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Development of k_0-standardized Neutron Activation Analysis using short-lived radionuclides at Dalat Research Reactor

Doanh Van Ho[1], Thien Quang Tran[2], Dung Manh Ho[1]*

[1]Center for Nuclear Technologies (CNT), VINATOM, Ho Chi Minh, Vietnam

[2]Dalat Nuclear Research Institute (DNRI), VINATOM, Dalat, Vietnam

Outline

1. Introduction
2. Materials & Methods
3. Results & Discussion
4. Conclusion

1. Introduction

Neutron activation analysis (NAA):

- Highly sensitive & non-destructive/minimum invasive: Quantitative multi-element analysis for environmental, geological, and biological samples.
- Key advantages: Minimal sample preparation, simultaneous multi-element detection, trace-element precision.
- In NAA, radionuclides are typically classified into three groups based on their half-lives: short-lived (~ seconds to minutes), medium-lived (minutes to hours), and long-lived (days to years).

1. Introduction *(con't)*

Conventional NAA vs. SLRN-based NAA:

Conventional NAA	SLRN-based NAA
Uses medium/ long-lived radio-nuclides (e.g., ^{24}Na, ^{42}K, ^{60}Co, ^{152}Eu)	Uses short-lived radionuclides (e.g., ^{77m}Se, ^{28}Al, ($T_{1/2}$ ~ seconds to minutes)
Long turnaround (days/weeks)	Rapid results (<1 hour or during day)
Limited by decay delays	Requires precise timing protocols and cyclic activation (CNAA)

Nuclear data for SLRNs used in NAA

No	Elements	Reactions	Radio-nuclides	$T_{1/2}$ (s)	σ_0 (barn)	I_0 (barn)	Main E_γ (keV)
1	Sb	$^{123}Sb(n,\gamma)^{124m}Sb$	^{124m}Sb	93.0	0.035	0.93	645.9
2	Ir	$^{191}Ir(n,\gamma)^{192m}Ir$	^{192m}Ir	87.0	300	1060	58.0
3	Dy	$^{164}Dy(n,\gamma)^{165m}Dy$	^{165m}Dy	75.6	1698	425	515.5
4	Rb	$^{85}Rb(n,\gamma)^{86m}Rb$	^{86m}Rb	61.2	0.05	1275	556.2
5	Ce	$^{138}Ce(n,\gamma)^{139m}Ge$	^{139m}Ce	56.4	0.15	2	757.0
6	Ge	$^{74}Ge(n,\gamma)^{75m}Ge$	^{75m}Ge	48.0	0.143	0.35	139.6
7	Rh	$^{103}Rh(n,\gamma)^{104}Rh$	^{104}Rh	42.3	134	112	555.8
8	O	$^{18}O(n,\gamma)^{19}O$	^{19}O	26.9	0.00016	0.00081	197.1
9	Ag	$^{109}Ag(n,\gamma)^{110}Ag$	^{110}Ag	24.6	89	0.2	657.8
10	Pd	$^{106}Pd(n,\gamma)^{107m}Pd$	^{107m}Pd	20.9	0.013	1.16	214.9
11	Sc	$^{46}Sc(n,\gamma)^{46m}Sc$	^{46m}Sc	18.75	9.6	--	142.5
12	Hf	$^{178}Hf(n,\gamma)^{179m}Hf$	^{179m}Hf	18.68	53	1039	216.0
13	Se	$^{76}Se(n,\gamma)^{77m}Se$	^{77m}Se	17.45	21	16	161.9
14	In	$^{115}In(n,\gamma)\ ^{116m}In$	^{116m}In	14.1	87	--	1293.6
15	Pt	$^{198}Pt(n,\gamma)^{199m}Pt$	^{199m}Pt	13.6	0.3	--	319.0
16	F	$^{19}F(n,\gamma)^{20}F$	^{20}F	11.02	0.0095	0.039	1633.6
17	Yb	$^{176}Yb(n,\gamma)^{177m}Yb$	^{177m}Yb	6.41	3.8	--	104.0
18	W	$^{182}W(n,\gamma)^{183m}W$	^{183m}W	5.65	20	600	107.9
19	Er	$^{166}Er(n,\gamma)^{167m}Er$	^{167m}Er	2.27	15	10	207.8
20	Pb	$^{206}Pb(n,\gamma)^{207m}Pb$	^{207m}Pb	0.8	0.03	0.1	570.0

1. Introduction *(con't)*

SLRN challenges in NAA:

- Radionuclides decay rapidly (e.g., ^{20}F: $T_{1/2} = 11$ s).
- Requires ms precision in irradiation, transfer & counting.

