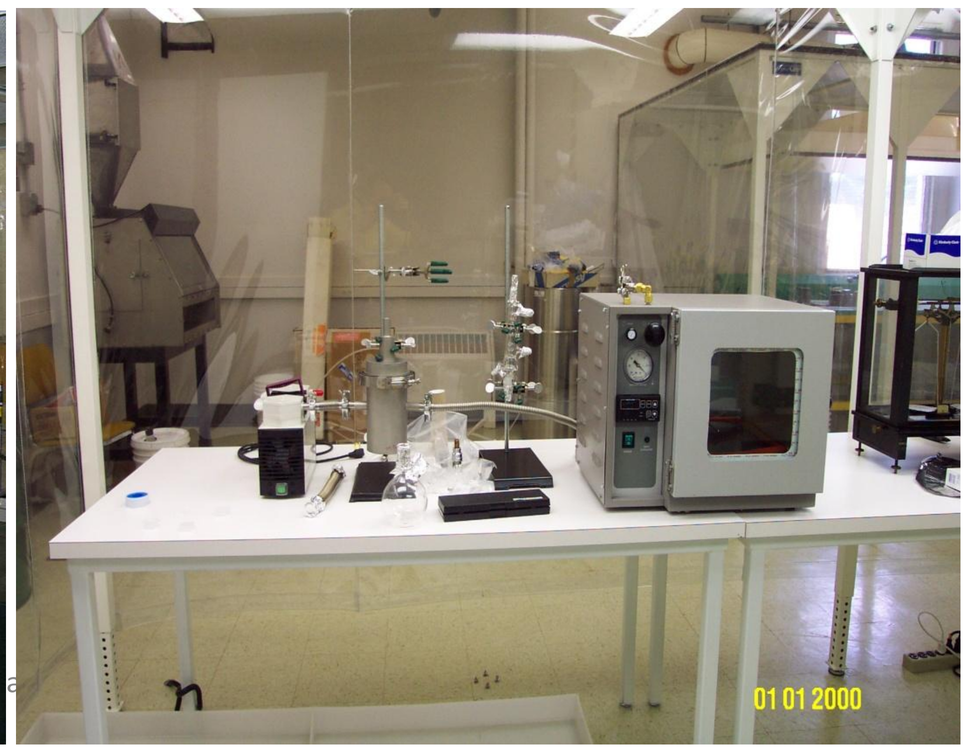
- Sensitivity can be increased by removing uninteresting side activities before counting.
- For liquid scintillator samples, post-radiation chemistry increased sensitivity to 10^{-15}g/g U/Th at UA.
- A group at TU Munich achieved 10^{-16}g/g U/Th utilizing chemistry and an onsite reactor.
- Only feasible for screening limited number of materials as each requires custom tailored chemical processes.

Neutron Activation Analysis

- Any neutron activation analysis requires a sample handling facility to handle considerable amounts of open radioactivity.
- Consistent pre-analysis treatment is critical to avoid surface contamination
  - At UA this is done in a class 500 clean room
  - High purity solvents, acids, and water for cleaning
- Activation costs for UA 1000--2000USD.
- Sample counting on, ideally multiple, large Ge detectors to combat decaying signal. Low background detectors are not required (except RNAA).

Procedure at UA

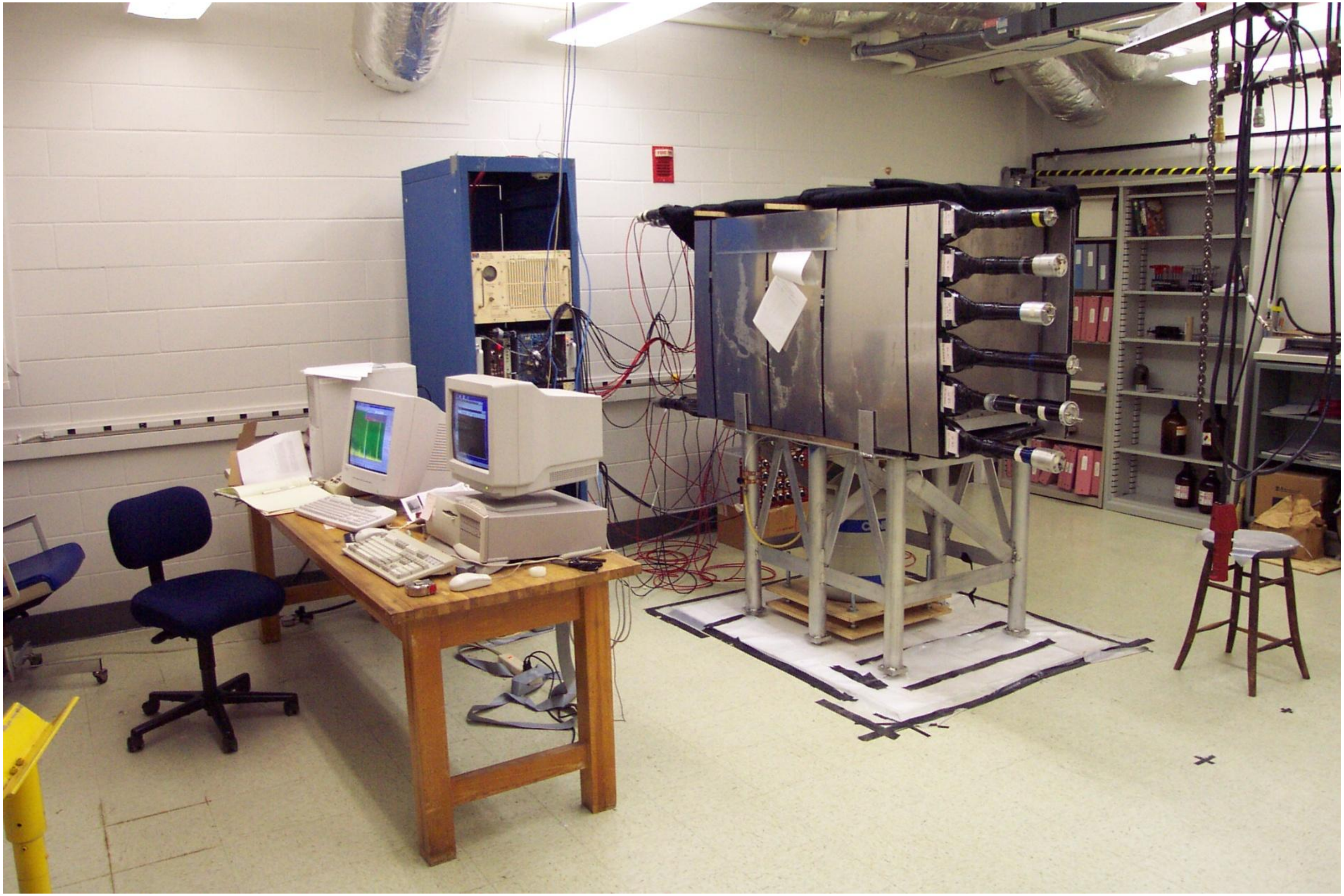
- Sample preparation in class 500 clean room.
- All lab supplies etched with ultra pure acid and rinsed with purified water.
- Depending on the required irradiation duration, sample activated in a small PE bottle or miniature quartz vial.



