Budget Uncertainty and Minimum Detectable Concentrations for the INAA Laboratory of the ETRR-2

M.Y. Khalil and Kh. A. Elsakhawy

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Accreditation

- ***Management Requirements:***
  - *organization chart of the laboratory,*
  - *quality assurance system,*
  - *document control,*
  - *review of requests,*
  - *tenders and contracts,*
  - *purchasing services and supplies,*
  - *service to the client,*
  - *corrective action,*
  - *preventive action,*
  - *control of records.*

(Accreditation)

- ***Technical Requirements:***
  - *personnel training,*
  - *environmental conditions,*
  - *method validation,*
  - *equipment,*
  - *estimation of uncertainty of measurements,*
  - *measurement traceability,*
  - *sampling,*
  - *assuring the quality of test and calibration results,*
  - *reporting the results*

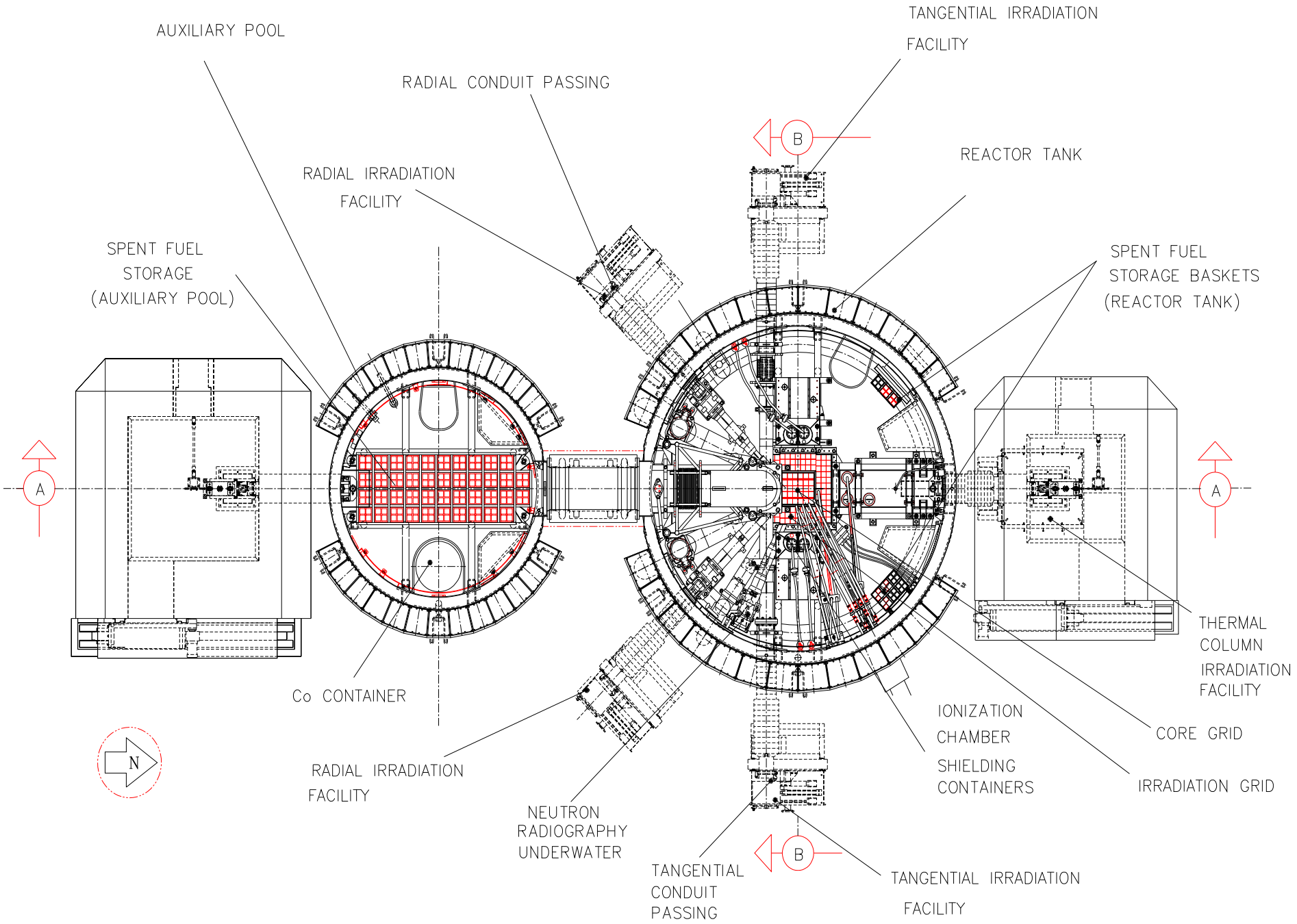
What we wanted to do

- **To fulfill some of the technical requirements for accreditation of ETRR-2 Instrumental Neutron Activation Analysis (INAA) laboratory**
  - ***Estimate the budget uncertainty***
  - ***Traceability measurements (Minimum Detectable Concentration)***

ETRR-2

Main Neutronic Parameters of the ETRR-2 Core

Fuel loading (MTR-plate fuel type)	29 fuel assembly each contain 19 plates
Average core cycle length	19 full power days
Core maximum thermal flux	2.3×10^{14} n/cm^2.sec
Average power density	140 W/cm^3
Maximum power peaking factor	2.43


AUXILIARY POOL
TANGENTIAL IRRADIATION FACILITY
RADIAL CONDUIT PASSING
B
REACTOR TANK
RADIAL IRRADIATION FACILITY
