

# Trace element analysis by means of neutron activation

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Many low rate, low energy experiments have very strict requirements for their content of radio-nuclides.

Natural radioactivity K, Th, and U is omnipresent. Many experiments go through great length to identify and certify their construction materials regarding these impurities.

This is often a time consuming and costly process.

If DUSL would offer a centralized site for high-sensitivity radio activity testing, duplication of work could be avoided. For our community this would enhance competitiveness (through time saving) and certainly result in cost savings (avoid duplication of lab effort).

This talk will focus on natural radio activity.

Techniques typically utilized:

- Direct  $\gamma$ - and to some extent  $\alpha$ - and  $\beta$ -counting. Typically requires large samples ( $10^{-14}$  g/g U gives 1 decay per day per 100 kg sample) and long counting time to obtain sufficient sensitivity. Measures the background creating members of the decay series directly.
- Mass spectroscopy can yield K, Th, U sensitivities of  $10^{-9}$  and  $10^{-12}/10^{-13}$  g/g. Requires only g-size samples done typically within a week. Applicable to materials that can be digested in acids (ICPMS) or are conductors (GDMS). Measures Th, U: material qualification involves the *assumption* of chain equilibrium.

- Neutron activation analysis achieves sensitivities (K, Th, U) of  $10^{-9}$  and  $10^{-12}/10^{-13}$  g/g for straight  $\gamma$ -counting of g-size samples following activation at a research reactor. At UA we work with the reactor at MIT. In the past we have also utilized the high flux reactor at ORNL. This involves typically a one day delay for initial cool-down and shipping by UPS/FEDEX. This technique is only applicable to matrix materials with low neutron capture cross section. Many metals are unsuited. High sensitivity analysis can take up to one month due the long half life of the Th activation product  $^{233}\text{Pa}$ .

- To enhance sensitivity by removing unwanted side activities (and thus reducing source related background) the sensitivity can be boosted.

For liquid scintillator samples such post-radiation chemistry yielded Th/U sensitivities of  $10^{-15}$  g/g at UA. A group at the TU Munich, utilizing an on-site reactor, even reported  $10^{-16}$  g/g.

This approach is usually only feasible for the screening of a limited number of material types as the chemistry has to be custom tailored and its effectiveness experimentally verified. This add some overhead.

Both flavors of NAA require a sample handling facility suited for handling of considerable amounts of open radio activity.

In all cases a consistent pre-analysis treatment is important. This is to avoid surface contamination and thus spurious readings when converting measurements on small samples to a large amount of construction materials.

A centralized analysis facility will thus also need clean room space with the capability to handle chemicals (organic solvents, pure acids, high purity water) for pre-analysis treatment. At UA a class 500 clean area is sufficient to obtain the sensitivities listed before.

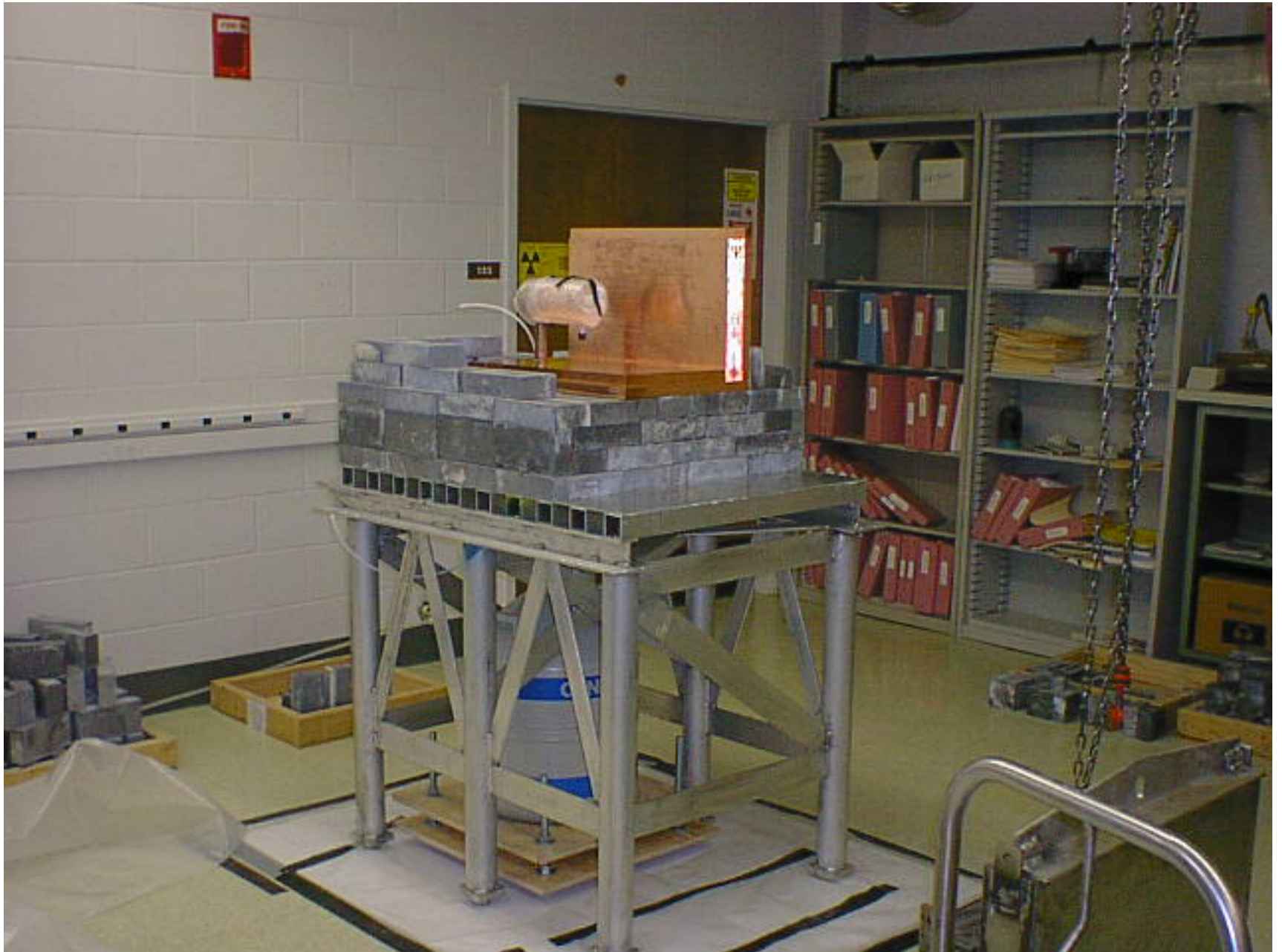
Depending on the activation length irradiation reactor costs of the order \$2000 per run (UA: up to 5 samples).