A few examples of reactors

Thermal neutron flux of some reactors with pneumatic sample insertion:

Reactor	Location	Thermal n Flux ($cm^{-2}s^{-1}$)
MITR	MIT	$5x10^{13}$
HFIR	Oak Ridge	$4x10^{14}$
MURR	University of Missouri	$6x10^{14}$
RFM II	TU Munich	$8x10^{14}$



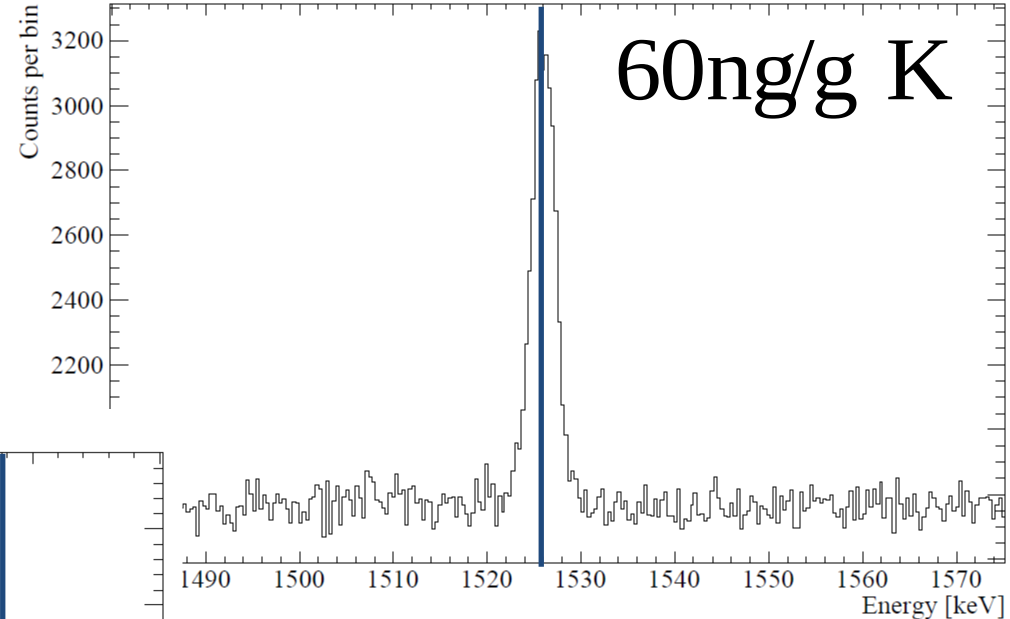
NA Analysis

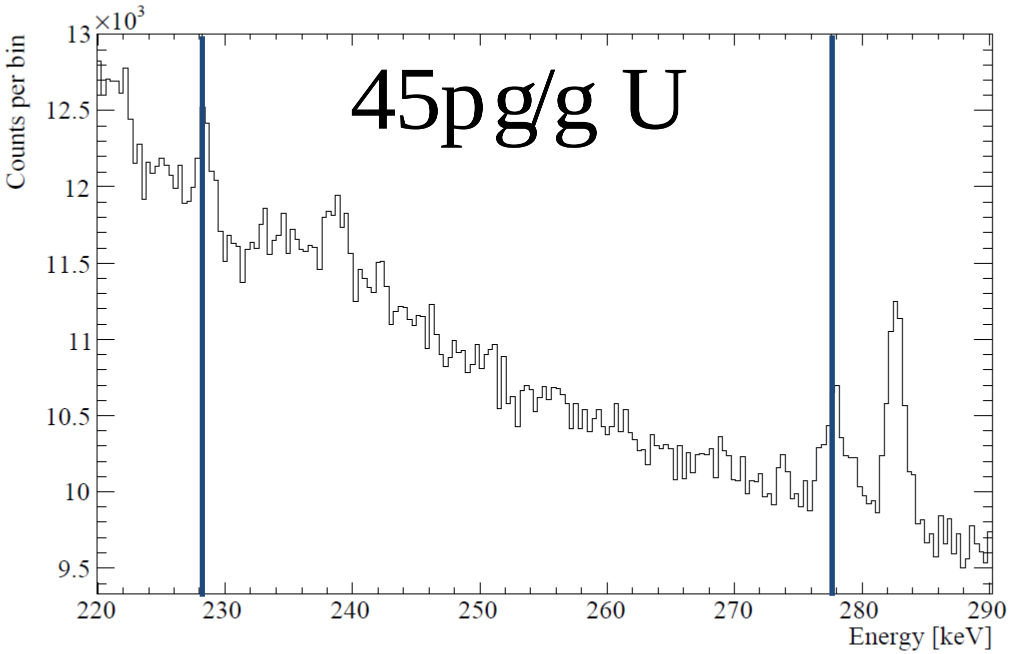
$$n + {}^{41}K \rightarrow {}^{42}K \xrightarrow[12.36h]{\beta^-} {}^{42}Ca, \quad \sigma_{th} = 1.2\ b$$