SPENT FUEL STORAGE (AUXILIARY POOL)
SPENT FUEL STORAGE BASKETS (REACTOR TANK)
A
A
THERMAL COLUMN IRRADIATION FACILITY
Co CONTAINER
CORE GRID
IONIZATION CHAMBER
SHIELDING CONTAINERS
IRRADIATION GRID
N
RADIAL IRRADIATION FACILITY
NEUTRON RADIOGRAPHY UNDERWATER
TANGENTIAL CONDUIT PASSING
B
TANGENTIAL IRRADIATION FACILITY

ETRR-2 Utilization

- Neutron Radiography
- Activation Analysis
- Isotopes Production
- Cold Neutrons Production
- BNCT
- Silicon Doping

What have been done

Apparatus

- **High pure germanium detector (HPGD):**
  - **Relative efficiency of the detector is 100%**
  - **Energy resolution(FWHM) at Co-60 line 1.33 MeV is 2.1**
  - **Cooling with liquid nitrogen at temperature (-186C).**

- **ETRR-2 as a neutron sources:**

  1- rabbit system (short irradiation).

  2- irradiation grid position (long irradiation).

Techniques

- Relative method
  - Sample and element standards are irradiated simultaneously
- Absolute method
  - In this case the thermal and epithermal neutron flux must be carefully measured, and their detector efficiency must be very well known
- K0 method
  - The rate of production of radioactive atoms from stable atoms of interest is related to the activation rate of gold through the so-called k0-constant

Materials

- **A- Reference materials:**
  - *rock-type P1,*
  - *CRM,*

Both from the proficiency test study under the AFRA IV-7 project

- **B- Flux monitors:**

-Gold (pure and diluted with aluminum (gold 0.1% and the aluminum 99.9%))
-Zirconium (99.99%).
-Nickel (99.99).
-Cadmium (cover sheet and tube 99.99%).