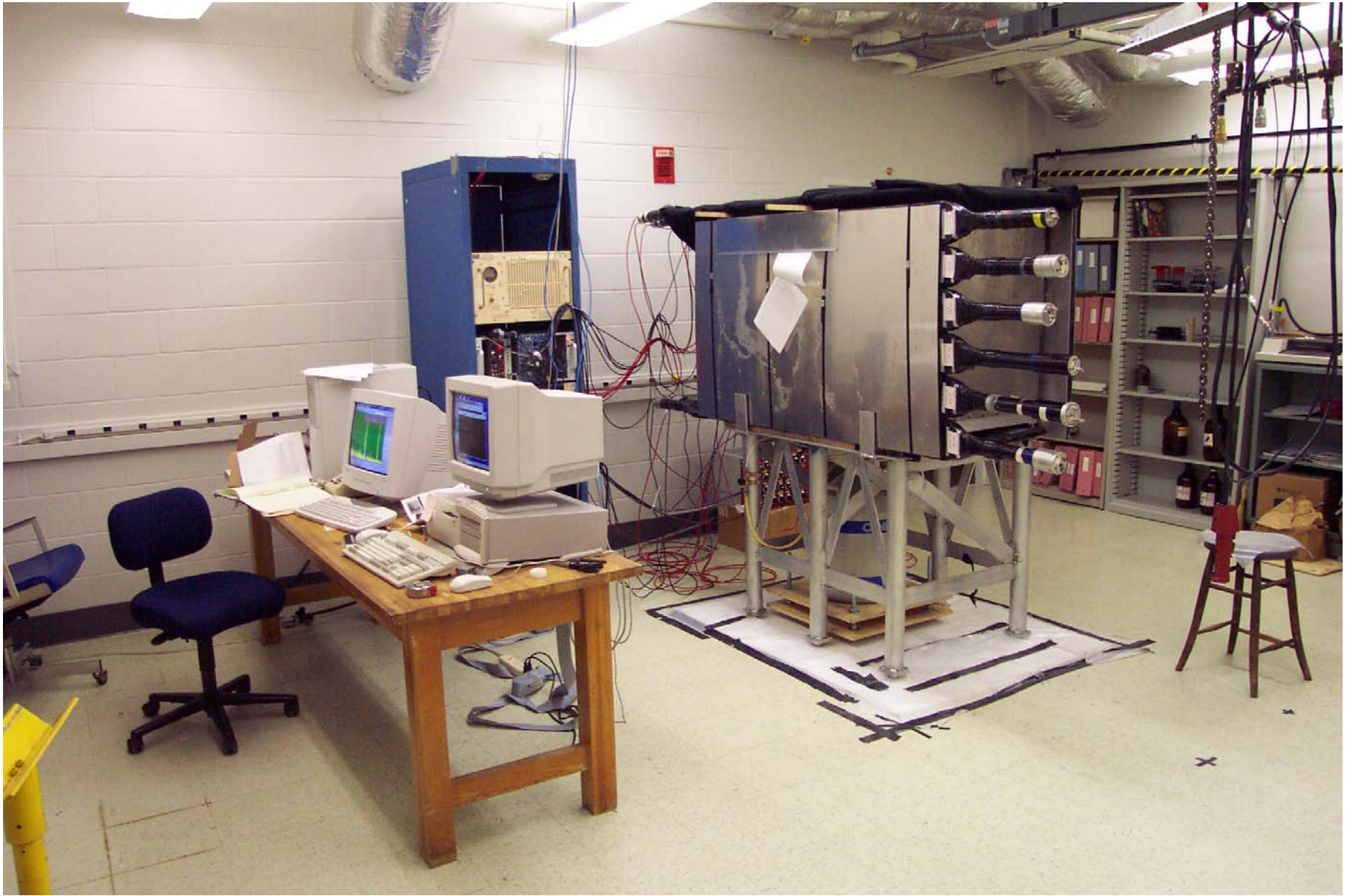
Sample counting requires multiple large Ge detectors to avoid decay. In most cases low background detectors are not needed (except RNAA and Th analysis).

