UNCERTAINTIES CALCULUS IN NEUTRON ACTIVATION ANALYSIS – IMPROVING THE METHOD PRECISION

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1. INTRODUCTION

- The KRAGTEN method (spreadsheet method) is recommended for complex expressions to simplify calculations.
- This procedure uses an approximate numerical method of differentiation and requires only knowledge of the calculations used to obtain the final result (including any necessary correction factors), the numerical values of the parameters, and their uncertainties.
- It assumes either that the measurement model is linear in the input variables or that the uncertainty of the corresponding input quantity is small compared to its value.
- These assumptions are not always fully observed. However, the method provides acceptable accuracy for practical purposes when it is considered with the necessary approximations made in estimating the uncertainties.
- The advantage of the KRAGTEN method is that the correlation between the variables can be easily included by adding suitable additional members to the spreadsheet.

1. INTRODUCTION, cont.

- Neutron Activation Analysis, k0 standardization is a powerful tool in calculation of elemental concentration of different kind of samples;
- Using the TRIGA research reactor as a thermal neutron source, the samples are irradiated and subsequently measured using a hyper-pure geramanium crystal detector;
- The deconvolution of gamma ray spectra is made using GENIE2000 software;
- The uncertainties calculation of the obtained results has been done using KRAGTEN method, as a tool for neutron activation analysis equation;
- The paper underlines the key role of expressing a high precision of results.

2. EXPERIMENTAL – irradiation facility

- The samples irradiation was conducted inside TRIGA SSR reactor operated at a power of 12 MW;
- The 132 grid positions are used as follows:
  - 29 Low Enriched Uranium fuel bundles (LEU);
  - 8 control rods;
  - 48 beryllium blocks;
  - 5 experimental locations inside the core;
  - 42 empty locations with plugs

Fig. 2. TRIGA SSR 14 MW in operation

Fig 3. TRIGA SSR 14 MW present core configuration

2. EXPERIMENTAL – NAA-K0 standardization

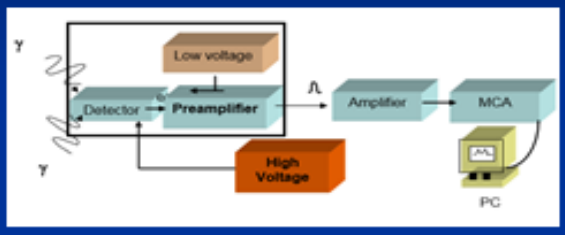
- NAA-k_0 standardization method is an analytic technique based on measuring the number and energy of gamma radiation emitted by the radioactive isotopes produced in the sample's matrix by irradiation in a thermal neutrons flux produced by a nuclear reactor;

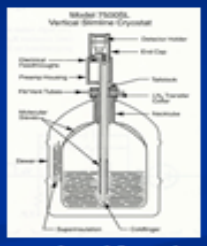
$$\rho_a(\mu g/g) = \frac{\left(\frac{N_p/t_c}{SDCW}\right)_a}{A_{sp,a}} \cdot \frac{1}{k_{0,m}(a)} \cdot \frac{G_{th,m}\cdot f + G_{e,m}\cdot Q_{0,a}(\alpha)}{G_{th,a}\cdot f + G_{e,a}\cdot Q_{0,a}(\alpha)} \cdot \frac{\varepsilon_{p,m}}{\varepsilon_{p,a}} \cdot 10^6$$

- k_0 nuclear data and decay correction scheme (all are available in open literature)
- Reactor's parameters: f and α (Høgdahl Convention, well-described)
- Detection efficiency calculated for absorption in sample (according with Moens)
- Irradiation and measurement aspects: neutron self-absorbtion in sample, dead time, counting time, coincidence correction, etc.

2. EXPERIMENTAL – HPGe spectrometry system

- The HPGe detector-based gamma spectroscopy system consists of an HPGe detector, high voltage power supply, preamplifier (which is usually sold as part of the detector), amplifier, Analogue to Digital Converter (ADC), and Multi-Channel Analyzer (MCA).





- The function of the electronic system is the collection of the electrons produced from the signal pulses and the processing of those pulses and sorting them by height or energy. This process can be described by the following steps:
  - Photon interacts with the detector crystal, produces burst of electrons;
  - Applied bias voltage sweeps electrons from crystal;
  - Current produced by electrons forms signal pulse;
  - Pulse size is increased with a preamplifier;
  - Pulse is further intensified and shaped with amplifier;
  - Pulse intensity is converted into numerical value using ADC;
  - Numerical values are sent to MCA;

2. EXPERIMENTAL – HPGe spectrometry system, cont.

- For measuring the spectra and detecting the chemical elements in the most effective way, the detector needs to be well calibrated in both energy and efficiency. For this purpose we used a certified calibration set of point sources: Co^{60}, Ba^{133}, Cs^{137}, Eu^{152} and Am^{241}.