Mitigation:

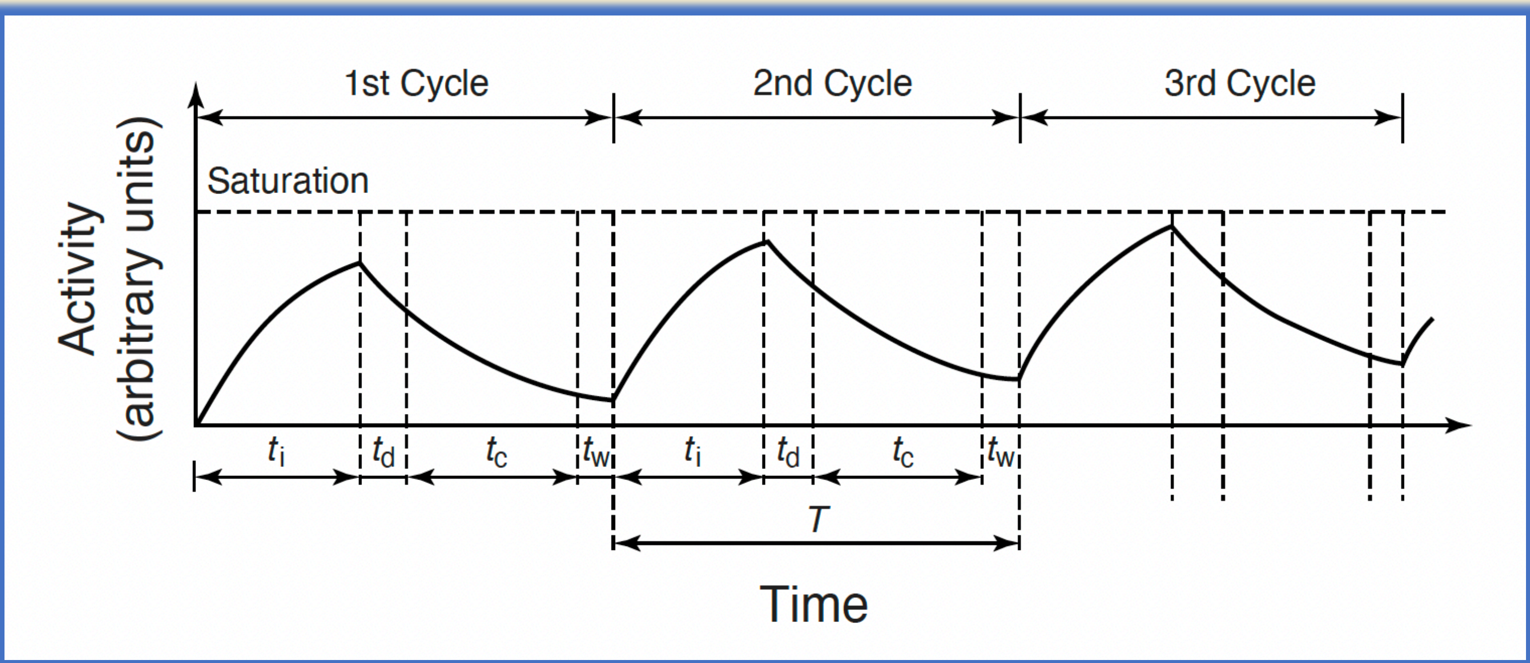
- ⏱️ Fast pneumatic transfer systems (e.g., ~1 s transfer).
- ⚡ Automated cyclic activation (CNAA) to accumulate activity.

1. Introduction *(con't)*

Overcoming SLRN challenges at DRR:

- **Developed Cyclic NAA (CNAA):**
  Repeated irradiation-decay-counting cycles enhance sensitivity and lower detection limits.
- **Applied k_0-Standardization:**
  Eliminates comparator standards; uses nuclear constants and reactor flux parameters (e.g., fluxes, α, f, T_n, etc).
- **Improvements & corrections:**
  Cyclic parameter optimization (irradiation/decay times), dead-time corrections, spectral interference mitigation.

Principle of Cyclic NAA



Elemental concentration in sample is calculated by k_0-CNAA

$$\rho = \frac{N_{pc}/t_m}{S.D.C.W} \cdot \frac{1}{k_0 F_c} \cdot \frac{1}{[G_{th}\varphi_{th}\sigma_0 + G_e\varphi_e I_0(\alpha)]} \cdot \frac{1}{\varepsilon_p}$$

$$N_{pc} = k\varphi_n \sum_{n'=1}^{n} \frac{1-e^{-n'\lambda T}}{1-e^{-\lambda T}} \cdot \frac{\varphi_{n+1-n'}}{\varphi_n}$$

Cumulated net peak area

$$F_c = \left[\frac{n}{(1-e^{-\lambda T})} - \frac{e^{-\lambda T}(1-e^{-n\lambda T})}{(1-e^{-\lambda T})}\right]$$