- Preparation of samples in class 500 clean room.
- All containers etched with ultra pure acid.
- Rinsed with purified water.
- Depending on  $t_i$  and type of sample use small PE bottle or miniature quartz vial. Both melted shut before irradiation.



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# Conclusion

NAA has been successfully used in the preparation of previous low background experiments: SNO, KamLAND, Borexino. It offers ppt to sub ppt-sensitivity depending effort made to avoid source related background.

It nicely complements the capabilities of ICPMS in being well suited for most plastics that are inaccessible to MS.

If DUSL wants to go beyond an underground counting facility and become a center for trace analysis NAA has to be in its repertoire.

Thermal neutron fluxes of some reactors that have pneumatic sample insertion facilities:

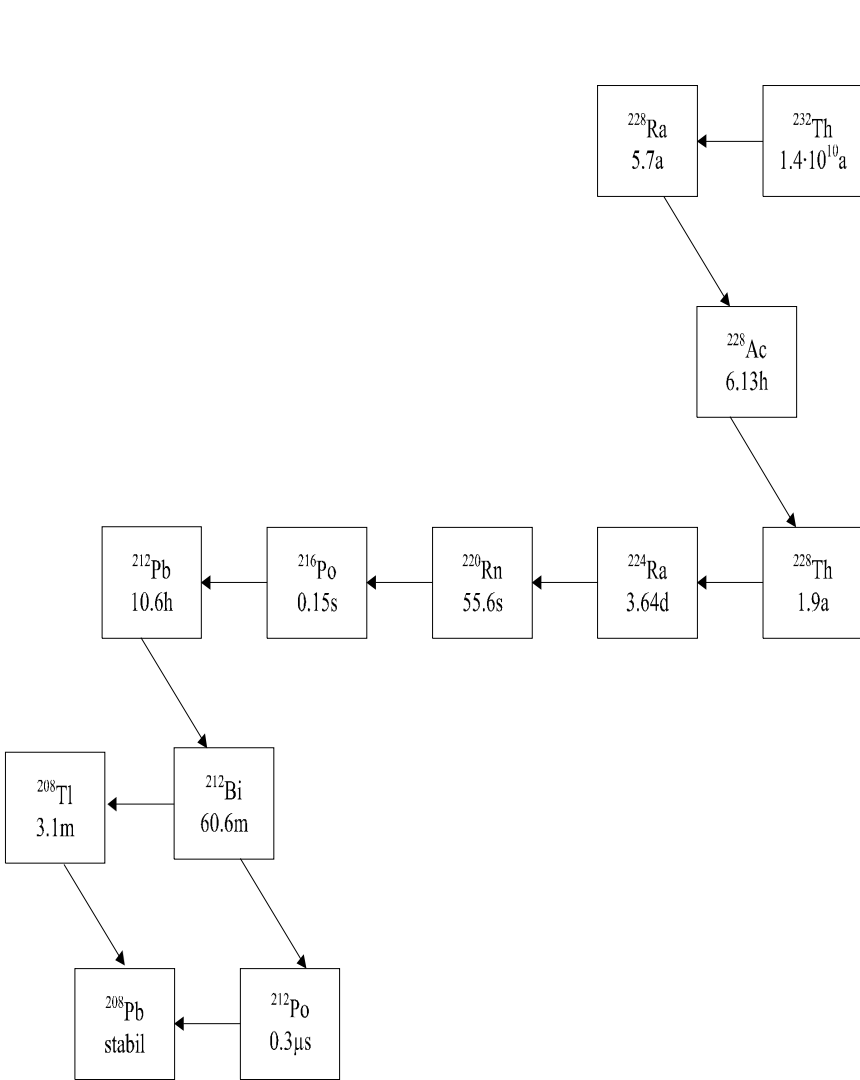
MITR:  $5 \cdot 10^{13}$  n/cm<sup>2</sup> s