Blanks

Were used to:

- 1. Eliminates the Effect of instrumental background,
- 2. Determines the spectrum baseline, which is the sum of the instrumental background and any signal due to interfering species,
- 3. Eliminates the Effect of contamination or the impurities in the vials

Method

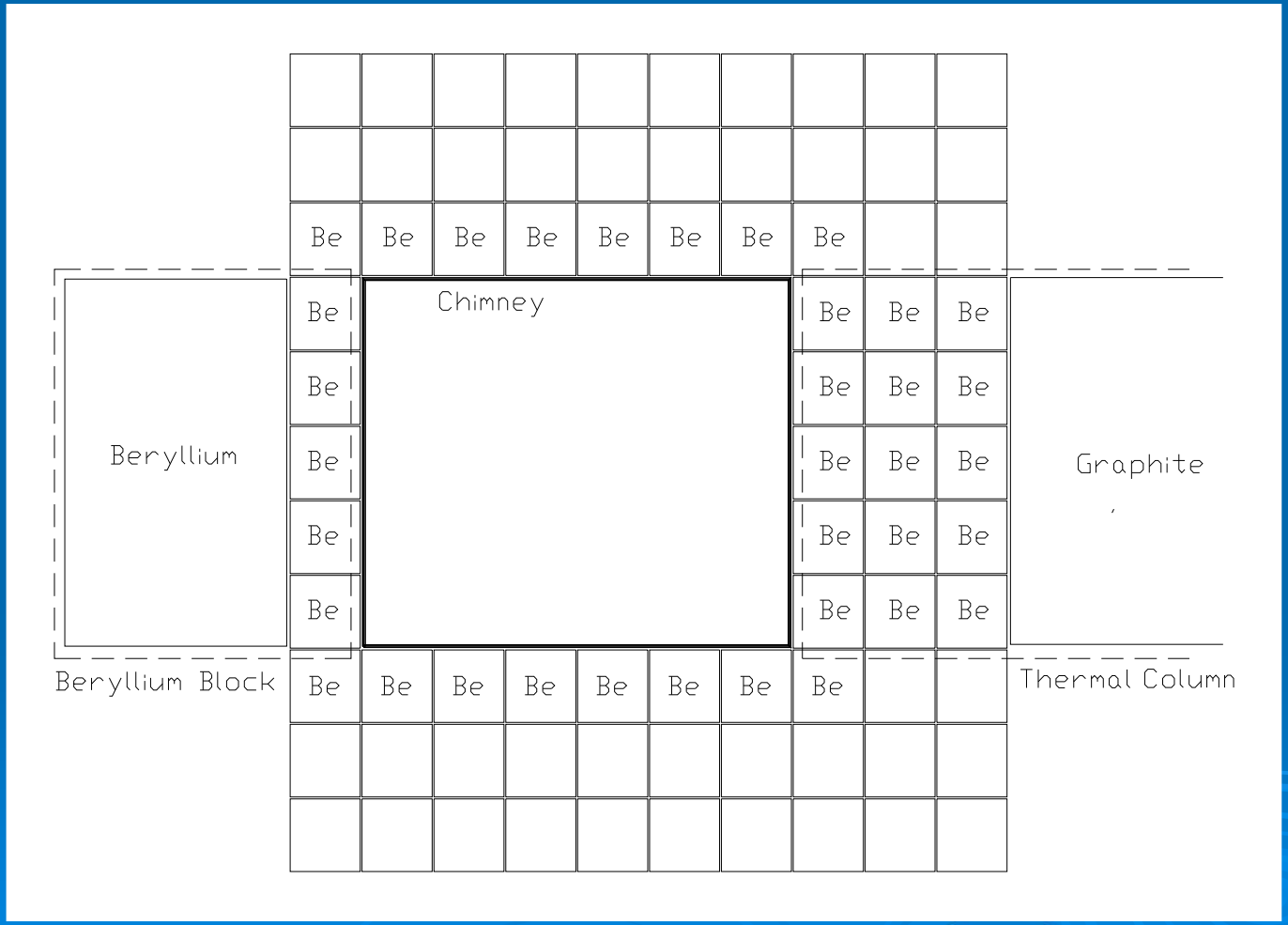
Short irradiation(Rabbit system)

- **Sample Irradiated for 60 second with aid of pneumatic facility.**
- **Blank polyethylene vial used to control the effect of the background.**

