CHARACTERISTICS OF A NEW PNEUMATIC TRANSFER SYSTEM FOR A NEUTRON ACTIVATION ANALYSIS AT THE HANARO RESEARCH REACTOR

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A rapid pneumatic transfer system (PTS) for an instrumental neutron activation analysis (INAA) is developed as an automatic irradiation facility involving the measurement of a short half-life nuclide and a delayed neutron counting system. Three new PTS designs with improved functions were constructed at the HANARO research reactor in 2006. The new system is composed of a manual system and an automatic system for both an INAA and a delayed neutron activation analysis (DNAA). The design and basic conception of a modified PTS are described, and the functions of system operation and control, radiation protection and emissions of radioactive gas are improved. In addition, a form of capsule transportation of these systems is tested. The experimental results pertaining to the irradiation characteristics with variation of the neutron flux and the temperature of the irradiation position with the irradiation time are presented, as is an analysis of the reference material for analytical quality control and uncertainty assessments.

KEYWORDS : HANARO Research Reactor, Instrumental Neutron Activation Analysis, Neutron Irradiation Facility, Pneumatic Transfer System, Delayed Neutron Counting System, Analytical Quality Control, Measurement Uncertainty

1. INTRODUCTION

The pneumatic transfer system (PTS) of a research reactor is an important facility that is used for the irradiation of a target material for an instrumental neutron activation analysis (INAA). In particular, a rapid pneumatic transfer system is essential for accurate measurements of short half-life nuclides from 1 to 100 seconds and for accurate assessments of the delayed neutrons produced by fission reactions [1,2]. The new PTS was designed as a multipurpose system involving both a manual system (PTS #1, 3) and an automatic system (PTS #2) for a conventional INAA and a delayed neutron activation analysis (DNAA) for the analysis of the fissile materials ^{235}U and ^{239}Pu. These components were reconstructed in three NAA irradiation holes at the HANARO research reactor in 2006 [3-8].

This system consists of many devices and assemblies for sample loading, sending the irradiation capsules from the NAA laboratory to the three holes in the reflector tank of the reactor, retrieval of the irradiated capsules after irradiation, monitoring of system control, and the automatic counting of the background radiation level of delayed neutrons and gamma-rays. PTS #1 and PTS #2 were reinstalled with new designs for the promotion of operational safety, efficient maintenance and an improvement of the irradiation functions. The irradiation tube assemblies of the PTSs were installed at three NAA holes in the reflector tank of the reactor, as shown in Fig. 1. For its safety and to maintain the system, the temperature at the irradiation position involving the use of a polyethylene (PE) capsule must be limited to less than 80 °C because the melting point of PE is approximately 120 °C The temperature of the irradiation sites can be measured with the irradiation time using a thermo-label attached to the inside and surface of the capsule. Consequently, the optimum irradiation time for the capsule in the PTS will depend on the actual temperature of the irradiation period with the irradiation position. The basic requirement for irradiation of the PTSs is based on the parameters of the neutron flux and distribution, temperature, gamma heating of the irradiation position, the radiation dose rate, the types and materials of the irradiation capsule, and the type and size of the sample used in a quantitative elemental analysis under the safe operation of the reactor [9-13].

On the other hand, a quantitative analysis of major, minor and micro component elements as impurities in

