IMPROVEMENT OF SENSITIVITY FOR FOODSTUFF ELEMENTAL ANALYSIS THROUGH THERMAL AND/OR EPI-THERMAL NEUTRON ACTIVATION ANALYSIS.

By

PRESITA ANNA LERULELO

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Study Leader: Prof Dr Arnaud FAANHOF
Specialist Scientist*

Mentor: Mr. Deon KOTZE
Senior Scientist**

* Necsa: Research & Development
** Necsa: RadioAnalysis

ABSTRACT

Instrumental neutron activation analysis (INAA) for essential and toxic elements in foodstuffs commonly consumed by South Africans as routinely applied at NECSA could not give adequate sensitivity for a variety of elements as shown by previous research. Improvement in sensitivity can be achieved by considering the following aspects:

Evaluation of the dominant radionuclide in the irradiated foodstuffs - (extracted from the available data from the previous study).

Determination of the cadmium ratio (R_{cd}) through irradiation of element standards in the full neutron spectrum as well as irradiation in the cadmium shielded part of the neutron spectrum.

Determination of the advantage factors of elements of interest over dominant nuclides in the gamma-spectra of irradiated foodstuffs

Determination of the neutron flux and neutron spectrum stability for INAA in the irradiation channels used for instrumental neutron activation analysis at the South African Fundamental Atomic Reactor Installation 1 (**SAFARI-1**) to evaluate the possible influence on the advantage factors.

Research deduced that dominant radionuclide vary according to irradiation-delay-count (IDC) systems, both RINGAS and PRS data indicated that there is change in flux and spectral shift within specified limits and change outside specified limits could be due to various activities carried out at **SAFARI-1**

Advantage factors have been determined thus irradiation facility can be chosen to obtain optimal sensitivity for elements of interest in specific food matrices.

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I lift up mine eyes to thee(almighty) for bringing me this far for you allow nothing for not.

Ke mmina Tholo yo sa thlolweng!

Declaration of Authenticity

I, the undersigned PRESITA ANNA LERULELO, hereby certify that the work presented in this dissertation except where otherwise indicated, is my own original work and has not been submitted to any university for the purpose of obtaining a degree.

PRESITA ANNA LERULELO
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GENERAL ABBREVIATION AND SYMBOLS

AI	Adequate Intake
DRI	Dietary Reference Intake
EAR	Estimated Average Requirement
FAO	Food and Agriculture organization of United Nations
IAEA	International Atomic Energy Agency
IDC	Irradiation, Delay and Counting
INAA	Instrumental Neutron Activation Analysis
MDC	Minimum Detectable Concentration
NECSA	South African Nuclear Energy Corporation Pty Ltd.
ppm	parts per million
PTI	Provisional Tolerable Intake
RDA	Recommended Dietary Allowance
SAFARI	South African Fundamental Atomic Reactor Installation
UNC	Uncertainty
WHO	World Health Organization

1. INTRODUCTION

1.1 Overview

During the last decades biological, medical, criminological, archaeological and agricultural studies, amongst others, have emphasized the need for trace elements analysis [1, 4, 7, 11]. It was also proved that even minute amounts of elements have a major contribution on the metabolism of all living organisms [7].

A previous study [13] revealed that routine neutron activation analysis techniques applied at Necsa were not adequately sensitive to provide meaningful results in terms of essential and or toxic element intake by humans through consumption of some 23 common foodstuffs in South Africa.

1.2 Objectives

The main objective of this study is to evaluate possible improvements in the sensitivity of instrumental neutron activation analysis (INAA) of essential and toxic elements in a variety of foodstuffs commonly used in South Africa. The following aspects have been considered to reach the objectives:

- Evaluation of the dominant radionuclide in the irradiated foodstuffs according to a number of routine irradiation, decay and counting time-schemes (extracted from the available data from the previous study).
- Determination of the cadmium ratio (R_{cd}) through irradiation of element standards in the full neutron spectrum as well as irradiation in the cadmium shielded part of the neutron spectrum.
- Determination of the advantage factors of specific elements over the dominant radionuclides in the gamma-spectra of irradiated foodstuffs.
- Determination of the neutron flux and neutron spectrum stability for INAA in the irradiation channels used for instrumental neutron activation analysis at the SAFARI Research Reactor.
- Evaluation of the possible improvement in analysis sensitivity.

2 PREVIOUS FINDINGS

In a previous investigation [13] some twenty three foodstuffs have been analyzed by routine INAA as applied at Necsa to determine the analysis sensitivity for a suite of about thirty essential and toxic elements. Elements of interest were classified as:

- **Essential macro nutrients and trace elements:**
  Ba, Br, Ca, Cl, Co, Cr, Cs, Cu, Fe, I, K, Mg, Mn, Mo, Na, Rb, Se, Sc, Zn

- **Trace elements that are probably essential:**
  B, Ni, Si, V

- **Toxic elements:**
  F, Pb, Cd, Hg, Al, Li, Sn.

- **Radiotoxic elements:**
  Th, U, and their progeny.

Of these elements boron, lead and lithium could not be determined by neutron activation as no radionuclides are produced that emit suitable photons, while aluminum and silicon proved cumbersome due to aluminum interference from the irradiation channels and accordingly these elements will not be considered for this study.

The commonly consumed foodstuffs, which were considered included: fish, pork, pumpkin, beetroot, green beans, maize, sugar, coffee, tea, milk, spinach, potatoes, white bread, brown bread, rice, cabbage, tomatoes, margarine, onions, chicken meat and eggs.

Four irradiation, delay and counting time-schemes were applied, using the two available INAA systems at the SAFARI-1 reactor. Table 1 provides the data.

The elements of interest with their specific radionuclides used for the analyses are given in Table 2.

The overall evaluation of the element analysis sensitivity of routine INAA as applied at Necsa is shown in Table 3.

Hence it is notable from Table 3 that adequate sensitivity is achieved for nine elements while thirteen elements still need improvement. Therefore the focus of this study will be on these elements specifically. Future evaluation is required for the six elements for which no recommended dietary allowances are available to date.

Table 1 Respective routine time schemes applied in the previous study [13]

Time-scheme	Facility	Irradiation time	Delay time	Counting time
1	RINGAS	10sec	10min	10min
2	RINGAS	10-20sec	3hrs	15-30min
3	RINGAS	10-20sec	30hrs	15-30min
4	PRS	30min	7days	30-60min

Table 2 Properties of radionuclides used in INAA [13]

Radionuclide	Half-life	Nuclide and reaction	Energy (keV)
^{76}As	26.4h	^{75}As (n,γ)	559
^{131}Ba	11.5 d	^{130}Ba(n,γ)	496
^{82}Br	35.3h	^{81}Br(n,γ)	776
^{47}Ca	4.54 d	^{46}Ca(n,γ)	1297
^{115m}Cd	44.8 d	^{114}Cd(n,γ)	934, 1291
^{38}Cl	37.2 m	^{37}Cl(n,γ)	1642
^{60}Co	5.27 a	^{59}Co(n,γ)	1173, 1332
^{51}Cr	27.7 d	^{50}Cr(n,γ)	320
^{134}Cs	2.06 a	^{133}Cs(n,γ)	795
^{66}Cu	5.1 m	^{65}Cu(n,γ)	1039
^{20}F	11 s	^{19}F(n,γ)	1634
^{59}Fe	44.6 d	^{58}Fe(n,γ)	1099
^{203}Hg	44.6 d	^{202}Hg(n,γ)	279
^{128}I	25 m	^{127}I(n,γ)	443
^{42}K	36 h	^{41}K(n,γ)	1525
^{27}Mg	9.46 m	^{26}Mg(n,γ)	844, 1014
^{56}Mn	2.58 h	^{55}Mn(n,γ)	846
^{99}Mo	66 h	^{98}Mo(n,γ)	740, 181, 778, 140 (^{99m}Tc)
^{24}Na	15.03 h	^{23}Na(n,γ)	1369
^{58}Co	70.78 d	^{58}Ni(n,p)	811
^{86}Rb	18.7 d	^{85}Rb(n,γ)	1077
^{46}Sc	84 d	^{45}Sc(n,γ)	889
^{75}Se	120 d	^{74}Se(n,γ)	265
^{125m}Sn	9.5 m	^{124}Sn(n,γ)	332
^{233}Pa	27 d	^{232}Th(n,γ) + β^- decay	312
^{239}Np	2.35 d	^{238}U(n,γ) + β^- decay	228
^{52}V	3.75m	^{51}V(n,γ)	1434
^{65}Zn	244 d	^{64}Zn(n,γ)	1115

Table 3 The overall evaluation of the sensitivity of element analysis in foodstuffs by routine INAA as applied at Necsa [13]

Element	Sensitivity improvement required	Suggested improvement
As	x 10	$T_{irradiation} >, T_{counting} >, M_{sample} >$**
Ba	No RDA	
Br	No RDA	
Ca	x 5	$T_{irradiation} >, T_{counting} >, M_{sample} >$**
Cd	x 50	$T_{irradiation} >, T_{counting} >, M_{sample} >$**
Cl	Overall adequate	
Co	X 15	$T_{irradiation} >, T_{counting} >, M_{sample} >$**
Cr	x 15*	$T_{irradiation} >, T_{counting} >, M_{sample} >$**
Cs	No RDA	
Cu	x 500	Ashing, isotope choice
F	x 5	Semi-cyclic activation
Fe	X 5*	$T_{irradiation} >, T_{counting} >, M_{sample} >$**
Hg	X 10	$T_{irradiation} >, T_{counting} >, M_{sample} >$**
I	Overall adequate	
K	Overall adequate	
Mg	Overall adequate	
Mn	Overall adequate	
Mo	x 20*	$T_{irradiation} >, T_{counting} >, M_{sample} >$**
Na	Overall adequate	
Ni	No RDA	
Rb	No RDA	
Sc	No RDA	
Se	x 20	$T_{irradiation} >, T_{counting} >, M_{sample} >$**
Sn	Overall adequate	
Th	Overall adequate	
U	x 5	$T_{irradiation} >, T_{counting} >, M_{sample} >$**
V	x 5	$T_{irradiation} >, T_{counting} >, M_{sample} >$**
Zn	Overall adequate	

*For infants only
**Sample ashing is advised to increase the sample mass

3 WORK PERFORMED IN THIS STUDY

As stated in section 1.2 a number of possible actions to improve the sensitivity of a number of elements have been envisaged. The following actions have been executed:

3.1 Evaluation of predominant radionuclides in previously irradiated foodstuffs.

- In this study all gamma spectra of the previously irradiated foodstuffs obtained through the four irradiation/delay/counting time–schemes as given in Table 1 have been evaluated and the dominant radionuclides have been tabulated.
- Five tables have been made, each representing one of the specific time-schemes.
- Up to a maximum of three dominant radionuclides have been identified.

3.2 Determination of the neutron flux and neutron spectrum stability for INAA in the irradiation channels used for instrumental neutron activation analysis.

- The RINGAS (Routine Instrumental Neutron and Gamma Analysis System) system has been evaluated on a suite of uranium and thorium standards, irradiated in both the cadmium shielded and bare irradiation positions, and determined by delayed neutron counting. Uranium fission is predominantly caused by thermal neutron interaction with ^{235}U, while thorium will undergo fission mainly through the interaction of epithermal and fast neutrons with ^{232}Th.
- The PRS (Pneumatic Rabbit System) system has been evaluated through iron monitors. Interaction of thermal neutrons with iron will result in the production of ^{59}Fe through the ^{58}Fe (n,γ)-reaction while the interaction of epithermal and fast neutrons with ^{54}Fe will lead to ^{54}Mn through the (n,p)-reaction.
- Both systems have been evaluated over a period ofabout 4-months.

3.3 Determination of the cadmium ratio (R_{cd})

- The RINGAS system, which is fitted with a cadmium-shielded and bare irradiation position, makes it possible to determine the ratio of the induced activity for each element of interest when irradiated under the full neutron spectrum and the epithermal/fast part of the neutron spectrum. This will thus allow one to evaluate the possible gain in element analysis sensitivity through comparison of the R_{cd}-values of the nuclides of the elements of interest and the major nuclides in the γ-spectrum of the individual foodstuffs after irradiation.
- The R_{cd}-value is defined by:

$$R_{cd} = \frac{\text{Induced activity upon irradiation under the full neutron spectrum}}{\text{Induced activity upon irradiation under the cadmium shielded part of the neutron spectrum}}$$

3.4 Determination of the advantage factors

- Evaluation of the R_{cd}-values of the nuclides produced upon neutron irradiation with the major nuclides observed in the γ-spectra of the foodstuffs of interest will lead to a so-called "advantage factor, defined by:

$$A^{x:Int} = \frac{R_{cd}^{\text{nuclide of interest}}}{R_{cd}^{\text{major interfering nuclide in the spectrum of the foodstuff of interest}}}$$

- Accordingly there will be an advantage leading to an improved sensitivity if:

$A^{x:Int}$	>	1	Irradiated under the full neutron spectrum
$A^{x:Int}$	<	1	Irradiated under the cadmium shielded part of the spectrum

3.5 Evaluation of the possible improvement in sensitivity

- Evaluation of the advantage factors against the required improvement in element analysis sensitivity will lead to improved information to produce relevant information regarding the nutritive value of essential and toxic elements in foodstuffs.
- The existing database can be updated for intake of a wide variety of consumption goods like traditional herbal medicine, raw water sources, crops affected by industrial contaminants as well inhalation of industrial pollutants.

4 GENERAL THEORY OF NEUTRON ACTIVATION

A short overview is given below to provide the reader with the essential information required for this study. More detailed information is provided in Appendix A for those that are further interested in the technique.

When a material is exposed to neutrons a nuclear reaction may occur resulting in the formation of a radionuclide, which, if radioactive, decays to eventually a stable isotope. When a neutron is absorbed by a nucleus, a nuclide is formed, mostly in an excited energy state, which decays (fast) to the ground state normally by emission of γ-rays (photons) or neutrons [4]. This nuclide, again if radioactive, will decay with a characteristic disintegration constant ($\lambda = \ln 2 / T_{1/2}$) to form another nuclide, which, again if radioactive, decays with its own disintegration constant (which differs from the previous one), until a stable nuclide is formed. After decay the product nucleus will normally be in an excited energy state releasing its excess energy by emission of (in general nuclide specific) γ-rays. The reactions can be represented by,

Nuclide A (activated) $\rightarrow$Nuclide B (with $T_{1/2}$)$\rightarrow$ Nuclide X (stable)

The rate of formation of the radionuclides depends on,

- the neutron flux (ϕ), which is the number of neutrons passing through a unit area per second ($n.cm^{-2}s^{-1}$)
- the number of atoms of the element (N) being bombarded,
- the tendency to capture neutrons, also called the cross section (σ),
- the time-span of the bombardment, and
- the decay of the formed radionuclide during the time-span of the bombardment.

Accordingly, the production of the number of radioactive atoms is given by the rate of its formation ($N\phi\,\sigma$) minus the rate of disintegration (λN), which can be described by

$$dN_t / dt = N_0.\sigma.\phi - \lambda.N_t \qquad \ldots.(1)$$

Integration of the equation (1) we will obtain the number of radioactive atoms present at the end of bombardment, i.e.

$$N_t = N_0 . \phi . \sigma . 1/\lambda . (1-e^{-\lambda t_1}) \qquad \ldots(2)$$

Where :

N_t	the number of radionuclides formed
N_0	the number of initial nuclides
t_1	the irradiation time in seconds
σ	cross section usually expressed in barns (1 barn = 10^{-24} cm^2)
ϕ	the neutron flux ($n.cm^{-2}. s^{-1}$)
λ	the decay constant (s^{-1}),which is equal to $\ln 2/T_{1/2}$
$T_{1/2}$	the half-life of the radionuclide formed (s)

The activity of the specific source is directly related to the number of atoms available and the rate at which they decay, which again is the function of the half-life of the radionuclide concerned. The relation is given by

$$A_t = \lambda . N_t \qquad \ldots..(3)$$

Substitution of (2) into (3) gives the activity formed after neutron irradiation during time t

$$A_t = N_0 . \phi . \sigma . (1 - e^{-\lambda t_1}) \quad .....(4)$$

The number of atoms available could be calculated from the mass of the element in the sample, the atomic mass, the isotopic abundance and Avogadro's number

$$N_0 = (W . \Phi . N_{Av}) / M \quad .....(5)$$

where:

- N_0 the number of atoms
- W the element mass
- Φ the isotopic abundance
- N_{Av} Avogadro's number ($6.024 . 10^{23}$)
- M the atomic mass of the element

Substitution of (5) into (4) leads to:

$$A_t = [(W . \Phi . N_{Av}) / M] . \phi . \sigma . (1 - e^{-\lambda t_1}) \quad .....(6)$$

From equation (6) it is clear that the mass of the element (W) can be determined if other quantities are known. However the neutron flux and the isotopic neutron cross section are usually not known with sufficient accuracy [15, 11, 5]. Accordingly, element standards are normally irradiated along with samples to allow for comparative measurements .The activity of a specific nuclide at the beginning of counting period depends on the induced activity at the end of the irradiation as well as the delay time between the end of the irradiation and the start of measurement, which can be described by:

$$A_t = [(W . \Phi . N_{Av}) / M] . \phi . \sigma . (1 - e^{-\lambda t_1}) . e^{-\lambda t_2} \quad [4] \quad .....(7)$$

where t_1 is irradiation time and t_2 the time elapsed since the end of the irradiation.

As the activity is expressed in disintegrations per second (unit: Becquerel), the net number of counts observed in a photopeak (C_t) of a specific nuclide of interest will be a function of:

- the intensity of specific photon in decay process (I_γ)
- the geometric efficiency of the sample with regard to the detector(ε_g)
- detector efficiency for the gamma energy of interest (ε_γ),and
- the integrated time the sample is counted for (t_3)

which can be described by:

$$C_t = A_t . I_\gamma . \varepsilon_g . \varepsilon_\gamma . t_3 . [(1 - e^{-\lambda t_3}) / \lambda t_3] \quad [4] \quad .....(8)$$

where

- I_γ :the intensity of the specific photon in the decay process
- ε_g :the geometric efficiency of the sample with regard to detector
- ε_γ :the detector efficiency for the gamma energy of interest

The last term due to integrated number of counts, as the nuclide will decay during the counting time. It can be easily seen that if the half-life of the nuclide of interest is much larger than the counting time, the last term will be equal to 1.

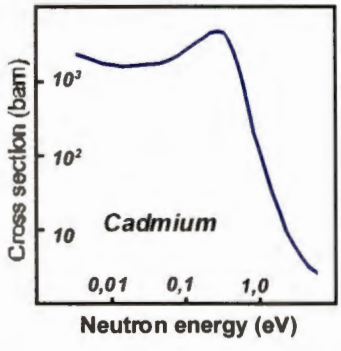
5 EXPERIMENTAL

5.1 Sensitivity

The sensitivity is associated with the lower limit of detection (LLD) or minimum detectable signal, to define the lowest or minimum detectable concentration (in μg/g or ppm) that could be detected in gamma spectroscopy [2]. The higher the sensitivity the lower the detection limit. To produce meaningful results the sensitivity should be adequate for every element of interest, i.e. if an element becomes toxic above 10 ng per gram of a foodstuff it does not make sense that an analytical method reports a detection limit of 100 ng/g; the analytical technique of choice should be able to determine the concentration of the specific toxic element at least down to a detection limit of 1 ng/g. The sensitivity of INAA is a function of the neutron density and neutron spectrum at the irradiation positions used at the Safari-1 research reactor (see also § 5.6)

5.2 Definition of Thermal and Epi-thermal Neutrons

Neutrons are neutral particles of the nucleus (no charge) hence able to interact with the entire atom without any force constraints. Their production in the reactor is induced by a process known as fission. Fission neutrons cover the entire range of the energy spectrum, i.e. thermal neutrons, epi-thermal neutrons and fast neutrons. The energy spectrum of these neutrons is called the fission neutron energy spectrum. Most of the research reactors contain within their core these distinct neutron flux components.
Fast neutrons are considered as those above 0.1 MeV, epi-thermal as those between 0.2 eV to 0.1 MeV and thermal are those below 0.2 eV [12]. The element cadmium has a high cross-section for thermal neutrons (see figure) and accordingly provides a tool to irradiate samples and element standards with epi-thermal and fast neutrons only.