Long irradiation

- **All samples divided into 4 aluminum cans**
  - **The cans placed in a sample holder and lowered into the irradiation position.**
  - **The samples were irradiated for 4 hour and 35 minutes.**
  - **The sample were kept in hot cell to reach safe handling level.**

Irradiation grid



What have we got

Samples irradiated in thermal column

	Irradiation time, sec	Cooling time, hr	Counting time, sec	Dead time %
SAMPLE #1	40	3.269	1800	0.41
Gold #1	40	20.356	1800	1.9
SAMPLE #2	60	3.688	1800	0.7
Gold #2	60	20.77	1800	8.6
SAMPLE #3	60	4.213	1800	0.76
Gold #3	60	21	1800	3.49
SAMPLE #7R	60	3.99	1800	0.22
Gold #7	60	9.21	1800	3.66
SAMPLE #8R	60	4.578	1800	1.18
Gold #8	60	9.65	1800	2.
Sample#9R	60	5.266	1800	1.12
Gold #9	60	10.155	1800	6.31

Net peak area

	Element	Isotope	Peak energy	Branching ratio	Net peak area
Sample #1	Manganese	Mn-56	846.76	99.87	41028±220
	Sodium	Na-24	1368.6	100	52755±242
	Potassium	K-42	1524.58	18.8	680±60
Gold#1	pure GOLD	Au-198	411.4	95.5	1277619±1150
Sample #7 R	Manganese	Mn-56	846.76	99.87	89537±332
	Sodium	Na-24	1368.6	100	176296±437
	Potassium	K-42	1524.58	18.8	990±100

	Element	Isotope	Peak energy	Branching ratio	Net peak area
Sample #2	Manganese	Mn-56	846.76	99.87	66589±280
	Sodium	Na-24	1368.6	100	93332±320
	Potassium	K-42	1524.58	18.8	1245±76
Gold #2	pure GOLD	Au-198	411.4	95.5	5517610±2404
Sample #8 R	Manganese	Mn-56	846.76	99.87	77941±312
	Sodium	Na-24	1368.6	100	176098±432
	Potassium	K-42	1524.58	18.8	966±98
Sample #3	Manganese	Mn-56	846.76	99.87	70166±287
	Sodium	Na-24	1368.6	100	104071±337
	Potassium	K-42	1524.58	18.8	1152±82
Gold#2	pure GOLD	Au-198	411.4	95.5	2351542±1561
Sample #9R	Manganese	Mn-56	846.76	99.87	94499±289
	Sodium	Na-24	1368.6	100	171407±432
	Potassium	K-42	1524.58	18.8	1071±89
Gold Cd	GOLD +Cd	Au-198	411.4	95.5	84336±296

Samples irradiated in irradiation grid

vail	Tirr	Ist counting			2nd counting			3rd counting		
		Td Day	Tc Sec	Tdead %	Td Day	Tc Sec	Tdead %	Td day	Tc Sec	Tdead %
5	4.35	10.5	1800	5.65	21.6	2400	3.44	33.6	2700	2.9
11	4.35	9.53	1800	8.98	21.6	2400	2.34	33.6	2700	1.87
6	4.35	6.53	1800	8.85	13.5	1800	6.48	21.6	2800	5.52
12	4.35	6.54	1800	8.2	13.5	1800	6.34	21.6	2800	3.97
24	4.35	13.6	1800	7.94	-	-	-	-	-	-
15	4.35	13.6	1800	5.27	-	-	-	-	-	-
18	4.35	7.6	1800	1.46	-	-	-	-	-	-
25	4.35	13.5	1800	12.55	-	-	-	-	-	-
16	4.35	14.6	1800	5.93	-	-	-	-	-	-
19	4.35	7.58	1800	1.77	-	-	-	-	-	-