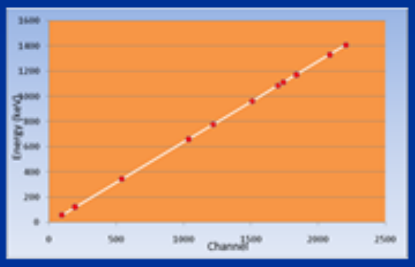


Fig. 4. Energy calibration curve of HPGe detector

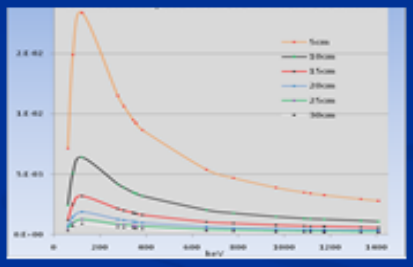
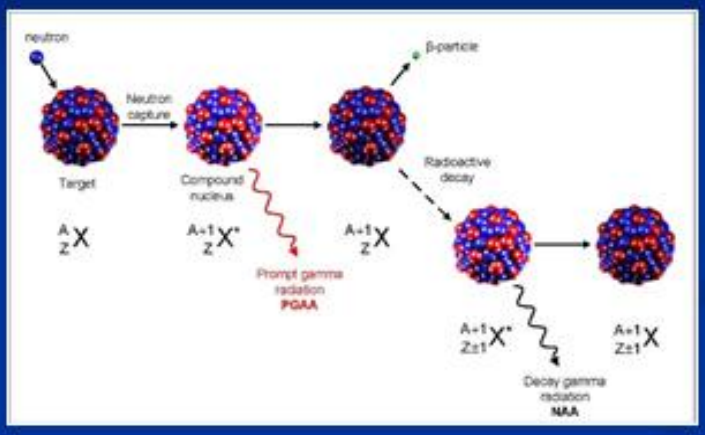
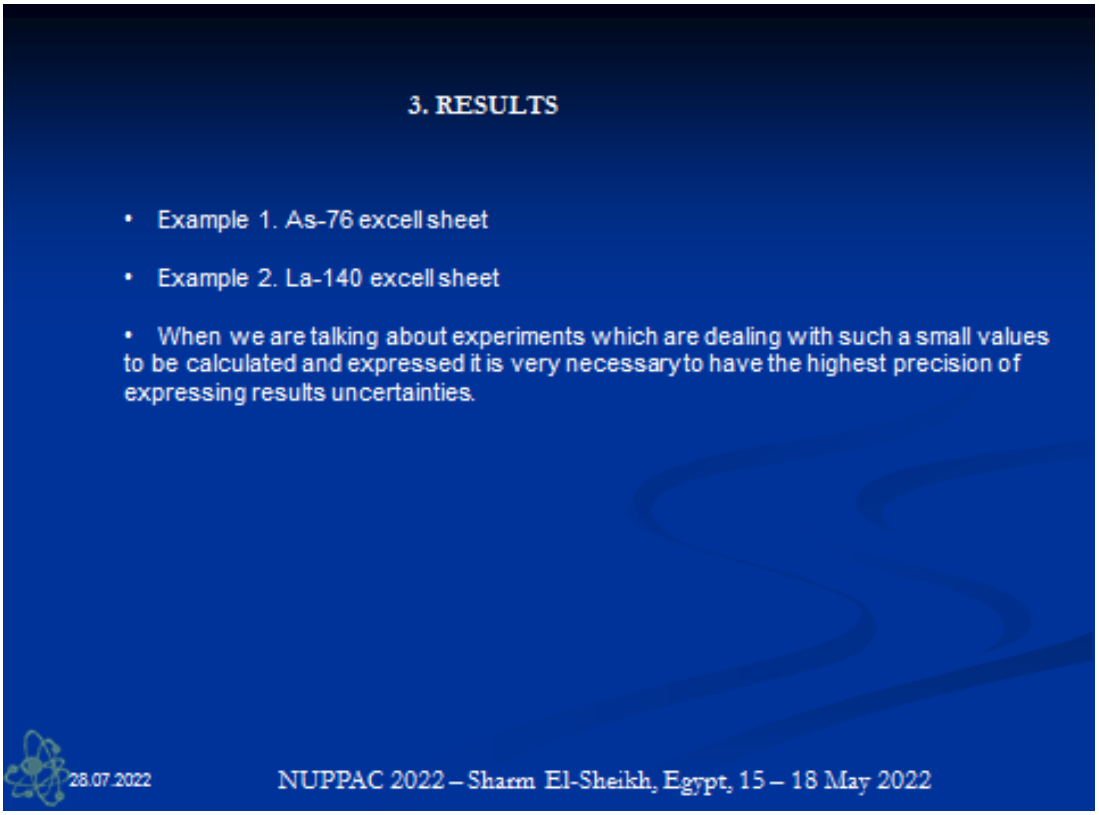
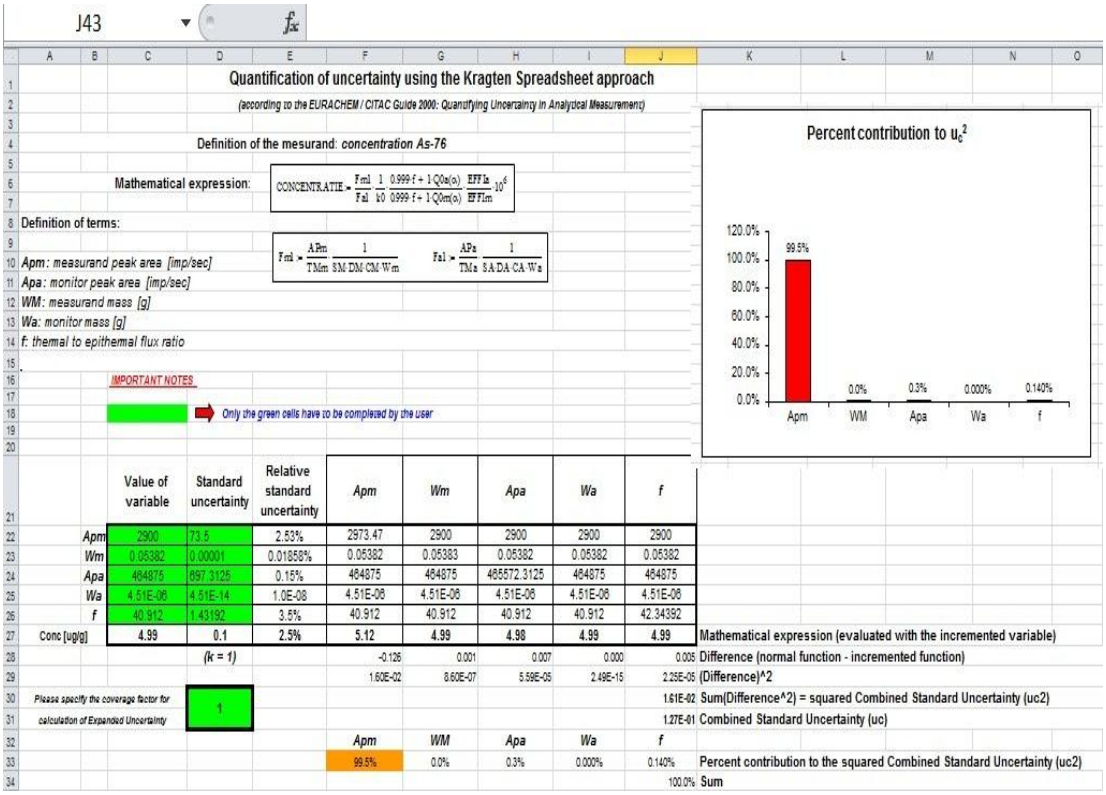


