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A new rapid neutron activation analysis system at Dalat nuclear research reactor

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Abstract: An auto-pneumatic transfer system has been installed at the Dalat research reactor for rapid instrument neutron activation analysis based on very short-lived nuclides. This system can be used to perform short irradiations in seconds either in the vertical channel 13-2 or in the horizontal thermal column of the reactor. The transferring time of sample from irradiation to measurement position is approximately 3.2 seconds. A loss-free counting system using HPGE detector has been also setup in compacting with the pneumatic transfer system for measurement of sample's activity, automatically starting for data acquisition at irradiated sample's arrival. This new facility was tested and shown to have high potential for the determination of short-lived nuclides with half-lives from 10 ÷ 100 seconds. This work presents the results of timing parameter measurements, characterization of irradiation facilities, and application of this system to determining Selenium concentration in several biological reference materials.

Keywords: *Auto-pneumatic transfer system, neutron activation analysis, short-lived nuclides.*

I. INTRODUCTION

Instrumental neutron activation analysis (INAA) has been developed and applied at the 500 kW Dalat research reactor (DNRR) since 1984. Until now, it is capable of analyzing more than 40 elements based on radionuclides with short, medium and long-lived time. For short-lived nuclides with half-lives from 2 minutes to 2.6 hours, samples are often irradiated at the neutron channel No.7-1 of Dalat research reactor through a semi-auto pneumatic transfer system (PTS) with valid irradiation time from 45 seconds to 20 minutes. Measurements are often performed using a gamma spectrometer coupled with a HPGe (GMX-30190), but with manual manipulation between loading and counting procedures. Therefore, the shortest-lived nuclides that could be detected are ^{28}Al ($T_{1/2}$ = 2.24 min), ^{52}V ($T_{1/2}$ = 3.75 min), and ^{51}Ti ($T_{1/2}$ = 5.76 min).

In the recent years, through the IAEA TC Project RER/4/028, a new automatic PTS for rapid neutron activation analysis based on short-lived nuclides has been developed. This facility consists of three main parts introduced in reference [1]. The first part, consisting of two aluminum irradiation tubes, which are inserted into the vertical channel No.13-2 and the horizontal thermal column (TC) of the reactor. The second part is a digital signal processing spectrometer connected to a 40% relative efficiency HPGe detector coupled with a transistor reset preamplifier. The third part is composed of pneumatic chambers, loading and sliding devices in Cabin-1 which facilitates the

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fully automatic irradiation-counting procedures. It has also a sample automatic loader for the sequential routing of the samples in multi-samples operation mode: when the measurement for one sample is finished, the next sample is loaded and sent to the irradiation and then counting positions. In this system, there is also has optical sensors for controlling the transport of the capsule as a sample carrier, and for accurately measuring the capsule flight time from irradiation position to detector. The installation diagram is shown in Fig. 1.

This PTS system can be used to perform short irradiations in seconds. The return time of sample from irradiation position to counting position is about 3.2 s. Timing information for both irradiation and counting will be instantly delivered to the activation analysis workstation computer. The digital gamma spectrometer is selected and tuned for accurate measurement at high and varying counting rates, using loss-free counting technology. Accordingly, shorter-lived nuclides (half-life < 1 min) such as ^{20}F, ^{77m}Se, ^{179m}Hf, ^{46m}Sc, and ^{110}Ag can be used for INAA at Dalat reactor, which the former system can not detect.

The main purpose of this work is to test the system for both mechanical and analytical reliability. A systematic study has been carried out including measurements for timing parameters of the system and neutron flux at irradiation positions, and the application of this system to determining of Selenium in a number of biological reference materials for validation purpose.