Net peak area

	Element	Isotope	Peak energy KeV	Branching ratio %	Net peak area
Sample #5	Calcium	Ca	159.38	67.9	10457±451
	Cobalt	Co	1332.5	99.9	41281±222
	Chromium	Cr	320.08	10.08	18971±373
	cesium	Cs	604.7	97.56	10233±369
	Iron	Fe	1099.25	56.5	222777±522
	rubidium	Rb	1076.6	8.78	4923±247
Sample #11 R	Calcium	Ca	159.38	67.9	19855±621
	Cobalt	Co	1332.5	99.9	75766±309
	Chromium	Cr	320.08	10.08	35404±507
	cesium	Cs	604.7	97.56	20486±501
	Iron	Fe	1099.25	56.5	411426±716
	rubidium	Rb	1076.6	8.78	9646±339
24	Au+Al	Au-198	411.4	95.5	2592853±1646
15	Nickel	Co-58	810	99.45	1013572±1025
18	Zirconium	Zr-95	756.73	54.5	335170±590
		Zr-97	743.33	97.9	12118±152

The relative standardization

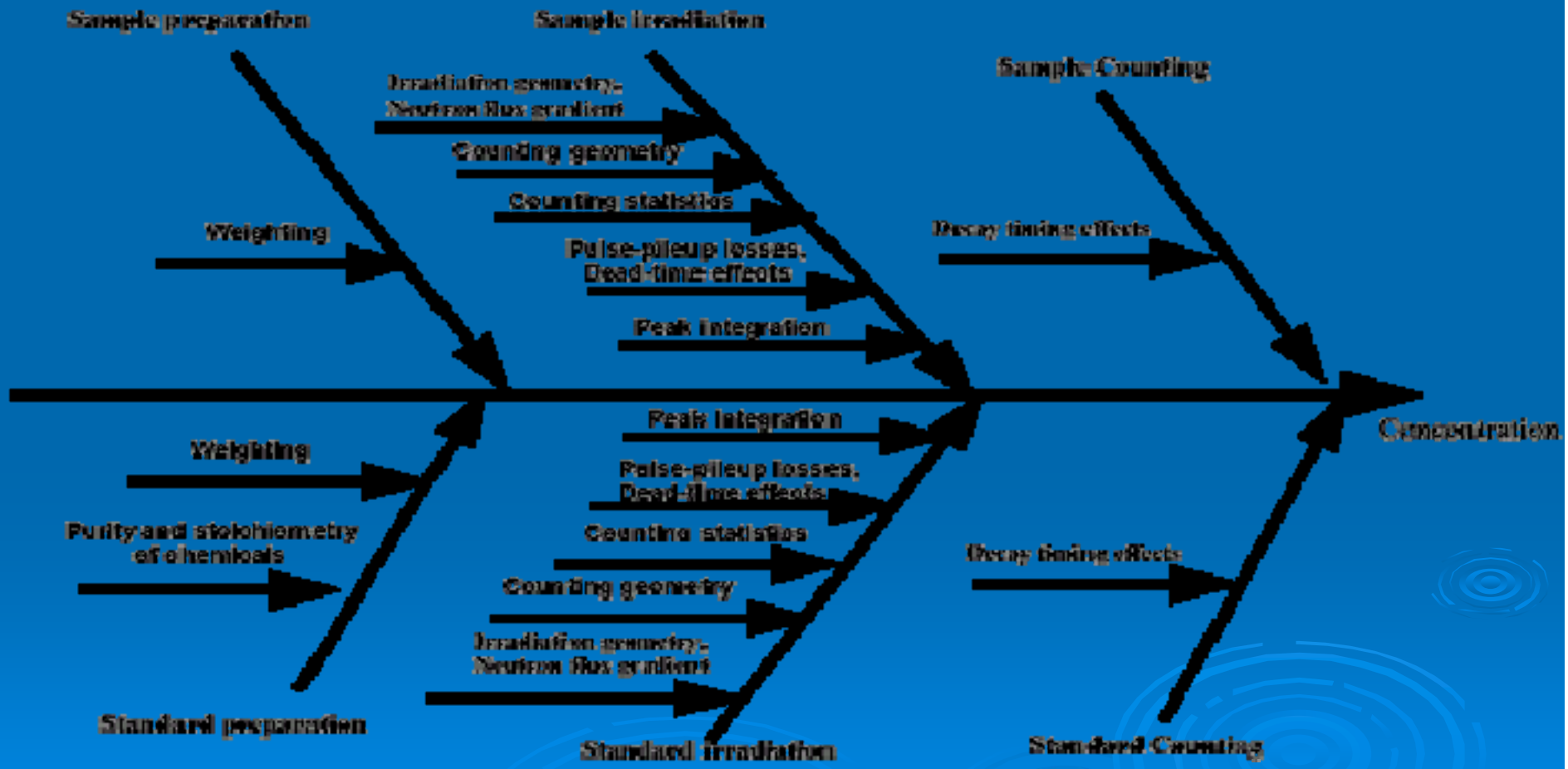
- The test and reference samples were irradiated simultaneously or sequentially in short half life under the same conditions.
- The concentration of the elements of interest calculated by caparison of the activity between the sample and reference standard .