Dead time and Pile-up correction

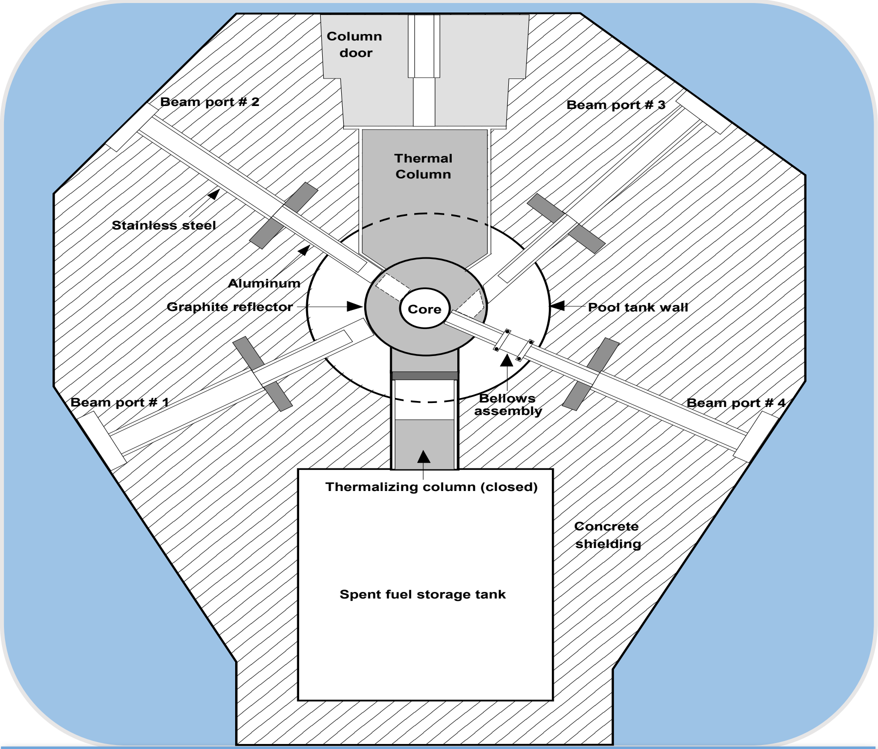
1. Introduction *(con't)*

Dalat Research Reactor (DRR):

- **Infrastructure Upgrades:**
  - Rapid Pneumatic Transfer System (PTS): ~0.65 s transit time.
  - Advanced Detection: DSPEC Pro spectrometer with Zero Dead-Time (ZDT) correction for high-activity.
- Apply the k_0-based CNAA protocols for SLRNs, e.g. ^{77m}Se (17.4s), ^{46m}Sc (18.75s) & ^{110}Ag (24.6s).
- Validate the method using SRMs/CRMs (biological, environmental, geological).

Facilities for NAA at Dalat Research Reactor

Dalat research reactor (DRR, 500 kW)



Layout of DRR horizontal beam ports

Lay out of the reactor

- Radial beam tubes: No #1: Oppened for fundamental studies; No #2: PGNAA; No #4: Total Cross-Section measurements
- Tangential beam tube: No #3: γ-γ coincidence spectrometer
- Thermal column: CNAA & nuclear data measurements

2. Materials and Methods

2.1. Irradiation Facility

Samples were irradiated at DRR with thermal neutron fluxes ~2.5×10^{12} $n \cdot cm^{-2} \cdot s^{-1}$ (Ch. 13-2); ~ 1.2×10^{11} $n \cdot cm^{-2} \cdot s^{-1}$ (TC).