5.3 Determination of the Cadmium Ratios (R_{cd})

Appropriate amounts of the elements discussed in section 1 have been irradiated according to respective time-schedules as previously applied for the foodstuffs as shown in Table 1. Irradiations have been done both in the cadmium shielded and bare irradiation positions. Element quantities have been prepared to give at least ten thousand counts in the principal photo-peak(s) of interest, to allow a statistical error in the counting results of ≤1% for each element. The R_{cd}-values have been calculated for each neutron induced reaction (i.e. separately for the (n,γ), (n,n'), (n,p) and (n,α) reactions).
The measured number of counts due to the induced activity in the cadmium shielded and bare irradiation positions for each element are defined using equation (8) as:

$$C_t^{cd} = (A_t)^{cd} . I_\gamma . \varepsilon_g^{cd} . \varepsilon_\gamma^{cd} . t_3^{cd} . [(1 - e^{-\lambda t_3})^{cd} / \lambda t_3^{cd}] \quad .....(9)$$

and

$$C_t^{ncd} = (A_t)^{ncd} . I_\gamma . \varepsilon_g^{ncd} . \varepsilon_\gamma^{ncd} . t_3^{ncd} . [(1 - e^{-\lambda t_3})^{ncd} / \lambda t_3^{ncd}] \quad .....(10)$$

With $(A_t)^{cd}$ and $(A_t)^{ncd}$ defined by equations (5) and (7), the induced activities can be defined by:

$$(A_t)^{cd} = N_0^{cd} . \phi^{cd} . \sigma^{cd} . (1 - e^{-\lambda t_1})^{cd} . (e^{-\lambda t_2})^{cd} \quad .....(11)$$

and

$$(A_t)^{ncd} = N_0^{ncd} \cdot \phi^{ncd} \cdot \sigma^{ncd} \cdot (1 - e^{-\lambda t_1})^{ncd} \cdot (e^{-\lambda t_2})^{ncd} \quad \text{.....(12)}$$

One then has to normalize for possible differences in element mass, irradiation time, decay time, counting time, sample geometry differences and detector efficiency. Accordingly, one arrives at specific activities expressed in Becquerel per gram (Bq/g) from which the cadmium ratio can be calculated as defined in section 3.3:

$$R_{cd} = (A_t)^{ncd} / (A_t)^{cd} \quad \text{.....(13)}$$

5.4 Determination of the Advantage Factors

If one takes the statement from the previous study [13] that sodium could be observed in most of the foodstuff samples analyzed, since ^{24}Na has prominent energy lines at 1368 keV and 1732 keV therefore elements which produce isotopes below these energy lines will be superimposed on the Compton continuum. This leads to a reduced minimum detectable signal, which can be seen from its definition:

$$C_{min} = f(B_u)^{(3)} \quad \text{...(14)}$$

Where,

C_{min} Minimum detectable number of counts
B_u Number of counts under the peak of interest

Hence the sensitivity of the element analyzed for will be decreased if their energy line(s) are superimposed on the ^{24}Na continuums. In order to be able to determine whether the sensitivity of an element other than sodium could be increased if the foodstuff would have been irradiated under the cadmium-shielded part of the neutron spectrum, one then has to evaluate to which extent the ^{24}Na activity will be reduced relative to the relevant radionuclides of other elements. As an example we take the required improvement in the sensitivity for arsenic (see Table 3). The following induced reactions apply:

$^{23}Na\ (n,\gamma)\ ^{24}Na \qquad E_\gamma =$ 1369 and 1732 keV [13]

and

$^{75}As\ (n,\gamma)\ ^{76}As \qquad E_\gamma =$ 559 keV. [13]

The question to be answered will be which of the induced activities will be reduced most, ^{24}Na or ^{76}As, if the sample is irradiated under the cadmium-shielded part compared to an irradiation under the full neutron spectrum? We can thus define the advantage factor according to section 3.4 as:

$$A^{As:Na} = R_{cd}(As) / R_{cd}(Na) \quad \text{.....(15)}$$

Accordingly we will have better sensitivity for arsenic in a sample where the dominant radionuclide in the gamma spectrum of the specific foodstuff is caused by sodium, to irradiate under the cadmium-shielded part of neutron spectrum only if:

$A^{As:Na} < 1$

Obviously, it will be more advantageous to irradiate under the full neutron spectrum if:

$A^{As:Na} > 1$

Note: As shown from the results (Table 4; section 6.1) there were many other dominant nuclides observed from the various food-spectra previously studied [10]. Hence screening of the predominant nuclides is necessary for individual matrices to optimize element sensitivities where applicable.

5.5 Neutron Flux and Spectrum Variations

Flux density and energy distribution are mostly important in reactors where activation analysis is carried out[14] Flux density in research reactors varies from about 10^{10} to 10^{15} $n.cm^{-2}.s^{-1}$ in smaller reactors (30 kW) up to high flux reactors (20 MW) respectively. As sensitivity of activation analysis is proportional to the flux density [equations (7) and (8)] neutron fluxes of about 10^{14} $n.cm^{-2}s^{-1}$ are required for the determination of very low element concentrations in the order of part per billion (ppb) to parts per million (ppm). Furthermore, the neutron flux should be uniform within a certain irradiation position in the reactor to avoid induced activity differences between samples and comparator element standards. The ratio of epi-thermal to thermal neutrons should preferably be fairly constant, especially if advantage factors are to be taken into consideration (section 5.4). High epi-thermal fluxes are of interest if activation of certain elements by resonance neutrons is required (e.g. ^{58}Ni (n,p) ^{58}Co, see Table 2).

High neutron flux variations are generated by the removal of regulating rods, which causes sudden substantial increases in neutron flux, which is reduced to a very small value (about 10 $n.cm^{-2}.s^{-1}$), because of the self-regulating properties of the reactor [15,12]

As described in section 3.2 the neutron flux and spectral variations have been monitored for the duration of about four months, during which special attention was paid to cover at least one major fuel element re-loading of the SAFARI-1 reactor. As we wanted to evaluate the thermal and epi-thermal flux variations, one needs to have elements that are preferably sensitive to either thermal or epi-thermal neutrons. For the RINGAS system element standards of uranium and thorium were used, which were irradiated alternatively in the cadmium-shielded and bare irradiation positions dedicated to INAA, followed by delayed neutron counting. Previous work [6] showed that the uranium ^{235}U(n.f)-reaction (neutron induced fission) is specifically sensitive to thermal neutrons, while the ^{232}Th(n,f) is mainly caused by epi-thermal and fast neutrons.

For the PRS system we used a number of iron element standards divided over the length of the irradiation capsule (≈ 10 cm) to evaluate the special distribution and flux variations of this irradiation channel. The ^{58}Fe(n,γ)^{59}Fe reaction and the ^{54}Fe(n,p)^{54}Mn reaction were used for the evaluation of the thermal and epi-thermal/fast flux variations respectively.

5.6 Evaluation of Neutron Flux and Spectral Stability

5.6.1 The RINGAS system

In addition to Chapter 4, one can also write the equation used for instrumental neutron activation in its extended form as [5]:

$$C_t = \gamma \cdot \left(\frac{a_b \cdot N_{Av}}{M}\right) \cdot K \cdot \left[\int_0^{\infty} \sigma(E) \cdot \frac{\varphi(E)}{K} \cdot dE\right] \cdot \left(\frac{A_p}{50 \cdot t_m}\right) \cdot (1 - e^{-\lambda t_1}) \cdot e^{-\lambda t_2} \cdot a_d \cdot \left[\frac{1 - e^{-\lambda t_m / (1 - \overline{D})}}{\lambda t_m / (1 - \overline{D})}\right] \cdot f^* \cdot G \cdot t_m$$

where,

C_t	=	Net peak area	[-]
γ	=	Sample-detector geometry factor	[-]
a_b	=	Isotopic abundance	[-]
N_{Av}	=	Avogadro's number	[Mol^{-1}]
M	=	Atomic Mass	[g]
K	=	Average neutron flux	[$cm^{-2} . s^{-1} . keV^{-1}$]
$\sigma(E)$	=	Cross-section per energy interval	[cm^2]
$\varphi(E)$	=	Neutron flux per energy-interval	[$cm^{-2} . s^{-1} . keV^{-1}$]
A_p	=	Pulser peak area	[-]
t_m	=	Measuring time	[s]
λ	=	Decay constant	[s^{-1}]
t_1	=	Irradiation time	[s]
t_2	=	Decay time	[s]
a_d	=	Abundance of the γ-ray	[-]
$\overline{D}$	=	Average total dead-time	[-]
f*	=	True element concentration in the sample	[$g . g^{-1}$]
G	=	Sample weight	[g]

$\left[\int_0^{\infty} \sigma(E) \cdot \frac{\varphi(E)}{K} \cdot dE\right]$ = Normalized neutron spectrum

From this it will be clear that the only really unknown parameter in comparative neutron activation analysis is the normalized neutron spectrum. If samples are irradiated under the full or cadmium-shielded part of the neutron spectrum one can divide the entire spectrum in two regions (see paragraph 5.2), i.e.

- Full spectrum — thermal, epi-thermal and fast neutrons
- Cadmium-shielded part — epi-thermal and fast neutrons

Equations (11) and (12) can then be rewritten as:

$$(A_t)^{cd} = [\phi^{epi}.\sigma^{epi} + \phi^{fast}.\sigma^{fast}] . N_0^{cd} . (1-e^{-\lambda t_1})^{cd} . (e^{-\lambda t_2})^{cd} \quad .....(16)$$

and

$$(A_t)^{ncd} = [\phi^{thermal}.\sigma^{thermal} + \phi^{epi}.\sigma^{epi} + \phi^{fast}.\sigma^{fast}] . N_0^{ncd} . (1-e^{-\lambda t_1})^{ncd} . (e^{-\lambda t_2})^{ncd} \quad .....(17)$$

For equal number of atoms, irradiation and decay times the cadmium ratio (equation 13) can then be defined as:

$$R_{cd} = (A_t)^{ncd} / (A_t)^{cd} = 1 + \frac{[\phi^{thermal}.\sigma^{thermal}]}{[\phi^{epi}.\sigma^{epi} + \phi^{fast}.\sigma^{fast}]} \quad .....(18)$$

For a research reactor with a stable neutron flux and spectrum the cadmium ratios should be constant and accordingly also the advantage factors. For the Safari-1 research reactor one would expect that the cadmium ratios over a short period (e.g. one day) will be

constant if no specific reloads or irradiation of molybdenum target plates occur. However, prior to and after such activities one may expect substantial differences in cadmium ratios and advantage factors, which provides an opportunity to evaluate the flux density and stability in the irradiation channels used for neutron activation analysis and the influences on the analytical results.

From equation (18) one now can define for two elements (x and y) the following relation:

$$\frac{[R_{cd}-1]^x}{[R_{cd}-1]^y} = \frac{[\phi^{thermal}.\sigma^{thermal}]^x}{[\phi^{thermal}.\sigma^{thermal}]^y} \cdot \frac{[\phi^{epi}.\sigma^{epi}+\phi^{fast}.\sigma^{fast}]^y}{[\phi^{epi}.\sigma^{epi}+\phi^{fast}.\sigma^{fast}]^x} \quad .....(19)$$

If the neutron spectrum between irradiations does not change the ratio above should be a constant, even when the neutron density is changed due to different power requirements of the reactor at the time of irradiation (the reactor may be operated at different power levels to accommodate the production requirements of fission molybdenum). However, if the neutron spectrum changes this ratio will be influenced and accordingly the variation in the ratio will provide a means to monitor the spectral stability at the irradiation positions used for neutron activation analysis.

5.6.2 The PRS system

As the PRS system is located outside the core and is surrounded by water the neutron spectrum is already well thermalized. An elegant solution to measure the spectral stability is using iron monitors and compare the ratios of the following neutron induced reactions:

Thermal neutrons	^{58}Fe (n,γ) ^{59}Fe
Epi-thermal neutrons	^{54}Fe (n,p) ^{54}Mn

Accordingly, the ratio between ^{59}Fe and ^{54}Mn will be a good value to monitor the spectrum stability, while the specific activity (Bq/g) of ^{59}Fe and ^{54}Mn provide a good indication of the power stability.

5.7 The South African Fundamental Atomic Reactor Installation 1 (SAFARI-1)

5.7.1 General information

Reactor type	Tank type, fully enriched (90%) uranium, light water moderated and cooled, beryllium reflected.
Purpose	Basic research, engineering tests, isotope production, fuel element development
Nominal power design	20 MW, initially 6.67 MW cooling capacity
Location	Pelindaba site, 30 km west of Pretoria, South Africa
Owner and operator	South African Nuclear Energy Corporation Ltd., originally the South African Atomic Energy Board [14]

5.7.2 Reactor physics

Neutron energy and lifetime: Thermal lifetime 6.9×10^{-5}sec, mean approximation is 10^{-4} s

Neutron flux Calculated in neutrons . cm^{-2} . sec^{-1} [14]

	6.67MW	20MW
Thermal (average)	5.0×10^{13}	1.8×10^{14}
Thermal (maximal)	1.3×10^{14}	4.0×10^{14}

5.7.3 Irradiation positions used for INAA at Safari-1

At Necsa's Safari-1 research reactor there are basically two irradiation facilities that can be used for neutron activation analysis. They are schematically indicated in Figure 1 and can be described as:

- **RINGAS** (Routine Instrumental Neutron and Gamma Analysis System)

Fitted with two irradiation positions located at two separated core positions of the reactor. One of these is lined with cadmium, which captures the thermal neutrons and accordingly samples and standards are irradiated mainly with epi-thermal and fast neutrons. This irradiation channel is positioned just above the core of the reactor since cadmium is not allowed inside the core, which would disturb the flux distribution of the core in an unacceptable way. In the other positions the full neutron energy spectrum is available for irradiation. As there are two positions one can not expect the neutron spectrum to be the same for both positions and thus no true cadmium-ratio (R_{cd}) can be determined for individual elements. This system is mainly used for short and intermediate-lived nuclides with half-lives between 11 seconds and 30 hours. Sample volumes are restricted to about 1ml which may limit the sensitivity of the analysis. [19]

- **PRS** (Pneumatic Rabbit System)

This irradiation position is located just outside the core of the reactor, inside the reactor pool. Due to its position, the total neutron flux will be lower than at the RINGAS irradiation position and the neutron spectrum will be more thermalized due the moderating effect of the pool-water between the core and the irradiation position. To determine the true cadmium value (R_{cd}) one has to use cadmium containers to shield the samples and standards from thermal neutrons. The disadvantage of this system is that the irradiation capsules have to be loaded and unloaded manually and the relative large amount of cadmium will cause high levels of activity as the cadmium itself is activated as well by neutrons. This system is mainly used for long-lived nuclides with half-lives larger than 30 hours. Sample volumes, routinely used, are also about 1ml to allow standardization of counting equipment. About ten samples can be irradiated at a time, which requires flux monitoring due to the special distribution of samples and element standards. However, single sample volumes of up to 20ml could be applied, which offers the opportunity to improve the sensitivity of element analysis in amongst others foodstuffs. [19]

- **The neutron energy and neutron flux at the irradiation positions**

Typical neutron fluxes neutron fluxes at 20 MW operational power are

RINGAS

Bare tube position

$1.46\text{-}1.73 \cdot 10^{14}$ neutrons/cm^2/s of epi-thermal and fast neutrons

$1.20\text{-}1.27 \cdot 10^{14}$ neutrons/cm^2/s of thermal neutrons

Cadmium shielded position

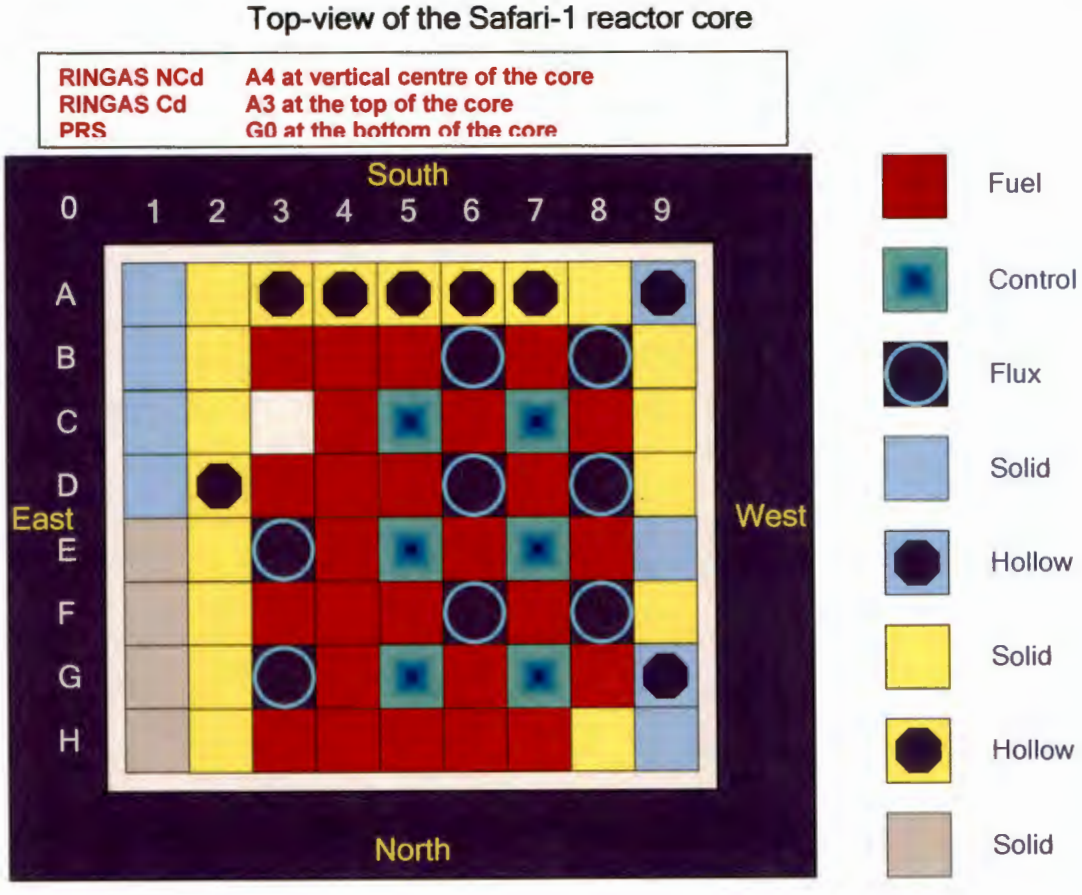
$\approx 5.3 \cdot 10^{13}$ neutrons/cm^2/s of epi-thermal and fast neutrons

PRS $\approx 3.9 \cdot 10^{13}$ neutrons/cm^2/s of thermal neutrons

As indicated above the neutrons are well moderated in this position. No epi-thermal flux-data are available at this point in time. However, where possible, we will determine the true R_{cd}-values for the elements of interest to evaluate the applicability

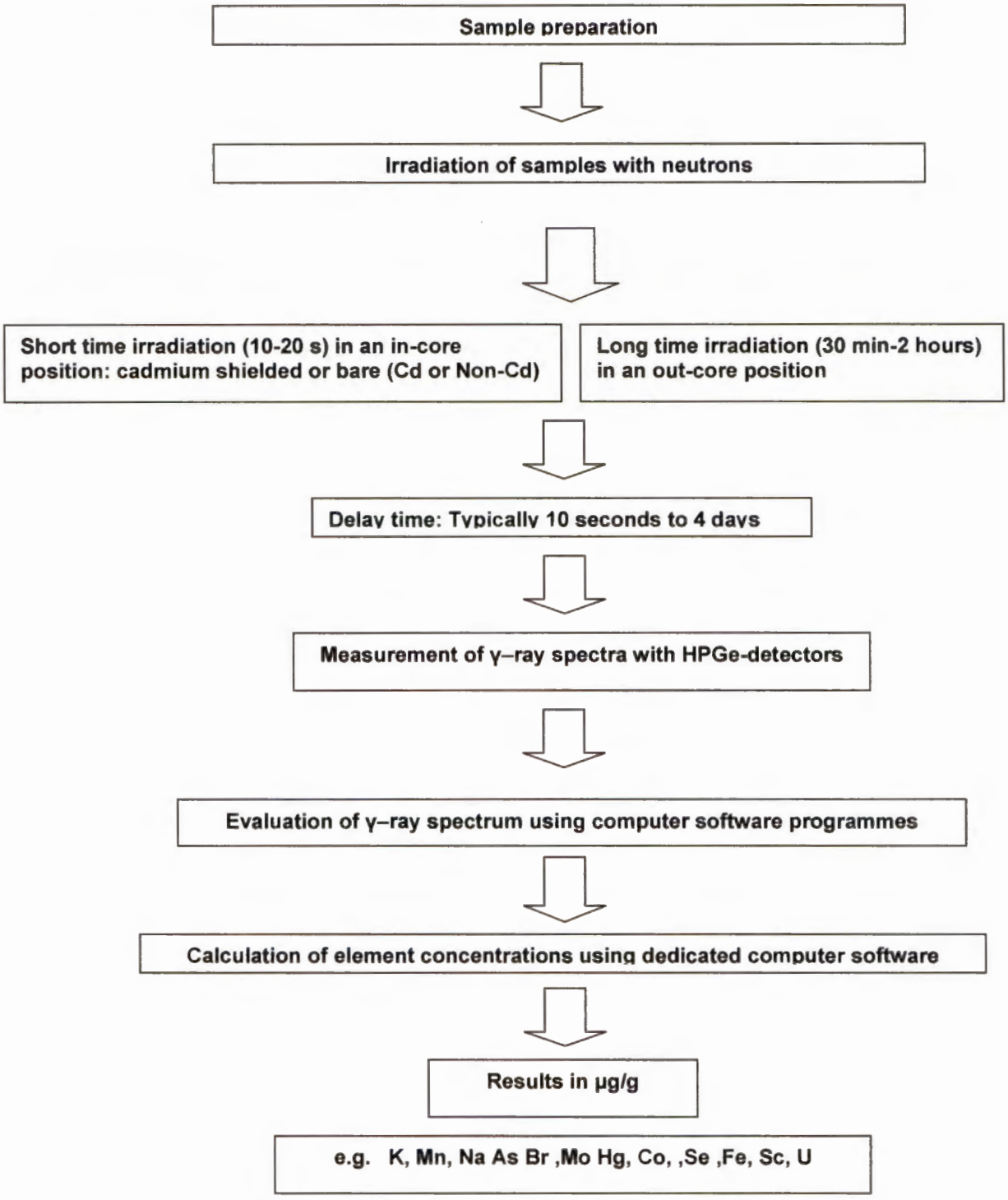
in food analysis, especially as larger volumes can be applied to improve the analysis sensitivity.