$$C_{unk} = C_{std} \frac{N_{p(unk)}}{N_{p(std)}} \times \frac{m_{std}\, e^{-\lambda t_{d-std}} (1 - e^{-\lambda t_{m-std}})}{m_{unk}\, e^{-\lambda t_{d-unk}} (1 - e^{-\lambda t_{m-unk}})}$$

Average concentrations:

Element	Concentration
Mn-54	612.56 mg/kg
Na-24	27.472 g/kg
K-42	13.485 g/kg
Sc-47	42.467 g/kg
Co-60	15.69 mg/kg
Cr-51	32.43 mg/kg
Fe-59	35.011 g/kg
Rb-86	39.017 mg/kg
Cs-134	1.85m g/kg

Uncertainty in relative standardization



Uncertainty sources

	$U_{mass\ S}$	$U_{mass\ Std}$	$U_{net\ area\ S}$	$U_{net\ area\ Std}$	U_{flx}	$U_{geometry\ difference}$	$U_{pulse\ pile\ up}$
Mn-54	0.15	0.15	0.243	0.232	0.8	3.95	-
Na-24	0.15	0.15	0.198	0.143	0.8	2.33	-
K-42	0.15	0.15	3.11	6.791	0.8	2.1	-
Sc-47	0.15	0.15	1.806	1.615	NON	7.3	3.2
Co-60	0.15	0.15	0.236	0.82		0.75	3.5
Cr-51	0.15	0.15	0.827	2.137		6.2	9
Fe-59	0.15	0.15	0.101	0.121		0.77	2.76
Rb-86	0.15	0.15	2.01	1.878		0.4	4.2
Cs-134	0.15	0.15	1.412	1.623		0.5	2.5

Expanded uncertainty

Element	Expanded uncertainty	Concentration
Mn-54	8.1	612.568 ±49.61
Na-24	4.96	27.472±1.36
K-42	15.6	13.485±2.1
Sc-47	16.66	42.467±7.10
Co-60	7.36	15.693±1.15
Cr-51	22.32	32.438±7.24
Fe-59	5.6	35.011±1.96
Rb-86	10.1	39.017±3.94
Cs-134	6.68	1.85±0.123

Minimum detectable concentration

Element	MDC
Sc-47	0.2017
Co-60	0.000838
Cr-51	0.0011433
Cs-134	0.00036
Fe-59	0.264
Mn-54	0.00226
Na-24	0.264
Rb-86	0.229
K-42	0.4734

The absolute standardization

- Sample and flux monitors are irradiated simultaneously.
- The neutron flux measured from the activity of the flux monitors.
- The concentration computed from the activation equation:

$$C = \frac{N_p \lambda M}{m\theta N_v \beta_i \varepsilon_i (\sigma \phi_t + I^* \phi_e)(1 - e^{-\lambda T_{irr}}) e^{-\lambda T_d} (1 - e^{-\lambda T_m})}$$