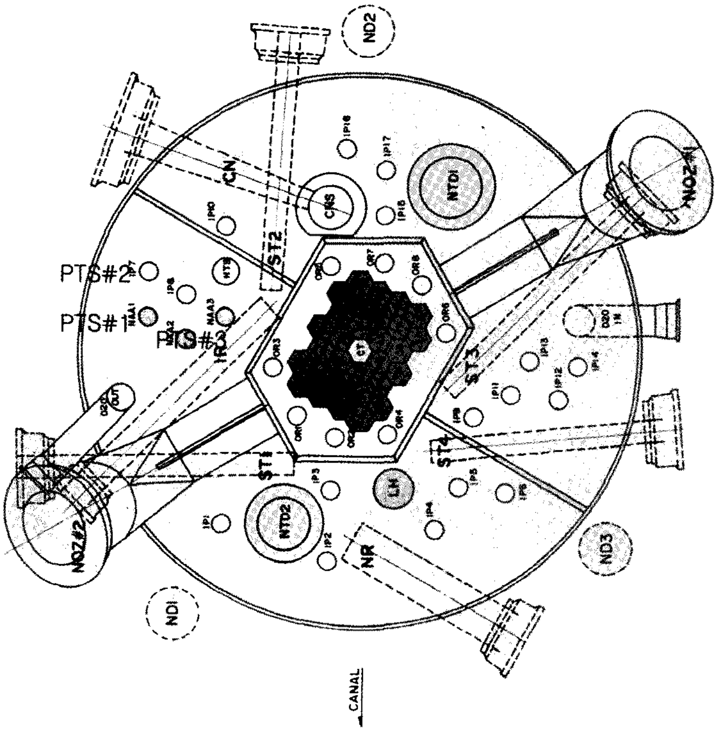

Fig. 1. Irradiation Sites of the PTS #1, 2, 3 Installed at the HANARO Research Reactor

materials is essential in many fields of basic science and technology as well as in commercial and industrial fields. In particular, a direct analysis of a sample offers a more effective investigation method in these fields. Reactor-based instrumental neutron activation analysis (INAA) has an inherent advantage of being a non-destructive, simultaneous multi-elemental analysis with high levels of accuracy and sensitivity. However, when the specific volume of a sample is to be analyzed using an INAA, the neutron flux distribution with the geometric position within a reactor irradiation hole, the irradiation capsule and the type of spatial matrix are two of the major sources of a measurement uncertainty in an activation method. Generally, the overall uncertainty of the analytical results, which is expressed as the root-sum-of-squares of an individual standard uncertainty in the measurement, contains several sources of random and systematic errors. These may be listed separately in the overall error analysis or in a combined uncertainty according to some defensible algorithms [14,15]. In addition, accurate irradiation characteristic data of an irradiation facility should be checked and monitored periodically as a type of analytical quality control for the management of the accreditation of the pertinent laboratory.

In the present work, the new designs of the PTS are initially described, and the irradiation facilities used for the irradiation of a target and for the operation and control of these systems are presented together with future applications. In addition, the experimental results of the irradiation characteristics as they pertain to the neutron flux and its distribution with the geometry of the sample in the capsule as well as the measurements of reference samples in three PTSs are reported as information to be used with analytical quality control and uncertainty assessments.

2. METHODS AND RESULTS

2.1 Design of a PTS

This system consists of many devices and assemblies for the loading of a sample capsule, the sending of irradiation capsules from the NAA laboratory into three irradiation holes in the reflector tank of the reactor, the retrieval of the irradiated capsules after irradiation of a sample normally and abnormally, and a change of the transferring path and the automatic counting of the radiation emitted from an activated sample. The system is operated in manual and automatic modes by a programmable logic controller (PLC) and a personal computer (PC) controller. As shown in Fig. 2 and Fig. 3, there are six components of a PTS, as follows: a) an irradiation and transfer system, b) a N_2 gas supply and exhaust systems, c) an emergency withdraw system, d) a radiation shielding system, e) a DNAA counting system, and f) an operational control system.