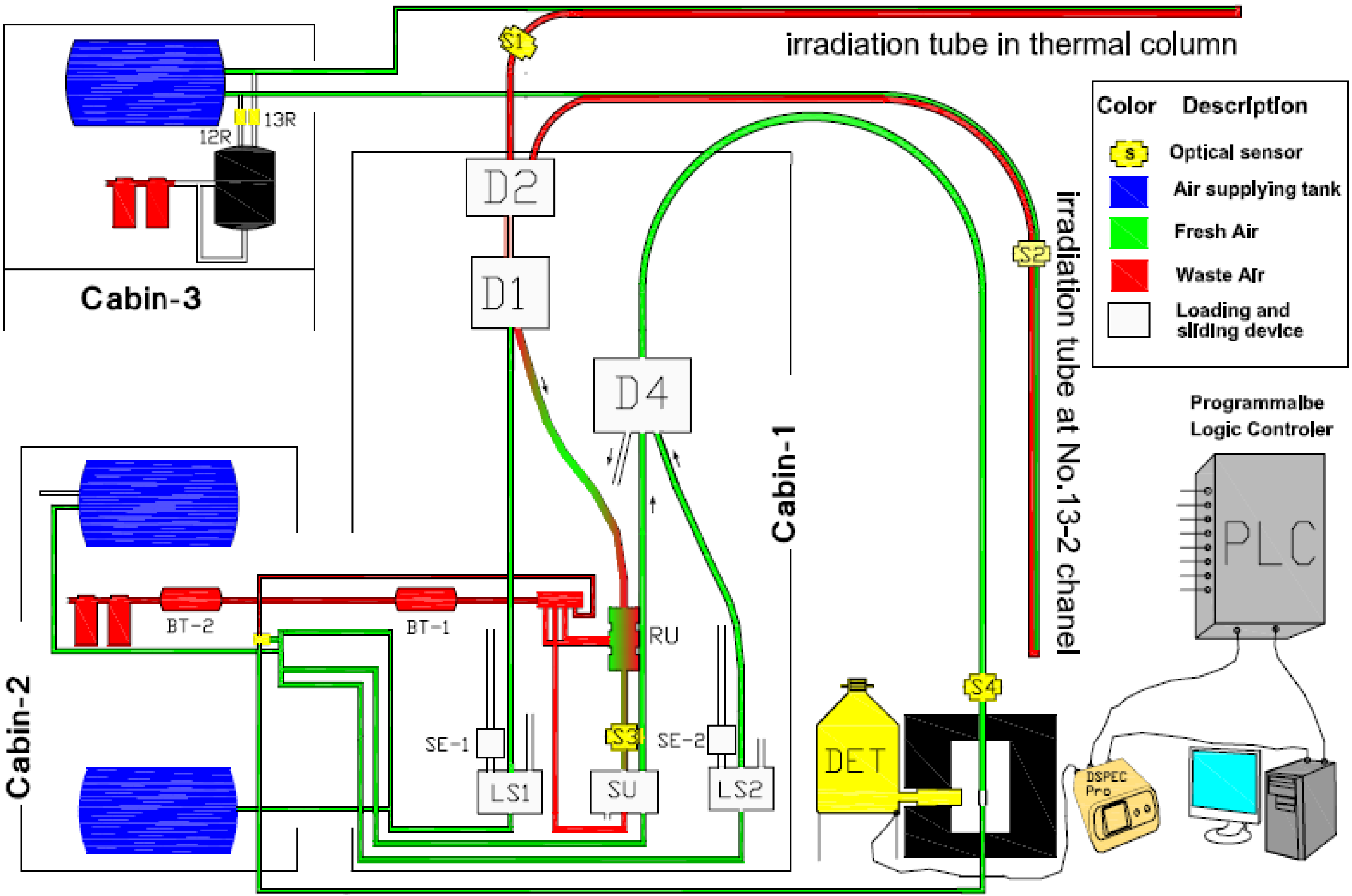
The flux parameters (α, $\varphi_{th}/\varphi_{epi}$, T_n) were monitored.

2.2. Cyclic Activation System

A fast pneumatic transfer system (PTS) enabled rapid sample transfer with transit times:
Channel 13-2: ~(0.63 ± 0.02) s and TC: ~(0.25 ± 0.02) s.

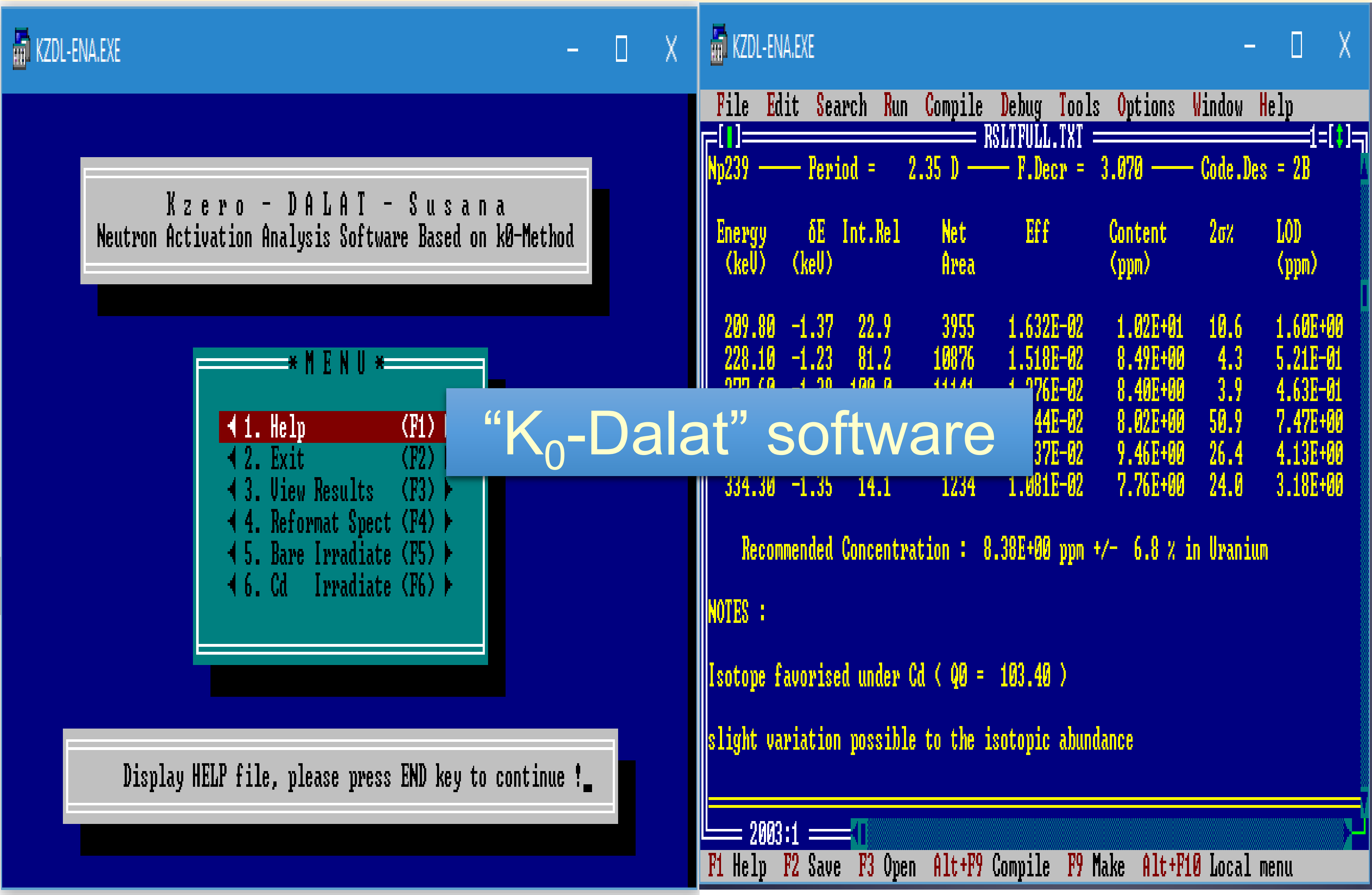
Period of cycles of irradiation, decay, counting and waiting:
$T = t_i + t_d + t_c + t_w$, automated.