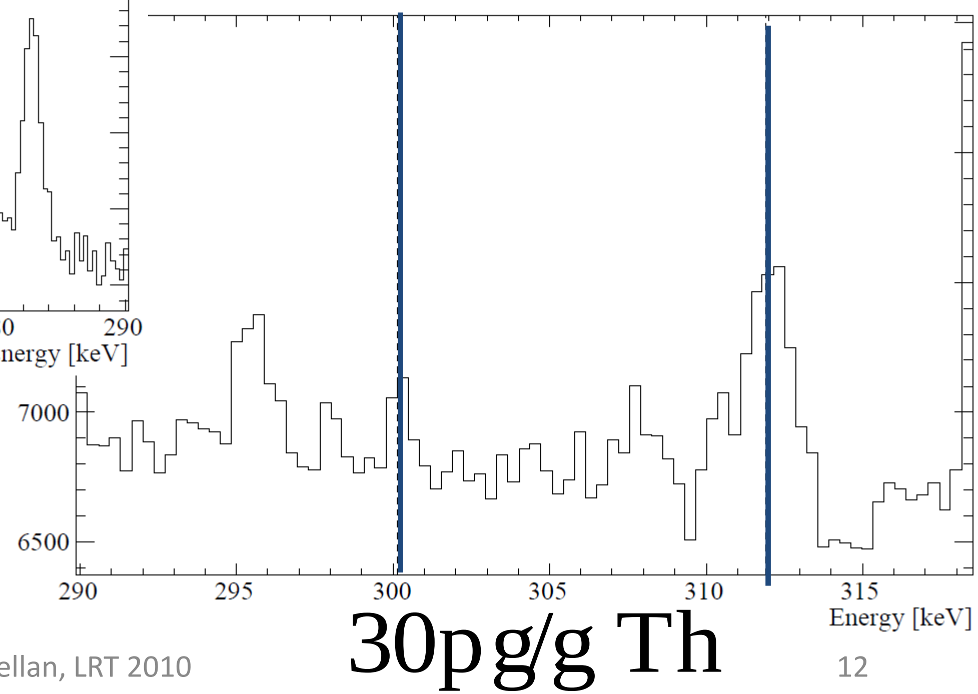
$$n + {}^{232}Th \rightarrow {}^{233}Th \xrightarrow[22.3min]{\beta^-} {}^{233}Pa \xrightarrow[27.0d]{\beta^-} {}^{233}U, \quad \sigma_{th} = 6.1\ b$$

$$n + {}^{238}U \rightarrow {}^{239}U \xrightarrow[23.5min]{\beta^-} {}^{239}Np \xrightarrow[2.36d]{\beta^-} {}^{239}Pu, \quad \sigma_{th} = 2.3\ b$$

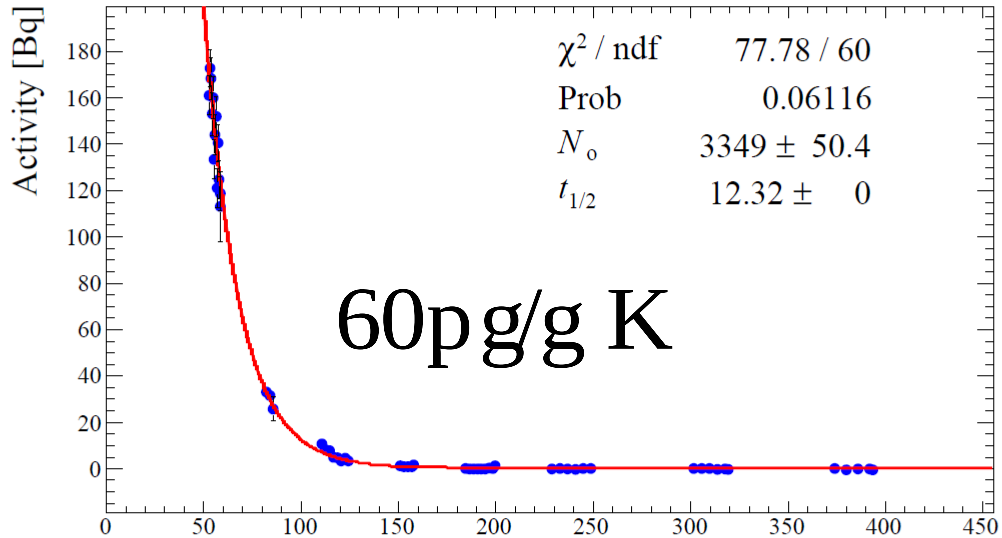
Evidence of natural radioactivity tracers

