2172-11

Joint ICTP-IAEA Workshop on Nuclear Data for Science and Technology: Analytical Applications

8 - 12 November 2010

The k0-Method of Instrumental Neutron Activation Analysis
Irradiation Facility Characterization

Menno BLAAUW
Technical University of Delft, Neutrons & Moessbauer Department, Mekelweg 15
2629 JB
Delft
THE NETHERLANDS

The k_0-method of Instrumental Neutron activation analysis

Irradiation facility characterization

Trieste, Nov 8-10, 2010

Delft University of Technology

Overview

- What we want to do in the end
- Standardization methods
- How to implement the k_0-method
  - Gamma-ray spectrometry
  - Detector characterization (Monday afternoon)
    - Coincidence-summing based detector calibration
    - Advanced topic I (Tuesday morning)
    - Advanced topic II (Tuesday morning)
  - Irradiation facility characterization (Tuesday afternoon)
- The interpretation step

What we want to do in the end

- The measure peak area *a* is linearly proportional to the elemental concentration *c*. The proportionality constant is *m.*
- *c* is what we want to know, *a* is what we measure, *m* is what we get from the calibration and/or standardization process.
- So we want to determine *m* first, and then analyze samples
- *m* is the peak area that would be measured if the elemental concentration was 1 kg/kg

$$a = mc$$

How to calculate *m*

$$m = \frac{w\theta N_{avogadro}}{M} R \frac{\left(1 - e^{-\lambda t_{ir}}\right)}{\lambda} e^{-\lambda t_d} \left(1 - e^{-\lambda t_m}\right) P$$

where w is the sample mass, θ the isotopic abundance, M the molar mass of the element, λ the decay constant, R the activation rate, and t_{ir}, t_d and t_m the irradiation, decay and measurement durations. P is the probability per disintegration of obtaining a count in the full-energy peak.

(This equation is only valid for the simplest activation case)

How to calculate R and P

- The activation rate R is calculated from the neutron spectrum shape parameters (Φ_s, Φ_e, α, T) and the capture cross-section parameters (σ, I_0, E_r, g(T)).
- The detection probability *P* is calculated from the detector's efficiency curves (full-energy and peak-to-total), the counting geometry and the decay scheme of the radionuclide.

Principles of the k_0 method

First conventional approach: adapted Høgdahl

$$R = \int_0^{v_{Cd}} \sigma(v)\Phi(v)\,\mathrm{d}v + \int_{v_{Cd}}^{\infty} \sigma(v)\Phi(v)\,\mathrm{d}v \approx$$

$$\int_0^{v_{Cd}} \frac{\sigma_0 v_0}{v}\Phi(v)\,\mathrm{d}v + \int_{E_{Cd}}^{\infty} \sigma(E)\Phi(E)\,\mathrm{d}E \approx$$

$$\int_0^{v_{Cd}} \sigma_0 v_0\,\mathrm{n}(v)\,\mathrm{d}v + \int_{E_{Cd}}^{\infty} \sigma(E)\frac{\Phi(E_{ref})}{\left(E/E_{ref}\right)^{1+\alpha}}\mathrm{d}E =$$

$$\sigma_0\Phi_s + I_0(\alpha)\Phi_e = \sigma_0\Phi_s\left(1 + Q_0(\alpha)/f\right)$$

Principles of the k_0 method

First conventional approach: adapted Høgdahl

$$I_0(\alpha) \equiv \int_{E_{Cd}}^{\infty} \sigma(E) \frac{E_{ref}^{\alpha}}{E^{1+\alpha}} dE$$

$$\Phi_e \equiv \Phi\left(E_{ref}\right) E_{ref}$$

$$I_0(\alpha) = E_{ref}^{\alpha} \left\{ \frac{I_0(0) - 0.429\sigma_0}{\bar{E}_r^{\alpha}} + \frac{0.429\sigma_0}{(2\alpha+1) E_{Cd}^{\alpha}} \right\}$$

$$f = \frac{\Phi_s}{\Phi_e}$$

Principles of the k_0 method

Second conventional approach: Westcott

Takes non 1/v (n,γ) reactions into account using g(T).

Principles of the k$_0$-IAEA software

Adapted^2 Høgdahl conventional approach: Blaauw

$$R=\int_0^\infty \frac{\sigma_0 v_0}{v}\Phi(v)\,\mathrm{d}v+\int_0^\infty\left\{\sigma(v)-\frac{\sigma_0 v_0}{v}\right\}\Phi(v)\,\mathrm{d}v$$

$$\approx\int_0^\infty \sigma_0 v_0\,\mathrm{n}(v)\,\mathrm{d}v+\int_0^\infty\left\{\sigma(E)-\frac{\sigma_0 v_0}{v}\right\}\frac{\Phi(E_{ref})}{\left(E/E_{ref}\right)^{1+\alpha}}\,\mathrm{d}E$$

$$=\sigma_0\Phi_t+I_0^*(\alpha)\Phi_e$$

$$=\sigma_0\Phi_t\left(1+Q_0^*(\alpha)/f^*\right)$$

Principles of the k_0-IAEA software

Adapted^2 Høgdahl conventional approach: Blaauw

$$\mathrm{I}_0^*(\alpha) \equiv \int_0^{\infty} \left\{ \sigma(E) - \frac{\sigma_0 v_0}{v} \right\} \frac{E_{ref}^{\alpha}}{E^{1+\alpha}} dE$$

$$\Phi_e \equiv \Phi\left(E_{ref} \right) E_{ref}$$

$$I_0^*(\alpha) = \frac{E_{ref}^{\alpha}}{\overline{E}_r^{\alpha}} \mathrm{I}_0^*(0)$$

$$f^* = \frac{\Phi_t}{\Phi_e}$$

Principles of the k_0-IAEA software

Conventional approach: Blaauw + Westcott +threshold

$$R = \sigma_0 g(T)\Phi_t + I_0^*(\alpha)\Phi_e + \sigma_{fast}\Phi_{fast}$$

$$= \sigma_0\Phi_t\left(g(T) + Q_0^*(\alpha)\Big/ f^*\right) + \sigma_{fast}\Phi_{fast}$$

$$g(T) = \frac{\int_0^\infty \sigma(v)\frac{2v^3}{v_T^4}e^{-\left(v/v_T\right)^2}dv}{\int_0^\infty \frac{\sigma_0 v_0}{v}\frac{2v^3}{v_T^4}e^{-\left(v/v_T\right)^2}dv}$$