HFIR:  $4 \cdot 10^{14}$  n/cm<sup>2</sup> s

MURR:  $8 \cdot 10^{13}$  n/cm<sup>2</sup> s

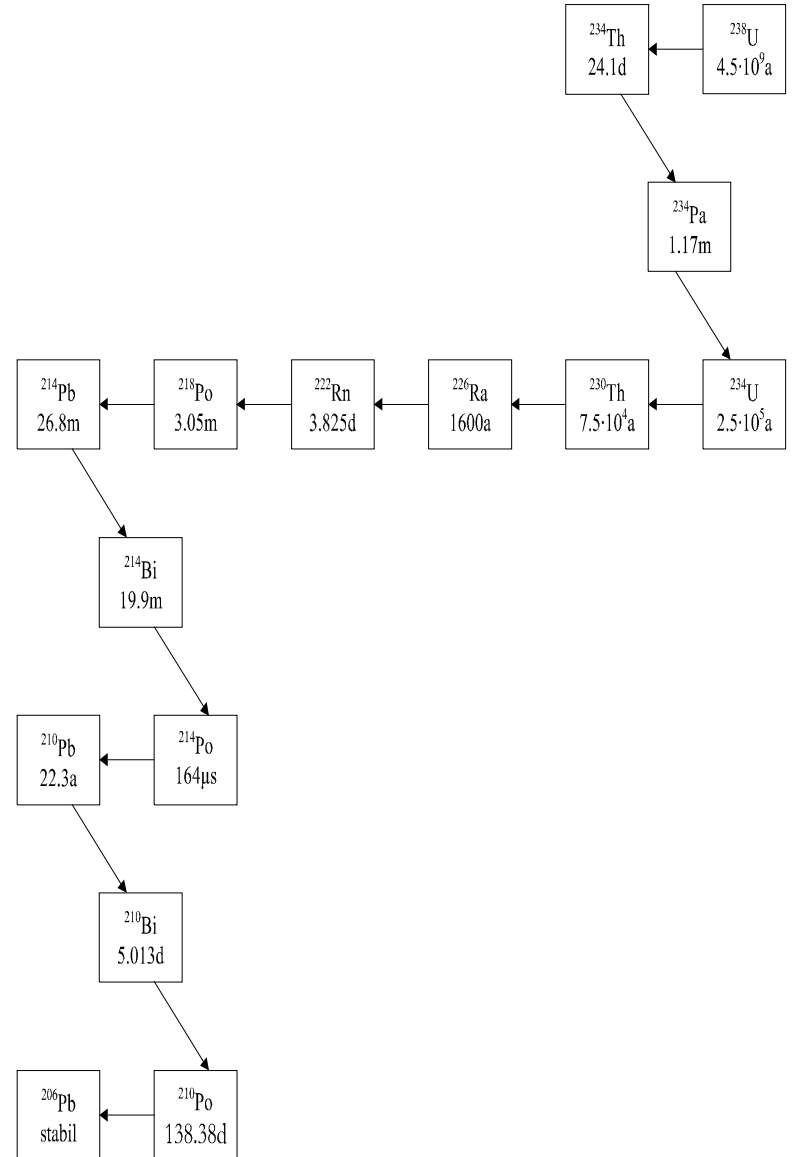
FRM II:  $8 \cdot 10^{14}$  n/cm<sup>2</sup> s

# Th chain



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# U chain



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# External Activities have less strict requirements

All external components are tested for activity  
By means of low background  $\gamma$ -ray spectroscopy.

Tests all members of the decay series

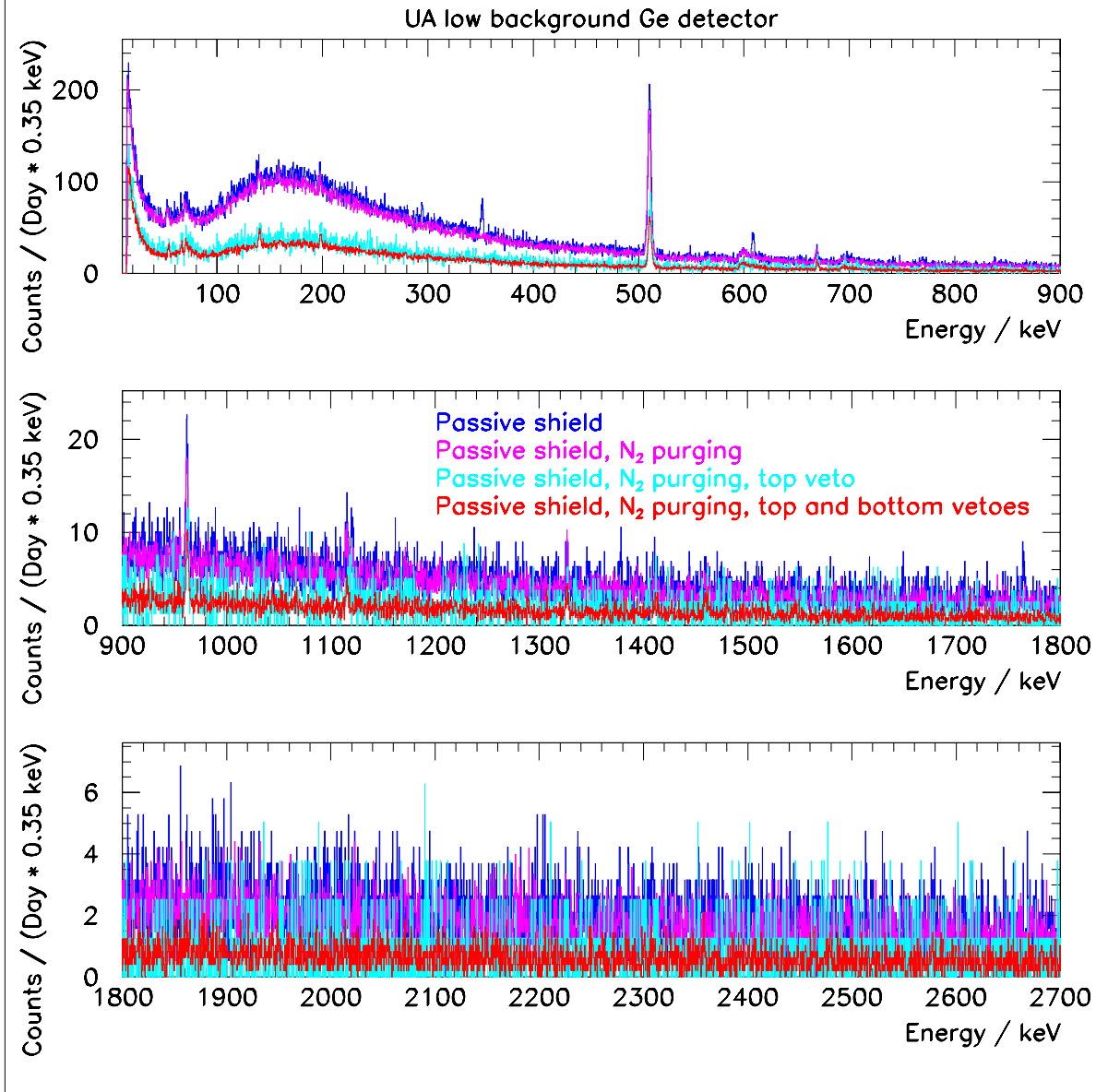
Use “above ground” set up.