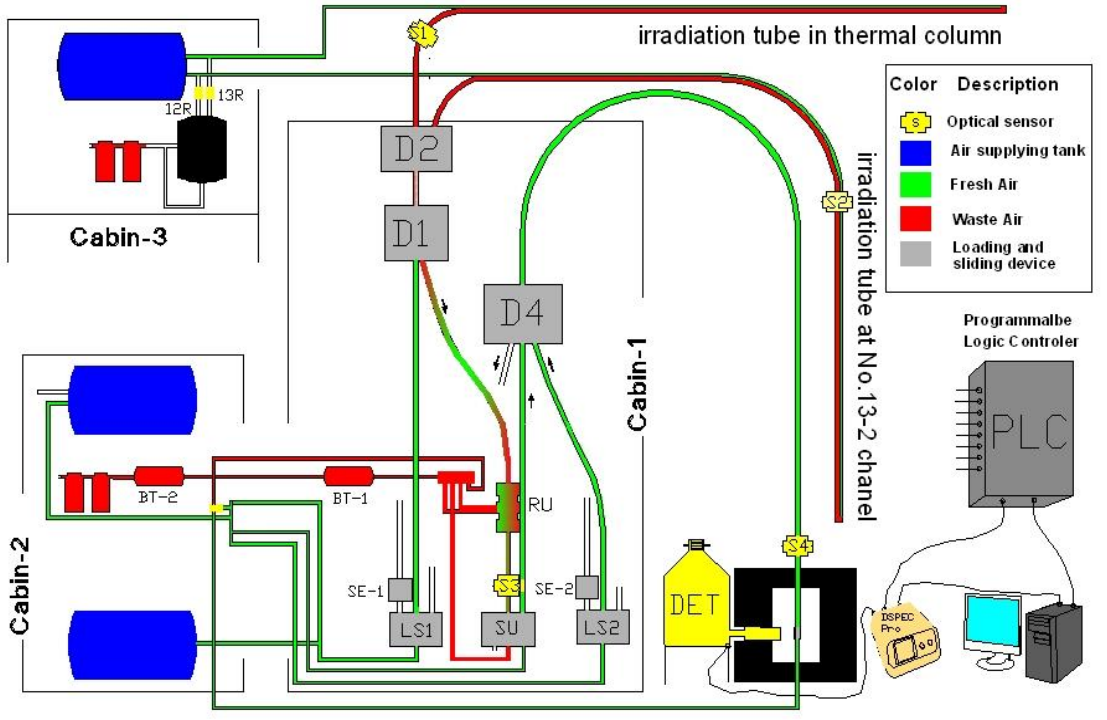

Fig. 1. Diagram of the auto-pneumatic transfer system installed at DNRR.

II. EXPERIMENT

A. Timing measurements

The accuracy of irradiation time of an irradiation facility should be checked and calibrated as a type of analytical qualify control [2]. In this experiment, the absolute irradiation time, the sample transferring time from entrance to irradiation position of aluminum tube (T_{in}) and the reverse movement (T_{out}) were determined. The experiments were done outside the reactor before installation of irradiation facilities inside the reactor. The arrangement of timing measurements is shown in Fig. 2. The setup includes two NaI(Tl) detectors placed at the top and bottom sides of the irradiation tube. The detectors detect gamma radiation pulses from a ^{131}I source inserted in a capsule while moving inside. The counters are set for running in Multi-Channel Scaler (MCS) mode; MCS mode records the counting rate of events as a function of time.

The return time from the irradiation to the measurement positions was determined by a series of irradiation (50 replicates) for a total weight (capsule, vial and sample) of about 4.4 gram and air pressure of 3.1 bars over a distance of 40 m for 13-2 channel and 36 m for thermal column. The return times were recorded at the computer via a signal from the fast solenoid providing the return gas (START of return time) and from the optical sensor at the detector station recording the arrival of capsule (END of return time).

B. Neutron spectrum parameters of irradiation position

The neutron spectrum parameters including thermal neutron flux ϕ_{th}, fast neutron flux ϕ_f, the thermal flux to epithermal neutron flux (ϕ_{epi}) ratio were measured at sample irradiation positions in channel No.13-2 and thermal column using Au, Zr, and Ni monitors. Monitors were inserted into a high purity polyethylene vial and loaded into rabbit (capsule) for irradiation. The activity measurements were carried out by a calibrated gamma-ray spectrometer combined with HPGe detector (GMX-30190). The measured spectrum were analyzed by using the k0-IAEA program. The irradiation, decay and counting times for each monitor are shown in Table I. Typically monitors with masses of 4 mg for Al-0.1%Au foil (IRMM-530R), 30 mg for pure Ni (wire), 10 mg for Zr (foil) were irradiated for 10 min at 13-2 channel (2 h at thermal column), and the decay time is 1 day for ^{97}Zr and 3 days for ^{198}Au, ^{95}Zr and ^{58}Co.