The k_0 method and the k_0-IAEA software

Relations between standard k_0 and k_0-IAEA parameters

$$Q_0^* = Q_0 - 0.429$$

$$f^* = f + 0.429$$

$$\Phi_t = \Phi_s + \Phi_e \left(\frac{1}{0.5+\alpha}\right)\sqrt{\frac{E_0}{E_{ref}}}\left(\frac{E_{ref}}{E_{Cd}}\right)^{(0.5+\alpha)}$$

$$= \Phi_s + 0.429\Phi_e \quad \text{if } \alpha = 0$$

and

$$E_0 = 25\text{ meV},\ E_{Cd} = 0.55\text{ eV},\ \mathrm{E}_{\mathrm{ref}} = 1\text{ eV}$$

How to implement the k_0 method

- 1- Characterize the detector first
  - find or determine the detector dimensions
  - measure peak-to-total ratio curve
  - measure full-energy efficiency curve
  - measure escape ratio curves (k_0-IAEA)
- 2- Characterize the irradiation facility second
  - determine T, f and α (and the fast flux)

How to implement the k_0 method
- irradiation facility characterization

- determine T, f and α (and the fast flux)
  - use a suitable mix of elements, some with low Q_0, some with high Q_0, with various E_r's, some showing (n,p) or other threshold reactions, some being non- 1/v
  - for example: Zr, Au, Ni, Lu.

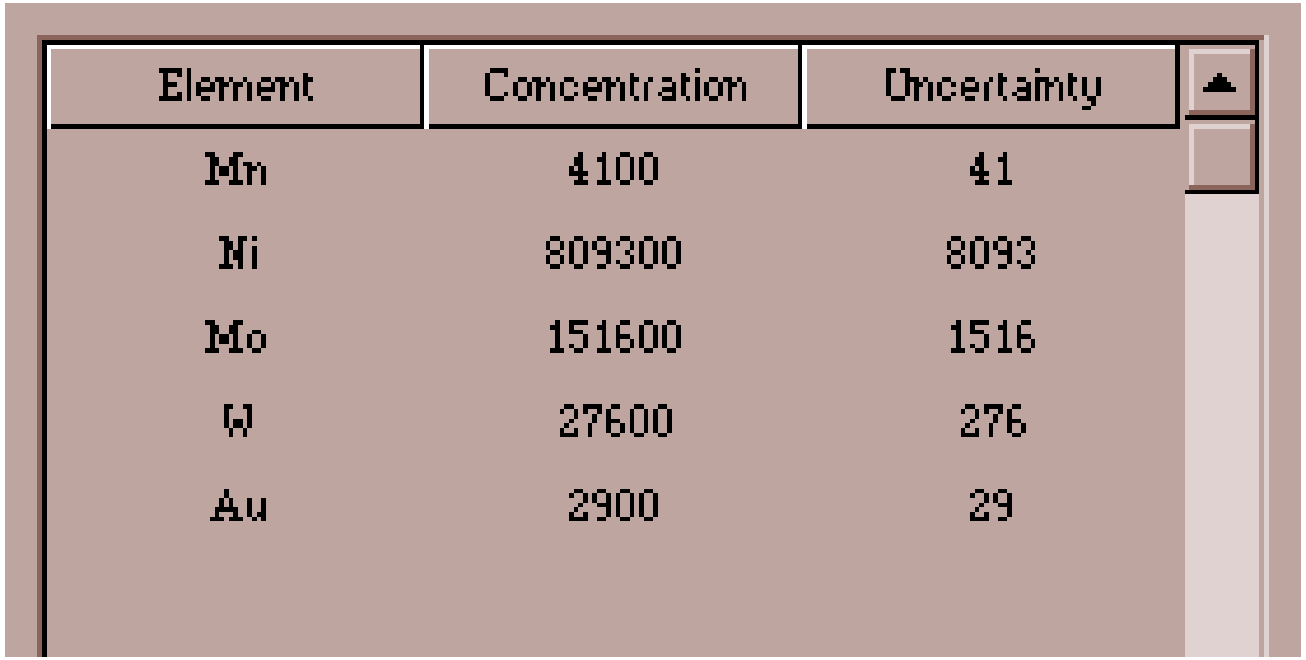
The trouble with the standard methods

- Cd-cover-method takes two samples of identical, complex composition - that's one too many.
- Bare triple-comparator method can only yield three parameters, we need five
- Adding Lu and e.g. Ni to the standard combination of Zr-Au seems a good option
- But foils and wires in one capsule are hard to count together (efficiency, decay, shielding)
- ^{97}Zr appear to be too extreme in its behavior, and not representative for the other high-Q nuclides

Possible alternatives

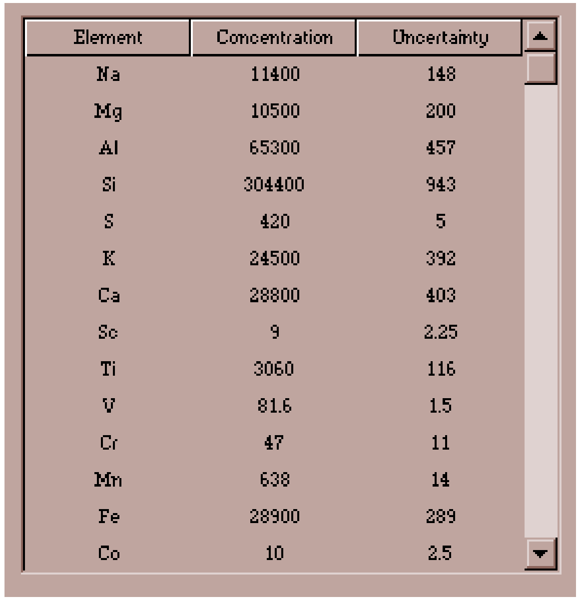
- Existing materials
  - available alloys that happen to be suitable
  - reference materials
  - SMELS
- Homemade mixtures
  - ground and mixed powders
  - solutions of suitable composition, pipetted on filter paper