$^{40}\text{K}$  : 8 mBq/kg or 30 ppt

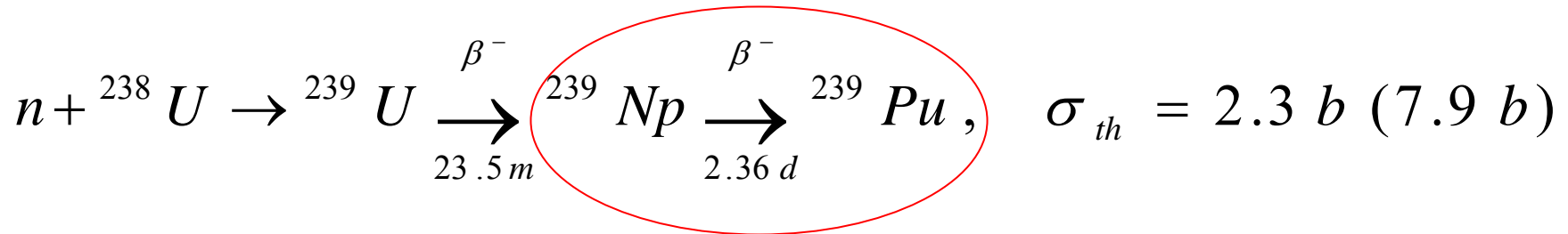
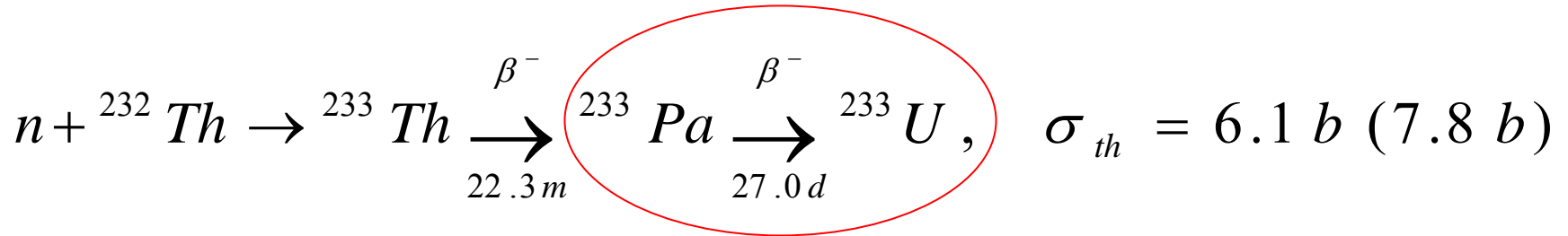
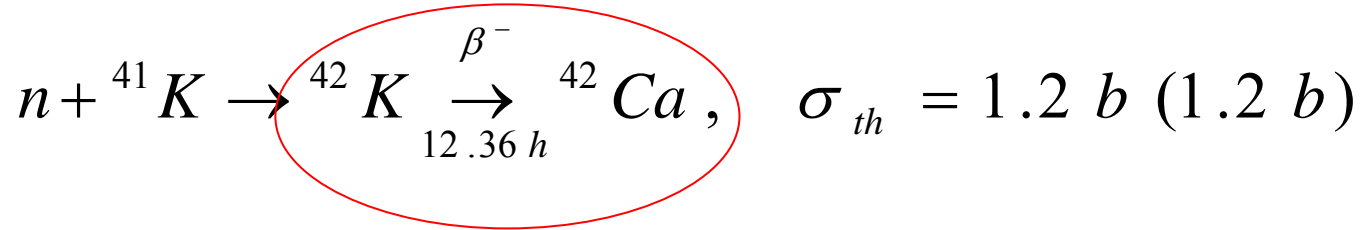
$^{232}\text{Th}$ : 3 mBq/kg or 700 ppt