Figure 1 Schematic representation of the Safari-1 reactor core configuration



5.7.4 Experimental procedure

The overview of routine instrumental neutron activation analysis procedure applied at Necsa is presented in the figure below.



6 RESULTS DISCUSSION

6.1 Evaluation of previous work done

- Section 2 (Table 2) presents all the elements analyzed, their half-lives, nuclear reactions and γ-ray energies applied in the analyses of foodstuffs.

- Table 4 presents previously analyzed foodstuffs according to the respective applied irradiation-delay-counting time-schemes (IDC) and the predominant radionuclides as extracted from the spectral data available from previous research [13]. It is reflected that for each IDC there are a number of nuclides that predominate.

Table 4 Predominant radionuclides

	Dominant Radionuclides											
Foodstuff	**RINGAS Ti/Td/Tm = 10s/30s/120s**			**RINGAS Ti/Td/Tm = 10-20s/3h/15-30m**			**RINGAS Ti/Td/Tm = 10-20s/30h/15-30m**			**PRS Ti/Td/Tm = 30m/7d/30-60m**		
Brown bread	Cl	Al		Na	Mn		Na			Na		
Cabbage	Br	Cu	Al	K	Na	Mn	Na	K	Br	Na		
Chicken	Cu	Br	Al	Na	K		Na	K	Br	Na		
Coffee	Br	Al		K	Na	Mn	Na	K	Br	Na		
Eggs	Br	Al	Cl	Br	Na		Na	Br		Na	Mo	
Maize	-	-	-	Br	Mn	K	Br	K		-	-	-
Margarine	Br	Cl	Na	Na			Na			Na	Cr	
Milk	Na	Br	Cu	K	Na		Na	K		Na	Cr	
Onion	Mn	Br	Cu	-	-	-	Na	K	Br			
Potatoes	Cu	Br	Cl	Na	Mn	Br	Na	K	Br			
Rice	Mn	Br	Cu	Mn			K	Br		La	K	
Spinach	Mn	Na	Al	Na	Mn		Na	Br		Na		
Sugar	Cu	Br	Al	Mn			Na			Cr		
Tea	Mn	Al		Mn	K		Na	K		Sc	Co	Na
Tomatoes	Cu	Br	Cl	K	Na	Mn	-	-	-	-	-	-
White bread	Cl	Al		Na	Mn		Na			Sc	Na	

From this it can be observed that the major predominant nuclides are from the following elements: Na, Br, K, Mn, Al, Cu, Cl, Cr, Sc, Mo, La and Co.

- Previous unpublished work [19] performed some two decades ago when the reactor was still operated at 5 MW also provided results that could be used as a first order insight in the cadmium ratios and advantage factors of a suite of elements. These results are shown in Table 5 and Table 6 respectively. It should be noted that the advantage factors are also to a certain extent energy dependent, which is not taken into account in Table 6.

- Accordingly, a first order estimate of a potential improvement in the sensitivity for the elements of interest according to Table 2 could be obtained. This evaluation is presented in Table 7.

Table 5 Previous results on elements irradiated in the RINGAS system

Element	Mass (µg)	Reaction	Isotope	E (keV)	Decay time 4 hours								Decay time 40 hours								R_{Cd}-values							
					Specific Activities (Bq/µg)																							
					Cd				NCd				Cd				NCd											
					X	Sx (%)	% Dt	FM	X	Sx (%)	% Dt	FM	X	Sx (%)	% Dt	FM	X	Sx (%)	% Dt	FM	X	Sx (%)	X	Sx (%)	X(avg)	Sx	**X(avg)**	**Sx**
Al	10	n,γ	28Al	1779																								
		n,α	24Na	1369	**0.298**	2			**0.0533**	2			**0.296**	2			**0.0617**				5.37	2.83	6.25	2.00	5.81	0.44	**5.81**	**0.44**
As	10	n,γ	76As	559	**46.3**	0.4	2.33	57.1	**6.44**	1.1	0.67	147.5	**46**	0.3	0.83	57.1	**6.49**	0.7	0.27	147.5	4.17	1.17	4.23	0.76	4.20	0.03	**4.35**	**0.12**
				657	**32.2**	1.8	2.33	57.1	**4.64**	5.4	0.67	147.5	**31.4**	1.3	0.83	57.1	**4.56**	2.9	0.27	147.5	4.32	5.69	4.36	3.18	4.34	0.02		
				1216	**124**	2.3	2.33	57.1	**17.5**	6.1	0.67	147.5	**121**	1.7	0.83	57.1	**19.2**	3.4	0.27	147.5	4.23	6.52	4.76	3.80	4.50	0.26		
Au	0.5	n,γ	198Au	412	**1890**	1.1	0.5	57.1	**264**	0.4	0.17	147.5	**1900**	0.6	0.43	57.1	**268**	0.2	0.17	147.5	4.19	1.17	4.23	0.63	4.21	0.02	**4.21**	**0.02**
Ba	10000	n,γ	135mBa	268	**1.8**	3.1	8.35	57.1	**0.115**	0.4	14.5	147.5	**1.88**	0.7	1.17	57.1	**0.13**	0.2	0.27	147.5	1.92	3.13	2.07	0.73	2.00	0.08	**2.00**	**0.08**
		n,γ	139Ba	166	**58.9**	0.1	8.35	57.1	**48**	0.1	14.5	147.5			1.17	57.1			0.27	147.5	24.45	0.14			24.45	0.03	**24.45**	**0.03**
		n,γ	131Ba	1048			8.35	57.1			14.5	147.5	**0.002**	2	1.17	57.1	**0.000**	2	0.27	147.5			4.14	2.83	4.14	0.12	**4.14**	**0.12**
Br	50	n,γ	82Br	554	**48.6**	1.4	12.02	57.1	**6.31**	0.5	2.34	147.5	**50.6**	0.8	4.17	57.1	**6.26**	0.3	0.7	147.5	3.90	1.49	3.71	0.85	3.80	0.09	**3.83**	**0.02**
				619	**45.5**	1.4	12.02	57.1	**5.78**	1	2.34	147.5	**47.4**	1.2	4.17	57.1	**6.13**	0.5	0.7	147.5	3.81	1.72	3.88	1.30	3.85	0.03		
				777	**62.4**	1.4	12.02	57.1	**8.22**	0.6	2.34	147.5	**64.7**	0.9	4.17	57.1	**8**	0.3	0.7	147.5	3.95	1.52	3.71	0.95	3.83	0.12		
Ca	10000	n,γ	47Sc	159	**6E-05**	2		52.2	**4E-05**	2		151.3				52.2				151.3	21.93	2.83			21.93	0.62	**21.93**	**0.62**
Cd	250	n,γ	115mIn	336	**3.99**	2.2	1.34	57.1	**0.34**	0.6	0.67	147.5	**7.78**	0.6	0.47	57.1	**0.936**	0.2	0.13	147.5	2.56	2.28	3.61	0.63	3.08	0.53	**3.44**	**0.25**
		n,γ	115Cd	492	**5.09**	4.8	1.34	57.1	**0.633**	1.4	0.67	147.5	**5.06**	2.1	0.47	57.1	**0.586**	0.8	0.13	147.5	3.73	5.00	3.47	2.25	3.60	0.13		
				528	**6.32**	2.2	1.34	57.1	**0.767**	0.7	0.67	147.5	**6.41**	1.1	0.47	57.1	**0.777**	0.4	0.13	147.5	3.64	2.31	3.64	1.17	3.64	0.00		
Ce	10000	n,γ	141Ce	145	**0.121**	0.4	5.51	57.1	**0.121**	0.3	2.83	147.5	**0.124**	0.2	1.77	57.1	**0.146**	0.1	1.4	147.5	30.00	0.50	35.32	0.22	32.66	2.66	**32.66**	**2.66**
		n,γ	143Ce	293	**0.499**	0.4	5.51	57.1	**0.333**	0.3	2.83	147.5	**0.507**	0.2	1.77	57.1	**0.334**	0.2	1.4	147.5	20.02	0.50	19.76	0.28	19.89	0.13	**19.89**	**0.13**
		n,γ	140La	329	**0.407**	0.4	5.51	57.1	**0.319**	0.3	2.83	147.5	**0.413**	0.2	1.77	57.1	**0.27**	0.2	1.4	147.5	23.51	0.50	19.61	0.28	21.56	1.95	**19.86**	**1.21**
				816			5.51	57.1			2.83	147.5	**0.0021**	2	1.77	57.1	**0.0013**	2	1.4	147.5			19.18	2.83	19.18	0.54		
				1596			5.51	57.1			2.83	147.5	**0.0038**	2	1.77	57.1	**0.0024**	2	1.4	147.5			18.84	2.83	18.84	0.53		
Co	10	n, α	56Mn	846	**0.222**	2		57.1	**0.0947**	2		147.5				57.1				147.5	12.80	2.83			12.80	0.36	**12.80**	**0.36**
Cs	2.5	n,γ	134mCs	127	**1400**	1.3	0.33	57.1	**215**	0.6	0.33	147.5			0.07	57.1			0.1	147.5	4.61	1.43			4.61	0.07	**4.61**	**0.07**
		n,γ	134Cs	605									**1.98**	2	0.07	57.1	**0.501**	2	0.1	147.5			7.59	2.83	7.59	0.21	**8.15**	**0.56**
				796									**2.39**	2	0.07	57.1	**0.694**	2	0.1	147.5			8.71	2.83	8.71	0.25		

Table 5 Previous results on elements irradiated in the RINGAS system (continued)

Element	Mass (µg)	Reaction	Isotope	E (keV)	Decay time 4 hours								Decay time 40 hours								R_{Cd}-values					
					Specific Activities (Bq/µg)																					
					Cd				NCd				Cd				NCd									
					X	Sx (%)	% Dt	FM	X	Sx (%)	% Dt	FM	X	Sx (%)	% Dt	FM	X	Sx (%)	% Dt	FM	X	Sx (%)	X	Sx (%)	X(avg)	Sx	X(avg)	Sx
Cu	100	n,γ	64Cu	511	**54.5**	0.4	1.33	57.1	**38.4**	0.4	1.33	147.5	**54.8**	0.5	0.17	57.1	**38.4**	0.5	0.3	147.5	21.14	0.57	21.02	0.71	21.08	0.06	**20.75**	**0.33**
				1346	**245**	6	1.33	57.1	**177**	4.2	1.33	147.5	**269**	5.6	0.17	57.1	**172**	5.5	0.3	147.5	21.67	7.32	19.18	7.85	20.43	1.25		
Dy	0.5	n,γ	165Dy	95	**16.5**	0.8	0.33	57.1	**29.4**	0.5	0.67	147.5			0.1	57.1			0.07	147.5	53.45	0.94			53.45	0.50	**50.64**	**2.82**
				362	**9.16**	5.1	0.33	57.1	**14.6**	3	0.67	147.5			0.1	57.1			0.07	147.5	47.82	5.92			47.82	2.83		
Er	50	n,γ	171Er	112	**28.5**	1.1	2.33	57.1	**8.54**	0.8	0.67	147.5	**28.7**	2.7	0.2	57.1	**8.56**	2.1	0.13	147.5	8.99	1.36	8.95	3.42	8.97	0.02	**9.18**	**0.16**
				296	**51.3**	1.1	2.33	57.1	**15.6**	0.7	0.67	147.5	**52.8**	2.5	0.2	57.1	**16.4**	2.1	0.13	147.5	9.12	1.30	9.32	3.26	9.22	0.10		
				308	**34.7**	0.8	2.33	57.1	**10.6**	0.5	0.67	147.5	**34.3**	1.9	0.2	57.1	**10.9**	1.6	0.13	147.5	9.16	0.94	9.53	2.48	9.35	0.18		
		n,p	167Ho	347	**17.6**	0.4	2.33	57.1	**4.17**	0.3	0.67	147.5			0.2	57.1			0.13	147.5	7.11	0.50			7.11	0.04	**7.11**	**0.04**
Eu	1	n,γ	152Eu	122	**5870**	0.4	3.83	57.1	**5810**	0.4	3.67	147.5	**6410**	0.9	0.27	57.1	**6050**	0.7	0.3	147.5	29.69	0.57	28.32	1.14	29.00	0.69	**29.52**	**0.40**
				841	**6780**	1.1	3.83	57.1	**6790**	1.1	3.67	147.5	**6820**	2.5	0.27	57.1	**6800**	1.8	0.3	147.5	30.04	1.56	29.91	3.08	29.98	0.07		
				963	**23300**	0.7	3.83	57.1	**23200**	0.8	3.67	147.5	**24500**	1.6	0.27	57.1	**23900**	1.2	0.3	147.5	29.87	1.06	29.27	2.00	29.57	0.30		
Fe	25000	n,p	56Mn	846	**0.672**	2		57.1	**0.0414**	2		147.5				57.1				147.5	1.85	2.83			1.85	0.05	**1.85**	**0.00**
				1811	**2.96**	2		57.1	**0.182**	2		147.5				57.1				147.5	1.84	2.83			1.84	0.05		
Ga	5	n,γ	72Ga	630	**106**	5.5	2	56.2	**25.7**	2.2	0.83	145.1	**120.8**	2	0.37	56.2	**26.4**	4.5	0.17	145.1	7.27	5.92	6.56	4.92	6.91	0.36	**6.78**	**0.13**
				834	**179**	2.1	2	56.2	**42**	0.9	0.83	145.1	**202**	0.8	0.37	56.2	**42.2**	1.8	0.17	145.1	7.04	2.28	6.27	1.97	8.65	0.39		
Ge	920	n,γ	77Ge	211	**1.34**	9.8	21.54	56.2		1.8	17.53	145.1	**1.578**	1.8	3.77	56.2	**0.1886**	10.1	3.67	145.1			3.59	10.26	3.59	0.37	**3.34**	**0.24**
				216	**1.37**	10.2	21.54	56.2		1.9	17.53	145.1	**1.586**	1.9	3.77	56.2	**0.164**	10.4	3.67	145.1			3.10	10.57	3.10	0.33		
		n,γ	79Ge	199	**142**	2.3	21.54	56.2	**82.5**	2.9	17.53	145.1			3.77	56.2			3.67	145.1	17.43	3.70			17.43	0.65	**16.04**	**1.39**
				264	**176**	0.4	21.54	56.2	**86**	2.9	17.53	145.1			3.77	56.2			3.67	145.1	14.66	2.93			14.66	0.43		
Gd	230	n,γ	159Gd	364	**83**	0.9	3.17	48.6	**8.37**	0.3	0.5	142.6	**84.4**	0.3	0.87	48.6	**8.42**	0.7	0.2	142.6	3.03	0.95	2.99	0.76	3.01	0.02	**3.01**	**0.02**
		n,γ	161Tb	75	**0.8**	0.5	3.17	48.6	**0.232**	0.3	0.5	142.6	**1.04**	2	0.87	48.6	**0.281**	2	0.2	142.6	8.70	0.58	8.11	2.83	8.40	0.30	**8.40**	**0.30**
Hf	970	n,γ	180mHf	215	**4.04**	1.1	10.52	56.2	**0.686**	0.4	2.17	145.1	**4.44**	2.6	1.3	56.2	**0.636**	6.9	0.6	145.1	5.09	1.17	4.30	7.37	4.70	0.40	**4.57**	**0.22**
				332	**1.14**	2.3	10.52	56.2	**0.177**	1.6	2.17	145.1	**1.29**	4.8	1.3	56.2	**0.1958**	15.7	0.6	145.1	4.66	2.80	4.55	16.42	4.61	0.05		
				443	**0.953**	2.6	10.52	56.2	**0.172**	2	2.17	145.1	**1.084**	6.1	1.3	56.2	**0.1506**	12.5	0.6	145.1	5.41	3.28	4.17	13.91	4.79	0.62		
				501	**7.48**	3.2	10.52	56.2	**1.05**	1.8	2.17	145.1	**7.88**	2	1.3	56.2	**1.102**	2	0.6	145.1	4.21	3.67	4.20	2.83	4.20	0.01		
		n,γ	181Hf	133	**0.612**	0.7	10.52	56.2	**0.26**	0.9	2.17	145.1	**0.616**	0.5	1.3	56.2	**0.234**	0.4	0.6	145.1	12.75	1.14	11.40	0.64	12.07	0.67	**11.99**	**0.08**
				482	**0.804**	1.2	10.52	56.2	**0.3**	1.2	2.17	145.1	**0.611**	0.4	1.3	56.2	**0.257**	0.5	0.6	145.1	11.19	1.70	12.62	0.64	11.91	0.71		

Table 5 Previous results on elements irradiated in the RINGAS system (continued)