An existing alloy

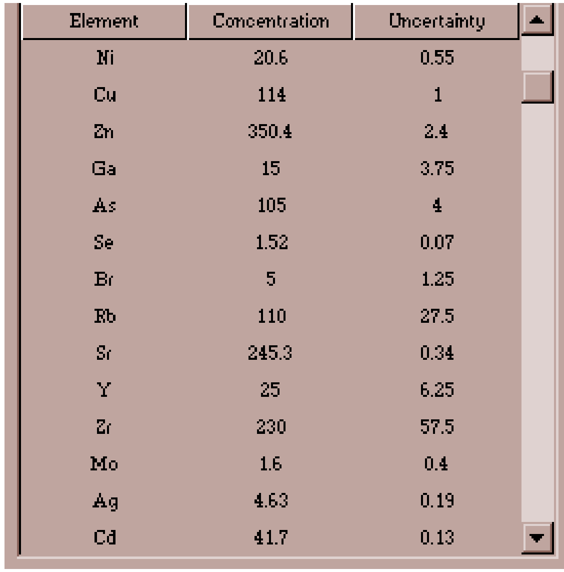


Element	Concentration	Uncertainty
Mn	4100	41
Ni	809300	8093
Mo	151600	1516
W	27600	276
Au	2900	29

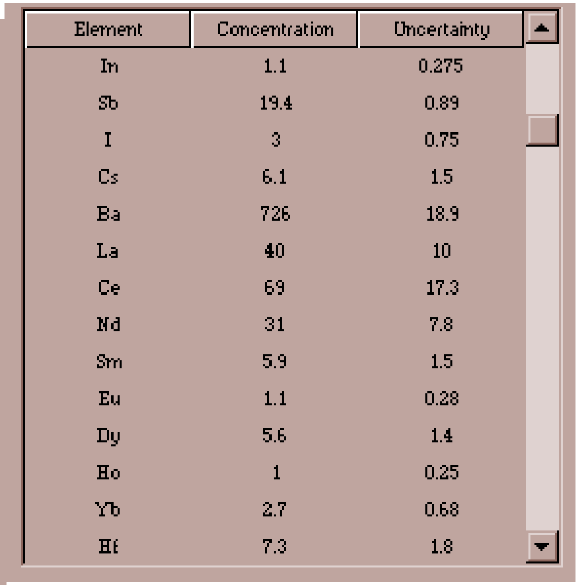
NIST Montana soil



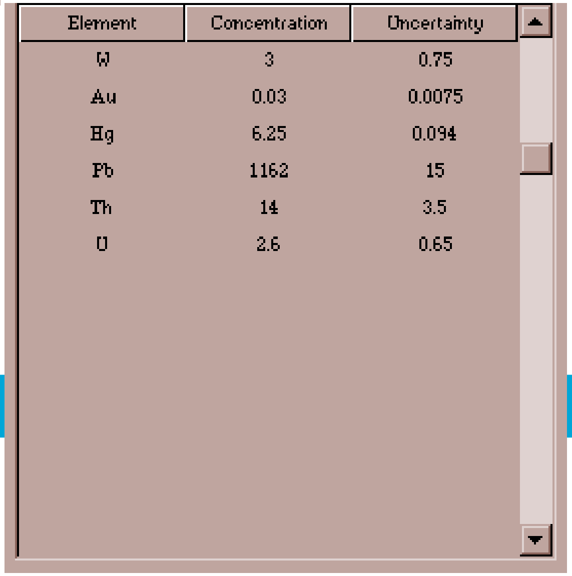
Element	Concentration	Uncertainty
Na	11400	148
Mg	10500	200
Al	65300	457
Si	304400	943
S	420	5
K	24500	392
Ca	28800	403
Sc	9	2.25
Ti	3060	116
V	81.6	1.5
Cr	47	11
Mn	638	14
Fe	28900	289
Co	10	2.5



Element	Concentration	Uncertainty
Ni	20.6	0.55
Cu	114	1
Zn	350.4	2.4
Ga	15	3.75
As	105	4
Se	1.52	0.07
Br	5	1.25
Rb	110	27.5
Sr	245.3	0.34
Y	25	6.25
Zr	230	57.5
Mo	1.6	0.4
Ag	4.63	0.19
Cd	41.7	0.13



Element	Concentration	Uncertainty
In	1.1	0.275
Sb	19.4	0.89
I	3	0.75
Cs	6.1	1.5
Ba	726	18.9
La	40	10
Ce	69	17.3
Nd	31	7.8
Sm	5.9	1.5
Eu	1.1	0.28
Dy	5.6	1.4
Ho	1	0.25
Yb	2.7	0.68
Hf	7.3	1.8



Element	Concentration	Uncertainty
W	3	0.75
Au	0.03	0.0075
Hg	6.25	0.094
Pb	1162	15
Th	14	3.5
U	2.6	0.65

Home-made ground and mixed powders

- Too risky because of segregation
- To be tried only by experienced, trained, professional reference-material makers
- Don’t even think of trying this at home!

Home-made solutions pipetted on filter paper

Delft - Sao Paolo example

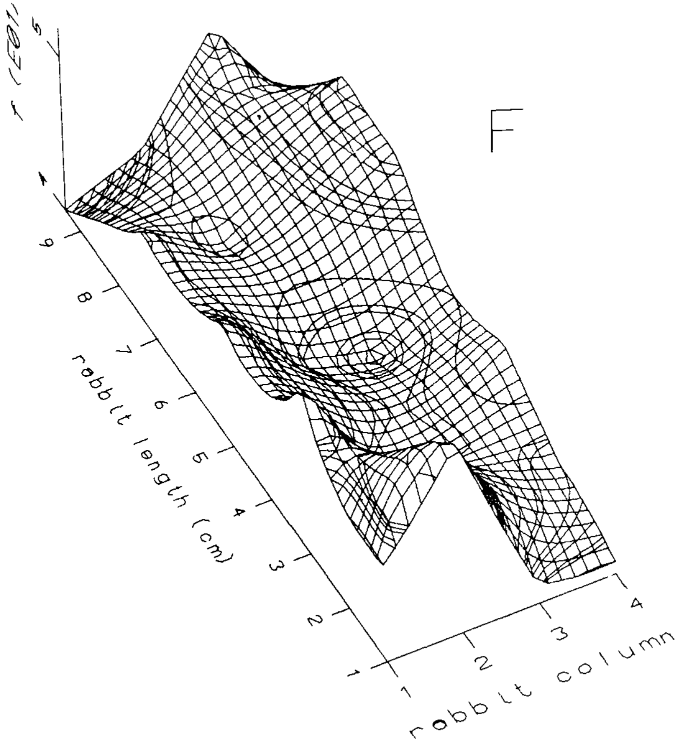
Alternative methods for Thermal and Epithermal Flux Monitoring