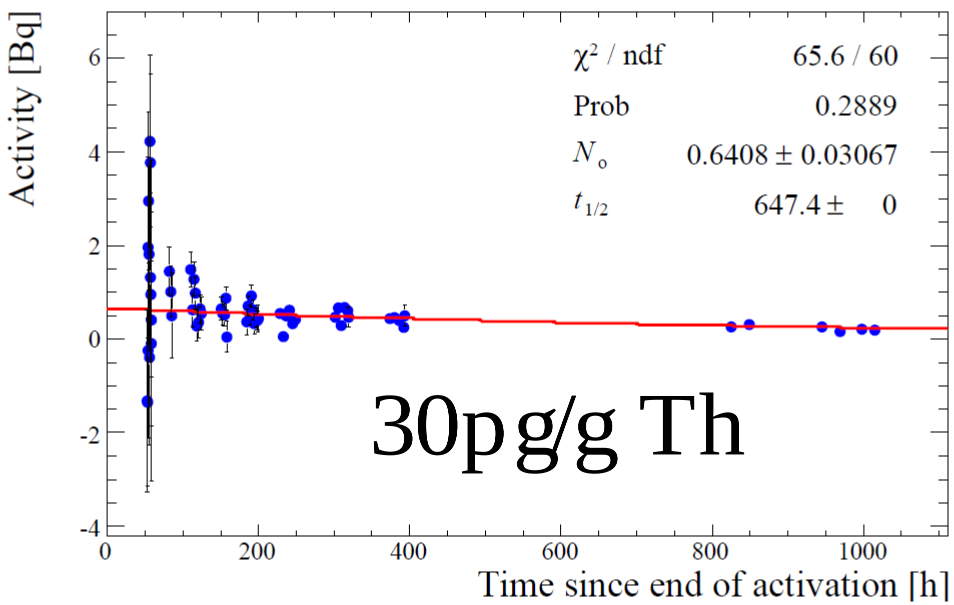


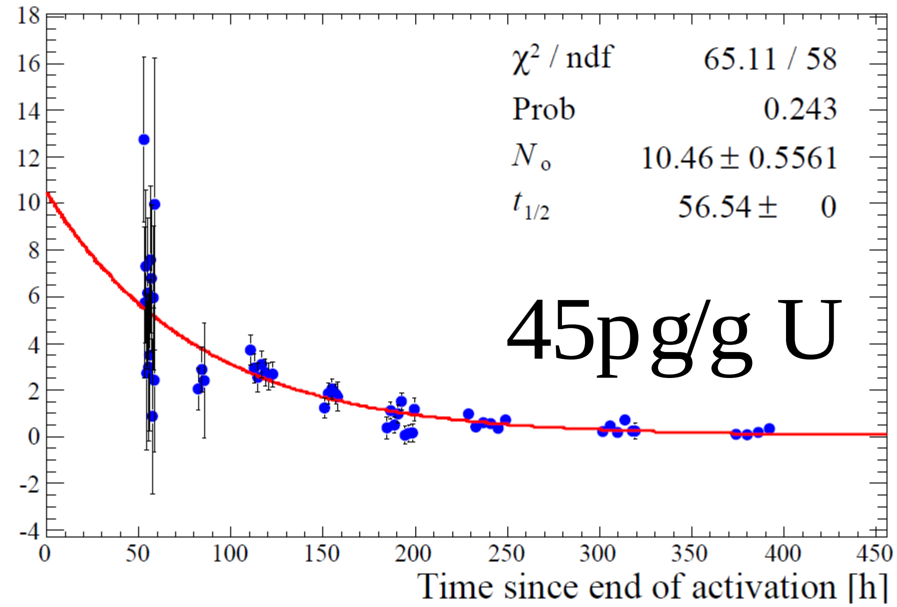




Time evolution of measured activity







Simply calculating constituents

Number of activated nuclei

Fraction remaining at time *t*

$$A = \phi_{\rm n}\sigma_{\rm tot}N(1 - e^{-\lambda t_{\rm irr}}) \times e^{-\lambda t}$$

Concentration of parent nuclei: $X[\mathsf{ng/g}] = 10^9 \frac{N}{N_{\rm A}} \frac{M}{x} / m_{\rm sample}$

Simply calculating constituents

Number of activated nuclei

Fraction remaining at time *t*

$$A = \phi_{\mathrm{n}}\sigma_{\mathrm{tot}}N(1 - e^{-\lambda t_{\mathrm{irr}}}) \times e^{-\lambda t}$$

Concentration of parent nuclei: $X[\mathsf{ng/g}] = 10^9 \frac{N}{N_{\mathrm{A}}} \frac{M}{x} / m_{\mathrm{sample}}$

For a two stage decay things get more complicated but still straight forwardly:

$$A = \phi_{\mathrm{n}}\sigma_{\mathrm{tot}}N \left(\frac{\lambda'}{\lambda' - \lambda} \frac{1 - e^{-\lambda t_{\mathrm{irr}}}}{\lambda} e^{-\lambda t} - \frac{\lambda}{\lambda' - \lambda} \frac{1 - e^{-\lambda' t_{\mathrm{irr}}}}{\lambda'} e^{-\lambda' t} \right) \lambda$$

Simply calculating constituents

Number of activated nuclei

Fraction remaining at time *t*

$$A = \phi_{\rm n}\sigma_{\rm tot}N(1 - e^{-\lambda t_{\rm irr}}) \times e^{-\lambda t}$$

Concentration of parent nuclei: $X[\mathsf{ng/g}] = 10^9 \frac{N}{N_{\rm A}}\frac{M}{x}/m_{\rm sample}$

For a two stage decay things get more complicated but still straight forwardly:

$$A = \phi_{\rm n}\sigma_{\rm tot}N\left(\frac{\lambda'}{\lambda' - \lambda}\frac{1 - e^{-\lambda t_{\rm irr}}}{\lambda}e^{-\lambda t} - \frac{\lambda}{\lambda' - \lambda}\frac{1 - e^{-\lambda' t_{\rm irr}}}{\lambda'}e^{-\lambda' t}\right)\lambda$$