Neutron flux at thermal column

Flux	Sample #1	Sample #2	Sample #3
Thermal	2.90E+11	2.89 E+11	2.82 E+11
Epithermal	2.6 E+8		

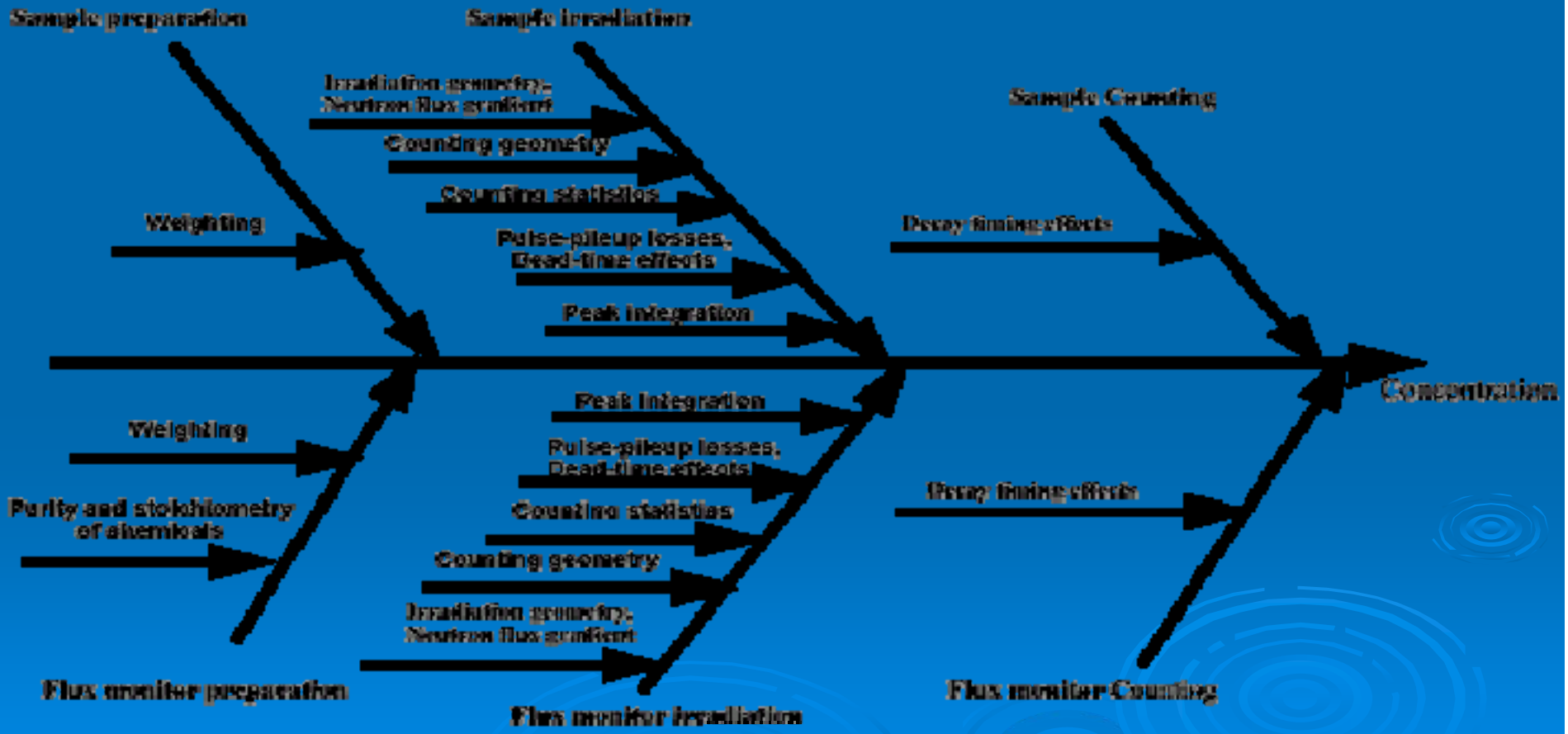
Neutron flux in irradiation grid

Flux	Sample #24	Sample #25
Thermal	9.29 E+13	9.23E13
Epithermal	2.41E+12	2.49E+12
Fast	3.62E+12	3.81E+12