$^{238}\text{U}$  : 4 mBq/kg or 300 ppt

**Sufficient for most external materials**



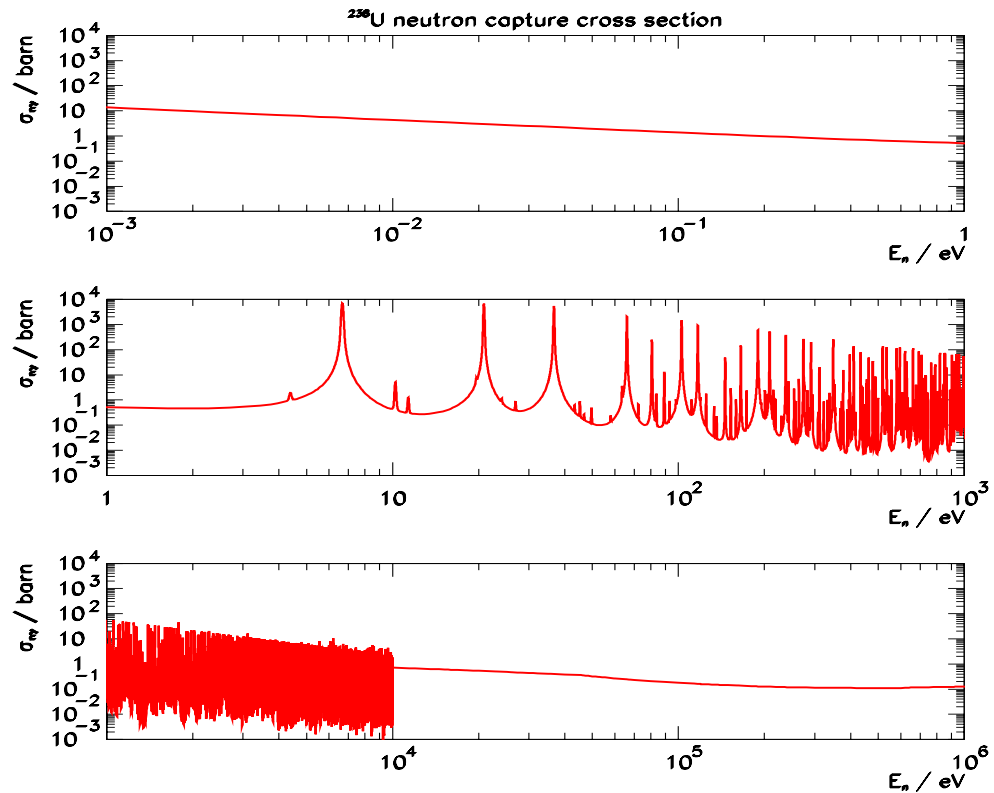
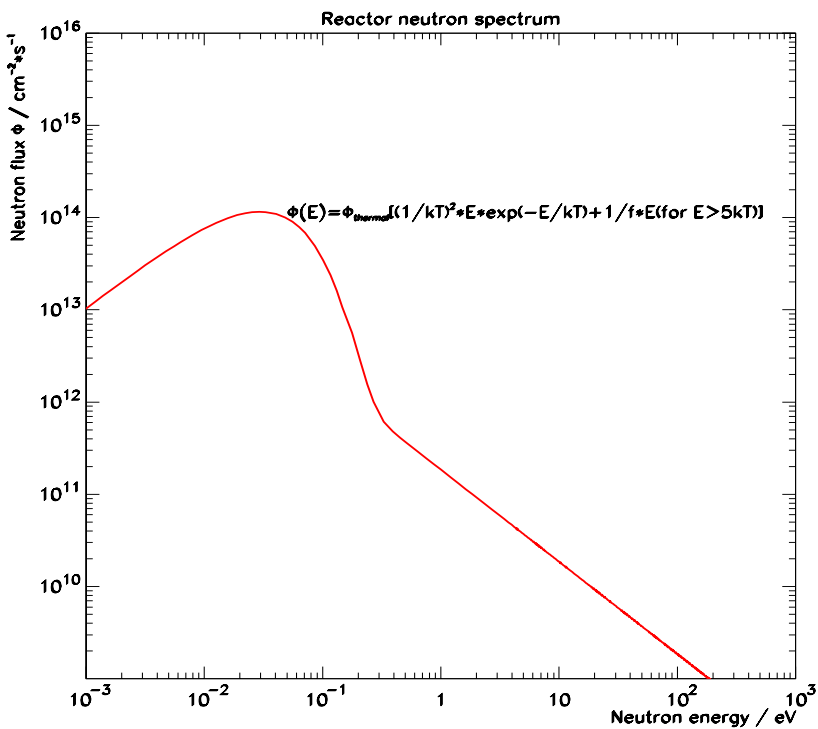
# Neutron activation analysis