Element	Mass (µg)	Reaction	Isotope	E (keV)	Decay time 4 hours								Decay time 40 hours								R_{Cd}-values							
					Specific Activities (Bq/µg)																							
					Cd				NCd				Cd				NCd											
					X	Sx (%)	% Dt	FM	X	Sx (%)	% Dt	FM	X	Sx (%)	% Dt	FM	X	Sx (%)	% Dt	FM	X	Sx (%)	X	Sx (%)	X(avg)	Sx	**X(avg)**	**Sx**
Hg	100	n,γ	197mHg	134	**0.229**	4	0.17	48.6	**0.409**	2.9	0.67	142.6	**0.232**	3.7	0.17	48.6	**0.461**	3.2	0.3	142.6	53.58	4.94	59.61	4.89	56.60	3.02	**57.34**	**0.67**
		n,γ	197Hg	68	**1.1**	2.7	0.17	48.6	**2.12**	3.3	0.67	142.6	**1.09**	1.8	0.17	48.6	**2.13**	3.5	0.3	142.6	57.82	4.26	58.62	3.94	58.22	0.40		
				78	**0.954**	1.3	0.17	48.6	**1.83**	1	0.67	142.6	**0.981**	2.6	0.17	48.6	**1.86**	0.7	0.3	142.6	57.55	1.64	56.88	2.69	57.21	0.33		
Ho	500	n,γ	166Ho	81	**16.3**	2.1	1.84	56.2	**2.9**	0.3	0.33	145.1	**17.72**	0.2	0.63	56.2	**2.9**	0.4	0.23	145.1	5.34	2.12	4.91	0.45	5.12	0.21	**5.37**	**0.24**
				1379	**44.5**	8.5	1.84	56.2	**8.6**	3.5	0.33	145.1	**47.6**	2.1	0.63	56.2	**8.6**	5.1	0.23	145.1	5.80	9.19	5.42	5.52	5.61	0.19		
In	20	n,γ	116mIn	138	**153000**	0.4	71	51.4	**24300**	0.4	45.5	145.3			0.07	51.4			0.1	145.3	4.76	0.57			4.76	0.03	**4.85**	**0.05**
				417	**118000**	0.5	71	51.4	**19200**	0.2	45.5	145.3			0.07	51.4			0.1	145.3	4.88	0.54			4.88	0.03		
				1097	**333000**	0.4	71	51.4	**54500**	0.4	45.5	145.3			0.07	51.4			0.1	145.3	4.91	0.57			4.91	0.03		
				1293	**365000**	0.4	71	51.4	**59000**	0.2	45.5	145.3			0.07	51.4			0.1	145.3	4.85	0.45			4.85	0.02		
Ir	5	n,γ	194Ir	294	**1490**	3.3	2.17	56.2	**234**	2	0.5	145.1	**1460**	1.6	0.7	56.2	**263**	2.5	0.27	145.1	4.71	3.86	5.40	2.97	5.06	0.35	**5.24**	**0.18**
				329	**2760**	0.7	2.17	56.2	**500**	0.3	0.5	145.1	**2790**	0.3	0.7	56.2	**502**	0.6	0.27	145.1	5.43	0.76	5.40	0.67	5.42	0.02		
K	10000	n,γ	42K	1525	**14.7**	0.4	12.33	56.2	**13.4**	0.5	15.53	145.1	**15.2**	0.6	1.57	56.2	**13.6**	0.4	2.77	145.1	27.35	0.64	26.84	0.72	27.09	0.25	**27.09**	**0.25**
La	100	n,γ	140La	329	**18**	0.9	4	56.2	**14**	0.8	3.17	145.1	**17.7**	0.5	2.03	56.2	**13.9**	0.5	1.8	145.1	23.33	1.20	23.56	0.71	23.45	0.11	**23.33**	**0.12**
				487	**20.8**	0.6	4	56.2	**15.9**	0.5	3.17	145.1	**20.3**	0.3	2.03	56.2	**15.8**	0.4	1.8	145.1	22.93	0.78	23.35	0.50	23.14	0.21		
				816	**60.2**	0.8	4	56.2	**46.8**	0.7	3.17	145.1	**60**	0.5	2.03	56.2	**47**	0.5	1.8	145.1	23.32	1.06	23.50	0.71	23.41	0.09		
				1596	**94.8**	0.7	4	56.2	**74**	0.7	3.17	145.1	**94.8**	0.4	2.03	56.2	**73.3**	0.4	1.8	145.1	23.42	0.99	23.20	0.57	23.31	0.11		
Lu	10	n,γ	176mLu	88	**8980**	0.3	7.01	50.3	**1030**	0.2	2	145.8	8492	0.5	0.27	50.3	974	0.5	0.37	145.8	3.44	0.36	3.44	0.71	3.44	0.00	**3.44**	**0.00**
		n,γ	177Lu	113	**23.7**	4.2	7.01	50.3	**37.7**	4.2	2	145.8	38.6	0.7	0.27	50.3	29.6	0.4	0.37	145.8	47.72	5.94	23.01	0.81	35.36	12.36	**33.40**	**1.97**
				208	**84.7**	1.9	7.01	50.3	**113**	1.9	2	145.8	113	0.6	0.27	50.3	86	0.5	0.37	145.8	40.02	2.69	22.83	0.78	31.43	8.60		
Mg	100	n,γ	24Na	1369	**0.685**	2		52.2	**0.0724**	2		151.3				52.2				151.3	3.17	2.83			3.17	0.09	**3.17**	**0.09**
Mn	8.65	n,γ	56Mn	847	**2760**	0.2	4	50.3	**2320**	0.3	8.68	145.8									25.22	0.36			25.22	0.09	**24.95**	**0.27**
				1811	**12400**	0.8	4	50.3	**10200**	1.2	8.68	145.8									24.68	1.44			24.68	0.36		
Mo	50	n,γ	99mTc	141	**1.89**	4.7	0.05	50.3	**0.0882**	0.9	0.17	145.8	4.08	0.3	0.2	50.3	0.369	1	0.3	145.8	1.40	4.79	2.71	1.04	2.06	0.66	**2.06**	**0.66**
Na	500	n,γ	24Na	1369	**51.1**	0.3	12.19	50.3	**52.7**	0.3	19.03	145.8	51.5	0.3	2.53	50.3	53.3	0.2	4	145.8	30.94	0.42	31.05	0.36	30.99	0.05	**30.99**	**0.05**

Table 5 Previous results on elements irradiated in the RINGAS system (continued)

Element	Mass (µg)	Reaction	Isotope	E (keV)	Decay time 4 hours								Decay time 40 hours								R_{Cd}-values					
					Specific Activities (Bq/µg)																					
					Cd				NCd				Cd				NCd									
					X	Sx (%)	% Dt	FM	X	Sx (%)	% Dt	FM	X	Sx (%)	% Dt	FM	X	Sx (%)	% Dt	FM	X	Sx (%)	X	Sx (%)	X(avg)	Sx	**X(avg)**	**Sx**
Nd	1250	n,γ	149Nd	114	**18.5**	0.7	9.67	50.3	**4.62**	0.8	7.01	145.8			1.17	50.3			0.6	145.8	7.49	1.06			7.49	0.08	**7.40**	**0.19**
				211	**103**	0.5	9.67	50.3	**26**	0.3	7.01	145.8			1.17	50.3			0.6	145.8	7.57	0.58			7.57	0.04		
				270	**117**	1.7	9.67	50.3	**27.8**	0.5	7.01	145.8			1.17	50.3			0.6	145.8	7.13	1.77			7.13	0.13		
		n,γ	151Pm	240			9.67	50.3			7.01	145.8	4.4	3.1	1.17	50.3	0.573	8.9	0.6	145.8			3.91	9.42	3.91	0.37	**4.11**	**0.20**
				340	**2.71**	1.5	9.67	50.3	**0.395**	2.9	7.01	145.8	2.77	0.7	1.17	50.3	0.391	1.9	0.6	145.8	4.37	3.26	4.23	2.02	4.30	0.07		
Ni	27500	n,γ	65Ni	366	**1.18**	0.8	3.51	59.5	**1.16**	0.6	4.51	154.7			1.03	59.5			0.17	154.7	29.49	1.00			29.49	0.29	**29.32**	**0.26**
				1115	**3.12**	0.7	3.51	59.5	**3.07**	0.5	4.51	154.7			1.17	59.5			0.6	154.7	29.52	0.86			29.52	0.25		
				1482	**6.86**	0.5	3.51	59.5	**6.62**	0.4	4.51	154.7			1.17	59.5			0.6	154.7	28.95	0.64			28.95	0.19		
		n,γ	58Co	511	**0.0144**	6.3	3.51	59.5			4.51	154.7	0.0176	0.7	1.17	59.5	0.0011	2.8	0.6	154.7	0.00	6.30	1.81	2.89	1.81	0.05	**1.76**	**0.05**
				811	**0.0642**	0.6	3.51	59.5	**0.0037**	5.1	4.51	154.7	0.0805	0.2	1.17	59.5	0.0045	0.9	0.6	154.7	1.73	5.14	1.69	0.92	1.71	0.02		
Pd	5	n,γ	109Pd	88	**409**	0.8	0.17	50.3	**47.7**	2.1	0.17	145.8	401	0.9	0.2	50.3	46.8	2.2	0.2	145.8	3.50	2.25	3.50	2.38	3.50	0.00	**3.50**	**0.00**
Pr	2650	n,γ	142Pr	1576	**530**	0.3	17.2	50.3	**324**	0.3	17.2	145.8	552	0.3	4.3	50.3	335	0.2	5.54	145.8	18.34	0.20	18.21	0.36	18.27	0.07	**18.27**	**0.07**
Pt	100	n,γ	197Pt	77	**4.55**	6.3	0.33	50.3	**0.941**	5.3	0.5	145.8			0.3	50.3	1.02	5.8	0.6	145.8	6.20	8.23			6.20	0.51	**6.42**	**0.22**
				191	**13.3**	2.1	0.33	50.3	**3.26**	5.2	0.5	145.8	15	1.9	0.3	50.3	2.96	3.8	0.6	145.8	7.35	5.61	5.92	4.25	6.64	0.72		
		n,p	199Au	158	**5.77**	0.6	0.33	50.3	**0.8**	1.7	0.5	145.8	5.74	0.3	0.3	50.3	0.827	0.8	0.6	145.8	4.16	1.80	4.32	0.85	4.24	0.08	**4.24**	**0.08**
Re	2.5	n,γ	186Re	137	**653**	0.7	0.33	50.3	**98.3**	1.8	0.5	145.8	652	0.3	0.33	50.3	98.9	0.9	0.23	145.8	4.52	1.93	4.55	0.95	4.53	0.02	**4.53**	**0.02**
		n,γ	188Re	155	**1350**	0.4	0.33	50.3	**445**	0.7	0.5	145.8	1350	0.5	0.33	50.3	443	0.7	0.6	145.8	9.89	0.81	9.84	0.86	9.87	0.02	**10.31**	**0.44**
				478	**366**	7.3	0.33	50.3	**134**	12.2	0.5	145.8	342	8.4	0.33	50.3	120	11.5	0.6	145.8	10.98	14.22	10.53	14.24	10.75	0.23		
Ru	20	n,γ	97Ru	215	**2.1**	3.5	0.17	50.3	**0.308**	10.7	0.17	145.8			0.2	50.3			0.17	145.8	4.40	11.26			4.40	0.50	**4.50**	**0.21**
		n,γ	105Ru	130	**48.4**	2	0.17	50.3	**7.58**	3.9	0.17	145.8			0.2	50.3			0.17	145.8	4.70	4.38			4.70	0.21	**4.52**	**0.22**
				317	**26.9**	4	0.17	50.3	**3.83**	8.1	0.17	145.8			0.2	50.3			0.17	145.8	4.27	9.03			4.27	0.39		
				469	**24.6**	2.9	0.17	50.3	**3.73**	5.9	0.17	145.8			0.2	50.3			0.17	145.8	4.55	6.57			4.55	0.30		
				676	**61.9**	2.4	0.17	50.3	**8.65**	5.4	0.17	145.8			0.2	50.3			0.17	145.8	4.19	5.91			4.19	0.25		
				724	**62.3**	1.4	0.17	50.3	**9.32**	2.9	0.17	145.8			0.2	50.3			0.17	145.8	4.49	3.22			4.49	0.14		
			105Rh	306	**4.8**	11.5	0.17	50.3			0.17	145.8	7.73	3.2	0.2	50.3	1.26	9.1	0.17	145.8			4.89	9.65	4.89	0.47		
				319	**4.38**	4.8	0.17	50.3			0.17	145.8	6.94	1.7	0.2	50.3	1.05	4.3	0.17	145.8			4.54	4.62	4.54	0.21		
Sb	25	n,γ	122Sb	564	**205**	0.3	1.84	52.2	**20.5**	0.9	0.5	151.3	204	0.2	1.3	52.2	20.5	0.5	0.3	151.3	3.00	0.95	3.01	0.54	3.01	0.01	**3.05**	**0.04**
				693	**132**	2.1	1.84	52.2	**12.3**	9.6	0.5	151.3	136	1.1	1.3	52.2	15.3	3	0.6	151.3	2.80	9.83	3.38	3.20	3.09	0.29		

Table 5 Previous results on elements irradiated in the RINGAS system (continued)

Element	Mass (µg)	Reaction	Isotope	E (keV)	Decay time 4 hours								Decay time 40 hours								R_{Cd}-values							
					Specific Activities (Bq/µg)																							
					Cd				NCd				Cd				NCd											
					X	Sx (%)	% Dt	FM	X	Sx (%)	% Dt	FM	X	Sx (%)	% Dt	FM	X	Sx (%)	% Dt	FM	X	Sx (%)	X	Sx (%)	X(avg)	Sx	**X(avg)**	**Sx**
Se	50	n,γ	75Se	265	**0.453**	2		52.2	**0.0753**	2		151.3	5.69	2		52.2	0.641	2		151.3	4.99	2.83	3.38	2.83	4.18	0.80	**4.18**	**0.80**
Sm	0.5	n,γ	153Sm	70	**357**	2.6	0.5	52.2	**53.3**	8.2	0.33	151.3	419	9	0.23	52.2	68.1	8.2	0.07	151.3	4.48	8.60	4.88	12.18	4.68	0.20	**4.71**	**0.04**
				103	**1060**	1.3	0.5	52.2	**167**	1.6	0.33	151.3	1030	0.3	0.23	52.2	164	0.8	0.6	151.3	4.73	2.06	4.78	0.85	4.75	0.03		
Sr	250	n,γ	87mSr	388	**18.9**	0.4	3	52.2	**6.44**	0.5	1.17	151.3			0.13	52.2			0.07	151.3	10.22	0.64			10.22	0.07	**10.08**	**0.14**
			85mSr?	151	**0.148**	0.5	3	52.2	**0.049**	2	1.17	151.3			0.13	52.2			0.07	151.3	9.93	2.06			9.93	0.20		
Te	1000	n,γ	127Te	418	**8.94**	4.9	21.67	56.2			17.53	145.1	9.66	8.8	3.47	56.2			3.37	145.1								
		n,γ	131mTe	150	**1.69**	2.4	21.67	56.2	**0.59**	5.5	17.53	145.1	0.242	5	3.47	56.2	0.074	13.8	3.37	145.1	10.47	6.00	9.17	14.68	9.82	0.65	**9.82**	**0.65**
		n,γ	129Te	460	**50.2**	1.2	21.67	56.2	**10.1**	4.8	17.53	145.1			3.47	56.2			3.37	145.1	6.04	4.95			6.04	0.30	**6.39**	**0.36**
				487	**77.8**	3.6	21.67	56.2	**17.5**	14.6	17.53	145.1			3.47	56.2			3.37	145.1	6.75	15.04			6.75	1.01		
		n,γ	131I	364	**0.0958**	3.9	21.67	56.2	**0.0553**	5.7	17.53	145.1	0.1132	0.8	3.47	56.2	0.0552	1.3	3.37	145.1	17.32	8.91	14.63	1.53	15.97	1.34	**15.97**	**1.34**
U	50	n,γ	239Np	103	**49.8**	1.6	7.67	52.2	**4.38**	1.6	2	151.3	50.8	1.6	4.37	52.2	**4.44**	1.7	0.6	151.3	2.64	2.26	2.62	2.33	2.63	0.01	**2.67**	**0.04**
				228	**889**	2.6	7.67	52.2	**81.1**	2.8	2	151.3	901	2.5	4.37	52.2	81.2	2.6	0.6	151.3	2.74	3.82	2.70	3.61	2.72	0.02		
				278	**406**	0.3	7.67	52.2	**36.7**	1	2	151.3	416	0.3	4.37	52.2	36.4	0.5	0.6	151.3	2.71	1.04	2.63	0.58	2.67	0.04		
W	25	n,γ	187W	134	**215**	4.1	5.18	52.2	**39.2**	4.5	1.17	151.3	219	4	1.67	52.2	31.3	2.8	0.47	151.3	5.47	6.09	4.29	4.88	4.88	0.59	**4.97**	**0.08**
				480	**55.9**	1.2	5.18	52.2	**9.51**	3	1.17	151.3	56.2	1	1.67	52.2	9.45	2.1	0.47	151.3	5.10	3.23	5.04	2.33	5.07	0.03		
				686	**725**	0.3	5.18	52.2	**119**	0.7	1.17	151.3	725	0.2	1.67	52.2	120	0.5	0.47	151.3	4.92	0.76	4.97	0.54	4.94	0.02		
Y	25000	n,γ	90mY	203	**0.557**	0.3	10.85	57.4	**0.127**	0.5	6.84	148.9			5.17	57.4			3.67	148.9	6.84	0.58			6.84	0.04	**6.92**	**0.08**
				480	**0.15**	1	10.85	57.4	**0.035**	1.9	6.84	148.9			5.17	57.4			3.67	148.9	7.00	2.15			7.00	0.15		
Yb	125	n,γ	175Yb	114	**23.7**	3.5	0.83	52.2	**25**	1.9	0.67	151.3	24.7	3.5	0.53	52.2	25.7	1.4	0.47	151.3	31.65	3.98	31.21	3.77	31.43	0.22	**38.48**	**4.99**
				283	**19**	3.7	0.83	52.2	**26.6**	2.2	0.67	151.3	18.4	2	0.53	52.2	25.3	0.9	0.47	151.3	42.00	4.30	41.25	2.19	41.63	0.38		
				396	**27.3**	0.9	0.83	52.2	**38.8**	0.7	0.67	151.3	27.5	0.4	0.53	52.2	38.6	0.3	0.47	151.3	42.64	1.14	42.11	0.50	42.37	0.26		
		n,γ	177Yb	122	**38.7**	1.9	0.83	52.2	**17.8**	2	0.67	151.3			0.53	52.2			0.47	151.3	13.80	2.76			13.80	0.38	**14.86**	**0.81**
				150	**44.3**	1.1	0.83	52.2	**21.2**	1.1	0.67	151.3			0.53	52.2			0.47	151.3	14.36	1.56			14.36	0.22		
				1080	**28.6**	7.8	0.83	52.2	**14.8**	8.3	0.67	151.3			0.53	52.2			0.47	151.3	15.52	11.39			15.52	1.77		
				1241	**89.4**	5.8	0.83	52.2	**47**	5.3	0.67	151.3			0.53	52.2			0.47	151.3	15.77	7.71			15.77	1.22		
		n,γ	169Yb	308	**14.9**	0.5	0.83	52.2	**0.896**		0.67	151.3			0.53	52.2			0.47	151.3	1.80	0.50			1.80	0.01	**1.80**	**0.01**

Table 5 Previous results on elements irradiated in the RINGAS system (continued)

Element	Mass (µg)	Reaction	Isotope	E (keV)	Decay time 4 hours								Decay time 40 hours								R_{Cd}-values						
					Specific Activities (Bq/µg)																						
					Cd				NCd				Cd				NCd										
					X	Sx (%)	% Dt	FM	X	Sx (%)	% Dt	FM	X	Sx (%)	% Dt	FM	X	Sx (%)	% Dt	FM	X	Sx (%)	X	Sx (%)	X(avg)	Sx	**X(avg)**	**Sx**
Zn	2500	**n,γ**	69mZn	439	**0.884**	0.3	2	52.2	**0.213**	0.7	2.67	151.3	0.887	0.4	0.33	52.2	0.217	0.7	0.13	151.3	7.23	0.76	7.34	0.81	7.28	0.06	**7.66**	**0.38**
			65Zn	1116	**1.13**	2	2	52.2	**0.303**	2	2.67	151.3			0.33	52.2			0.13	151.3	8.04	2.83			8.04	0.23	**8.04**	**0.00**
Zr	25000	**n,γ**	97Zr	508	**1.24**	1.4	27.5	52.2	**0.0932**	2.6	2.67	151.3	1.36	1.2	5.94	52.2	0.0949	2.3	0.67	151.3	2.25	2.95	2.09	2.59	2.17	0.08	**2.16**	**0.03**
				743	**1.95**	0.2	27.5	52.2	**0.148**	0.4	2.67	151.3	2.12	0.1	5.94	52.2	0.149	0.4	0.67	151.3	2.28	0.45	2.11	0.41	2.19	0.08		
				1148	**0.746**	2.9	27.5	52.2	**0.052**	11.3	2.67	151.3	0.813	2.2	5.94	52.2	0.06	6.7	0.67	151.3	2.09	11.67	2.21	7.05	2.15	0.06		
		n,γ	97Nb	658	**1.85**	0.1	27.5	52.2	**0.132**	0.4	2.67	151.3	2.14	0.1	5.94	52.2	0.149	0.3	0.67	151.3	2.14	0.41	2.09	0.32	2.11	0.03		