Rapid pneumatic transfer system (PTS) for CNAA

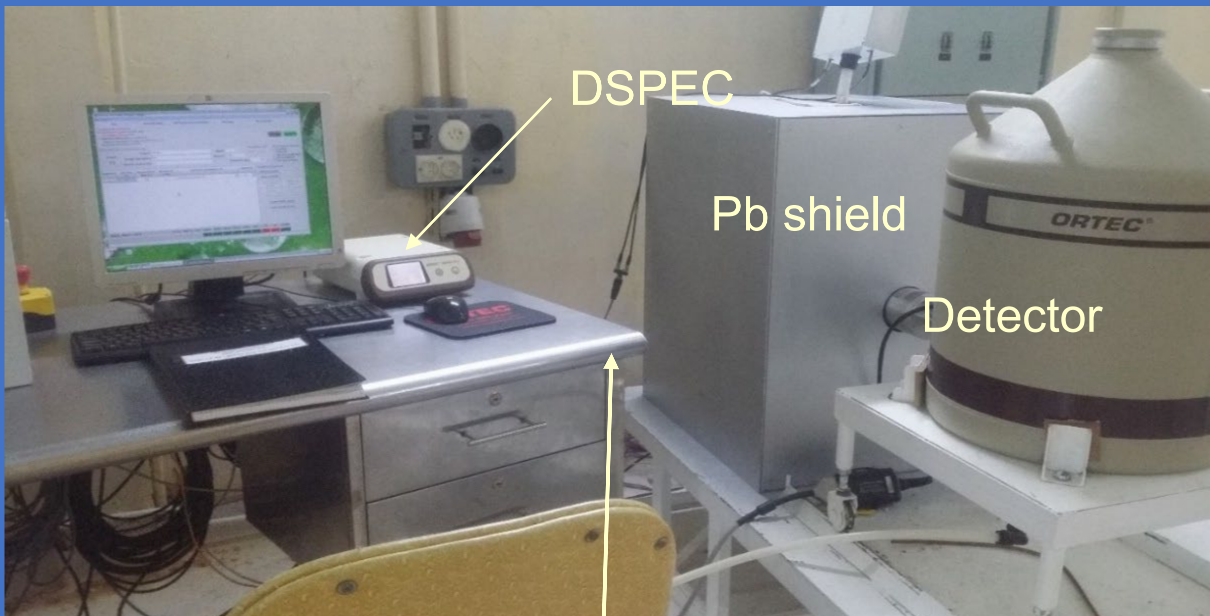


PTS for cyclic irradiations at Channel 13-2 or Thermal Column

Software for k_0-standardized (INAA, ENAA & CNAA)