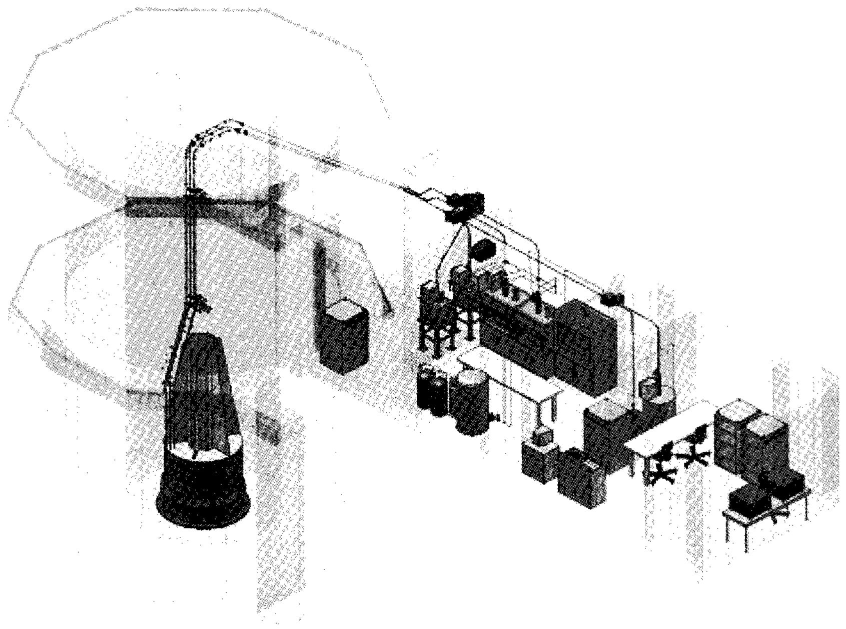

Fig. 2. Birdseye-view of the HANARO NAA Facility

2.2 Construction of a PTS

The new irradiation tubes of the PTS, which consist of Al6061 material, are installed at two NAA holes (NAA 1 & 2). To increase the water cooling efficiency, the structure of the irradiation tubes installed at PTS #1 and PTS #2 was changed to a single tube, which was in direct contact with the cooling water in the reflector tank of the reactor from the double jacket tubes of the old type. However, the double jacket type of irradiation tube of PTS #3, which is close to the reactor core, is only used for short irradiation times of less than 60 seconds due to the heating of the tube by neutrons and gamma-rays. The expanded length of the irradiation tube is approximately 14 m from the bottom of the core to the top of the reactor, but the length of the real irradiation part is about 50 cm from the irradiation end of the tube. For the safety analysis of the irradiation tube assembly of a PTS, which is designated as a seismic II class under the conditions of reactor operation, a structural safety analysis of the irradiation and transfer tubes has been performed. The allowable stress and Young modulus (Moduli of elasticity) for the irradiation tube assembly and the coefficient of thermal expansion between low- and high-temperature conditions (20 to 200 °C) were evaluated according to the given material properties for Al 6061-T6. [16] A rubber gasket made of ethylene-propylyne-diene rubber [17], used to connect the tubes in the reactor pool, was validated in quality assurance tests.

The length of the transfer tube, which consists of SUS304, has a range of 28 to 35 m between the end of the irradiation tube and each shielding receiver in the NAA laboratory. The outer and inner diameters of the transfer tube are 34.1 and 27.5 mm, respectively. After the installation of a PTS is completed, a leak test for all of the lines of the system is carried out at a pressure of 10 psi for 30 min.

The sending and receiving of a capsule in a PTS is controlled by a system controller connected to a programmable logic controller (PLC) and a personal computer with a preset timer, manually or automatically. In addition, PTS #2 for both an INAA and a DNAA is composed of an automatic irradiation and counter system which is operated by a PLC and a PC connected with a MCA coupled to eighteen ^{3}He proportional counters for the measurement of the delayed neutrons.

2.3 Transportation of a Capsule

The transportation of a capsule could be observed by photo-sensors (PS), which are located at the upper part of the loaders and the irradiation tubes and at the front of the transfer tube connected to the diverter, as shown in the layout of the PTS in Fig. 3. To obtain an accurate and precise irradiation time, the transfer time of a capsule is measured by an acoustic method in both the manual and automatic modes of the controller. The N_2 gas pressure of the PTS lines was adjusted to within a range of 20 to 35 psi. Irradiation capsules consist of high-purity high-density polyethylene, as shown in Fig. 4. The total weight and volume were 9.8 g and approximately 27 ml, respectively. The average sending time of a capsule filled with a weighted material (of about 5 g) to the reactor was in the range of 9.1~12.5 seconds. The average receiving