Table 6 Preliminary R_{CD}-values as obtained from previous data

Element	Mass (µg)	Reaction	Isotope	Advantage factors: Na		Br		K		Mn		Mo		La	
				30.99	0.05	3.83	0.02	27.09	0.25	24.95	0.27	2.06	0.66	23.33	0.12
				X(avg)	Sx	X(avg)	Sx	X(avg)	Sx	X(avg)	Sx	X(avg)	Sx	X(avg)	Sx
Al	10	n,α	24Na	0.187	0.014	1.518	0.116	0.214	0.017	0.233	0.018	2.825	0.927	0.249	0.019
As	10	n,γ	76As	0.140	0.004	1.136	0.032	0.160	0.005	0.174	0.005	2.113	0.677	0.186	0.005
Au	0.5	n,γ	198Au	0.136	0.001	1.101	0.007	0.155	0.002	0.169	0.002	2.048	0.654	0.181	0.001
Ba	10000	n,γ	135mBa	0.064	0.003	0.522	0.021	0.074	0.003	0.080	0.003	0.970	0.312	0.086	0.003
Br	50	n,γ	82Br	0.123	0.001	1.000	0.006	0.141	0.001	0.153	0.002	1.861	0.594	0.164	0.001
Ca	10000	n,γ	47Sc	0.707	0.020	5.730	0.164	0.809	0.024	0.879	0.027	10.661	3.417	0.940	0.027
Cd	250	n,γ	115mIn	0.111	0.008	0.899	0.066	0.127	0.009	0.138	0.010	1.673	0.548	0.148	0.011
Ce	10000	n,γ	141Ce	1.054	0.086	8.536	0.697	1.205	0.099	1.309	0.108	15.881	5.233	1.400	0.114
		n,γ	143Ce	0.642	0.004	5.198	0.041	0.734	0.008	0.797	0.010	9.672	3.089	0.853	0.007
Co	10	n,α	56Mn	0.413	0.012	3.344	0.096	0.472	0.014	0.513	0.016	6.222	1.994	0.549	0.016
Cs	2.5	n,γ	134mCs	0.149	0.002	1.204	0.018	0.170	0.003	0.185	0.003	2.240	0.716	0.198	0.003
		n,γ	134Cs	0.263	0.018	2.130	0.147	0.301	0.021	0.327	0.023	3.963	1.294	0.349	0.024
Cu	100	n,γ	64Cu	0.670	0.011	5.424	0.089	0.766	0.014	0.832	0.016	10.091	3.226	0.890	0.015
Dy	0.5	n,γ	165Dy	1.634	0.091	13.233	0.739	1.869	0.105	2.030	0.115	24.621	7.979	2.171	0.121
Er	50	n,γ	171Er	0.296	0.005	2.399	0.043	0.339	0.007	0.368	0.007	4.463	1.427	0.394	0.007
		n,p	167Ho	0.229	0.001	1.858	0.013	0.262	0.003	0.285	0.003	3.456	1.104	0.305	0.002
Eu	1	n,γ	152Eu	0.952	0.013	7.714	0.110	1.089	0.018	1.183	0.020	14.352	4.586	1.265	0.018
Fe	25000	n,p	56Mn	0.060	0.000	0.483	0.002	0.068	0.001	0.074	0.001	0.898	0.287	0.079	0.000
Ga	5	n,γ	72Ga	0.219	0.004	1.773	0.035	0.250	0.005	0.272	0.006	3.299	1.055	0.291	0.006
Ge	920	n,γ	77Ge	0.108	0.008	0.874	0.063	0.123	0.009	0.134	0.010	1.626	0.532	0.143	0.010
		n,γ	79Ge	0.518	0.045	4.193	0.363	0.592	0.051	0.643	0.056	7.801	2.580	0.688	0.059
Gd	230	n,γ	159Gd	0.097	0.001	0.786	0.006	0.111	0.001	0.121	0.001	1.463	0.467	0.129	0.001
		n,γ	161Tb	0.271	0.010	2.196	0.078	0.310	0.011	0.337	0.012	4.086	1.312	0.360	0.013
Hf	970	n,γ	180mHf	0.148	0.007	1.195	0.059	0.169	0.008	0.183	0.009	2.224	0.718	0.196	0.010
		n,γ	181Hf	0.387	0.003	3.133	0.026	0.442	0.005	0.481	0.006	5.829	1.862	0.514	0.004
Hg	100	n,γ	197mHg	1.850	0.022	14.986	0.188	2.116	0.032	2.299	0.037	27.883	8.908	2.458	0.031
		n,γ	197Hg	1.862	0.017	15.084	0.148	2.130	0.027	2.314	0.032	28.064	8.963	2.474	0.025
Ho	500	n,γ	166Ho	0.173	0.008	1.402	0.064	0.198	0.009	0.215	0.010	2.609	0.841	0.230	0.010
In	20	n,γ	116mIn	0.157	0.002	1.268	0.015	0.179	0.003	0.194	0.003	2.359	0.754	0.208	0.003
Ir	5	n,γ	194Ir	0.169	0.006	1.369	0.047	0.193	0.007	0.210	0.008	2.546	0.818	0.225	0.008
K	10000	n,γ	42K	0.874	0.008	7.081	0.073	1.000	0.013	1.086	0.016	13.174	4.208	1.162	0.012
La	100	n,γ	140La	0.753	0.004	6.096	0.042	0.861	0.009	0.935	0.011	11.342	3.622	1.000	0.007

Table 6 Preliminary R_{CD}-values as obtained from previous data (continued)

Element	Mass (µg)	Reaction	Isotope	Advantage factors											
				Na		Br		K		Mn		Mo		La	
				30.99	0.05	3.83	0.02	27.09	0.25	24.95	0.27	2.06	0.66	23.33	0.12
				X(avg)	Sx	X(avg)	Sx	X(avg)	Sx	X(avg)	Sx	X(avg)	Sx	X(avg)	Sx
Lu	10	n,γ	176mLu	0.111	0.000	0.899	0.004	0.127	0.001	0.138	0.001	1.673	0.534	0.148	0.001
		n,γ	177Lu	1.077	0.064	8.728	0.516	1.233	0.074	1.339	0.080	16.238	5.272	1.432	0.085
Mg	100	n,γ	24Na	0.102	0.003	0.829	0.024	0.117	0.003	0.127	0.004	1.542	0.494	0.136	0.004
Mn	8.65	n,γ	56Mn	0.805	0.009	6.520	0.077	0.921	0.013	1.000	0.015	12.130	3.875	1.069	0.013
Mo	50	n,γ	99mTc	0.066	0.021	0.537	0.172	0.076	0.024	0.082	0.026	1.000	0.452	0.088	0.028
Na	500	n,γ	24Na	1.000	0.002	8.100	0.040	1.144	0.011	1.242	0.014	15.070	4.812	1.329	0.007
Nd	1250	n,γ	149Nd	0.239	0.006	1.933	0.051	0.273	0.008	0.297	0.008	3.597	1.152	0.317	0.008
		n,γ	151Pm	0.132	0.006	1.073	0.052	0.152	0.007	0.165	0.008	1.996	0.645	0.176	0.009
Ni	27500	n,γ	65Ni	0.946	0.009	7.663	0.077	1.082	0.014	1.175	0.016	14.257	4.553	1.257	0.013
		n,p	58Co	0.057	0.002	0.459	0.013	0.065	0.002	0.070	0.002	0.855	0.274	0.075	0.002
Pd	5	n,γ	109Pd	0.113	0.000	0.915	0.004	0.129	0.001	0.140	0.002	1.702	0.543	0.150	0.001
Pr	2650	n,γ	142Pr	0.590	0.002	4.775	0.028	0.674	0.007	0.732	0.008	8.885	2.837	0.783	0.005
Pt	100	n,γ	197Pt	0.207	0.007	1.678	0.057	0.237	0.008	0.257	0.009	3.122	1.002	0.275	0.009
		n,p	199Au	0.137	0.003	1.108	0.022	0.157	0.003	0.170	0.004	2.062	0.660	0.182	0.004
Re	2.5	n,γ	186Re	0.146	0.001	1.185	0.007	0.167	0.002	0.182	0.002	2.204	0.704	0.194	0.001
		n,γ	188Re	0.333	0.014	2.695	0.117	0.381	0.017	0.413	0.018	5.013	1.615	0.442	0.019
Ru	20	n,γ	97Ru	0.145	0.007	1.177	0.055	0.166	0.008	0.181	0.009	2.190	0.707	0.193	0.009
		n,γ	105Ru	0.146	0.007	1.181	0.058	0.167	0.008	0.181	0.009	2.197	0.710	0.194	0.010
Sb	25	n,γ	122Sb	0.098	0.001	0.796	0.011	0.112	0.002	0.122	0.002	1.481	0.473	0.131	0.002
Se	50	n,γ	75Se	0.135	0.026	1.093	0.210	0.154	0.030	0.168	0.032	2.034	0.758	0.179	0.034
Sm	0.5	n,γ	153Sm	0.152	0.001	1.232	0.011	0.174	0.002	0.189	0.003	2.292	0.732	0.202	0.002
Sr	250	n,γ	87mSr	0.325	0.005	2.634	0.040	0.372	0.006	0.404	0.007	4.900	1.566	0.432	0.007
Te	1000	n,γ	129Te	0.206	0.011	1.670	0.093	0.236	0.013	0.256	0.015	3.108	1.007	0.274	0.015
		n,γ	131mTe	0.317	0.021	2.567	0.170	0.363	0.024	0.394	0.026	4.777	1.557	0.421	0.028
		n,γ	131I	0.515	0.043	4.174	0.352	0.590	0.050	0.640	0.054	7.767	2.564	0.685	0.058
U	50	n,γ	239Np	0.086	0.001	0.699	0.010	0.099	0.002	0.107	0.002	1.300	0.415	0.115	0.002
W	25	n,γ	187W	0.160	0.003	1.298	0.022	0.183	0.003	0.199	0.004	2.415	0.772	0.213	0.004
Y	25000	n,γ	90mY	0.223	0.003	1.808	0.022	0.255	0.004	0.277	0.004	3.365	1.075	0.297	0.004
Yb	125	n,γ	175Yb	1.241	0.161	10.055	1.305	1.420	0.185	1.542	0.201	18.708	6.447	1.649	0.214
		n,γ	177Yb	0.480	0.026	3.884	0.214	0.549	0.030	0.596	0.033	7.227	2.341	0.637	0.035
		n,γ	169Yb	0.058	0.000	0.471	0.003	0.067	0.001	0.072	0.001	0.877	0.280	0.077	0.001
Zn	2500	n,γ	69mZn	0.247	0.012	2.003	0.100	0.283	0.014	0.307	0.016	3.727	1.204	0.329	0.016
Zr	25000	n,γ	97Zr	0.070	0.001	0.564	0.008	0.080	0.001	0.087	0.001	1.050	0.335	0.093	0.001

Table 7 Potential improvement in sensitivity extracted from available data in analogy with the required information in Table 2

Element	Reaction	Isotope	E (keV)	R_{Cd}-values		Average advantage factors (Na, K, Mn)		Sensitivity improvement required	Sensitivity improvement expected
				X(avg)	Sx	27.68	2.50		
As	(n,γ)	76As	559	4.20	0.03	0.16	0.01	10	6.3
			657	4.34	0.02				
Ba	(n,γ)	135mBa	268	2.00	0.08	0.07	0.01	No RDA	13.8
	(n,γ)	131Ba	1048	4.14	0.12	0.15	0.01	No RDA	6.6
Br	(n,γ)	82Br	554	3.80	0.09	0.14	0.01	No RDA	7.2
			619	3.85	0.03				
Ca	(n,γ)	47Sc	159	21.93	0.62	0.80	0.07	5	1.3
Cd	(n,γ)	115mIn	336	3.08	0.53	0.13	0.01	50	8.0
	(n,γ)	115Cd	492	3.60	0.13				
			528	3.64	0.00				
Cl	(n,γ)	38Cl	1642					Overall adequate	
Co	(n,γ)	60Co	1173					15	
			1332						
Cr	(n,γ)	51Cr	320					15*	
Cs	(n,γ)	134mCs	127	4.61	0.07	0.17	0.01	No RDA	6.0
	(n,γ)	134Cs	605	7.59	0.21	0.30	0.03	No RDA	3.4
			796	8.71	0.25				
Cu	(n,γ)	64Cu	1346	20.43	1.25	0.76	0.07	500	1.3
F	(n,γ)	20F	1634					5	
Fe	(n,γ)	59Fe	1099					5*	
			1292						
	(n,p)	54Mn	835					5*	
Hg	(n,γ)	197mHg	134	56.60	3.02	2.09	0.18	10	0.5
	(n,γ)	197Hg	68	53.22	0.40	2.10	0.19	10	0.5
			78	57.21	0.33				
I	(n,γ)	128I	443					Overall adequate	
K	(n,γ)	42K	1525	27.09	0.25	0.99	0.09	Overall adequate	1.0
Mg	(n,γ)	24Na	1369	3.17	0.09	0.12	0.01	Overall adequate	8.7

Table 7 **Potential improvement in sensitivity extracted from available data in analogy with the required information in Table 2 (continued)**

Element	Reaction	Isotope	E (keV)	R_{Cd}-values		Average advantage factors (Na, K, Mn)		Sensitivity improvement required	Sensitivity improvement expected
				X(avg)	Sx	27.68	2.50		
Mn	(n,γ)	56Mn	847	25.22	0.09	0.91	0.08	Overall adequate	1.1
			1811	24.68	0.36				
Mo	(n,γ)	99mTc	141	2.06	0.66	0.07	0.01	20*	13.4
Na	(n,γ)	24Na	1369	30.99	0.05	1.13	0.10	Overall adequate	0.9
Ni	(n,γ)	65Ni	366	29.49	0.29	1.07	0.09	No RDA	0.9
			1115	29.52	0.25				
			1482	28.95	0.19				
	(n,γ)	58Co	811	1.71	0.02	0.06	0.01	No RDA	15.6
Rb	(n,γ)	86Rb	1077					No RDA	
Sc	(n,γ)	46Sc	889					No RDA	
			1121						
Se	(n,γ)	75Se	265	4.18	0.80	0.15	0.01	20	6.6
Sn	(n,γ)	113Sn	392					Overall adequate	
Th	(n,γ)	233Pa	312					Overall adequate	
U	(n,γ)	239Np	103	2.63	0.01	0.10	0.01	5	10.3
			228	2.72	0.02				
			278	2.67	0.04				
V	(n,γ)	52V	1434					5	
Zn	(n,γ)	69mZn	439	7.28	0.06	0.28	0.02	Overall adequate	3.6
	(n,γ)	65Zn	1116	8.04	0.23	0.29	0.03	Overall adequate	3.4

*For infants only

6.2 Additional information from the current work done

The only real change between the experiments done in 1986 and the work done in the current investigation is that the reactor power changed from 5 MW to 20 MW. The irradiation positions remained the same, the thickness of the cadmium layer did not change, the mass of the element standards remained the same while the irradiation time was reduced to adapt to the higher reactor power (from 20 s under 5 MW to 5 s under 20 MW).

6.2.1 Determination of R_{Cd}-values

In order to evaluate the previous data obtained for the R_{Cd}-values when the reactor was still operated at 5 MW and to verify the results of the elements of interest in this study (see Table 3 and Table 7) the following element standards were prepared according to specified concentration and volumes.

Element	Nuclide of interest	Half-life	Type	Stock	Amount used
Hg	^{203}Hg / ^{197}Hg	46.6 d / 64,1 h	Long-lived / intermediate	100 µg/ml	20 µl
Mo	^{99}Mo	66 h	Intermediate	1000 µg/ml	50 µl
Se	^{75}Se / ^{77m}Se	120 d / 17,5 s	Long-lived / short-lived	1000 µg/ml	50 µl
U	^{239}Np	2.355 d	Long-lived	1000 µg/ml	50 µl
Na	^{24}Na	15.03 h	Intermediate	1000 µg/ml	50 µl
K	^{42}K	36 h	Intermediate	K_2O	24.83 mg
Sc	^{46}Sc / ^{46m}Sc	84 d / 18,7 s	Long-lived / short-lived	100 µg/ml	20 µl
Co	^{60}Co / ^{60m}Co	70.78 d / 10,5 m	Long-lived / short-lived	1000 µg/ml	50 µl
Ca	^{47}Sc	4.54 d	Long-lived	CaO	0.02798g
Cr	^{51}Cr	27.7 d	Long-lived	1000 µg/ml	250 µl
Fe	^{59}Fe	44.6 d	Long-lived	Iron ring	0.0280g

6.2.2 Evaluation of the RINGAS system

6.2.2.1 Neutron spectrum and flux stability of the RINGAS system

Around 50 element standards of each uranium and thorium were prepared and irradiated and counted for delayed neutrons due to the induced fission reactions. As it is known that the fission reaction on ^{235}U is mainly induced by thermal neutrons and the reaction on ^{232}Th is mainly induced by epithermal and fast neutrons, this provides an elegant way to test the neutron spectrum as well as the neutron flux stability over a period of time. The standards were processed at regular intervals from the middle of May to the middle of September 2006.

Table 8 provides the experimental results (empty cells indicate that either the cadmium or non-cadmium irradiation had mechanical problems, and accordingly no results were obtained). During this period the neutron spectrum and flux was monitored specifically during the removal of a molybdenum target from the reactor core to evaluate if abnormal spectrum changes are observed in the RINGAS irradiation positions (the coloured band in Tables 8 and 9). Graphic representation of the specific activities measured in the bare and cadmium shielded irradiation positions for uranium and thorium during the monitoring period are shown in Figure 2. The following conclusions can be drawn:

- During the monitoring period the flux of epi-thermal/fast neutrons in the in-core irradiation position (= bare tube, located at the vertical mid-point of the reactor core) decreased by about 17% while the thermal neutron flux increased slightly by about 4%.
- During the same monitoring period the flux of thermal neutrons in the out-core irradiation position (= cadmium lined tube, located at the top of the reactor core, which is located some 30 cm up from the bare tube position) increased by about 34% while the epi-thermal/fast neutron flux increased by about 4,5%.
- During the replacement period of a molybdenum target plate no significant changes could be observed in the neutron spectrum as well as the neutron flux at the irradiation positions of the RINGAS system.

6.2.2.2 R_{Cd} stability of the RINGAS system

The respective relative R_{Cd}-values for thorium and uranium can be calculated and are given in Table 9 and are graphically represented in Figure 3 (again, empty cells indicate that either the cadmium or non-cadmium irradiations had mechanical problems, and accordingly no results were obtained). As the neutron spectrum and flux varied over the monitoring period one does not necessarily expect the R_{Cd}-values to change during this period if the relative changes are constant. However, from the observed variations in the thermal and epi-thermal/fast neutron fluxes in the two irradiation positions one would expect the R_{Cd}-values to decline over the monitoring period. It is observed that:

- the R_{Cd}-value for thorium declined about 17%
- the R_{Cd}-value for uranium declined about 25%.

6.2.2.3 Additional neutron spectrum stability evaluation of the RINGAS system

As indicated in paragraph 5.6.1 one can define for two elements (x and y) the following relation:

$$\frac{[R_{cd}-1]^x}{[R_{cd}-1]^y} = \frac{[\phi^{thermal}.\sigma^{thermal}]^x}{[\phi^{thermal}.\sigma^{thermal}]^y} \cdot \frac{[\phi^{epi}.\sigma^{epi}+\phi^{fast}.\sigma^{fast}]^y}{[\phi^{epi}.\sigma^{epi}+\phi^{fast}.\sigma^{fast}]^x} \quad \text{.....(19)}$$

If the neutron spectrum between irradiations does not change this ratio should be a constant, even when the neutron density is changed due to different power requirements of the reactor at the time of irradiation. However, if the neutron spectrum changes this ratio will be affected and accordingly the variation in the ratio will provide a means to monitor the spectral stability at the irradiation positions used for neutron activation analysis. The results are given in Table 10, and are graphically represented in Figure 4 (also here, empty cells indicate that either the cadmium or non-cadmium irradiations had mechanical problems, and accordingly no results were obtained). From this it is observed that:

- The average spectral stability constant of 1,362 is measured with an uncertainty of about 19% while maximum and minimum values of 1,79 and 0,85 are obtained respectively.
- Over the monitoring period a decline of about 17% is observed, which indicates that the R_{Cd}-value of uranium declined more than the R_{Cd}-value of thorium which already is indicated in the previous paragraph.