2.3. Gamma-Ray Spectrometry



Gamma-ray spectrometer for CNAA at DRR

An open-sided Pb shield containing
GMX-4076 detector

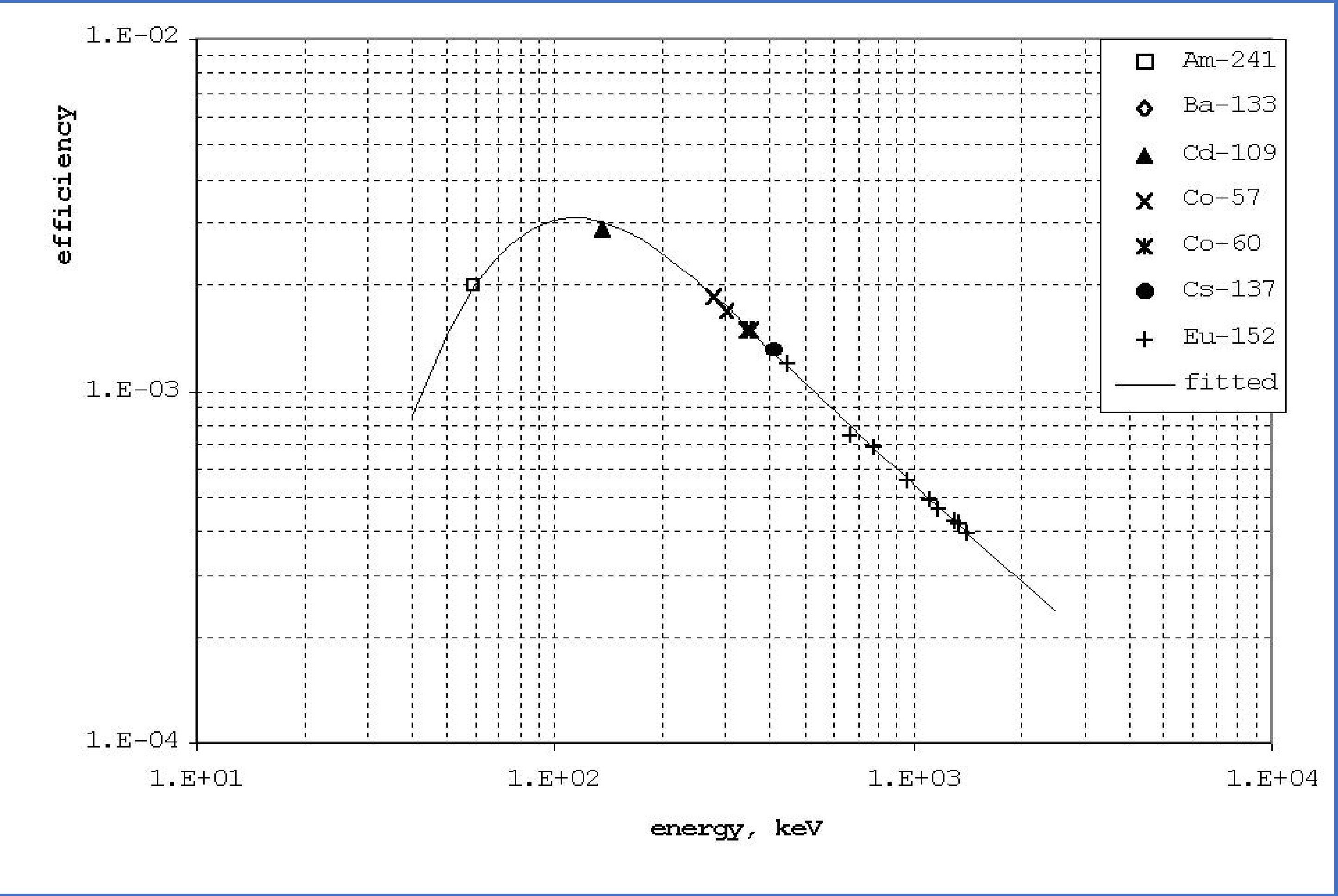
3. Results and Discussion

3.1. Characterization of neutron parameters

Neutron Parameters	Thermal Column	Channel 13-2
Thermal Flux *($\times 10^{12}$ n.cm^2.s^{-1})*	0.11 ± 0.01	4.09 ± 0.09
Epithermal Flux *($\times 10^{9}$ n.cm^2.s^{-1})*	0.56 ± 0.01	3.78 ± 0.20
Fast Flux *($\times 10^{11}$ n.cm^2.s^{-1})*	0.84 ± 0.06	6.60 ± 0.22
α	0.092 ± 0.035	-0.038 ± 0.006
f	203 ± 25	10.9 ± 0.6
T_n (K)	298 ± 28	312 ± 31