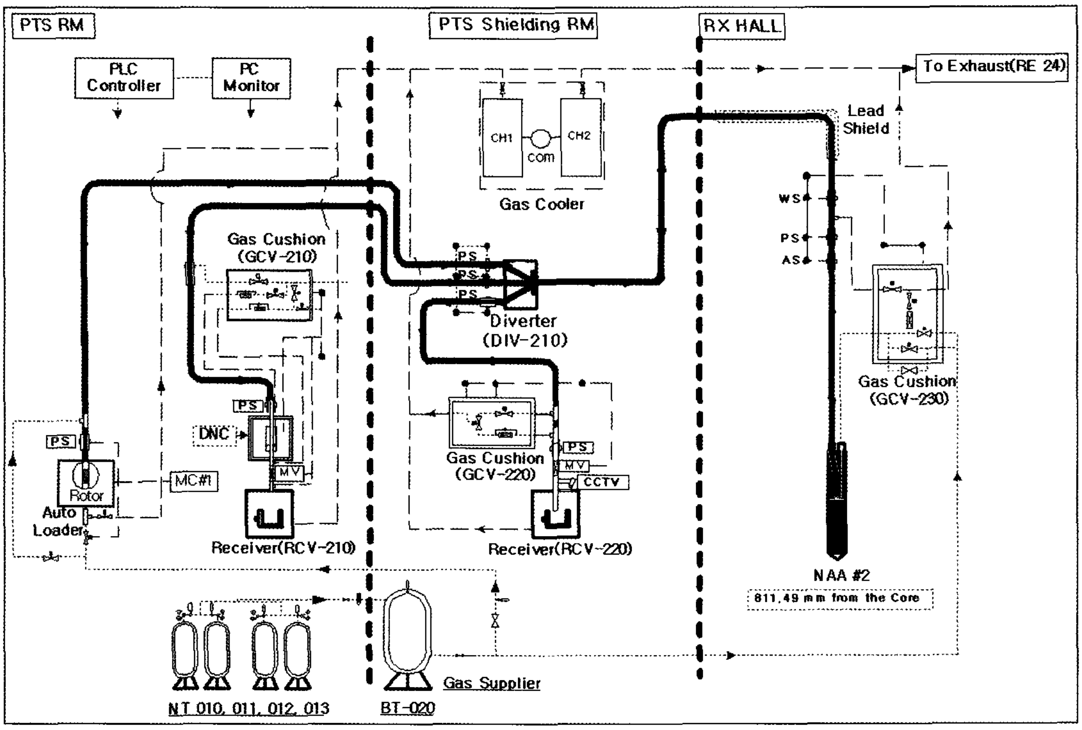

Fig. 3. Layout of PTS #2 at the HANARO Research Reactor

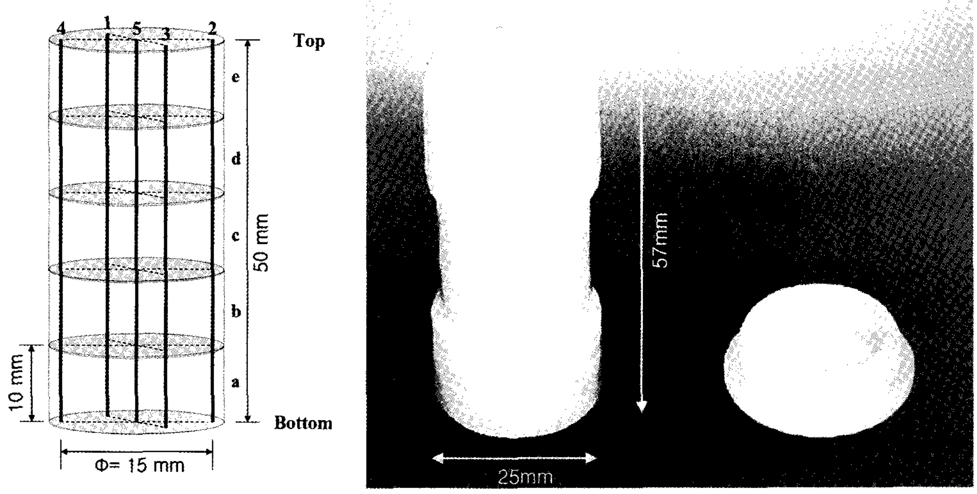

Fig. 4. Geometries of the Flux Monitor and Irradiation Capsule Used

time upon its return to the receiver was in the range of 5.7~6.1 seconds. The standard deviations of the sending and receiving times were less than 1.6% and 1.4%, respectively. The sending time is longer than the receiving time because the capsule is slowed down by air cushion devices to protect against an impact of the capsule in the irradiation tube within the reactor.

2.4 Characteristics of the Irradiation Positions

The requirements for neutron irradiation in the PTS are based on the parameters of the neutron flux and distribution, the temperature, the gamma heating of the irradiation site, the radiation dose rate, the types and materials of the capsule, and the type of sample for the safe operation of the reactor [12,13]. Therefore, information pertaining to the condition and damage to an irradiation tube and a transfer tube in the reactor pool is very important for the safe operation of the reactor. The gamma heating rate in the irradiation tube under an air condition was estimated to be close to 5 Watts·g^{-1}.