6.2.2.4 Overall evaluation of the results of the RINGAS system

Looking further at the results one would expect the R_{Cd}-value of uranium to be much larger as the fission reaction is mainly caused by thermal neutrons while epithermal neutrons mainly induce the fission of thorium. However, the R_{Cd}-value of uranium is only about 10% more than the value of thorium. This illustrate that the cadmium lining is burned-up or in other words, the ^{113}Cd nuclide, which is mainly responsible for the capture of thermal neutrons, has been transformed by the (n,γ) reaction to ^{114}Cd, leaving only ^{110}Cd, ^{111}Cd and ^{112}Cd to absorb thermal neutrons at a rate of 500 times less efficient than ^{113}Cd.

Accordingly, the cadmium lined position of the RINGAS system had to be replaced before further work could be done for the determination of the advantage factors so as to evaluate the potential improvement in sensitivity using epi-thermal neutron activation analysis.

As the relative standard deviation in the R_{Cd}-values is 20,5% and 11,4% for thorium and uranium respectively, while counting statistics do not contribute substantially to this uncertainty, it may indicate that other uncertainties have been introduced due to other factors beyond research's scope (i.e. not caused by spectrum and flux variations at the irradiation positions). The observed decline and incline of the thermal and epi-thermal/fast neutron flux is due to reactor fuel burn-up whereas major uncertainty could be due to irreproducible preparation of standards and/or malfunctioning of the RINGAS (counting?) system.

As the evaluation of the RINGAS system showed that the cadmium shielded position had to be replaced (semi) R_{Cd}-values for the prepared element standards could not be done.

Table 8 **Experimental data on the neutron spectrum and flux changes in the RINGAS irradiation positions**

		Specific activities (counts per milligram)							
Date	Time	Th-NC	Uncert.	Th-CD	Uncert.	U-NC	Uncert.	U-CD	Uncert.
11-May-06	01:50 PM			57037	343	1147396	3636		
22-May-06	11:14 AM	132761	525	45831	309	1330536	3599	398808	1982
31-May-06	03:58 PM	136049	531	47249	313	1271210	3580	419615	1986
01-Jun-06	03:51 PM	127631	518	41436	296	1301581	3489	395759	1892
08-Jun-06	02:00 PM	19809	205	7579	125	1269911	3363	455169	1962
09-Jun-06	10:49 AM	117978	500	52464	334	1310662	3936	480107	2386
12-Jun-06	03:27 PM	112151	485	49854	323	1299678	3938	494215	2406
13-Jun-06	03:24 PM	112926	489	51803	332	1266381	3935	480025	2434
04-Jul-06	03:58 PM	105495	475	52106	334	1183869	3754	468934	2331
06-Jul-06	04:05 PM	102041	469			1026827	2947		
06-Jul-06	04:07 PM	111415	487			1218595	3777		
07-Jul-06	02:34 PM	130966	521	51060	326	1230808	3796	480558	2209
10-Jul-06	02:07 PM	125055	509	59296	351	1432051	4062	479606	2174
11-Jul-06	08:27 AM	113532	489	49007	322	1209338	3432	511908	2193
12-Jul-06	10:28 AM	103831	469	49085	322	1281502	3594	496989	2120
13-Jul-06	11:49 AM	102694	469	59643	357	1110296	3065	493634	2406
14-Jul-06	08:46 AM					1215012	3803	516327	2446
21-Jul-06	10:49 AM	107388	477	45149	310	1210857	3454	439754	2269
31-Jul-06	03:25 PM	111516	505			1309690	3878	424799	2243
01-Aug-06	01:40 PM	111056	504	39925	301	1340517	3923	453608	2309
10-Aug-06	10:30 AM	108978	502	46984	327	1265049	3933	508371	2450
10-Aug-06	10:45 AM	106147	492	47087	336	1301498	4013	461827	2356
10-Aug-06	11:00 AM	103204	487	47101	330	1295938	3947	493069	2436
10-Aug-06	11:16 AM	110989	497	45412	328	1293474	3896	505772	2451
14-Aug-06	09:45 AM	103495	504	49226	342			554939	2592
14-Aug-06	10:00 AM	108840	504	48017	339	1279008	3886	528386	2486
14-Aug-06	10:15 AM	102850	483	48874	341	1271289	3895	541251	2540
14-Aug-06	10:30 AM	103861	509	47498	331	1259175	3909	519685	2510
15-Aug-06	09:45 AM	105081	494	49845	339	1270024	3916	545527	2541
15-Aug-06	10:00 AM	100929	498	47588	346	1283184	3941	545847	2499
15-Aug-06	10:15 AM	102127	506	47103	341	1281692	3840	546399	2524
15-Aug-06	10:30 AM	106811	488	46831	325	1276378	3972	538977	2547
16-Aug-06	09:45 AM	102171	480	48520	329	1260470	3950	565257	2632
16-Aug-06	10:00 AM	103956	487	48826	331	1282214	3907	561269	2568
16-Aug-06	10:17 AM	93803	460	48953	330	1264244	3910	560451	2580
16-Aug-06	10:31 AM	108992	504	48731	334	1283022	3946	553186	2536
17-Aug-06	09:45 AM	107955	495	52267	352	1255268	3868	559727	2577
17-Aug-06	10:00 AM	101365	481	49152	336	1263342	3984	559601	2601
17-Aug-06	10:15 AM	103713	486	48427	334	1259136	3759	549410	2626
17-Aug-06	10:30 AM	102851	484	50638	343	1259037	3943	544970	2567
18-Aug-06	09:47 AM	104839	485	47499	335	1235490	3775	555249	2597
18-Aug-06	10:00 AM	98349	472	48931	332	1240178	3833	548589	2594
18-Aug-06	10:15 AM	106638	490	47655	327	1221040	3799	558279	2592
18-Aug-06	10:30 AM	100509	478	45620	321	1228523	3722	550863	2485
19-Aug-06	10:00 AM	103526	486			1336091	4059	557149	2541
19-Aug-06	10:15 AM	103891	490	50186	337	1254340	3916	569823	2594

Table 8 Experimental data on the neutron spectrum and flux changes in the RINGAS irradiation positions (continued)

		Specific activities (counts per milligram)							
Date	**Time**	**Th-NC**	**Uncert.**	**Th-CD**	**Uncert.**	**U-NC**	**Uncert.**	**U-CD**	**Uncert.**
19-Aug-06	10:30 AM	102042	482	52243	354	1249369	3932	565517	2607
19-Aug-06	07:45 PM	105947	494	53637	352	1270851	3904	565042	2608
20-Aug-06	06:10 AM	103143	479	54122	358	1253134	3835	570511	2603
20-Aug-06	06:17 AM	102644	502	51691	350	1238988	3911	576889	2643
20-Aug-06	06:30 AM	107655	502	52960	356	1281145	3889	586211	2618
20-Aug-06	06:45 AM	102605	482	51957	351	1235561	3840		
20-Aug-06	07:00 AM	99965	499	51034	344	1263592	3916	574940	2635
20-Aug-06	07:15 AM	98709	479	53201	350	1283176	3937		
20-Aug-06	08:15 AM	106239	511	51046	359	1228753	3857	549176	2507
20-Aug-06	08:30 AM	104827	513	51958	358	1228861	3760	607855	2662
20-Aug-06	08:50 AM	104085	481	52799	345	1271211	3964	598989	2685
20-Aug-06	09:05 AM	98363	471			1251250	3935	577365	2660
20-Aug-06	09:20 AM	103862	486	53926	348	1257643	3869	606016	2669
20-Aug-06	09:35 AM	108744	496	53447	345	1238537	3870	601271	2672
20-Aug-06	09:50 AM	107448	505	54911	354	1253571	3856	573932	2685
20-Aug-06	10:00 AM	104385	489	51088	341	1277037	3930	571212	2532
20-Aug-06	10:20 AM	100915	482	49925	337	1253112	3965	577000	2669
12-Sep-06	01:58 PM	107387	497	45094	321	1306161	4048	533012	2565
13-Sep-06	02:30 PM	103743	489	48173	331	1315171	4010	510272	2454
13-Sep-06	02:45 PM	105269	490	48832	342	1287178	3991	531707	2528
13-Sep-06	03:11 PM	111135	506	48971	336	1291331	3935	505223	2466
13-Sep-06	03:30 PM	111463	514	49067	347	1273717	3889	517741	2528
14-Sep-06	12:45 PM	111024	509	48684	341	1299256	3917	529170	2488
14-Sep-06	02:00 PM	105990	490	39651	315	1293461	3929	532876	2543
14-Sep-06	02:15 PM	108608	501	48135	334	1298836	3968	549855	2577
14-Sep-06	02:30 PM	104968	508	49052	352	1289165	3951	554245	2518
15-Sep-06	02:17 PM	106116	516	49302	349	1363498	3961	544860	2520
15-Sep-06	02:30 PM	118185	513	47743	328	1304339	4015	521733	2506
15-Sep-06	02:45 PM	109880	498	46893	323	1300990	4013	538676	2569
15-Sep-06	03:00 PM	104821	489	47872	327	1264917	3881		
Average / Standard deviation		107404	7650	49498	3557	1265462	51760	529214	47725
Standard deviation (%)			7.1		7.2		4.1		9.0
Mo-target removal period	Prior	102994	2.8%	52287	2.1%	1254907	1.6%	571545	2.2%
	During	104530	2.9%	53021	2.4%	1254016	1.2%	590949	2.5%

Empty cells indicate that the cadmium or non-cadmium irradiation had mechanical problems, and accordingly no result was obtained

Linear Regression

Th-NCd	y = - 160,34 x + 122114	Relative uncertainty:	slope (17,0%)	constant (5,2%)
Th-Cd	y = - 18,24 x + 51164	Relative uncertainty:	slope (79,9%)	constant (7,0%)
U-NCd	y = 397,42 x + 1229991	Relative uncertainty:	slope (49,6%)	constant (4,2%)
U-Cd	y = 1146,3 x + 425257	Relative uncertainty:	slope (13,3%)	constant (8,4%)

where,

y = Specific activity in counts per milligram
x = time in days

Table 9 **Experimental data on the relative R_{Cd}-values in the RINGAS irradiation positions**

		Ratios			
DATE	TIME	Th-NCd/Cd	Uncert.	U-NCd/Cd	Uncert.
11-May-06	01:50 PM				
22-May-06	11:14 AM	2.90	0.02	3.34	0.02
31-May-06	03:58 PM	2.88	0.02	3.03	0.02
01-Jun-06	03:51 PM	3.08	0.03	3.29	0.02
08-Jun-06	02:00 PM			2.79	0.01
09-Jun-06	10:49 AM	2.25	0.02	2.73	0.02
12-Jun-06	03:27 PM	2.25	0.02	2.63	0.02
13-Jun-06	03:24 PM	2.18	0.02	2.64	0.02
04-Jul-06	03:58 PM	2.02	0.02	2.52	0.01
06-Jul-06	04:05 PM				
06-Jul-06	04:07 PM				
07-Jul-06	02:34 PM	2.56	0.02	2.56	0.01
10-Jul-06	02:07 PM	2.11	0.02	2.99	0.02
11-Jul-06	08:27 AM	2.32	0.02	2.36	0.01
12-Jul-06	10:28 AM	2.12	0.02	2.58	0.01
13-Jul-06	11:49 AM	1.72	0.01	2.25	0.01
14-Jul-06	08:46 AM			2.35	0.01
21-Jul-06	10:49 AM	2.38	0.02	2.75	0.02
31-Jul-06	03:25 PM			3.08	0.02
01-Aug-06	01:40 PM	2.78	0.02	2.96	0.02
10-Aug-06	10:30 AM	2.32	0.02	2.49	0.01
10-Aug-06	10:45 AM	2.25	0.02	2.82	0.02
10-Aug-06	11:00 AM	2.19	0.02	2.63	0.02
10-Aug-06	11:16 AM	2.44	0.02	2.56	0.01
14-Aug-06	09:45 AM	2.10	0.02		
14-Aug-06	10:00 AM	2.27	0.02	2.42	0.01
14-Aug-06	10:15 AM	2.10	0.02	2.35	0.01
14-Aug-06	10:30 AM	2.19	0.02	2.42	0.01
15-Aug-06	09:45 AM	2.11	0.02	2.33	0.01
15-Aug-06	10:00 AM	2.12	0.02	2.35	0.01
15-Aug-06	10:15 AM	2.17	0.02	2.35	0.01
15-Aug-06	10:30 AM	2.28	0.02	2.37	0.01
16-Aug-06	09:45 AM	2.11	0.02	2.23	0.01
16-Aug-06	10:00 AM	2.13	0.02	2.28	0.01
16-Aug-06	10:17 AM	1.92	0.02	2.26	0.01
16-Aug-06	10:31 AM	2.24	0.02	2.32	0.01
17-Aug-06	09:45 AM	2.07	0.02	2.24	0.01
17-Aug-06	10:00 AM	2.06	0.02	2.26	0.01
17-Aug-06	10:15 AM	2.14	0.02	2.29	0.01
17-Aug-06	10:30 AM	2.03	0.02	2.31	0.01
18-Aug-06	09:47 AM	2.21	0.02	2.23	0.01
18-Aug-06	10:00 AM	2.01	0.02	2.26	0.01
18-Aug-06	10:15 AM	2.24	0.02	2.19	0.01
18-Aug-06	10:30 AM	2.20	0.02	2.23	0.01
19-Aug-06	10:00 AM			2.40	0.01
19-Aug-06	10:15 AM	2.07	0.02	2.20	0.01

Table 9 **Experimental data on the relative R_{Cd}-values in the RINGAS irradiation positions (continued)**

		Ratios			
DATE	TIME	Th-NCd/Cd	Uncert.	U-NCd/Cd	Uncert.
19-Aug-06	10:30 AM	1.95	0.02	2.21	0.01
19-Aug-06	07:45 PM	1.98	0.02	2.25	0.01
20-Aug-06	06:10 AM	1.91	0.02	2.20	0.01
20-Aug-06	06:17 AM	1.99	0.02	2.15	0.01
20-Aug-06	06:30 AM	2.03	0.02	2.19	0.01
20-Aug-06	06:45 AM	1.97	0.02		
20-Aug-06	07:00 AM	1.96	0.02	2.20	0.01
20-Aug-06	07:15 AM	1.86	0.02		
20-Aug-06	08:15 AM	2.08	0.02	2.24	0.01
20-Aug-06	08:30 AM	2.02	0.02	2.02	0.01
20-Aug-06	08:50 AM	1.97	0.02	2.12	0.01
20-Aug-06	09:05 AM			2.17	0.01
20-Aug-06	09:20 AM	1.93	0.02	2.08	0.01
20-Aug-06	09:35 AM	2.03	0.02	2.06	0.01
20-Aug-06	09:50 AM	1.96	0.02	2.18	0.01
20-Aug-06	10:00 AM	2.04	0.02	2.24	0.01
20-Aug-06	10:20 AM	2.02	0.02	2.17	0.01
12-Sep-06	01:58 PM	2.38	0.02	2.45	0.01
13-Sep-06	02:30 PM	2.15	0.02	2.58	0.01
13-Sep-06	02:45 PM	2.16	0.02	2.42	0.01
13-Sep-06	03:11 PM	2.27	0.02	2.56	0.01
13-Sep-06	03:30 PM	2.27	0.02	2.46	0.01
14-Sep-06	12:45 PM	2.28	0.02	2.46	0.01
14-Sep-06	02:00 PM	2.67	0.02	2.43	0.01
14-Sep-06	02:15 PM	2.26	0.02	2.36	0.01
14-Sep-06	02:30 PM	2.14	0.02	2.33	0.01
15-Sep-06	02:17 PM	2.15	0.02	2.50	0.01
15-Sep-06	02:30 PM	2.48	0.02	2.50	0.01
15-Sep-06	02:45 PM	2.34	0.02	2.42	0.01
15-Sep-06	03:00 PM	2.19	0.02		
Average / Standard deviation		2.13	0.44	2.43	0.28
Standard deviation (%)			20.5		11.4
Mo-target removal period	Prior	1.97	3.5%	2.19	1.3%
	During	1.99	2.2%	2.12	3.3%

Empty cells indicate that the cadmium or non-cadmium irradiation had mechanical problems, and accordingly no result was obtained

Linear Regression

R_{Cd}-Th	y = - 0,00334 x + 2.500	Relative uncertainty:	slope (30,3%)	constant (9,2%)
R_{Cd}-U	y = - 0,00591 x + 2.964	Relative uncertainty:	slope (16,2%)	constant (7,6%)

where,

y = Specific activity ratio between the bare tube and the Cd-shielded irradiation positions of the RINGAS system
x = time in days

Figure 2 Variation in time for thorium and uranium in the bare and cadmium shielded irradiation positions of the RINGAS system

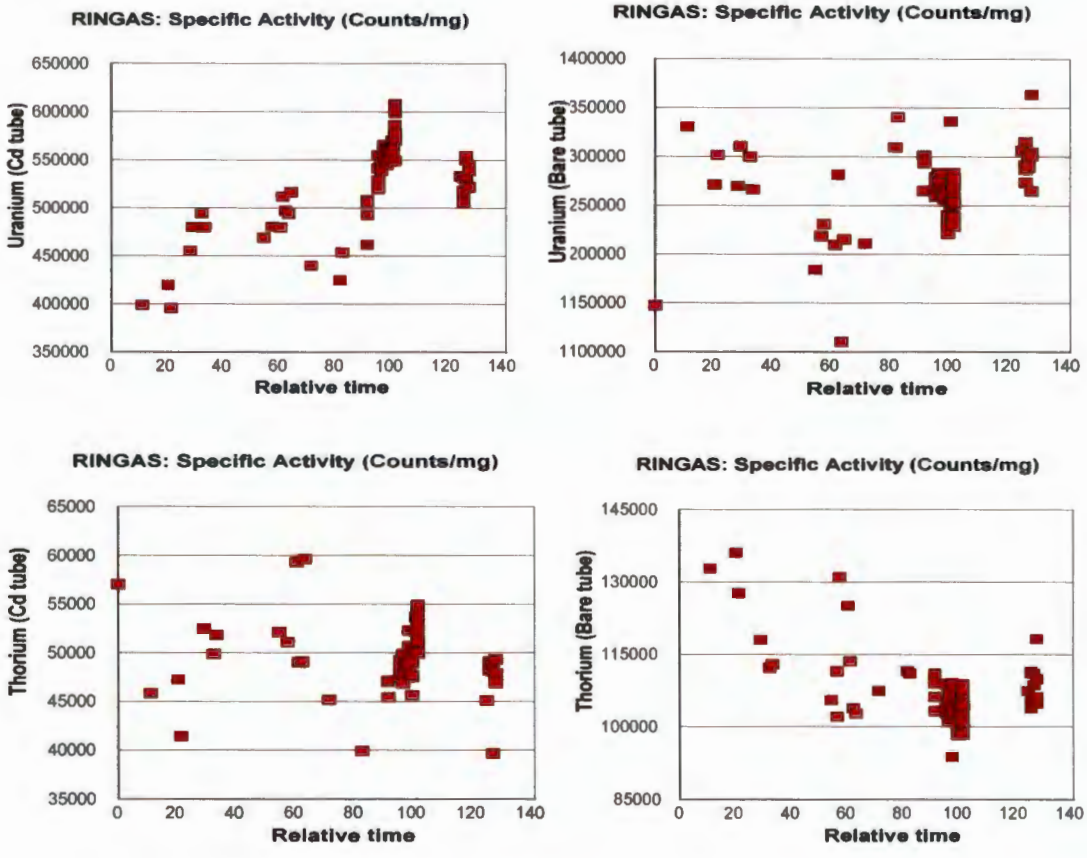


Figure 3 Variation in time for relative cadmium ratios for thorium and uranium in the bare and cadmium shielded irradiation positions of the RINGAS system