3. Results and Discussion

3.2. Calibration of detector

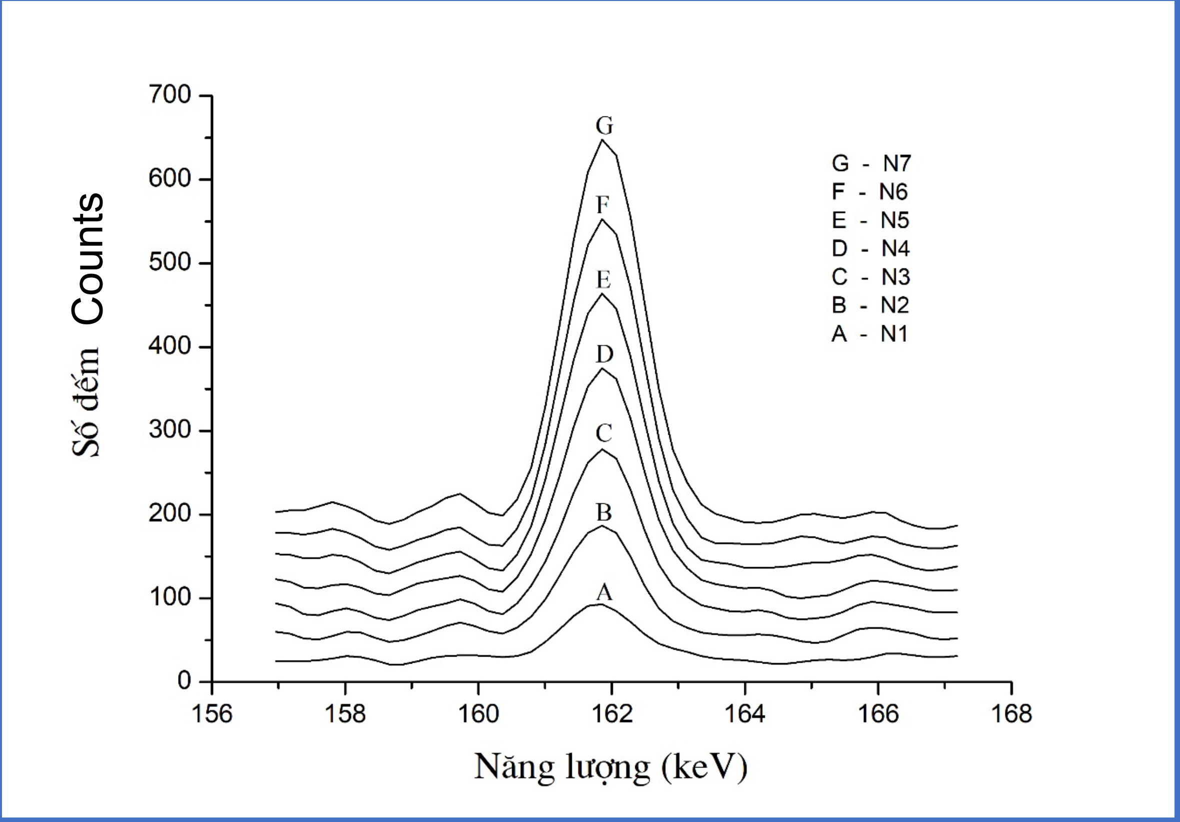


Efficiency curve of the detector at 16 cm.

3. Results and Discussion

3.2. Method Optimization

Saturated activity at 4-9 cycles; t_i = 5-10 s; LOD = 0.06 µg for ^{77m}Se in NIST-SRM-1566b.



Gamma peak at 161.9 keV of ^{77m}Se accumulated over cycles.

3. Results and Discussion

3.3. Method Validation: Using NIST-SRM-1566b (oyster)

Elements	$T_{1/2}$	Analysed (mg/kg)	Certified (mg/kg)	Analysed/ Certified	u-score
^{77m}Se	17.45 s	2.02 ± 0,23	2.06 ± 0,15	0.98 ± 0.13	-0.15
^{110}Ag	24.6 s	0.628 ± 0,082	0.660 ± 0,010	0.95 ± 0.12	-0.39
^{66}Cu	5.12 min	56.6 ± 18,7	71.6 ± 16,0	0.79 ± 0.31	-0.61
^{49}Ca	8.718 min	941 ± 282	838 ± 20	1.12 ± 0.34	0.36
^{52}V	3.75 min	0.56 ± 0,11	0.58 ± 0,02	0.97 ± 0.19	-0.16
^{28}Al	2.24 min	219 ± 46	197 ± 6	1.11 ± 0.24	0.47
^{27}Mg	9.46 min	1314 ± 123	1085 ± 23	1.21 ± 0.12	1.83
^{56}Mn	2.579 h	20.4 ± 2,0	18.5 ± 0,2	1.10 ± 0.11	0.95
^{38}Cl	37.24 min	5656 ± 735	5140 ± 100	1.10 ± 0.14	0.70
^{42}K	12.36 h	6637 ± 31	6520 ± 90	1.02 ± 0.01	1.23
^{24}Na	14.96 h	3467 ± 350	3297 ± 53	1.05 ± 0.11	0.48
^{20}F	11.02 s	278 ± 20	-	-	-
^{179m}Hf	18.68 s	0.026 ± 0,006	-	-	-
^{46m}Sc	18.75 s	0.030 ± 0,011	-	-	-
^{104}Rh	42.3 s	0.050 ± 0,021	-	-	-