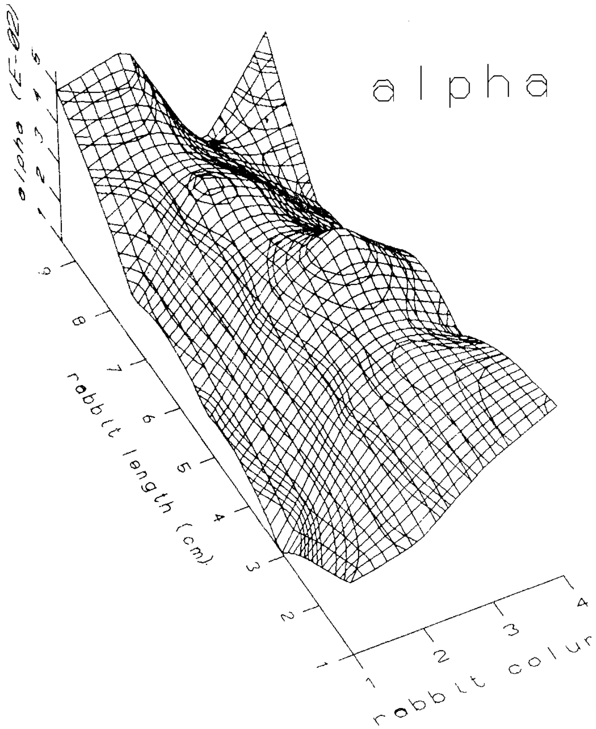
Anneke Koster-Ammerlaan, Marcio Bacchi,
Peter Bode, Elisabete A.De Nadai Fernandes

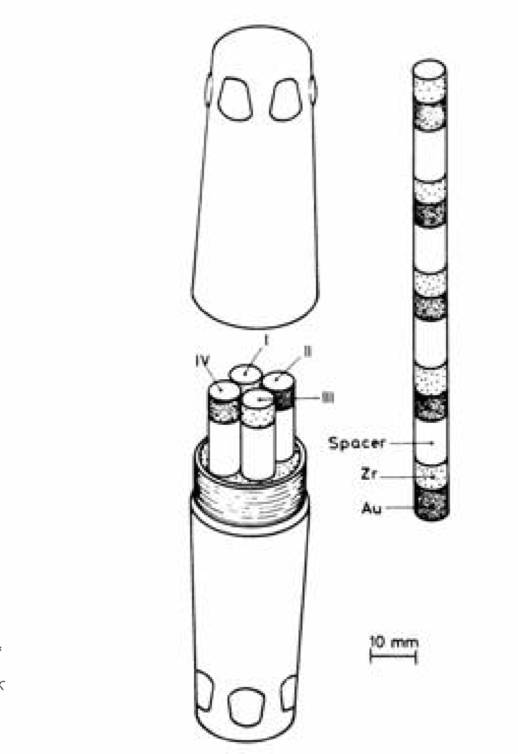
Trieste, Nov 8-10, 2010

Delft University of Technology

Thermal and Epithermal Flux Monitoring







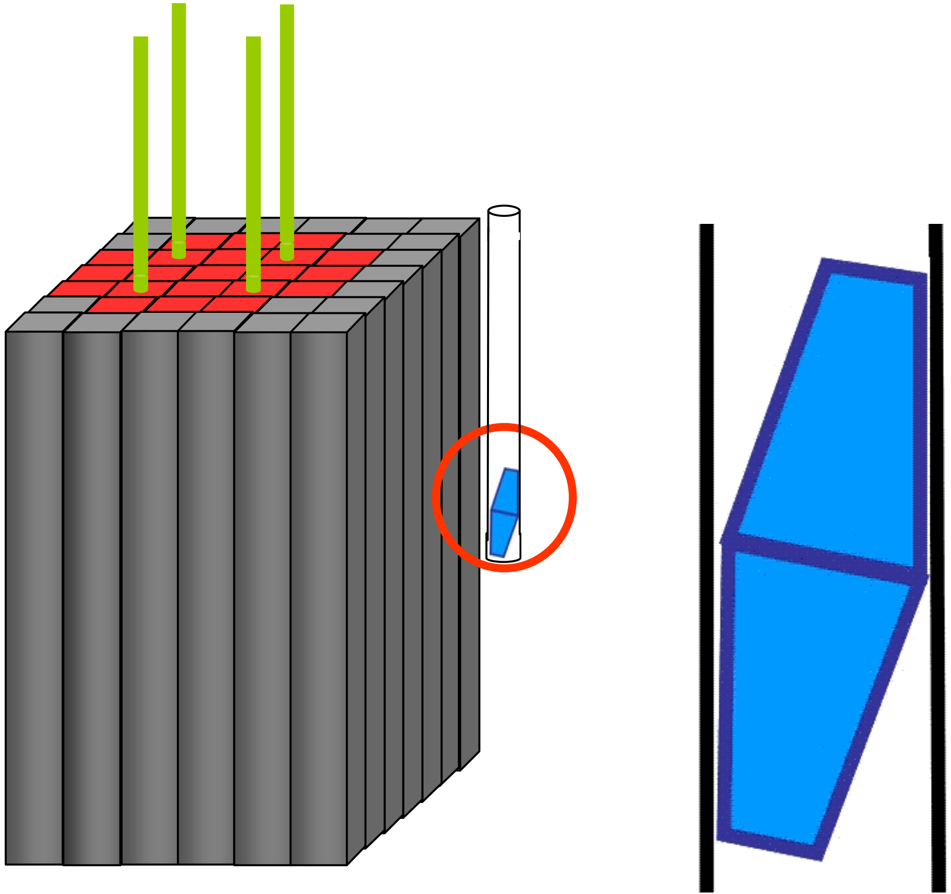
Thermal and Epithermal Flux Monitoring

Poor reproducibility of f and α in consecutive irradiations under stable reactor conditions:

	1st series (n=5)	2nd series (n=5)
f	50 - 63	64 - 90
α	0,07 - 0,11	0,02 - 0,08

Temporarily solved at IRI by choosing $\alpha = 0.10$ and calculating the corresponding f values

Thermal and Epithermal Flux Monitoring



Thermal and Epithermal Flux Monitoring

Conclusions:

- Metrology requires to determine spectrum parameters in every irradiation for every position inside the rabbit.