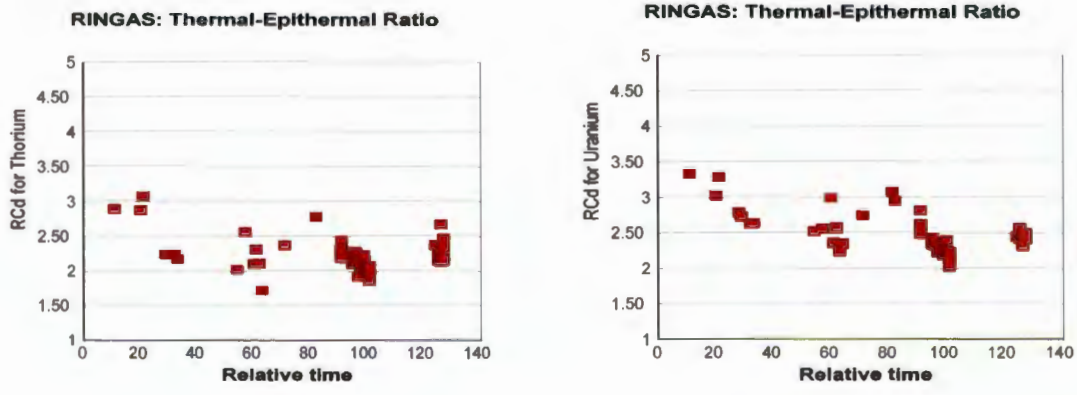


Table 10 Spectral stability of the RINGAS system

DATE	TIME	Activation ratio = Constant Ratio	Uncert.
11-May-06	01:50 PM		
22-May-06	11:14 AM	1.23	0.01
31-May-06	03:58 PM	1.08	0.01
01-Jun-06	03:51 PM	1.10	0.01
08-Jun-06	02:00 PM		
09-Jun-06	10:49 AM	1.39	0.01
12-Jun-06	03:27 PM	1.30	0.01
13-Jun-06	03:24 PM	1.39	0.01
04-Jul-06	03:58 PM	1.49	0.02
06-Jul-06	04:05 PM		
06-Jul-06	04:07 PM		
07-Jul-06	02:34 PM	1.00	0.01
10-Jul-06	02:07 PM	1.79	0.02
11-Jul-06	08:27 AM	1.03	0.01
12-Jul-06	10:28 AM	1.42	0.02
13-Jul-06	11:49 AM	1.73	0.02
14-Jul-06	08:46 AM		
21-Jul-06	10:49 AM	1.27	0.01
31-Jul-06	03:25 PM		
01-Aug-06	01:40 PM	1.10	0.01
10-Aug-06	10:30 AM	1.13	0.01
10-Aug-06	10:45 AM	1.45	0.02
10-Aug-06	11:00 AM	1.37	0.02
10-Aug-06	11:16 AM	1.08	0.01
14-Aug-06	09:45 AM		
14-Aug-06	10:00 AM	1.12	0.01
14-Aug-06	10:15 AM	1.22	0.01
14-Aug-06	10:30 AM	1.20	0.01
15-Aug-06	09:45 AM	1.20	0.01
15-Aug-06	10:00 AM	1.21	0.01
15-Aug-06	10:15 AM	1.15	0.01
15-Aug-06	10:30 AM	1.07	0.01
16-Aug-06	09:45 AM	1.11	0.01
16-Aug-06	10:00 AM	1.14	0.01
16-Aug-06	10:17 AM	1.37	0.02
16-Aug-06	10:31 AM	1.07	0.01
17-Aug-06	09:45 AM	1.17	0.01
17-Aug-06	10:00 AM	1.18	0.01
17-Aug-06	10:15 AM	1.13	0.01
17-Aug-06	10:30 AM	1.27	0.01
18-Aug-06	09:47 AM	1.01	0.01
18-Aug-06	10:00 AM	1.25	0.01
18-Aug-06	10:15 AM	0.96	0.01
18-Aug-06	10:30 AM	1.02	0.01
19-Aug-06	10:00 AM		
19-Aug-06	10:15 AM	1.12	0.01
19-Aug-06	10:30 AM	1.27	0.01
19-Aug-06	07:45 PM	1.28	0.01
20-Aug-06	06:10 AM	1.32	0.01
20-Aug-06	06:17 AM	1.16	0.01
20-Aug-06	06:30 AM	1.15	0.01
20-Aug-06	06:45 AM		

Table 10 Spectral stability of the RINGAS system (continued)

DATE	TIME	Activation ratio = Constant Ratio	Uncert.
20-Aug-06	07:00 AM	1.25	0.01
20-Aug-06	07:15 AM		
20-Aug-06	08:15 AM	1.14	0.01
20-Aug-06	08:30 AM	1.00	0.01
20-Aug-06	08:50 AM	1.16	0.01
20-Aug-06	09:05 AM		
20-Aug-06	09:20 AM	1.16	0.01
20-Aug-06	09:35 AM	1.02	0.01
20-Aug-06	09:50 AM	1.24	0.01
20-Aug-06	10:00 AM	1.18	0.01
20-Aug-06	10:20 AM	1.15	0.01
12-Sep-06	01:58 PM	1.05	0.01
13-Sep-06	02:30 PM	1.37	0.02
13-Sep-06	02:45 PM	1.23	0.01
13-Sep-06	03:11 PM	1.23	0.01
13-Sep-06	03:30 PM	1.15	0.01
14-Sep-06	12:45 PM	1.14	0.01
14-Sep-06	02:00 PM	0.85	0.01
14-Sep-06	02:15 PM	1.08	0.01
14-Sep-06	02:30 PM	1.16	0.01
15-Sep-06	02:17 PM	1.30	0.01
15-Sep-06	02:30 PM	1.02	0.01
15-Sep-06	02:45 PM	1.05	0.01
15-Sep-06	03:00 PM		
Average / Standard		1.18	0.22
Standard deviation (%)			18.7

Mo-target removal period	Prior	1.21	5.7%
	During	1.13	7.5%

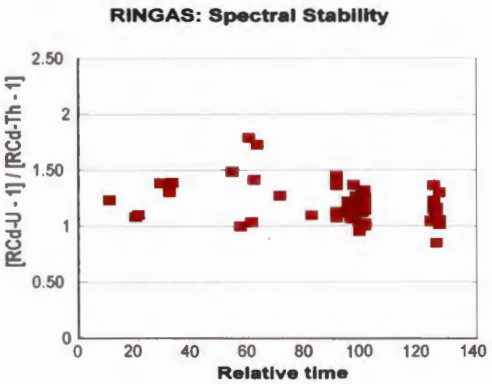
Empty cells indicate that the cadmium or non-cadmium irradiation had mechanical problems, and accordingly no result was obtained

Linear Regression

$[R_{Cd} - 1]^x / [R_{Cd} - 1]^y$ $p = - 0{,}00182\ q + 1{,}362$ Relative uncertainty: slope (38,0%) constant (11,4%)

where,
x = Uranium
y = Thorium
p = Spectral stability as expressed in equation (19)
q = time in days

Figure 4 Spectral stability of the RINGAS system



6.2.3 Evaluation of the PRS system

6.2.3.1 Neutron spectrum and flux stability of the PRS system

As indicated in paragraph 5.6.2 the PRS system is located outside the core and is surrounded by water and accordingly the neutron spectrum is already well thermalized. An elegant solution to measure the spectral stability is using iron monitors and compare the ratios of the following neutron induced reactions:

Thermal neutrons	^{58}Fe (n,γ) ^{59}Fe
Epi-thermal neutrons	^{54}Fe (n,p) ^{54}Mn

From the middle of May 2006 till the end of October 2006 41 iron ring flux monitors were irradiated at regular intervals in the PRS system. The rabbit provides an inside space of around 10 cm and to allow for measurement at the different positions inside the rabbit iron ring monitors were packed at 2,5 cm intervals providing 4 measurements per irradiation. The results are shown in Table 11 and Figure 5 (empty cells indicates lost data). From this data the $^{59}Fe/^{54}Mn$ induced activity ratio was calculated and is presented in Table12 and Figure 6.

From Table 11 and Figure 5 it is observed that the specific activity for both ^{54}Mn and ^{59}Fe decreased during the monitoring period. However, the decrease in the specific activity of both nuclides seems to be at more or less the same rate, which indicates that the neutron spectrum is relatively stable at the PRS position. Linear regression of the specific activities of ^{54}Mn and ^{59}Fe against time gave the following results,

^{59}Fe (Specific Activity)	=	(- 447,2 ± 49,3) . T	+	150694 ± 12540
^{59}Fe (normalized to specific activity of 54Mn)	=	(- 5,605 ± 0,618) . T	+	1888,7 ± 157,2
^{54}Mn (Specific Activity)	=	(- 5,681 ± 0,690) . T	+	1894,3 ± 175,4

where T = relative time in days

This also means that the neutron flux diminished by about 50%, which is substantially more then observed for the RINGAS irradiation positions. Here it was observed that over the

period middle of May till 3rd-week of September the thermal neutron flux at the side-top of the reactor core increased by about 34% while for the same period the thermal flux at the PRS position dropped by about 36%. This can only be explained by the steady burn-up of the reactor fuel and accordingly the shift in the control rod positions. It should also be noted that the RINGAS system is situated at the top-end of the reactor while the PRS system is situated at the bottom-end, and the control rods are moved upwards in time to compensate for burn-up.

The relative stability of the spectral distribution at the PRS system is also observed from Table 12 and Figure 6. Linear regression of the $^{59}Fe/^{54}Mn$-ratio (Specific Activity) against time, shown hereunder, also shows that the ratio is basically constant over the monitoring period.

$^{59}Fe/^{54}Mn$-ratio (Specific Activity) = $(- 0{,}00065 \pm 0{,}00590) \cdot T + 79.52 \pm 1{,}50$

where T = relative time in days

The fact that no serious additional uncertainties were observed can be attributed to the fact that pre-cut iron rings were used of almost uniform mass, which was again fine-tuned by weighing of the individual rings. However, it should be noted that some data were omitted from the results where it was clear that due to the sorting procedure two or no iron monitors were added to the counting vials.

Again the general decrease in the neutron flux during the monitoring period can be accounted for by the burn-up of the reactor fuel.

6.2.3.2 Overall evaluation of the results of the PRS system

The mere fact that the neutron flux dropped with around 50% over the period middle of May till end of October 2006, but that basically no change was observed in $^{54}Mn/^{59}Fe$-activity ratios raises the suspicion that the observed ^{54}Mn activity is not caused by the epithermal and/or fast part of the neutron spectrum, but that the neutrons at the position of the PRS system are highly thermalized and that ^{54}Mn is produced by interaction of ^{54}Fe with thermal neutrons. Accordingly, one does not have to worry about spectral changes but only monitor the neutron flux.

The average spatial distribution of the neutron flux over the rabbit (about 10 cm) seems to be constant and no statistical variation could be observed from both the ^{54}Mn and ^{59}Fe data. However, variations between the specific activities of the 4 monitors could be observed and varied between 0,4% and 11,3% for the ^{59}Fe activity and between 0,6% and 12,0% for ^{54}Mn. As there is a fair correlation between the variations in the ^{59}Fe and ^{54}Mn activities ($R^2 = 0{,}71$) it is suspected that material may have been lost from the iron monitors during transfer from the irradiation vials to the counting vials. This could have been caused due to loss of rust from the surface of the iron metal monitors.

As the evaluation of the PRS system showed that the neutron spectrum is highly thermalized at that position it was not meaningful to determine R_{Cd}-values for the relevant element standards prepared for this study.

Table 11 ^{54}Mn and ^{59}Fe Data of the PRS system

				MN-54									FE-59									
			Tim	Position 1		Position 2		Position 3		Position 4		Average	Unc	Position 1		Position 2		Position 3		Position 4		Average	Unc
Date	Time	Sample ID	(min)	Bq/g	Unc	Bq/g	Unc	Bq/g	Unc	Bq/g	Unc			Bq/g	Unc	Bq/g	Unc	Bq/g	Unc	Bq/g	Unc		
11-May-06	5:03:00 PM	FE01-04	30	1450	99	1451	120	1479	108	1556	115	1484	43	115200	2121	116520	2145	119160	2192	118800	2184	117420	1633
23-May-06	6:09:00 PM	FE05-08	30	1679	118	1687	124	1874	131	1748	121	1747	78	138000	2514	138600	2528	136140	2483	128400	2344	135285	4077
30-May-06	5:30:00 PM	FE09-12	30	1700	79	1549	87	1926	87	1673	76	1712	136	139620	2516	132480	2389	143580	2585	136080	2455	137940	4120
01-Jun-06	5:00:00 PM	FE17-20	30	1799	77	1705	80	1725	79	1723	79	1738	36	132240	2391	137340	2482	136260	2462	134340	2428	135045	1943
08-Jun-06	1:57:00 PM	FE13-I6	30	1802	77	1536	81	1771	89	1604	82	1679	112	138840	2506	141600	2556	141360	2552	129240	2336	137760	5036
09-Jun-06	5:04:00 PM	FE21-24	30	1966	84	1818	80	1908	80	1766	77	1864	77	140460	2534	144300	2603	144900	2614	142680	2573	143085	1719
12-Jun-06	5:57:00 PM	FE25-28	30	1873	63	1943	65	2018	60	1964	61	1950	52	152070	2716	154860	2766	157200	2807	162750	2913	156720	3927
13-Jun-06	5:00:00 PM	FE29-32	30	1662	86	1744	76	1524	89	1589	74	1630	82	133140	2401	135900	2451	134760	2431	127020	2293	132705	3426
05-Jul-06	5:00:00 PM	FE49-52	30	1880	83	1837	91	1838	80	1927	87	1870	37	148440	2672	150840	2715	150600	2711	149880	2699	149940	935
06-Jul-06	5:00:00 PM	FE45-48	30	1692	72	1651	90	1771	79	1676	78	1698	45	131880	2380	136140	2455	136860	2467	125760	2272	132660	4415
07-Jul-06	5:00:00 PM	FE41-44	30	1853	86	1746	86	1642	75	1594	81	1709	100	138180	2489	139680	2516	131340	2369	136800	2466	136500	3148
09-Jul-06	5:04:00 PM	FE153-156	30	1628	24	1250	9	1308	16	1221	9	1352	163	127654	2376	98636	1841	101683	1898	99470	1857	106861	12057
10-Jul-06	5:00:00 PM	FE33-36	30	1701	36	1787	37	1743	37	1779	37	1753	34	133900	2378	133700	2376	134800	2395	133400	2370	133950	522
11-Jul-06	5:00:00 PM	FE53-56	30	1799	25	1728	22	1667	21	1790	22	1746	53	134300	2381	137500	2438	135100	2394	140600	2492	136875	2452
12-Jul-06	5:00:00 PM	FE57-58	30	1771	22	1732	22					1752	20	137400	2436	139200	2467					138300	900
13-Jul-06	5:00:00 PM	FE61-63	30	1750	21	1716	21	1623	20			1696	54	137800	2442	138700	2459	127200	2254			134567	5222
15-Jul-06	5:00:00 PM	FE69-72	30	1661	21	1664	22	1691	25	1896	22	1728	98	135400	2399	134300	2380	133100	2358	146700	2600	137375	5445
16-Jul-06	5:00:00 PM	FE73-76	30	1719	23	1688	23	1688	27	1784	24	1720	39	132400	2347	137400	2435	123900	2196	141700	2510	133850	6621
17-Jul-06	5:00:00 PM	FE77-80	30	1679	22	1561	36	1605	21	1883	24	1682	123	135000	2391	126500	2242	123600	2189	147800	2618	133225	9400
18-Jul-06	5:00:00 PM	FE81-84	30	1856	28	1836	23	1858	22	1436	13	1747	179	144600	2561	150300	2663	146800	2600	117000	2129	139675	13248
31-Jul-06	5:00:00 PM	FE93-96	30	1405	15	1338	15	1308	8	1308	8	1340	40	113300	2053	109200	1977	106900	1932	102300	1852	107925	3975
01-Aug-06	6:00:00 PM	FE97-100	30	1402	15	1300	15	1229	15	1275	15	1302	63	108100	1958	104600	1889	103400	1873	102000	1843	104525	2260
02-Aug-06	5:10:00 PM	FE101-104	30	1341	15	1305	14			1307	15	1318	17	105700	1910	106700	1930			103100	1862	105167	1517
03-Aug-06	5:00:00 PM	FE105-108	30	1471	16	1401	16	1473	16	1447	16	1448	29	118500	2139	115200	2080	117300	2118	116800	2113	116950	1184
04-Aug-06	5:39:00 PM	FE109-112	30	1286	15	1318	15	1419	15	1423	15	1362	61	105800	1909	109100	1968	110500	1998	110200	1993	108900	1864
06-Aug-06	5:07:00 PM	FE177-180	30	1180	8	1136	18	1177	21	1080	14	1143	40	98402	2057	87558	1588	88631	1614	88961	1615	90888	4369
15-Aug-06	5:01:00 PM	FE165-168	30	1187	8	1222	14	1223	9	1276	15	1227	32	97310	1766	93250	1693	97130	1763	99210	1800	96725	2165
16-Aug-06	5:00:00 PM	FE157-160	30	1166	8	1223	8	1083	8	1043	8	1129	70	90890	1649	95580	1734	88050	1598	84730	1538	89813	3980
17-Aug-06	5:00:00 PM	FE161-164	30	1143	9	1262	9	1248	15	1310	11	1241	61	93170	1690	97570	1769	97110	1761	108700	1969	99138	5780
19-Aug-06	5:00:00 PM	FE173-176	30	1190	9	1241	9	1251	15	1140	8	1206	44	92310	1673	95290	1727	97600	1769	92640	1679	94460	2150
22-Aug-06	5:00:00 PM	FE113-116	30	1262	15	1292	16	1146	14	1169	14	1217	61	103700	1863	104200	1870	93980	1687	95280	1713	99290	4686
23-Aug-06	5:00:00 PM	FE117-120	30	1297	16	1314	16	1012	13	1151	15	1194	122	105600	1894	106400	1913	82770	1486	96010	1726	97695	9538
24-Aug-06	5:35:00 PM	FE193-196	30	1267	15	1227	8	1203	8	1236	10	1233	23	99300	1799	101600	1841	98500	1784	98710	1788	99528	1232
12-Sep-06	5:02:00 PM	FE137-140	30	1184	9	974	16	1222	18	1025	8	1101	104	93000	1671	72430	1302	94090	1690	81720	1469	85310	8874
13-Sep-06	5:05:00 PM	FE129-132	30	1192	9	1194	9	1101	9	1151	15	1159	38	94550	1698	92896	1654	88394	1573	86691	1543	90633	3202
22-Sep-06	5:00:00 PM	FE205-208	30	1216	17	1236	18	1110	16			1187	55	101800	1826	100165	1813	87249	1580			96405	6508
26-Sep-06	5:00:00 PM	FE89-92	30	1194	18	1030	16	1113	17	1170	18	1127	63	97430	1736	83530	1487	95980	1708	91940	1638	92220	5405
26-Sep-06	5:36:00 PM	FE189-192	30	1154	22	1089	9	969	17	1156	21	1092	76	88979	1593	87404	1571	78134	1407	88683	1596	85800	4465
29-Sep-06	4:55:00 PM	FE185-188	30	1115	9	1153	18	1122	9	1059	16	1112	34	89620	1605	87970	1575	90840	1626	85870	1538	88575	1864
29-Sep-06	5:30:00 PM	FE181-184	30	1103	29	1105	18	1087	9	1095	16	1098	7	86002	1537	84080	1504	86810	1553	85650	1532	85636	992
29-Oct-06	6:00:00 PM	FE133-136	30	1247	23	1208	21	1075	22	1098	12	1157	72	91570	1626	93340	1658	80730	1435	89160	1585	88700	4835
Average / Uncertainty				**1486**	**274**	**1444**	**265**	**1442**	**307**	**1435**	**293**	**1452**	**20**	**117111**	**2034**	**115916**	**2269**	**113909**	**2298**	**113862**	**2271**	**115200**	**1380**
Uncertainty (%)					**18%**		**18%**		**21%**		**20%**		**1.4%**		**17%**		**20%**		**20%**		**20%**		**1.2%**