3. Results and Discussion

3.3. Method Validation: Using NIST-SRM-2711a (soil)

Radionuclides	Analysed (mg/kg)		Certified (mg/kg)		Analysed/ Certified	*u*-score
	Conc.	*Unc*	*Conc.*	*Unc*		
^{165m}Dy	4.69	0.75	5.00	0.63	0.94	-0,32
^{46m}Sc	8.19	2.21	8.50	0.10	0.96	-0,14
^{179m}Hf	8.03	0.96	9.20	0.20	0.87	-1,20
^{51}Ti	3100	651	3170	80	0.98	-0,11
^{56}Mn	631	63	675	18	0.93	-0,68
^{52}V	75	4	81	6	0.93	-0,88
^{28}Al	60400	3020	67200	600	0.90	-2,20
^{49}Ca	20600	2260	24200	847	0.85	-1,51

4. Conclusion

- CNAA facility at DRR significant upgraded to enhance its analytical capabilities, particularly for short-lived radionuclides.
- K_0-Dalat software improved for k_0-CNAA data processing.
- Dead Time/Pile-Up Correction introduced with novel techniques (ZDT, LFC) to recover 95% of counts lost at ≤50% dead time, improving data reliability.
- Method validated mostly achieved <10% deviation from certified values in reference materials (NIST-1566b, NIST-2117a); LOD of Se ~0.1–0.3 mg/kg in biological samples.
- Neutron fluxes parameters at Channel 13-2 and TC characterized and Detectors calibrated for k_0-CNAA.

Publications

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The upgrading of the cyclic neutron activation analysis facility at the Dalat research reactor

Van Doanh Ho[1] · Manh Dung Ho[1] · Thanh Viet Ha[2] · Quang Thien Tran[1] · Dong Vu Cao[1]

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Abstract
The cyclic neutron activation analysis (CNAA) facility based on a pneumatic transfer system for short irradiation and rapid counting has recently been upgraded at the Dalat research reactor. The original facility was only designed for single irradiation. Therefore, this work has aimed to upgrade both hardware and software for the cyclic irradiation. In this paper, the upgrading of the facility for CNAA was described. Irradiation time of the facility were calibrated, thereby reducing irradiation time to seconds with precision. The accuracy and sensitivity of CNAA based-on the upgraded facility were assessed by determination of some short-lived nuclides.

Keywords Cyclic neutron activation analysis · k0-CNAA · Dalat research reactor

Introduction

Instrumental neutron activation analysis (INAA) has been developed and applied at the 500 kW Dalat research reactor (DRR) since 1984 [1]. Until now, it is capable of analyzing more than 40 elements in various materials based on radionuclides with short, medium and long half-lives

(PTS 13-2/TC) for rapid neutron activation analysis has been equipped to analyze very short-lived nuclides through the IAEA Technical Cooperation Project RER/4/028. The installation diagram was shown in Fig. 1. This facility consists of three main parts, designed by S.S. Ismail which was introduced in Ref. [4]. The first part consists of two aluminum irradiation tubes, which were inserted into the

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Combination and optimization of the cyclic NAA modes at the Dalat research reactor for determination of selenium in biological materials using ^{77m}Se

Van Doanh Ho[1] · Manh Dung Ho[1] · Quang Thien Tran[1] · Thi Sy Nguyen[1] · Nhi Dien Nguyen[1]

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Abstract The combination and optimization of the cyclic modes in neutron activation analysis (NAA) at the Dalat research reactor were performed to determine short-lived nuclides. This work focused on determination of selenium in biological materials using ^{77m}Se ($T_{1/2}$ = 17.4 s) as a case in point. The detection limit was significantly improved by a factor of 3–4 times in comparison with the conventional NAA. The precision was typically within the range of 4–6 % at the third or fourth cycle. It is a reliable technique for determination of selenium in biological materials at sub-ppm concentration levels.

Keywords Cyclic · Pseudo-cyclic · Cumulative · Replicate · Selenium

Introduction

^{77m}Se ($T_{1/2}$ = 17.4 s). However, the precision and detection limit (DL) were unsatisfied at sub-ppm concentration levels. Therefore, attempts are being made to solve this problem by applying cyclic modes in neutron activation analysis (NAA), including cyclic, pseudo-cyclic, cumulative NAA and combining cumulative NAA with CNAA or PCNAA. The principle of CNAA has been reported in detail [1]. In addition, the k0-based cyclic NAA has recently developed for the determination of a few short-lived nuclides [2]. In cyclic NAA, the sample is irradiated for a short period of time. After a short decay time the gamma-ray emitted is counted for a short period of time, then the sample is irradiated again. The entire process repeats for a number of cycles [3, 4]. In the case of sample with high dead time, a pseudo-cyclic NAA (PCNAA) technique was alternatively developed based on CNAA by adjusting the CNAA timing parameters [5–8], especially

Recent Collaborations

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Thank you for your attention!