Fig. 5. Efficiency calibration curve of HPGe detector for different sample-end cap distances

2. EXPERIMENTAL – Neutron Activation mechanism







Quantification of uncertainty using the Kragten Spreadsheet approach

(according to the EURACHEM / CITAC Guide 2000: Quantifying Uncertainty in Analytical Measurement)

Definition of the mesurand: *concentration As-76*

Mathematical expression: $CONCENTRATIE = \frac{Fml}{Fal} \cdot \frac{1}{k0} \cdot \frac{0.999\,f + 1.Q0a(\alpha)}{0.999\,f + 1.Q0m(\alpha)} \cdot \frac{EFFla}{EFFlm} \cdot 10^6$

Definition of terms:

$Fml = \frac{APm}{TMm} \cdot \frac{1}{SM \cdot DM \cdot CM \cdot Wm}$ $Fal = \frac{APa}{TMa} \cdot \frac{1}{SA \cdot DA \cdot CA \cdot Wa}$

Apm: measurand peak area [imp/sec]
Apa: monitor peak area [imp/sec]
WM: measurand mass [g]
Wa: monitor mass [g]
f: thermal to epithermal flux ratio

IMPORTANT NOTES

Only the green cells have to be complexed by the user

		Value of variable	Standard uncertainty	Relative standard uncertainty	Apm	Wm	Apa	Wa	f	
	Apm	2900	73.5	2.53%	2973.47	2900	2900	2900	2900	
	Wm	0.05382	0.00001	0.01858%	0.05382	0.05383	0.05382	0.05382	0.05382	
	Apa	464875	697.3125	0.15%	464875	464875	465572.3125	464875	464875	
	Wa	4.51E-06	4.51E-14	1.0E-08	4.51E-06	4.51E-06	4.51E-06	4.51E-06	4.51E-06	
	f	40.912	1.43192	3.5%	40.912	40.912	40.912	40.912	42.34392	
Conc [ug/g]		4.99	0.1	2.5%	5.12	4.99	4.98	4.99	4.99	Mathematical expression (evaluated with the incremented variable)
			(k = 1)		-0.126	0.001	0.007	0.000	0.005	Difference (normal function - incremented function)
					1.60E-02	8.60E-07	5.59E-05	2.49E-15	2.25E-05	(Difference)^2
Please specify the coverage factor for calculation of Expanded Uncertainty			1						1.61E-02	Sum(Difference^2) = squared Combined Standard Uncertainty (uc2)
									1.27E-01	Combined Standard Uncertainty (uc)
					Apm	WM	Apa	Wa	f	
					99.5%	0.0%	0.3%	0.000%	0.140%	Percent contribution to the squared Combined Standard Uncertainty (uc2)
									100.0%	Sum

Chart 1

Quantification of uncertainty using the Kragten Spreadsheet approach

(according to the EURACHEM / CITAC Guide 2000: Quantifying Uncertainty in Analytical Measurement)

Definition of the mensurand: *concentratie Sm-153*

Mathematical expression: $CONCENTRATIE = \frac{Fmi}{Fal} \frac{1}{k0} \frac{0.999\,f + 1 \cdot Q0a(\alpha)}{0.999\,f + 1 \cdot Q0m(\alpha)} \frac{EFFla}{EFFlm} 10^6$

Definition of terms:

$Fmi = \frac{APm}{TMm} \frac{1}{SM \cdot DM \cdot CM \cdot Wm}$ $Fal = \frac{APa}{TMa} \frac{1}{SA \cdot DA \cdot CA \cdot Wa}$

Apm: measurand peak area [imp/sec]
Apa: monitor peak area [imp/sec]
WM: measurand mass [g]
Wa: monitor mass [g]
f: thermal to epithermal neutron flux ratio

IMPORTANT NOTES

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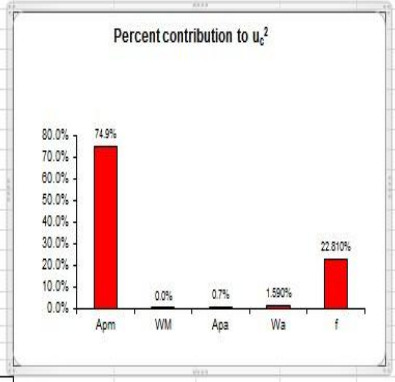

	Value of variable	Standard uncertainty	Relative standard uncertainty	Apm	Wm	Apa	Wa	f	
Apm	4780	72.7	1.52%	4852.72	4780	4780	4780	4780	
Wm	0.05382	0.00001	0.01858%	0.05382	0.05383	0.05382	0.05382	0.05382	
Apa	464875	697.3125	0.15%	464875	464875	465572.3125	464875	464875	
Wa	4.51E-06	1.00E-08	0.2%	4.51E-06	4.51E-06	4.51E-06	4.52E-06	4.51E-06	
f	40.912	1.43192	3.5%	40.912	40.912	40.912	40.912	42.34392	
Conc [ug/g]	9.52	0.2	1.8%	9.67	9.52	9.51	9.54	9.44	Mathematical expression (evaluated with the incremented variable)
		(k = 1)		-0.145	0.002	0.014	-0.021	0.080	Difference (normal function - incremented function)
				2.10E-02	3.13E-06	2.03E-04	4.46E-04	6.39E-03	(Difference)^2
Please specify the coverage factor for calculation of Expanded Uncertainty		1						2.80E-02	Sum(Difference^2) = squared Combined Standard Uncertainty (uc2)
								1.67E-01	Combined Standard Uncertainty (uc)

Apm	WM	Apa	Wa	f	
74.9%	0.0%	0.7%	1.590%	22.810%	Percent contribution to the squared Combined Standard Uncertainty (uc2)
				100.0%	Sum