2.4.1 Temperature and Transfer Time

Measurement of the temperature at the irradiation position should be checked for the safety of the PE capsule. As the melting point of the PE capsule rabbit is close to 120 °C, the temperature at the irradiation position has to be held below 80 °C within the given irradiation period. The temperature at the irradiation positions at 30MW of thermal power was measured with the irradiation time using a thermo-label attached to the inside and surface of the capsule. In PTS #1 and #2, when the capsule is irradiated for 1 to 200 min., the measured temperature was in the range of 50 to 65 °C The possible irradiation time of a sample was estimated to be 3 hours. The water cooling efficiency of the irradiation tube was improved by a change of the structure from a double jacket tube to a single tube. In the old PTS #3 tube, when the capsule is irradiated for 10 sec. to 80 sec., the measured temperature was in the range of 50 to 80 °C and the normal irradiation time was limited to less then 60 s. That is, the optimum irradiation time for a PTS depends on the actual temperature at the irradiation position in conjunction with the irradiation period.

2.4.2 Radiation Monitoring

The emission of the radioactive gas used in a PTS should be minimized to control radiation from entering the environmental. Thus, a radiation monitoring system of the noble gases and the work area was implemented. The sample loader of the new PTS is always purged using N_2 gas to remove any existing air before the irradiation of a sample for the control of gas activation. The radiation level of the noble gas exhausted from the PTS is checked by the radiation monitoring system (RMS) of HANARO, and the value is maintained at less than 10^{-5} μ Ci/cm^3 at the stack monitor (RE024). When an irradiated capsule is returned, the surveyed radiation dose rate at the area outside of the receiver was in the range of 20 to 70 μSvh^{-1}, whereas that of the transfer line in the reactor hall was less than 15 μSvh^{-1}. Hence, radiation protection in the work area was assured.

2.4.3 Neutron Flux Monitoring

Neutron flux monitoring is carried out regularly for analytical quality control. When a sample is irradiated with neutrons, the activation rates depend on the geometry effect due to the irradiation position within the capsule, the flux variation, and the differences with the irradiation site and thermal power of the reactor [12,13]. Particularly, the main sources of measurement uncertainty in an INAA are parameters such as neutron self-shielding and self-absorption, flux variation within a sample, the irradiation geometry in the capsule, and the irradiation position. When the *ko*-NAA standardization method is applied for the quantitative analysis, the information on the neutron flux and its distribution in the reactor are necessary. Relevant parameters for the irradiation of the sample must be obtained experimentally from measurements in the irradiation location of the reactor. These are used for the determination of a sample [18,19].

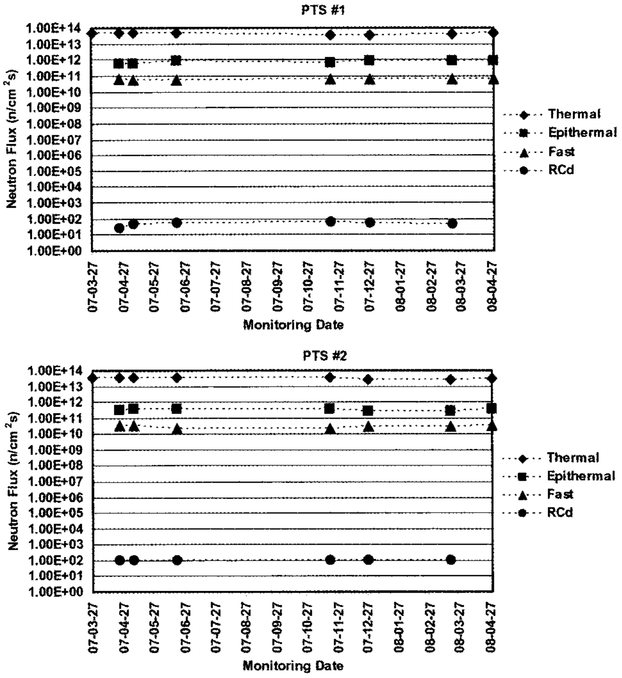

Fig. 5. Neutron Flux Monitoring with Operation Cycles of Irradiation sites

For neutron flux monitoring and the measurement of the cadmium ratio, activation wires (R/X activation wire, Reactor Exp. Inc.) of Au-Al, Co, Fe, Ni and Zr and Cd box were used. The measurements were carried out using a calibrated gamma-ray spectrometer (HP-Ge detector, GEM 35185P and 919A MCB, Gamma Vision software, EG&G ORTEC, USA). The calculation was carried out using the new Windows PC-code, Labview software of KAERI with a nuclear data library [20,21]. This program was developed by the NAA laboratory for rapid and simple data treatment for a gamma-ray spectrum obtained from preset detection conditions.