Table 12 The $^{59}Fe/^{54}Mn$ induced activity ratio

Fe/Mn Induced Activity Ratio									
Position									
1		2		3		4		Average	
Ratio	Uncert.	Ratio	Uncert.	Ratio	Uncert.	Ratio	Uncert.	Ratio	Uncert.
79.4	5.6	80.3	6.8	80.6	6.1	76.3	5.8	79.2	1.7
82.2	6.0	82.2	6.2	72.7	5.2	73.5	5.3	77.6	4.6
82.1	4.1	85.5	5.0	74.5	3.6	81.3	4.0	80.9	4.0
73.5	3.4	80.6	4.1	79.0	3.9	78.0	3.8	77.8	2.6
77.0	3.6	92.2	5.1	79.8	4.3	80.6	4.3	82.4	5.8
71.5	3.3	79.4	3.8	75.9	3.5	80.8	3.8	76.9	3.6
81.2	3.1	79.7	3.0	77.9	2.7	82.8	3.0	80.4	1.8
80.1	4.4	77.9	3.7	88.4	5.4	79.9	4.0	81.6	4.0
79.0	3.8	82.1	4.3	81.9	3.9	77.8	3.8	80.2	1.9
77.9	3.6	82.5	4.7	77.3	3.7	75.0	3.7	78.2	2.7
74.6	3.7	80.0	4.2	80.0	3.9	85.8	4.6	80.1	4.0
78.4	1.9	78.9	1.6	77.7	1.7	81.5	1.6	79.1	1.4
78.7	2.2	74.8	2.0	77.3	2.1	75.0	2.0	76.5	1.6
74.7	1.7	79.6	1.7	81.0	1.8	78.5	1.7	78.5	2.4
77.6	1.7	80.4	1.7					79.0	1.4
78.7	1.7	80.8	1.7	78.4	1.7			79.3	1.1
81.5	1.8	80.7	1.8	78.7	1.8	77.4	1.6	79.6	1.6
77.0	1.7	81.4	1.8	73.4	1.8	79.4	1.8	77.8	3.0
80.4	1.8	81.0	2.4	77.0	1.7	78.5	1.7	79.2	1.6
77.9	1.8	81.9	1.8	79.0	1.7	81.5	1.7	80.1	1.7
80.6	1.7	81.6	1.7	81.7	1.6	78.2	1.5	80.5	1.4
77.1	1.6	80.5	1.7	84.1	1.8	80.0	1.7	80.4	2.5
78.8	1.7	81.8	1.7			78.9	1.7	79.8	1.4
80.6	1.7	82.2	1.7	79.6	1.7	80.7	1.7	80.8	0.9
82.3	1.8	82.8	1.8	77.9	1.6	77.4	1.6	80.1	2.4
83.4	1.8	77.1	1.9	75.3	1.9	82.4	1.8	79.5	3.4
82.0	1.6	76.3	1.6	79.4	1.5	77.8	1.7	78.9	2.1
78.0	1.5	78.2	1.5	81.3	1.6	81.2	1.6	79.7	1.6
81.5	1.6	77.3	1.5	77.8	1.7	83.0	1.7	79.9	2.4
77.6	1.5	76.8	1.5	78.0	1.7	81.3	1.6	78.4	1.7
82.2	1.8	80.7	1.8	82.0	1.8	81.5	1.8	81.6	0.6
81.4	1.8	81.0	1.8	81.8	1.8	83.4	1.9	81.9	0.9
78.4	1.7	82.8	1.6	81.9	1.6	79.9	1.6	80.7	1.7
78.5	1.5	74.3	1.8	77.0	1.8	79.7	1.6	77.4	2.0
79.3	1.5	77.8	1.5	80.3	1.6	75.3	1.7	78.2	1.9
83.7	1.9	81.0	1.9	78.6	1.8			81.1	2.1
81.6	1.9	81.1	1.9	86.2	2.0	78.6	1.8	81.9	2.8
77.1	2.0	80.3	1.6	80.6	2.0	76.7	2.0	78.7	1.8
80.4	1.6	76.3	1.8	81.0	1.6	81.1	1.9	79.7	2.0
78.0	2.5	76.1	1.8	79.9	1.6	78.2	1.8	78.0	1.3
73.4	1.9	77.3	1.9	75.1	2.0	81.2	1.7	76.8	2.9
78.8	2.8	80.3	3.2	79.3	3.3	79.6	2.5	79.5	1.5
	3.5%		4.0%		4.2%		3.2%		1.8%

Figure 5 Change in ^{54}Mn and ^{59}Fe specific activity with time in the PRS irradiation position

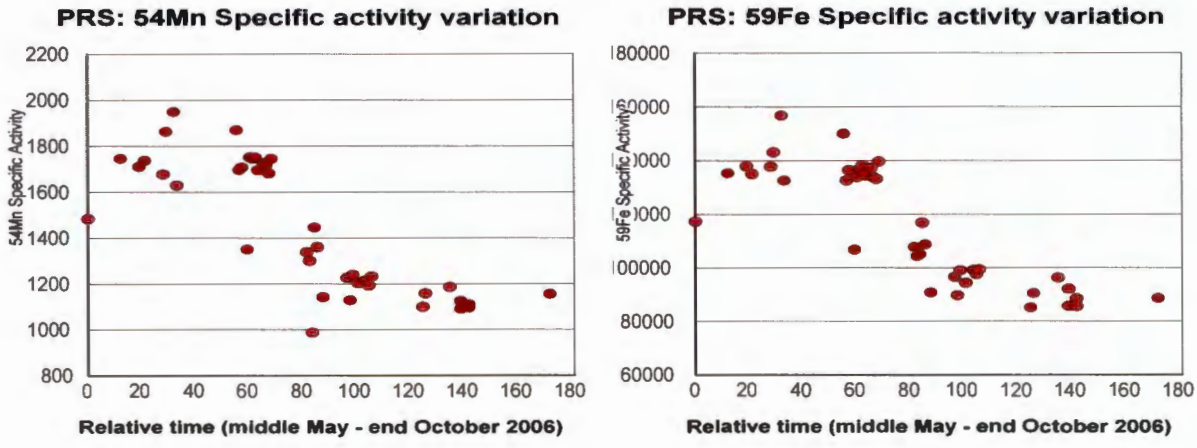
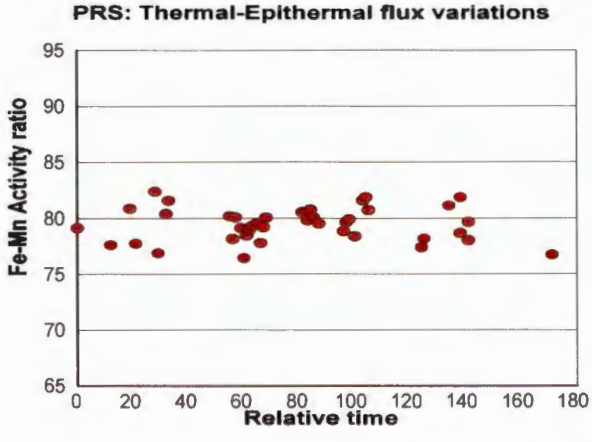


Figure 6 Change in ^{54}Mn and ^{59}Fe specific activity ratios with time in the PRS irradiation position



7. CONCLUSION

As the main objective of this study is to evaluate possible improvements in the sensitivity of instrumental neutron activation analysis (INAA) of essential and toxic elements in a variety of foodstuffs commonly used in South Africa, the following conclusions can be drawn:

- Na, K and Mn seem to be the major elements of which the radionuclides formed upon neutron irradiation dominate the gamma-spectrum. Relevant improvement in the sensitivity upon irradiation with epithermal/fast neutrons is to be expected for As, Ba, Br, Cd, Cs, Mg, Mo, Ni, Se, U and Zn. However, the required improvement in sensitivity to produce meaningful results in foodstuffs analysis is only to be expected for U and possibly Mo. Consequently one has now to look further into changes in the irradiation-delay-counting time scheme applied routinely at Necsa, increasing the sample mass or using more efficient gamma detectors.

- The former conclusion is based on data obtained two decades ago when the reactor was operated at 5 MW compared to the 20 MW operation during this study. No new cadmium ratio values (R_{Cd}) could be obtained from the experiments as it appeared from this study that the cadmium cover was "Burned-up" and first had to be replaced before future work could continue.

- Valuable information was obtained on the change in the neutron flux and energy over time in the three irradiation positions used for neutron activation analysis, indicating that the comparator technique (using element standards with each batch of samples) is still the preferred way for INAA with the Safari-1 reactor. However, the neutron spectrum was relatively stable thus the change in the neutron flux is the major contributing factor, which can provide a means to add a single monitor to each batch of samples and apply a single correction factor for all other elements.

8. REFERENCES

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9. GLOSSARY

Compton continuum	A smooth distribution of energy deposited in a gamma detector caused by partial absorption of energy from processes such as Compton scattering or bremstrahlung
Decay	The disintegration of the nucleus of an unstable atom by spontaneous fission, by the spontaneous emission of an alpha or beta particle, isomeric transitions, or by electron capture.
Detector	A device sensitive to radiation, which produces a current or voltage pulse which may or may not correspond to the energy deposited by an individual photon or particle.
Fission	The splitting of a heavy nucleus into two parts accompanied by release of a relatively large amount of energy and generally one or more neutrons.
Geometric Errors	Errors regarding the detector-to-sample distance, size and shape of the detector and the sample, as well as any shielding, all of which affect the radiation seen by detector.
Matrix	The nature of a sample material.
Flux	The number of neutrons passing through an area per second.
Flux Gradient	The difference in flux intensity or distribution.
Neutron	An uncharged particle with mass slightly larger than that of a proton, both of which are found in the nucleus of every atom except hydrogen.
Resolution	The ability of a spectrometry system to differentiate between two energy lines (peaks) that are close together.
Spectrometry	A process of counting the emission of radiation of a specific energy or range of energies to the exclusion of all other energies.
Self absorption	Absorption of the photons emitted by the radioactive nuclides in the sample by the sample material itself.

Appendix A PRINCIPLES OF NEUTRON ACTIVATION ANALYSIS

A 1 Introduction

The basic principle is that a stable isotope when exposed to neutrons can undergo nuclear transformation to produce a radioactive nuclide, which is usually formed in a higher energy state (excited state) [17]. To achieve stability it will emit gamma rays that can be captured by a detector to characterize qualitatively and quantitatively the elemental composition of the target material. This technique is applied in many fields of research such as e.g. certification of standards reference materials, nutrition [composite diets, food], environmental sciences [dust, plants and tree-leaves], and industrial products [fertilizers, coatings].

Thermal and epi-thermal neutrons from nuclear research reactors are commonly used for element determination by neutron activation analysis (NAA) [10]. This technique is capable of detecting many elements at extremely low concentrations. After exposure to neutrons the sample is allowed to decay and thereafter is counted using lithium drifted or high purity germanium detectors absorbing the emitted gamma-rays, resulting in gamma-ray spectra. The position of each peak in the spectrum is related to the energy of the gamma ray, which identifies the radionuclide and accordingly the respective element, while the area under the peak is proportional to its concentration. There are two matrix effects that can influence the results,

- if a sample has a high concentration of an element with a large cross section, neutrons may be absorbed rendering other product nuclei less radioactive than they theoretically should be,
- if a sample has a high density (e.g. 5 g/cm^3 or more) gamma-rays, especially those of lower energy, may be absorbed by the sample itself and accordingly will not reach the detector [9].

A 2 Reactions

Stable isotopes can undergo numerous nuclear transformations. The reaction that is commonly used in instrumental neutron activation is the neutron-gamma reaction (n,γ), which is illustrated in equation A.1 and A.2.

$$_{33}As^{75} + {}_0n^1 \rightarrow {}_{33}As^{76} + {}_0\gamma^0 \quad \ldots(A.1)$$

$$_{55}Cs^{133} + {}_0n^1 \rightarrow {}_{55}Cs^{134} + {}_0\gamma^0 \quad \ldots(A.2)$$

The above reactions are normally expressed as

$$^{75}As\ (n,\gamma)\ ^{76}As \quad \ldots(A.3)$$

$$^{133}Cs\ (n,\gamma)\ ^{134}Cs \quad \ldots(A.4)$$

The (n,γ) reaction is normally caused by thermal neutrons (~0.025 eV) although it can occur with neutrons of different energies. A neutron is captured by the atom and one or more gamma rays are emitted immediately (prompt γ). If the product nucleus is radioactive it will dacay, with a nuclide specific decay constant, to an other nuclide, which can then also emit gamma-rays (delayed γ). Although the (n,γ) reaction is widely used a number of other reactions are also important. In the (n,p) reactions, which require higher than thermal

energies, a neutron enters an atom with sufficient energy resulting in subsequent proton release and reduction of atomic number by 1, thereby converting the target atom into different element . An example is,

$$_{16}S^{32}\,(n, p)\,_{15}P^{32} \qquad \ldots(A.5)$$

The (n,α) reaction like the (n,p) reaction, usually requires higher energy neutrons. In the (n,α) reaction a neutron enters the target atom and causes an alpha particle (α) to be emitted. The atomic number of target atom reduces by 2 as shown below

$$_{13}Al^{27}\,(n,\gamma)\,_{11}Na^{24} \qquad \ldots(A.6)$$

In our current investigation the focus will be on the (n,γ) reaction [7]

A 3 Gamma-ray Spectroscopy

There are various methods involved when gamma rays interact with matter, but in spectroscopy only three events are of importance i.e. photoelectric effect, Compton scattering and pair production, which are dependent on the energy of the photon and the atomic number of the material.

A 3.1 Photoelectric Effect

This is the absorption of the photon by a material and the subsequent emission of a photoelectron from one of its (outer) shells. This process can not take place with the presence of free electrons. This process provides a possible means of measuring the original photon, since the total kinetic energy of the emitted electron is equal to that of the gamma ray energy. This event contributes to the full energy peak in the recorded pulse height spectrum [2,7]. *These peaks are the characteristics that make gamma spectrometry a useful tool.*

A 3.2 Compton Scattering

The incoming photon collides with an electron in the absorbing material, partially transferring its energy to the electron and scattering through an angle different to its original direction. The probability of scattering depends upon the number of available electrons and therefore increases linearly with the atomic number of the material. This energy distribution leads to the Compton continuum of the spectrum [2,7].

A 3.3 Pair Production

This occurs when a photon interacts with the nucleus of an atom and uses up part of its energy to produce an electron-positron pair. Due to the combined energy of the electron-positron pair of 1022 keV only gamma-rays higher than 1022 keV can lead to pair production. However, the electron-positron pair will annihilate and produce two gamma-rays of 511 keV. If one of these photons escapes the detector without any interaction a so-called single escape peak is formed (the original energy of the gamma-ray minus 511 keV) and if both photons escape undetected the double escape peak arises (the original energy of the gamma-ray minus 1022 keV). This enhances the characteristic spectral features [2,7].

A 4 Sample Packaging and Dimensions

A 4.1 Sample material / container

Sample packaging should consider the inherent safety of the procedure for neutron activation analysis as well as minimization of sample materials from contamination from other samples and/or the transport system in and out of the reactor. Hence, two containments are used at Necsa. High density polyethylene (HDPE) is a preferred material due to its durability and purity. It is not expensive except for some high purity qualities and costs of moulds to manufacture vial types that are specially made for application in trace analysis. Containers come in different forms such as snap-cap vials, screw-cap bottle, welded tubing and bags [(2)]. Examples are given in Figure A1. HDPE may easily be worked on by cutting and melting, its resistance to acid and basic media is very good, but polyethylene can easily be penetrated by many organic vapours and mercury. Because of its low melting point, it can only be used in irradiation facilities with temperatures below 80 ^{O}C. It is easily destroyed upon long irradiation times and high neutron fluxes, although the damage is not directly from neutrons but caused mainly by gamma radiation. At a neutron flux of about 10^{13} neutron.cm^{-2}.s^{-1} they should not be irradiated for more than two hours.

Figure A1 **Irradiation vials and the PRS rabbit**

A 4.2 Sample size

The usual size of samples is between 50 and 500 milligrams. However, for some materials this may prove to be too small due to inhomogeneities in element composition (e.g. gold in uranium ores). On the other hand samples of 10-100 grams require a suitable irradiation facility and counting infrastructure. With such large samples problems of neutron self shielding, gamma-ray self absorption and the neutron flux gradient over the sample become very stringent and specific experimental setups are required and procedures must be carefully elaborated to eliminate the errors introduced by these effects. Voluminous samples are mainly applied for activation analysis with isotopic neutron sources having a

low neutron flux and procedures of high precision and accuracy have been developed on that basis [2].

A 4.3 Sample shape

Instrumental neutron activation analysis can treat samples of different shapes as long as their dimensions do not exceed those of the irradiation containers. There are restrictions, since large pieces are not uniformly activated due to neutron flux gradient in the reactor or due to neutron self shielding. Asymmetric sample shapes have drawbacks that lead to geometric errors of gamma ray measurement which can not be precisely calculated [2]. The use of well-type crystals eliminates this problem as long as the sample is relatively small compared to the depth of the well.

A 5 Fast Rabbit Tube

To minimize delay between the end of the irradiation time and start of the measuring time samples have to be transported rapidly. Therefore pressure driven sample containers (rabbits) are usually used. Setbacks only arise from high accelerating forces when the rabbits are started and stopped in the irradiation and measuring positions. Malfunctioning of mechanical or opto-electronic sensing elements is often caused by a high radiation dose rate, mainly near the in-core stations. Fast rabbit tube systems provide automatic and fast separation of the sample (or inner pure container) from the transport capsule [2].

A 6 Reliability of Analytical Data

A valuable asset of a laboratory to determine its errors is the availability of Standard Reference Material (SRM). These reference materials offer several advantages and to a certain extent provide a check for sample handling. The IAEA provides laboratories with means to asses the analytical quality of data through the manufacturing of certified reference materials and participation in intercomparison studies. For daily quality control a laboratory usually develops its own in-house control standards [18].

A. 7 Measurement of Gamma Rays

The gamma-ray spectrometer is a fully integrated system for the qualitative and quantitative determination of gamma emitters [8]. A system should comprise of a high purity germanium detector (20-25% relative efficiency and full width at half max of around 2 keV), a lead shield (about 10 cm thickness) and spectrum accumulation and deconvolution hard- and software. The crystal should be operated at liquid nitrogen temperature, in order to improve the resolution but these detectors can be stored at room temperature. Because of their operational advantages they are now replacing lithium-drifted germanium detectors in nearly all applications. It is a highly sensitive and versatile research tool that can detect gamma-rays over a wide energy range of approximately 10 kiloelectronvolts (keV) to several megaelectronvolts (MeV). Most detectors have excellent energy resolution of about 180-200 electron volts (eV). To maximize the detection efficiency for low gamma-energies, the cryostat end-cap has a thin entrance window of beryllium or carbon fiber. Normally the detectors are designed to take advantage of the yield of low energy photons, while relatively small volumes are used to provide a low sensitivity to higher energy gamma rays of radioisotopes and background [2]. A gamma-ray spectrometer is subjected to performance control at least once a week. The routine involves the measurement of calibration sources and blank capsules. The spectrometer system may also be fitted with a sample changer to increase the counting capacity [15]. Detectors may be used in passive measurements (a ratemeter recording the radiation flux) or as flow-through detectors detecting radiation passing through the sensitive detection volume [16].The spectrometer system used in this work is provided with an automatic sample changer, a lead shielded store for up to 50 samples, an electro-

mechanically driven claw to feed the sample from the store to the counting position inside a 20% efficient well type detector and a cylindrical lead shield around the detector with a hole on top to allow access to the well. The irradiated samples, element standards, flux monitors and blanks are loaded in aluminium tubes, about 15mm Φ × 90mm long, with a rim on top (for the claws to catch the tube).The actual samples to be counted are sealed in counting containers (polyethylene vials), which are then fitted in the bottom of the aluminium tube. The transport arm is moved vertically and horizontally by a stepper-motor. Figure A2 shows the part of the sample storage and changing mechanism.

A.8 Quality assurance

Quality assurance entails quality control, quality assessment and maintenance of quality standards. Internal and external quality-assurance programmes are important in keeping satisfactory levels of performance (as a measure of quality assessment and control) in any laboratory [14]. The following are characteristics of quality assurance in INAA facilities:

- full documentation of standard operating procedures,
- database of previous experimental conditions,
- analyzed results, materials, detection limit,
- internal audits, customer satisfaction evaluation and non-conformance evaluation [15]

A.9 Calibration

^{152}Eu and ^{22}Na are used for the energy calibration of the spectrometers. Moreover well-type detectors require an experimental determination of the intensity ratios of all peaks since tabulated ratios can not be used due to coincidence summing effects. Calibrations are done using primary or secondary standards.[15]

Figure A2 **The sample storage and changing mechanism used in this investigation**