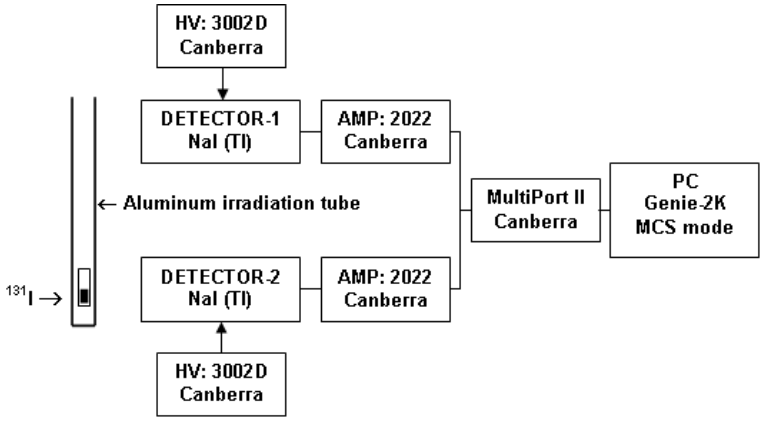

Fig 2. Arrangement for experiment of timing

Table I. The irradiation, decay and counting times for the monitors.

Time/position irradiation (monitor, mass)	Decay time	Counting time (combination)	Measured radionuclides ($T_{1/2}$, γ-rays in keV)
10 min/ 13-2 channel 2 hours/ thermal column (Al-0.1% Au, ~ 4 mg) (99.8% Zr, ~ 10 mg) (99.98% Ni, ~ 30 mg)	~ 1 d	1 ÷ 2 h	^{97m}Nb (60 s, 743.4)*; ^{97}Nb (16.7 h, 657.9)
	~ 3 d	0.5 ÷ 3 h (5 h)	^{198}Au (2.7 d, 411.8); ^{95}Zr (64 d, 765.8); ^{58}Co (70.8 d, 810.8)

* Nuclide ^{97m}Nb is decayed from nuclide ^{97}Zr with half-life of 16.7h.

C. Determination of Selenium

A variety of reference materials (Tuna Fish IAEA-436, Oyster tissue NIST 1566b, Bovine Liver NIST 1577, Bovine Liver NIST 1577b) were selected to assess reliability of this system on the short-time activation application. All of the samples were irradiated at a neutron flux of 4.2×10^{12} n.cm^{-2}.s^{-1} in the 13-2 channel and counted on the calibrated HPGe gamma-ray spectrometer (GMX40-76-PL).

In order to evaluate the limit of detection of Se in biological samples, two 200mg replicates of each material (IAEA 436 and NIST 1566b) were weighed and packed in high purity polyethylene bags. The samples were irradiated for 5, 10, 15, 20, 25, 30, 35 and 40 s. After a delay of 3.2 s (including both transferring time of sample from irradiation position to detector and the time required to start the detector). Each sample were counted for 20 s at a distance of 10 cm from detector.

To test accuracy for the analysis of the Se concentration in biological reference materials, four 200 mg replicates of each material (IAEA 436, NIST 1566b, NIST 1577 and NIST 1577b) were weighed. The samples were irradiated for 25 s, allowed 20 s delay time to eliminate interference of ^{116m}In with a half-life of 2.18 s [3, 4]) and counted for 25 s at a distance of 10 cm from the detector (GMX40-76-PL). The concentrations of Selenium were determined by both k-zero and relative methods.

III. RESULTS AND DISSCUSION

A. Timing measurements

The results for average transferring time of sample from the top to bottom of the aluminum irradiation tube (T_{in}) is (0.628 ± 0.021) s for the channel No.13-2 irradiation tube (a length of 6 m) and T_{out} is (0.323 ± 0.030) s (averaged for 90 runs over the three days). For thermal column irradiation tube (a length of 2.8 m), T_{in} is (0.248 ± 0.019) s and T_{out} is (0.146 ± 0.004) s, as shown in Table II. The result obtained for measuring the return time from the irradiation position to the measurement position was found to be (3.165 ± 0.002) s for channel No.13-2. That for thermal column was (3.025 ± 0.013) s. It should be noted that this timing parameters are included in the time required to start the detector after receiving the start signal.

Table II. The result of time measurements.