Average concentrations

Element	Concentration
Mn-54	541 mg/kg
Na-24	26.5180 g/kg
K-42	18.780 g/kg
Co-60	13.149 mg/kg
Cr-51	37.426 m g/kg
Fe-59	38.2020 g/kg
Cs-134	2.462 mg/kg
Rb-86	39.949 mg/kg

Uncertainty in absolute standardization



Uncertainty sources

	Uflx M	U S M	Uflx Conc.	Uflx Net area	U S Net area 0.2427	U geometry diff.	U pulse pile up	U effici - ency
Mn-54	0.15	0.15	0.01	0.0251	0.1979	2.1	-	2.4
Na-24	0.15	0.15	0.01	0.0251	3.1	2.475	-	2.4
K-42	0.15	0.15	0.01	0.0251	0.3105	10.1	-	2.4
Co-60	0.15	0.15	0.01	0.037	0.3105	0.32	5.7	2.4
Cr-51	0.15	0.15	0.01	0.037	1.1352	7.83	10.75	2.4
Cs-134	0.15	0.15	0.01	0.037	2.0819	3.17	0.98	2.4
Fe-59	0.15	0.15	0.01	0.037	0.1353	0.32	11.05	2.4
Rb-86	0.15	0.15	0.01	0.037	5.03	5.03	1.12	2.4

Expanded Uncertainty

Element	Expanded uncertainty	Concentration
Mn-54	6.4	541.247 ± 34.6 mg/kg
Na-24	6.92	26.5180 ± 1.830 g/kg
K-42	21.66	18.780 ± 4.060 g/kg
Co-60	12.4	13.149 ± 1.630 mg/kg
Cr-51	27.12	37.426 ± 10.15 m g/kg
Fe-59	22.64	38.2020 ± 8.650 g/kg
Cs-134	9	2.462 ± 0.220 mg/kg
Rb-86	12.76	39.949 ± 5.100 mg/kg

Minimum detectable concentration

Element	MDC
Co-60	0.000118
Cr-51	0.001486
Cs-134	0.000667
Fe-59	0.116
Mn-54	0.0015
Na-24	0.593
Rb-86	2.24
K-42	8.641

The k_0-IAEA standardization software .

- Single comparator standardization.
- The rate of production of radioactive atoms from stable atoms of interest is related to the activation rate of gold through the so-called k0-constant.

$$k_0 = \frac{M_C \theta_a \sigma_a \beta_a}{M_a \theta_c \sigma_c \beta_c}$$

- relative production rate depends on the specific irradiation facilities and gamma-ray detectors used
- Detector calibration.
- Facility characterization.
- Sample analysis

Average concentration

Element	Concentration	MDC, ppm
Ca	32.27±3.22 g/kg	0.48
Co	11.65±1.4 mg/kg	0.000762
Cr	31.81±4.14 mg/kg	0.0196
Cs	1.755±0.31 mg/kg	0.000791
Fe	34.91±1.43 g/kg	0.782
Mn	600.1±20.40 mg/kg	0.0041
Na	30.24±1.54 g/kg	0.0109
Rb	40.81±1.38 mg/kg	0.6126
K	13.28±5.4 g/kg	0.315

Conclusions

- The uncertainty budget in the relative standardization ranges from 2% to 11%.
- The uncertainty budget in k0-method ranges from 3% to 15%.
- uncertainty budget in the absolute method ranges from 6% to 21%.
- The MDC in relative standardization is the lowest and then the k0-IAEA standardization and the absolute standardization is the largest MDC

Thank you