Simply calculating constituents

Number of activated nuclei

Fraction remaining at time *t*

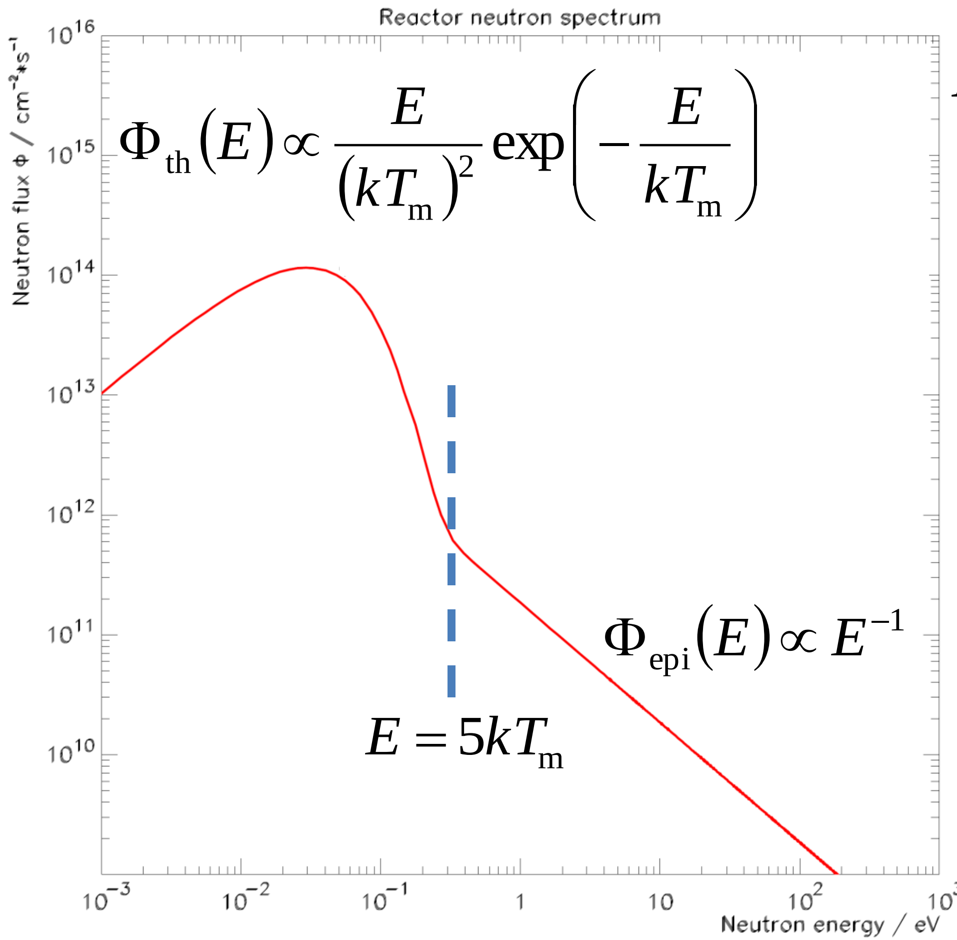
$$A = \phi_{\mathrm{n}}\sigma_{\mathrm{tot}} N(1 - e^{-\lambda t_{\mathrm{irr}}}) \times e^{-\lambda t}$$

Concentration of parent nuclei: $X[\mathsf{ng/g}] = 10^9 \frac{N}{N_{\mathrm{A}}} \frac{M}{x} / m_{\mathrm{sample}}$

For a two stage decay things get more complicated but still straight forwardly:

$$A = \phi_{\mathrm{n}}\sigma_{\mathrm{tot}} N \left(\frac{\lambda'}{\lambda' - \lambda} \frac{1 - e^{-\lambda t_{\mathrm{irr}}}}{\lambda} e^{-\lambda t} - \frac{\lambda}{\lambda' - \lambda} \frac{1 - e^{-\lambda' t_{\mathrm{irr}}}}{\lambda'} e^{-\lambda' t} \right) \lambda$$

Parametrize the neutron flux: thermal vs. epithermal neutrons



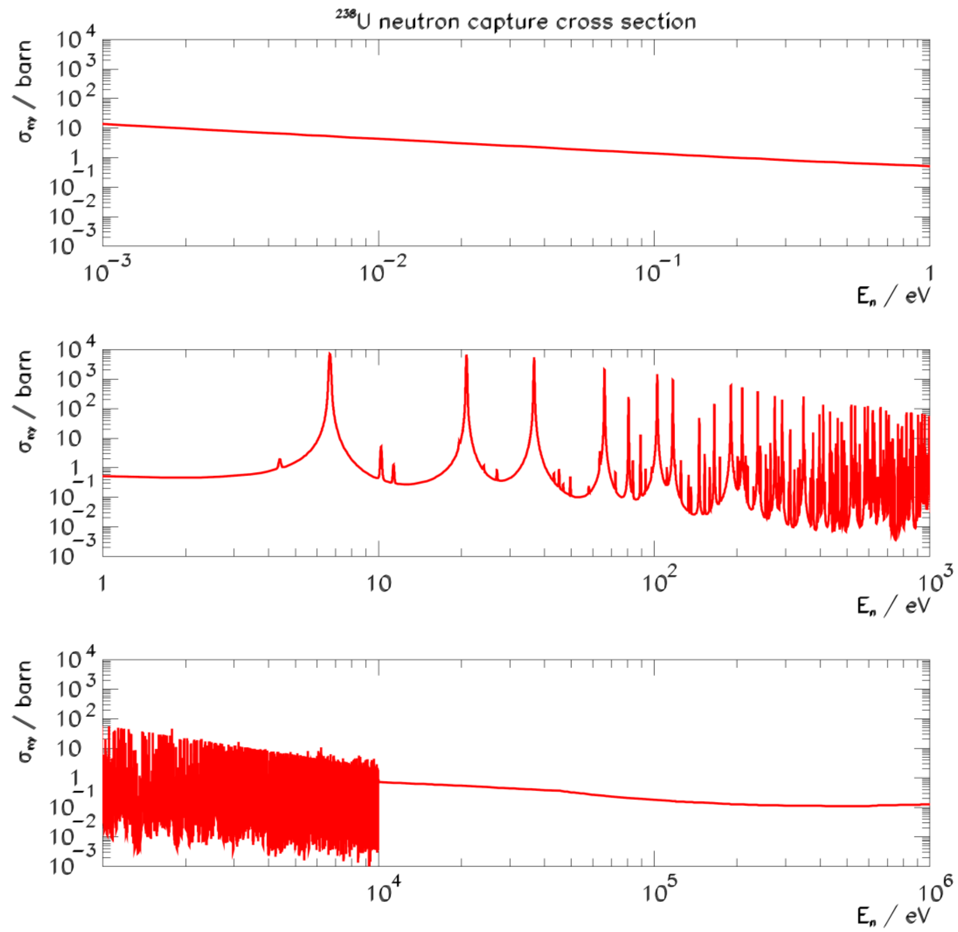
$$\Phi_{\text{th}}(E) \propto \frac{E}{(kT_{\text{m}})^2} \exp\left(-\frac{E}{kT_{\text{m}}}\right)$$

$$E = 5kT_{\text{m}}$$

$$\Phi_{\text{epi}}(E) \propto E^{-1}$$

At MITR and HFIR $T_{\text{m}} \approx 340\text{K}$

Parametrize the product of neutron flux and cross section; thermal vs. epithermal neutrons