Irradiation position	The transferring time throughout aluminum irradiation tube (second)		The return time from irradiation position to detector position (second)	
	T_{in}	T_{out}	This word	Manufacturer*
13-2 channel	0.628 ± 0.021	0.323 ± 0.030	3.165 ± 0.002	3.301 ± 0.013
Thermal column	0.248 ± 0.019	0.146 ± 0.004	3.025 ± 0.013	3.261 ± 0.022

** Sample weight: ~ 8 g for thermal column tube and ~ 6 g for 13-2 channel tube, operation air pressure: ~3.1 bars, distance: 30 meters.*

There are significant differences between this work and that of the manufacturer in capsule sample weight and distance from irradiation position to measurement position. Hence, there are differences (~ 7%) in the result of the return time from irradiation position to detector position. However, it is not a problem for analytical measurements.

Results for absolute irradiation time at channel No.13-2 and thermal column were determined by a series of irradiations ranging from 1 to 30 s (3 replicates), as shown in Fig. 3 and Fig. 4. The relative error of irradiation time in the first second is 16.02% for channel No.13-2 and 26.43% for thermal column, and those for irradiation time of 2 s is 1.5% for channel No.13-2 and 4.91% for thermal column. The relative error is less than 1% at irradiation time of 5 s for channel No.13-2, and 10 s for thermal column. The large error for the first second is due to delay of the system in starting the irradiation timer and in ejecting the capsule once the "end of irradiation" signal has been received.

This timing delay problem can be adjusted through the control unit and the software package for managing optimal operation and the analytical procedures. However, it is not a problem for INAA because the time parameters remain unchanged for all samples, standards, and control material

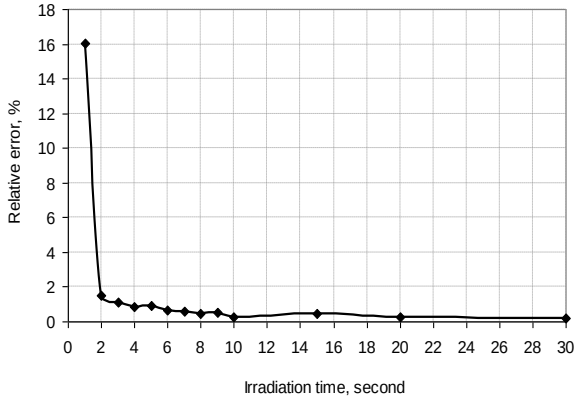

Fig. 3. The relative error of irradiation time for 13-2 channel.

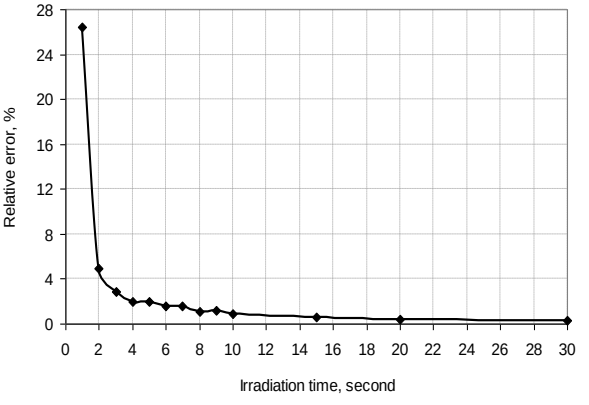

Fig. 4. The relative error of irradiation time for thermal column.

B. Neutron spectrum parameters of irradiation positions

The results of the determination of neutron spectra parameters are shown in Table III. This table includes data obtained for the thermal, fast neutron flux, the ratio of thermal to epithermal neutron flux (ϕ_{th}/ϕ_{epi}). The thermal neutron flux at the irradiation position in the channel No. 13-2 is 4.2E+12 n.cm^2.s^{-1}, and associated with 0.5 times of epithermal. The integral fast neutron flux is 6.61E+12 n.cm^{-2}.s^{-1} for all neutrons above 2.9MeV in energy [5], measured using the ^{58}Ni(n,p)^{58}Co nuclear reaction. The thermal neutron flux at the irradiation position in the thermal column is 1.25E+11 n.cm^2.s^{-1}, associated with much lower fast and epithermal neutron flux. Hence, thermal column is a useful irradiation channel for eliminating interference reactions induced by fast neutron, in which sample is irradiated in an extremely well thermalized neutron field [6].

Table III. The results of neutron spectra parameters at irradiation positions in the channel No.13-2 and thermal column of DNRR.