Thermal and Epithermal Flux Monitoring

Cd-covered Zr-Au method:

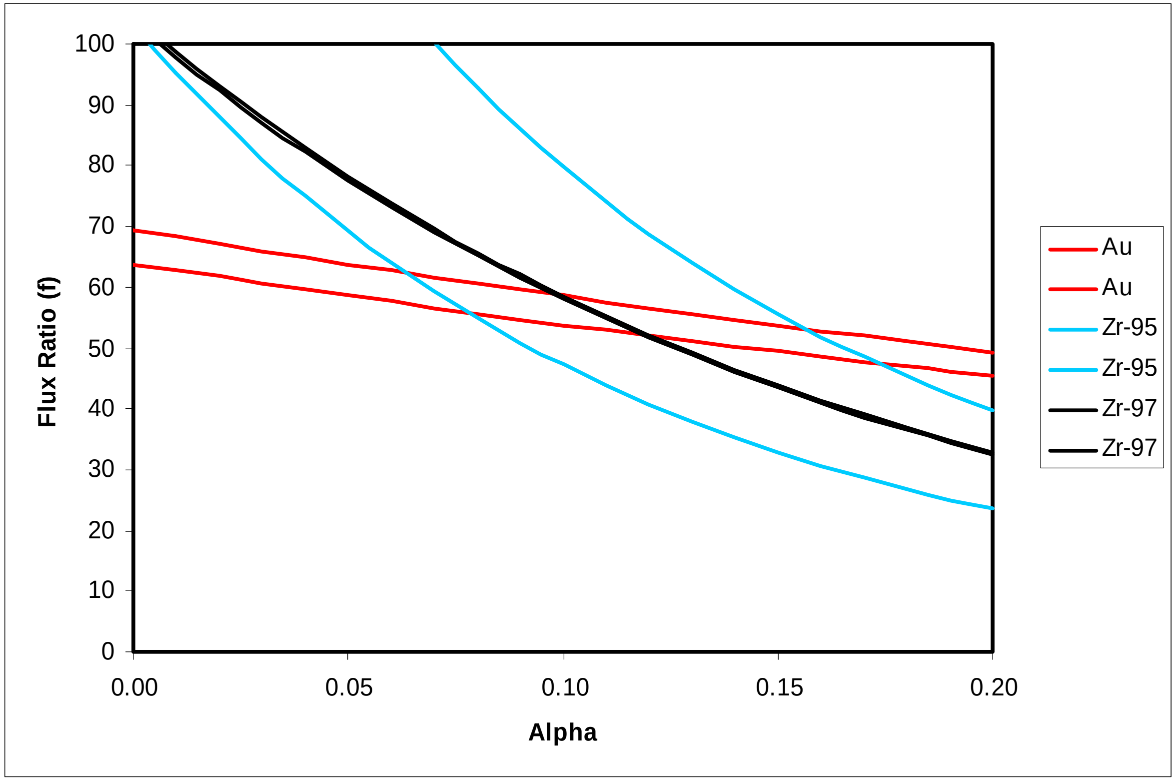
- Inapplicable due to thermal heating of Cd with a serious risk of damage to the plastic irradiation container.
- Inapplicable due to flux depressions in the real samples

Thermal and Epithermal Flux Monitoring

Bare triple method (Zr-Au):

- Poor counting statistics for ^{97}Zr under routine INAA conditions (t_{irr}: 1-4 h, t_d: 3-5 days, t_c: 1-4 h)
- Strong influence of counting statistics

Thermal and Epithermal Flux Monitoring



Thermal and Epithermal Flux Monitoring

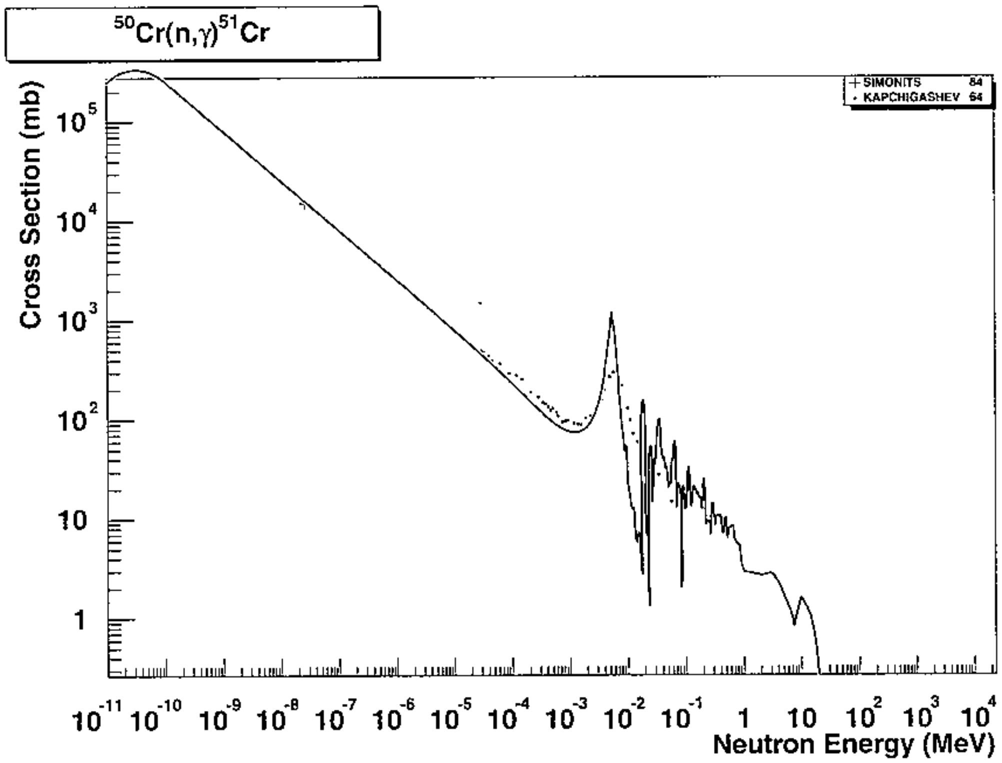
Search for alternative pairs of flux and spectrum monitors

Needed:

- High Q_0, low E_r + High Q_0, high E_r
- High σ_0
- No spectral interferences; minimal coincidence summing
- Easy to prepare in large batches
- $t_{1/2} > 1$ d