The monitoring data for the neutron flux and the distribution of the irradiation site comprise the basic requirements for an INAA due to the variable conditions of the reactor operation. Fig. 5 shows the variation of the flux monitoring for the thermal, epithermal and fast neutrons along with the cadmium ratio of the irradiation sites at PTS #1 and PTS # 2 within the operation cycles.

Table 1. Average neutron Fluxes and Cadmium Ratio Measured at the NAA Irradiation Holes (30 MW)

Irradiation Hole (PTS#)	Neutron Flux (n/cm^{2}s)			Cadmium Ratio (Au)
	Thermal, Φ_t	Epithermal, Φ_e	Fast, Φ_f	
NAA 1 (PTS #1)	$4.80 \pm 0.02 \times 10^{13}$	$7.80 \pm 0.22 \times 10^{11}$	$6.38 \pm 0.49 \times 10^{10}$	48±2
NAA 2 (PTS #2)	$3.30 \pm 0.09 \times 10^{13}$	$3.44 \pm 0.29 \times 10^{11}$	$3.27 \pm 0.47 \times 10^{10}$	100±2
NAA 3 (PTS #3)	$1.53 \pm 0.06 \times 10^{14}$	$1.01 \pm 0.07 \times 10^{12}$	$9.78 \pm 0.05 \times 10^{11}$	10±1

The standard deviations of the thermal, epithermal and fast neutron fluxes measured in PTS #1 within the monitoring period were 0.30, 0.28 and 7.71%, respectively. The standard deviations of the thermal, epithermal and fast neutron fluxes measured in PTS #2 within the monitoring period were 2.98, 0.84 and 2.48%, respectively. The variations of the neutron fluxes are reasonable considering the given the capsule geometric error. The thermal, epithermal and fast neutron flux together with the cadmium ratio, R_{Cd}, of PTS #1, PTS #2 and PTS #3 at a thermal power of 30 MW were measured, and the average values with the standard deviations are summarized in Table 1. These results of the neutron flux distribution can be used for an evaluation of the standard uncertainty. Moreover, when a sample is irradiated, estimation of the proper irradiation time with the sample matrix and geometry correction of a sample can be used.

2.4.4 Neutron Variation within the Capsule

An evaluation of the thermal neutron flux variation along with the height (axial gradient) and width (radial gradient) of the geometry for an irradiation capsule was performed. That is, the flux distribution of the capsule at 10 mm intervals in length and 5 mm intervals in width were measured using an 0.01% Au-Al wire monitor (5.6 mg, R/X activation wire, Reactor Exp. Inc.), as shown Fig. 4. The measured values for the flux variation within the capsule geometry at the irradiation positions of three pneumatic transfer systems are shown in Fig. 6. The radial deviations for PTS #1, PTS #2 and PTS #3 in the PE matrix are 2.84, 11.6 and 2.84%, respectively. The radial positions are indicated arbitrarily as it was impossible to control the angular orientation of a sample during the transport of a capsule. The axial deviations for PTS #1, PTS #2 and PTS #3 in the PE matrix are 4.20, 3.84 and 1.71%, respectively. The thermal neutron flux of the radial gradient is somewhat higher than the axial gradient for the irradiation positions in a capsule. The differences in the average thermal neutron flux values for PTS #1, PTS #2 and PTS #3 with the PE matrix were 4.4, 1.2 and 7.4%, respectively. These values are less than those without a PE matrix (as a blank). These geometric error results in a capsule can be used for evaluations of the standard uncertainty given the sample volume and size.