Sizable cross sections and long enough half lives for delayed counting

# Reaction rate relates to concentration of parent nuclide

$$R = N \cdot \int \Phi(E_n) \sigma(E_n) dE_n$$



$$R = \Phi_{th} \left( \langle \sigma_{th} \rangle + \frac{1}{f} \langle \sigma_{epi} \rangle \right) \cdot N$$

# Decay rate of activation product relates to reaction rate

$$R_y = \frac{N_y}{\tau_y} = \Phi_{th} \left( \langle \sigma_{th} \rangle + \frac{1}{f} \langle \sigma_{epi} \rangle \right) \cdot N_x \cdot \left( 1 - e^{-t_i/\tau_y} \right) \cdot e^{-t_s/\tau_y}$$

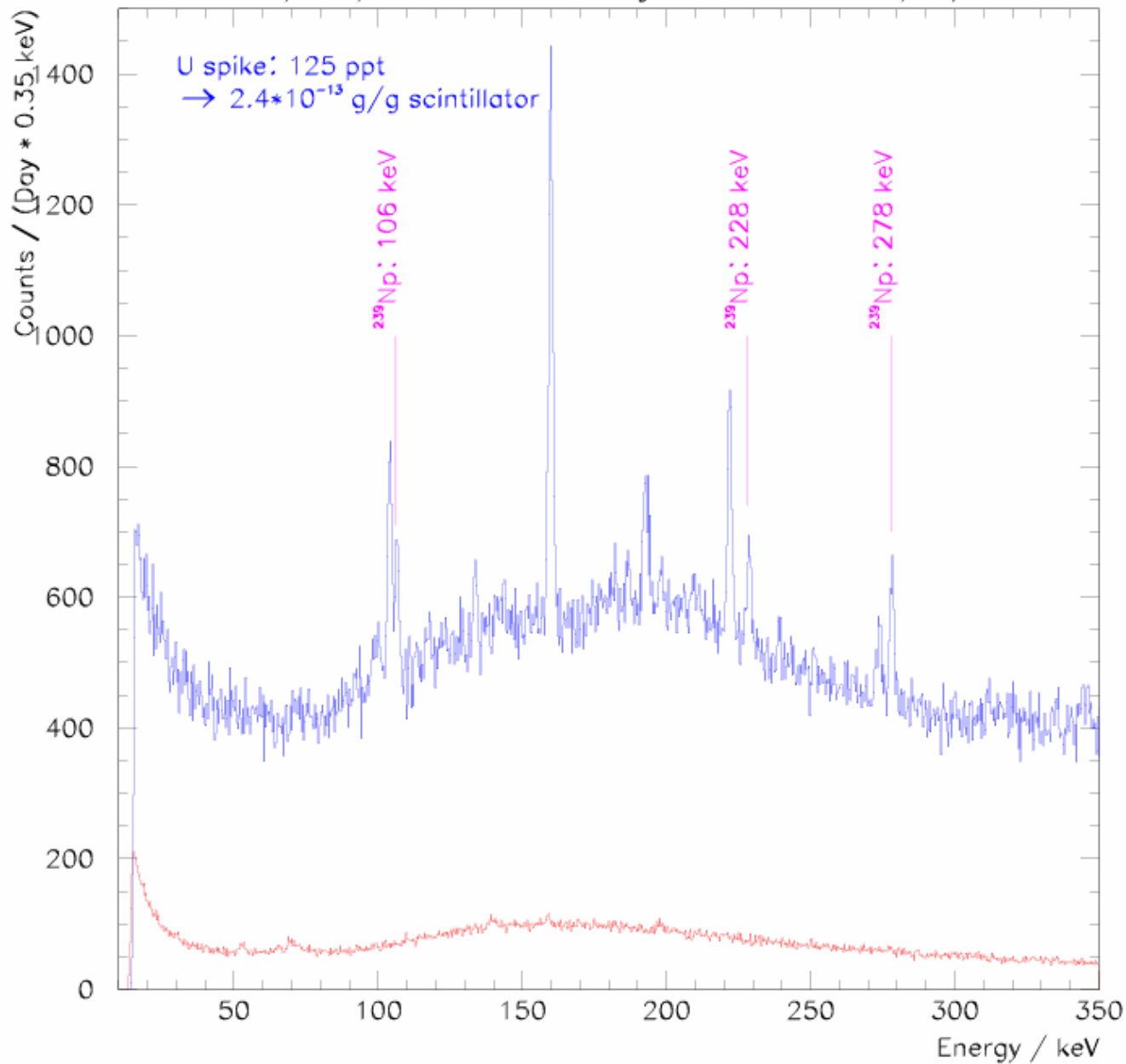
To achieve high sensitivity (large  $R_y$ ):