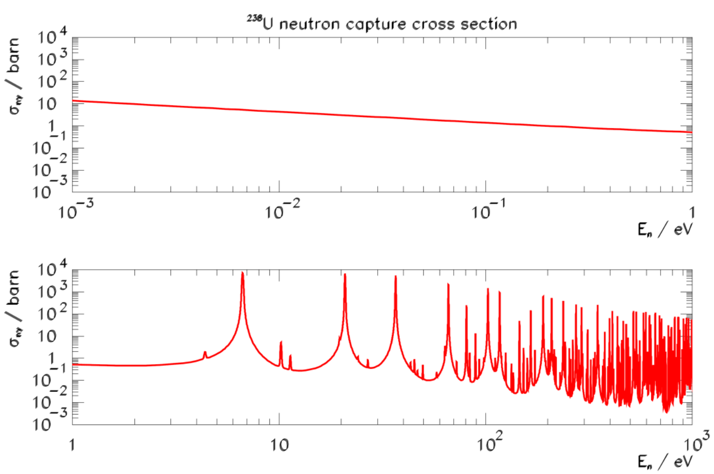
Parametrize the product of neutron flux and cross section; thermal vs. epithermal neutrons

$$\int \sigma(E)\Phi(E)\mathrm{d}E \approx \phi_{\mathrm{th}}\left(\langle\sigma_{\mathrm{th}}\rangle + \frac{1}{f}\langle\sigma_{\mathrm{epi}}\rangle\right) \equiv \phi_{\mathrm{n}}\sigma_{\mathrm{tot}}$$

Thermal neutron flux: $\phi_{\mathrm{n}} \equiv \phi_{\mathrm{th}}$

Epithermal flux suppression factor: $f \equiv \dfrac{\phi_{\mathrm{th}}}{\phi_{\mathrm{epi}}}$

$$\langle\sigma_{\mathrm{th/epi}}\rangle = \frac{\int_{E_{\mathrm{th/epi}}} \sigma(E)\Phi(E)\mathrm{d}E}{\int_{E_{\mathrm{th/epi}}} \Phi(E)\mathrm{d}E}$$



$$\phi_{\mathrm{th/epi}} = \int_{E_{\mathrm{th/epi}}} \Phi(E)\mathrm{d}E$$

National Institute of Standards & Technology

Certificate of Analysis

Standard Reference Material® 1633b

Constituent Elements in Coal Fly Ash

Al	15.05	±	0.27
Ca	1.51	±	0.06
Fe	7.78	±	0.23
Mg	0.482	±	0.008
K	1.95	±	0.03
Si	23.02	±	0.08
Na	0.201	±	0.003
S	0.2075	±	0.0011
Ti	0.791	±	0.014

As	136.2	±	2.6
Ba	709	±	27
Cd	0.784	±	0.006
Cr	198.2	±	4.7
Cu	112.8	±	2.6
Pb	68.2	±	1.1
Mn	131.8	±	1.7
Hg	0.1431	±	0.0018
Ni	120.6	±	1.8
Se	10.26	±	0.17
Sr	1041	±	14
Th	25.7	±	1.3
U	8.79	±	0.36
V	295.7	±	3.6

Global χ^2 minimization of

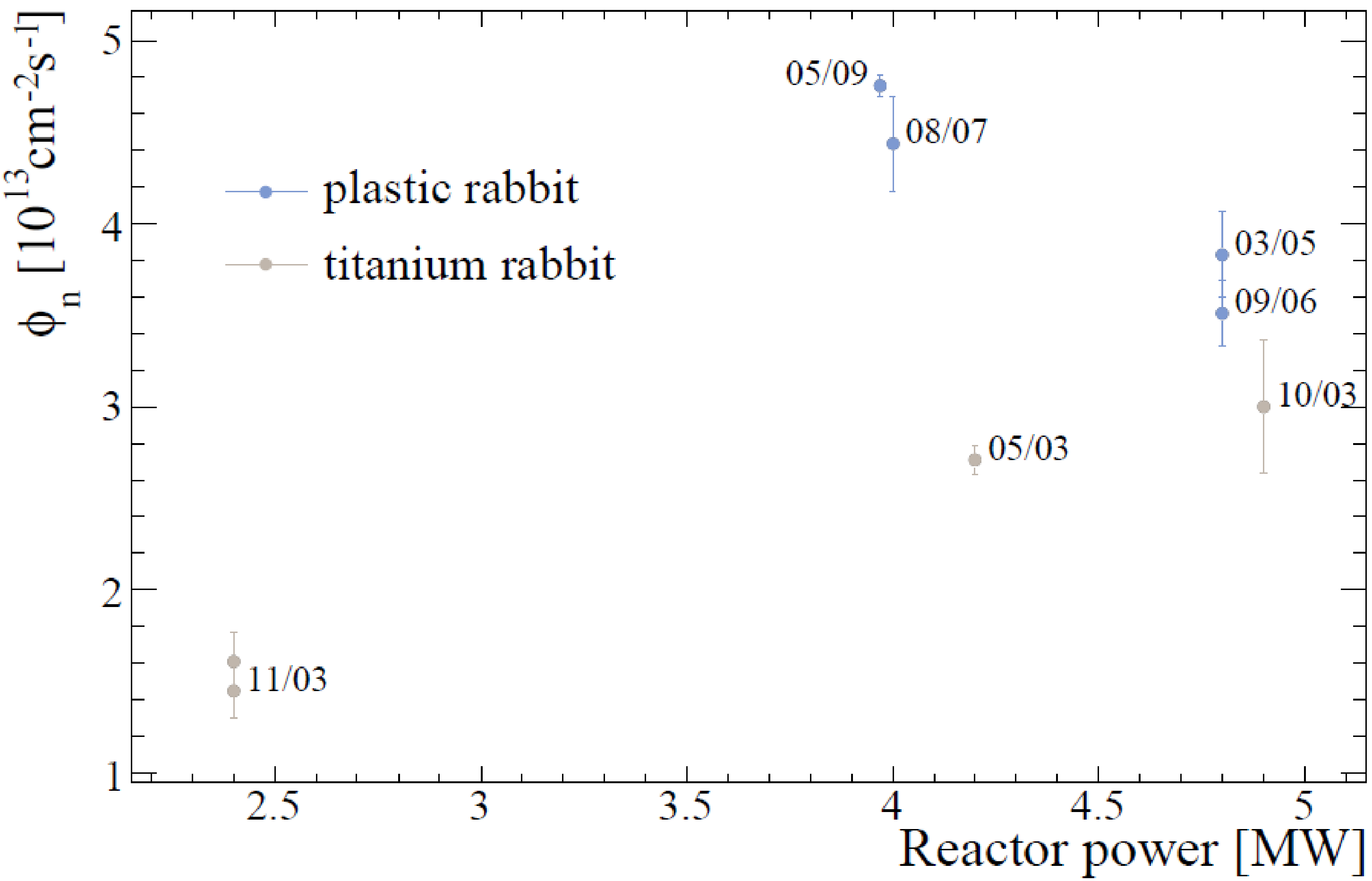
$$A = \phi_{\rm n}\sigma_{\rm tot}N(1 - e^{-\lambda t_{\rm irr}}) \times e^{-\lambda t}$$