Thermal and Epithermal Flux Monitoring

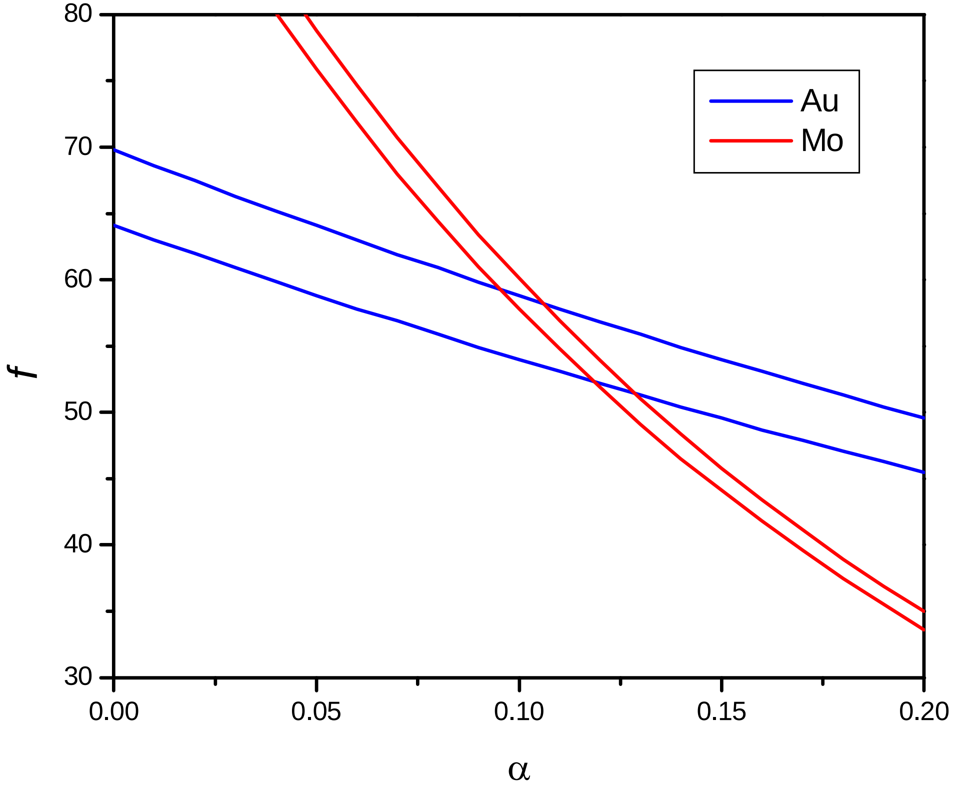
Epithermal flux
can be neglected
if f>50



Thermal and Epithermal Flux Monitoring

Alternative set of monitors:

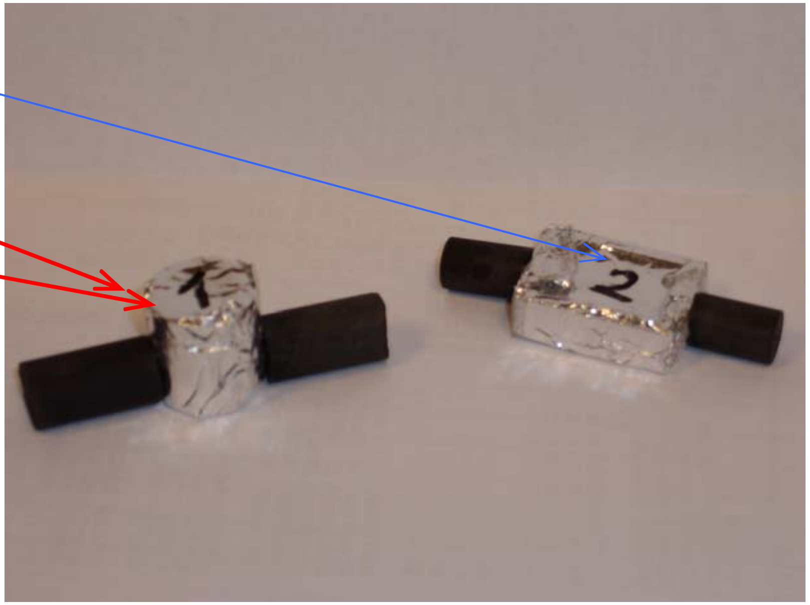
^{51}Cr, ^{99}Mo, ^{198}Au



Thermal and Epithermal Flux Monitoring

Verification in pool-side facility and Al-containers

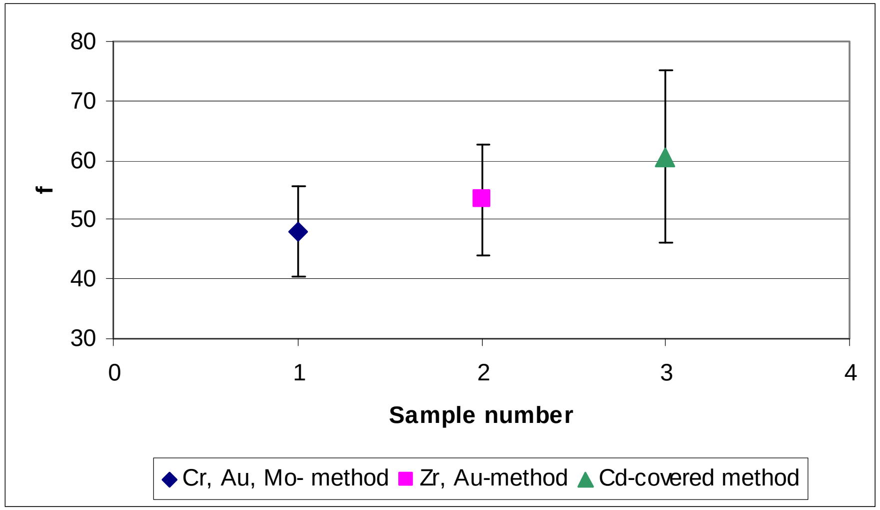
- Cd-covered Zr-Au-monitor
- Cr, Mo, Au monitor
- Zr, Au-monitor