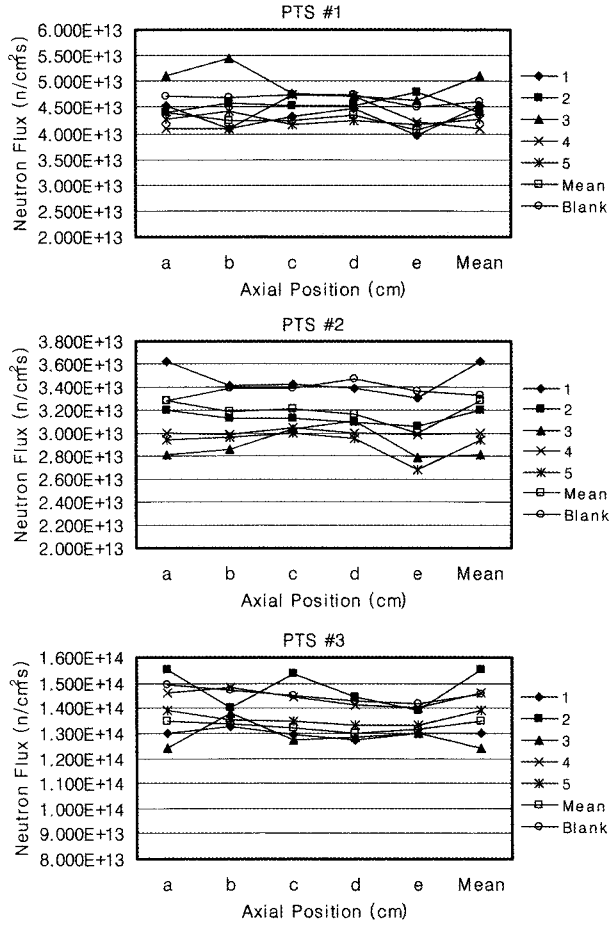

Fig. 6. Radial and Axial Flux Gradient within the Irradiation Capsule in the PTS

2.4.5 Measurement of Sample

For an evaluation of the accuracy and precision of a measurement, a repetitive determination of a selected short half-lived nuclide was carried out using certified reference materials (NIST SRM 1400- Bone ash). Powdered reference samples of approximately 20~30 mg in weight were prepared. Under the same irradiation and measurement conditions (Ti: 20s, Td: 5~10m, Tc: 5~10m), the amounts of

Table 2. Determination of Selected Short Half-lived Nuclide Using Certified Reference Materials. (NIST SRM 1400, Bone ash)

Element	Nuclide	Half-life	Elemental Concentration (mg/kg)						Certified value± (SD)
			PTS#1		PTS#2		PTS#3		
			Mean±SD(RSD)	RE(%)	Mean±SD(RSD)	RE(%)	Mean±SD(RSD)	RE(%)	
Al	^{28}Al	2.24 m	519±23(4.46)	2.06	510±9(1.69)	3.75	545±31(5.73)	2.82	530
Ca	^{49}Ca	8.72 m	364700±26100(7.15)	4.48	369500±6740(1.82)	3.23	366800±11710(3.19)	3.92	381800±1300
Mg	^{27}Mg	9.46 m	6460±470(7.33)	5.60	6840±220(3.16)	0.04	6840±440(6.38)	0.01	6840±130
Na	^{24}Na	14.96 h	6140±650(10.6)	2.40	6360±140(2.15)	5.94	6330±170(2.67)	5.44	6000

^{28}Al, ^{49}Ca, ^{27}Mg, and ^{24}Na in the samples were analyzed. These results are presented in Table 2. The maximum of the standard deviation and the relative error of the measured elements using the new irradiation systems were found to be less than 11% and 6%, respectively. It was assumed that there is a slight difference in the systematic error and reproducibility between the three irradiation systems, and the analytical results including the predominant uncertainties of the transfer time, neutron fluences and geometric parameters are reasonable for the quality control of the system.

3. CONCLUSION

To promote the effective use of a new irradiation facility through an improvement of the PTS for a NAA at the HANARO research reactor, a functional test of the system and an irradiation characteristic examination with regard to the irradiation positions were carried out. In addition, the measured results of the parameters of the transfer time, the neutron flux, the temperatures of the irradiation position with the irradiation time, and the radiation dose rate when the irradiation capsule is returned were reported for an analytical quality control and uncertainty assessment of the irradiation system. The results may be used as a reference or as a correction factor for the calculation of accurate and precise elemental contents. They may also have a wider range of application for a NAA in many fields [20]. It is concluded that the geometric effect of a sample in an irradiation capsule must be considered for a more accurate and precise quantitative analysis whenever a sample is irradiated.

However, the new PTS will be used in an instrumental neutron activation analysis which consists of a delayed neutron activation analysis during a nuclear fission reaction. In addition, the system can be used for the production of a radioactive tracer, in an irradiation test of several materials, or as a nuclear fission tracking method.

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