N measured vs N expected

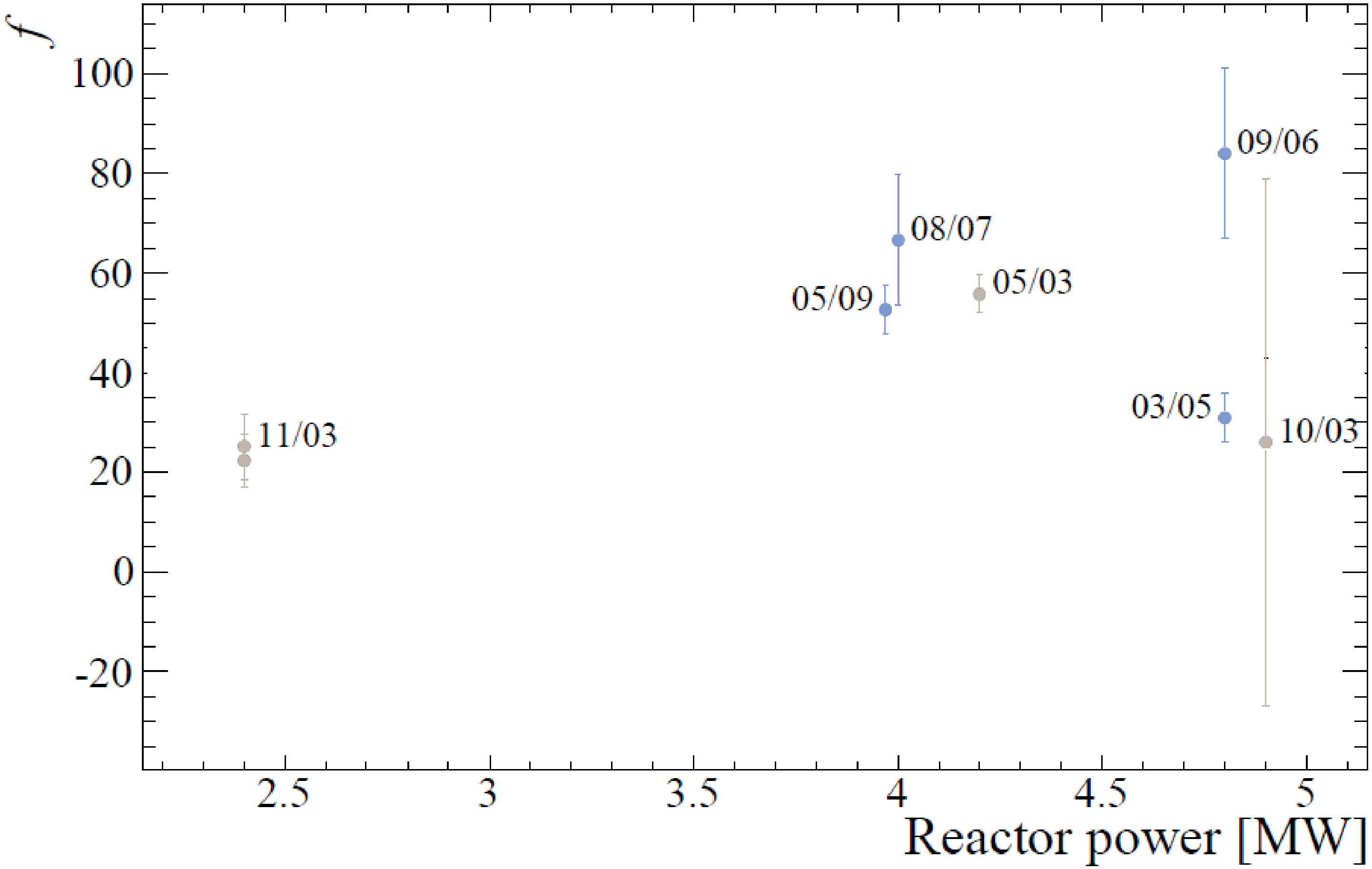
$$N = N_{\rm A}\frac{x}{M}m_{\rm sample}m_{\rm frac}$$

yields $\phi_{\rm n}$ and f

MITR ϕ_n as a function of reactor power



MITR *f* as a function of reactor power



Summary

- NAA has been successfully used in the preparation of previous successful low background experiments. It offers ppt to sub ppt-sensitivity depending effort made to avoid source related background.
- Complements ICP-MS technique. Well suited for most plastics that are inaccessible to traditional MS.