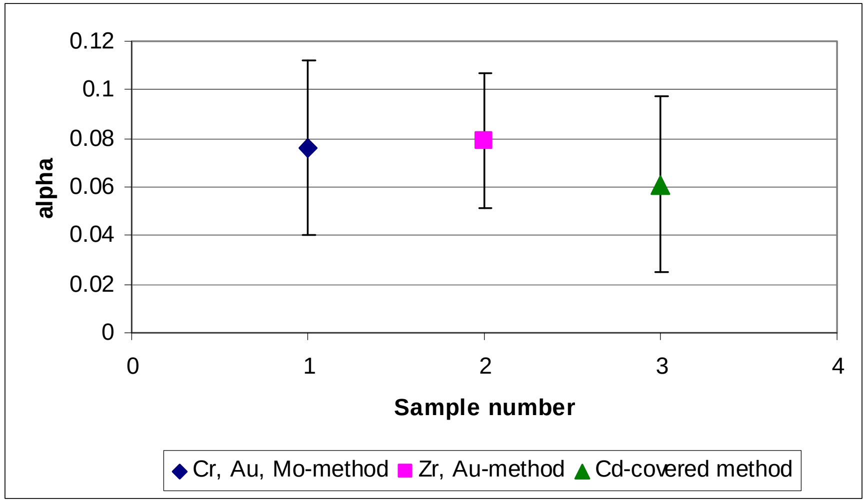
Irradiation time: 30 minutes
(2 separate irradiations)
Decay time: 4 days
Counting time: 2 hours
Detector: 35 % Ge-detector