- High neutron flux  $\Phi \rightarrow$  use high flux research reactor

Oak Ridge National Lab


MIT

UA36 Th/U-spiked after ion exchange. Activated at MIT 6/10/2001



# Activation Analysis of KamLAND Scintillator

**Goal: Reliable, Independent monitor of Purity and Backgrounds**

Clean  


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”

Hot  


5.  $\gamma$  Counting (HPGe)

6. Analysis and Results



## Samples contain various activities after irradiation

- Separate sample from container using hot  $\text{HNO}_3$ .
- Separate Pa and Np from side activities by ion exchange:  
Use commercial CMPO resin dissolved in TBP

CMPO: Octylphenyl-N, N-di-ISO-butyl Carbamoylphosphin Oxide

TBP: Tri-N-Butyl Phosphate

→ red. factor for  $^{24}\text{Na} > 1000$  (large)

$^{82}\text{Br} \sim 7$  (large)

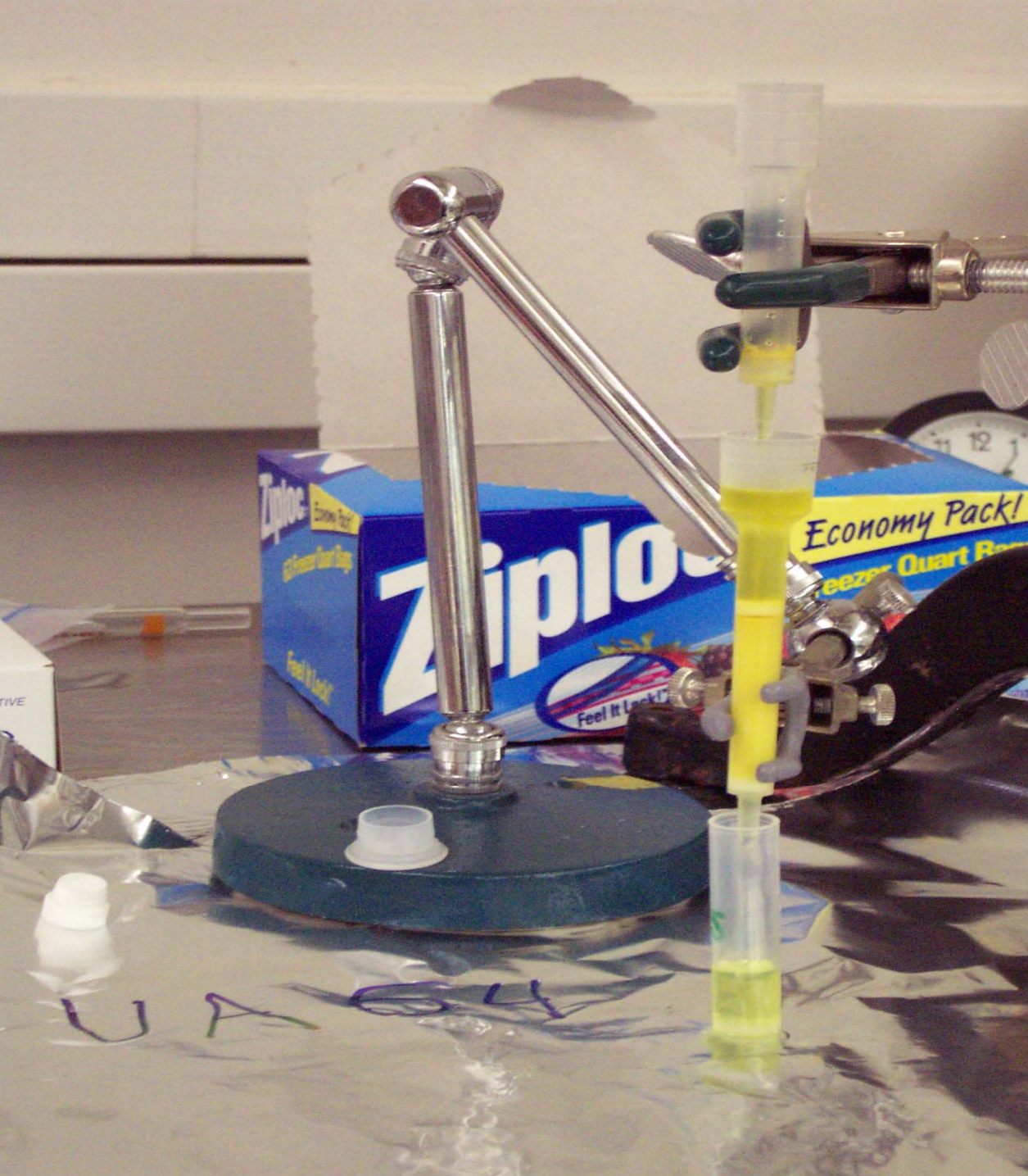
$^{46}\text{Sc} \sim 2$

$^{51}\text{Cr} \sim 500$

$^{65}\text{Zn} \sim 300$

$^{198}\text{Au} \sim 5$

$^{233}\text{Pa}, ^{239}\text{Np} \sim 1.3$  → **organic tracer in PPO**  
**inorganic form: 1.0**



Just count the ion exchange column