One final note: document surveys

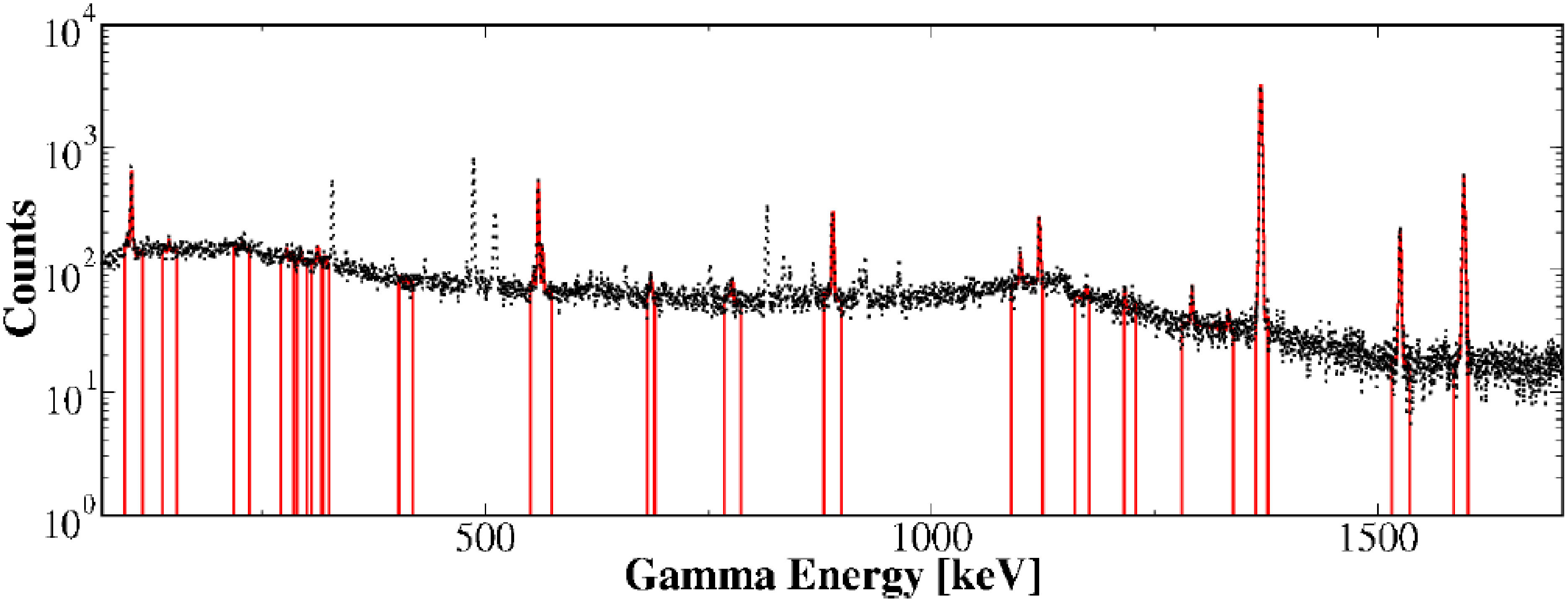
Systematic study of trace radioactive
impurities in candidate construction materials
for EXO-200

10 pages of table (small font) listing all materials characterization for EXO-200 construction (useful and otherwise). Most cited EXO paper to date!

D. S. Leonard *et al.*, Systematic study of trace radioactive impurities in candidate construction materials for EXO-200, Nucl. Instrum. Meth. A **591**, 490 (2008) [arXiv:0709.4524]

UA KamLAND scintillator analysis

1. Collection of Scintillator
   - Ship samples to US
2. Preconcentration
   - Remove Reactor-unfriendly organics
3. Irradiation at Reactor (ORNL/MITR)
4. Post-chemistry
   - Separate “Signal” isotopes from “Background”
5. γ Counting (HPGe)
6. Analysis and Results



10^4
10^3
10^2
10^1
10^0
Counts
500
1000
1500
Gamma Energy [keV]

Status	Radionuclide	Activity	Activity error	Model prediction	χ^2 contribution
on	^{24}Na	13640	200	13537.6	0.2
on	^{42}K	6320	110	6508.4	2.8
on	^{59}Fe	2220	130	1810.4	9.7
on	^{51}Cr	1650	60	1601.9	0.6
off	^{76}As	10760	210	13052.4	119.0
on	^{203}Hg	10.5	3.6	0.4	7.9
on	^{233}Pa	654	36	639.4	0.2
on	^{239}Np	770	50	769.7	0.0
off	^{124}Sb	36.2	2.4	44.8	12.3
off	^{46}Sc	5750	60	5715.3	0.4
off	^{60}Co	335.8	1.8	316.9	106.6
off	^{65}Zn	129.9	3.3	93.3	123.0
off	^{82}Br	142	6	121.6	13.3
off	^{140}La	11572	31	12324.2	598.6
off	^{181}Hf	93	10	77.5	2.4
off	^{182}Ta	59.6	2.9	58.5	0.2

Effective Neutron Capture Cross Sections

Andreas Piepke
Department of Physics and Astronomy
University of Alabama

April 18, 2001

Isotope	Tabulated [barn]			This evaluation [barn]		
	σ 293.6 K	$\langle\sigma_{th}\rangle$ 293.6 K	$\langle\sigma_{epi}\rangle$	$\langle\sigma_{th}\rangle$ 293.6 K	$\langle\sigma_{th}\rangle$ 340 K	$\langle\sigma_{epi}\rangle$
^{23}Na	0.531	0.471	0.311	0.467	0.434	0.349
^{41}K	1.459	1.294	1.58	1.298	1.206	1.664
^{50}Cr	15.92	14.12	7.38	14.13	13.13	8.30
^{58}Fe	1.300	1.153	1.357	1.020	0.948	1.561
^{59}Co	37.18	32.96	75.51	33.07	30.72	76.68
^{64}Zn				0.677	0.629	1.564
^{81}Br	2.690	2.385	46.63	2.385	2.217	51.03
^{197}Au				88.02	81.99	1566.7
^{232}Th	7.400	6.532	84.35	6.567	6.095	86.91
^{238}U	2.717	2.414	278.1	2.421	2.251	278.3