Thermal and Epithermal Flux Monitoring

f

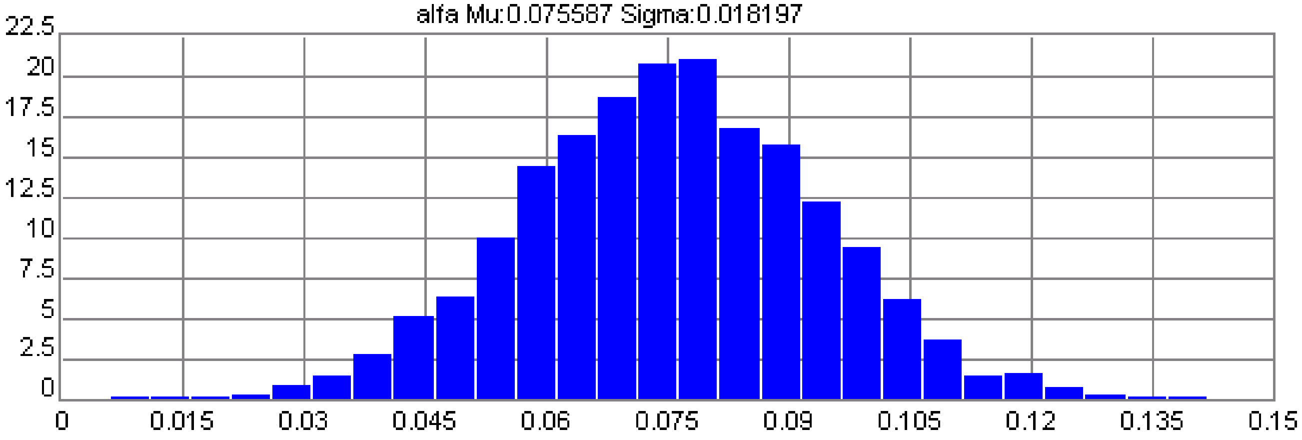


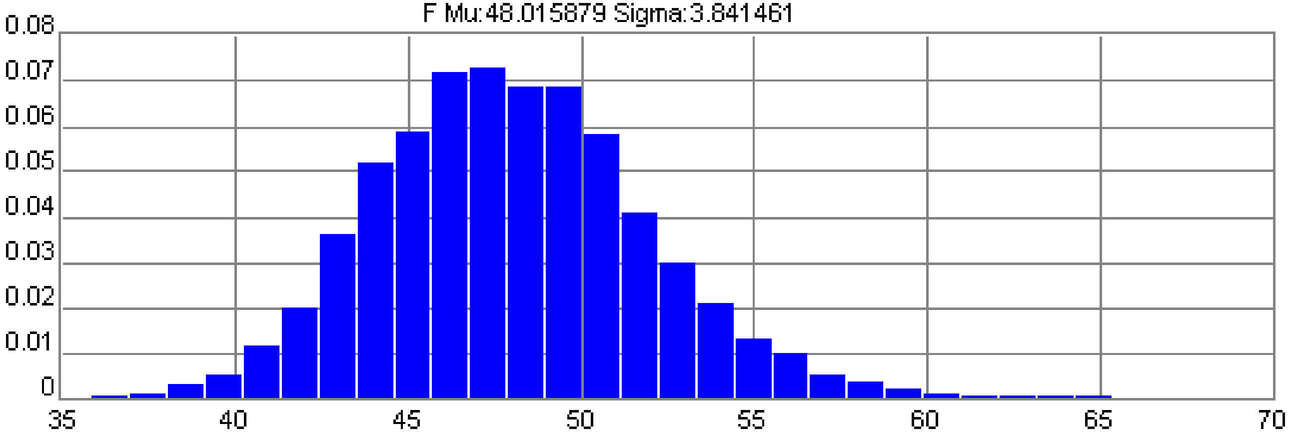
α



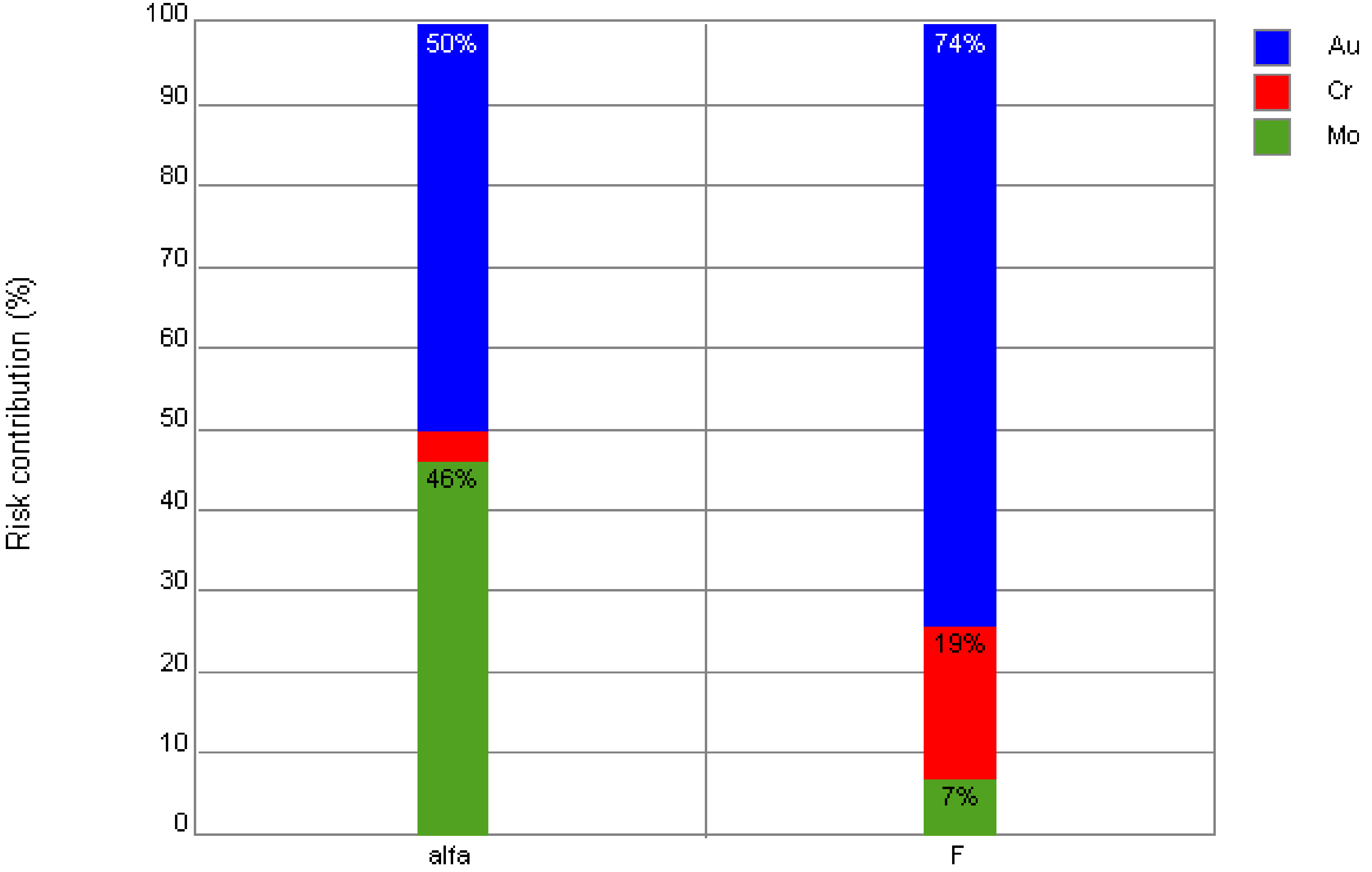
Thermal and Epithermal Flux Monitoring

	Activity uncertainty (%)
^{51}Cr	0.7
^{99}Mo	2.0
^{198}Au	1.1





Thermal and Epithermal Flux Monitoring



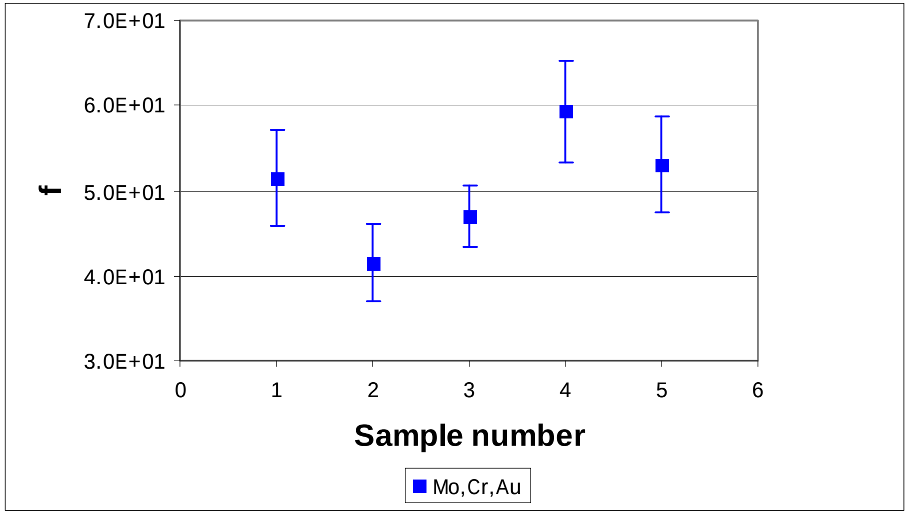
Thermal and Epithermal Flux Monitoring

Characterization of pneumatic facility

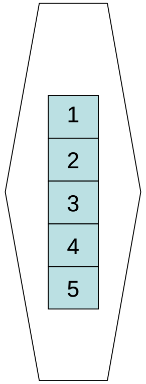
5 Cr, Mo, Au-monitors

Irradiation time: 1 hour
Decay time: 4 days
Counting time: 1 hours
Detector: 35 % Ge-detector

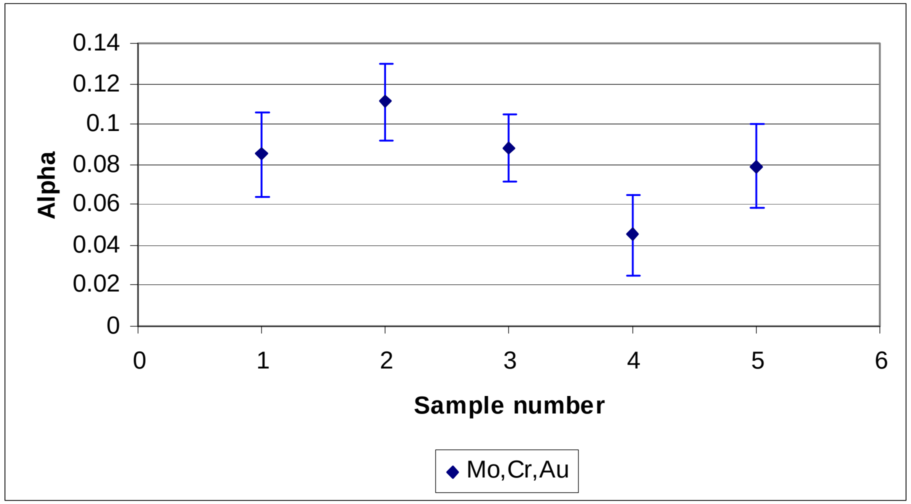
Thermal and Epithermal Flux Monitoring



f



α



Thermal and Epithermal Flux Monitoring

Conclusions:

- The Mo, Au, Cr- monitor is the solution for spatial f and α monitoring
- The monitor is easy to prepare in large batches
- The monitor will be used in routine INAA