Irradiation position	ϕ_{th} (n/cm^2/s)	ϕ_F (n/cm^2/s)	ϕ_{th} / ϕ_{epi}
13-2 channel	(4.2 ± 0.1) x 10^{12}	(6.6 ± 0.9) x 10^{12}	10.7 ± 2.4
Thermal column	(1.24 ± 0.03) x 10^{11}	(8.4 ± 0.5) x 10^8	195 ± 4

C. Determination of Selenium

Finally, measurements of detection limits of Se in IAEA 436 and NIST 1566b samples were performed. The results for these measurements are presented in Fig 4. The obtained results confirm that in irradiation from 15 s to 25 s at irradiation position of the channel No.13-2 coupled with counting for roughly 20 s at 10 cm distance from detector, the detection limits for Se is within the range 0.5 ÷ 0.7 ppm, depending on the sample composition. It provides adequate analytical sensitivities for Se rapid determination in a variety of biological matrices.

The accuracy for determination of Selenium using the short-lived nuclide ^{77m}Se was evaluated by analyzing a number of certified reference materials with different levels of Se (IAEA 436, NIST 1566b, NIST 1577 and NIST 1577b). The agreement between measured and certified values was generally very good with u-score < 1.64, as shown in Table IV.

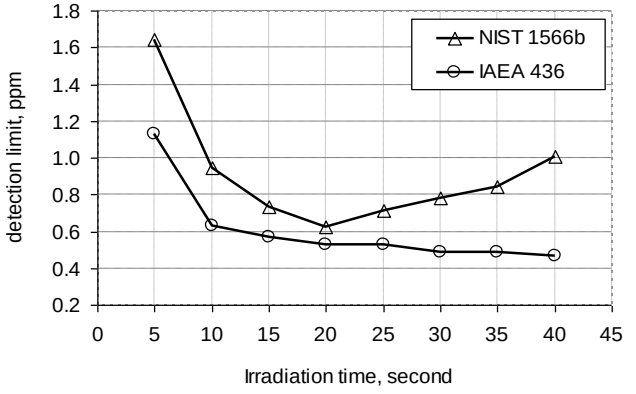

Fig. 4. The detection limits of Se in IEAE 436 and 1566b.

For the determination of the Selenium by the instrumental neutron activation analysis, the long-lived nuclide ^{75}Se or the short-lived nuclide ^{77m}Se can be used [7]. With the short-lived nuclide, not only completion times are a distinct advantage but analytical sensitivities are also improved. The data for procedures are listed in Table V.

Table IV. The results of concentration analysis for Se in biological reference materials.

Reference material	Certificated value (in ppm)	k-zero method		The relative method	
		This work (in ppm)	u-score	This work (in ppm)	u-score
IAEA 463	4.63 ± 0.48	4.55 ± 0.50	0.12	4.19 ± 0.46	0.66
NIST 1566b	2.06 ± 0.15	2.48 ± 0.57	0.71	2.18 ± 0.42	0.27
NIST 1577	1.10 ± 0.10	1.24 ± 0.31	0.43	1.17 ± 0.22	0.29
NIST 1577b	0.73 ± 0.06	0.70 ± 0.11	0.24	0.80 ± 0.17	0.39

Table V. Parameters were used for INAA analysis of Selenium in biological sample by using ^{77m}Se and ^{75}Se isotopes.

Radionuclide	^{75}Se	^{77m}Se
Half-life	120 d	17.4 s
Activation	20 h at 3.5 x 10^{12} (n/cm^2/s)	15÷25 s at 4.2 x 10^{12} (n/cm^2/s)
Decay time	20 d	20 s
Counting time	2÷3 h	25 s
Detection limit	1.4 ppm	0.6 ppm
Sample: IAEA 436	4.63 ± 0.48 ppm	4.63 ± 0.48 ppm
The results	4.35 ± 1.1 ppm	4.19 ± 0.46 ppm

IV. CONCLUSION

A fast pneumatic sample transfer system for analyzing of extremely short-lived nuclides by neutron activation analysis has been installed and operated at Dalat nuclear research reactor. In this study, time parameters of the system were calibrated, thereby reducing irradiation time to seconds with precision. Neutron spectra parameters of the thermal column and channel No.13-2 were also determined in order to establish analytical procedures using the k0-NAA method. The system was applied to determine the concentration of Se in the biological sample by using the short-lived nuclide ^{77m}Se. The results obtained through this research have opened a new possibility on using INAA technique for measurement of extremely short-lived nuclides at Nuclear Research Institute.

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