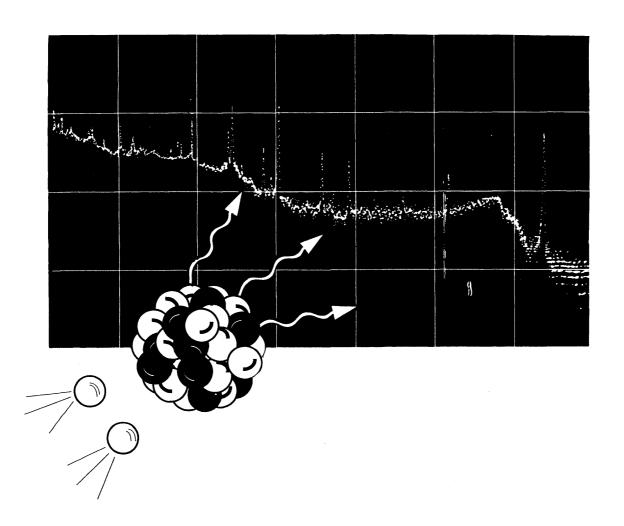
# 放射化分析

No.19



放射化分析研究会

# 放射化分析研究会 Japan Association of Activation Analysis (JA³)

会誌 「Activation Analysis」

表紙図案は 岡田往子さん(武蔵工大・工学部)によるものです。

# 放射化分析 No.19

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# 特集

### 韓国原子力研究所

#### はじめに

首都大学東京都市教養学部 大浦泰嗣

ここ数年毎年、アジア各国の原子力研究所の研究炉を見学していますが、アジア各国での放射化分析の役割はまだまだ大変大きいと感じます。見学した中で韓国原子力研究所(KAERI)は、日本に勝るとも劣らないもっとも充実した施設でした。2003 年に韓国放射化分析研究会(KA3)が発足し、最近、JA3-KA3 の協力について検討されています。また、京大炉の休止にともない KAERI の研究炉 HANARO を利用した共同利用が計画されています。そこで、海外の放射化分析関連施設紹介の第一弾として、韓国原子力研究所の紹介を企画しました。

本特集に当たり KAERI の NAA グループリーダーである Yong-Sam Chung 氏に紹介内 容の検討や執筆者の人選をしていただき、編集には NAA グループの Hyun-Je Cho 氏と Jong-Hwa Moon 氏が協力してくださいました.

本特集は下記8論文からなります.

- 1. 韓国放射化分析研究会(KA3)の紹介. Jin-Hong Lee
- 2. 韓国研究炉 HANARO の放射化分析施設とその利用. Jong-Hwa Moon et al.
- 3. 韓国研究炉 HANARO の即発γ線分析装置. Hyun-Je Cho et al.
- 4. 中性子放射化分析による老齢韓国人の食事の研究. Seung Y. Cho et al.
- 5. 中性子放射化分析を用いた韓国 Katap-Ri の屋根瓦と陶器の出所. Kwang-Yong Chung et al.
- 6. XRFと INAA を用いたポリエチレン固体中の環境有害元素の定量分析.

Chan-Soo Park et al.

- 7. 機器的中性子放射化分析法を用いた韓国における微小大気浮遊粒子中の微量元素 Jong-Myoing Lim et al.
- 8. HANARO 研究炉における PGAA の応用 Hyun-Je Cho et al.

1は、韓国放射化分析研究会の活動状況の紹介で、2と3は、HANAROでの放射化分析施設と即発γ線分析装置の紹介です。4~7はNAAの健康学、考古学、環境科学への応用で、4は60歳以上の老人の食事を分析し、食事から一日に摂取する各元素の量を調べていま

す. 5 は韓国南部で出土した屋根瓦と陶器の製造地を元素組成から推定しています. 6 はポリエチレン標準試料を作製し、その濃度と均一性を INAA で調べた後、XRF 法を適用し、その精度と確度を調べています. 7 は KEARI のある大田市で 2000 年から 2001 年に採集した大気浮遊粒子 PM2.5 の元素組成を定量し、因子分析 と濃縮係数分析を行なっています. 8 は PGA の応用で、生体試料と金属試料の水素とホウ素の定量について検討しています.

本特集はJA3 とKA3 の初めてのコラボレーションです.少なくとも HANARO 共同利用申請の参考になると幸いです.

#### **Introduction to Korean Association of Activation Analysis (KA3)**

Prof. Jin-Hong Lee

Chairman, Korean Association of Activation Analysis

Department of Environmental Engineering, Chungnam National University, Daejeon 305-764, Korea

The utilization of Neutron Activation Analysis (NAA) in Korea was implemented after the operation of the TRIGA Mark II (100 kW) research reactor, KAERI, Seoul in 1962. The Korean Association of Neutron Activation Analysis was actually commenced by the management of the NAA laboratory in KAERI as a study circle from 1993. At that time, a research association for activation analysis supported by STEPI, Ministry of Science and Technology (MOST), Korea, was formulated to promote the standardization, practical utilization and industrial application of NAA by the participation of domestic experts in a variety of application fields. From 1993 to 2003, the activities of the research association, so called the NAA Peer Group in the HANARO Application Center, KAERI, has been concentrated on the development of advanced neutron activation analysis technologies, enhancement of their utilization, expansion of the NAA users and so on. By the end of 2003, the NAA peer group was changed to the Korean Association of Activation Analysis (KA3) and an advisory group was reorganized. The members of KA3 consists of more than 25 expert members and they are divided into six specific research and application fields such as the environment, industry, human health, standard reference material, social and cultural applications and prompt gamma activation analysis (PGAA). The objectives of KA3 have been fulfilled so that it can contribute to advertising as well as promoting research and development with respect to NAA and the related fields. Major activities conducted by KA3 over the past several years can be summarized as follows; (a) issuance of a newsletter or collected scientific articles with NAA and the related methods, semi-annually (b) regular meetings including seminars and training (c) information exchange of scientific issues between the members, and the provision of suggestions for the progresses of KA3 (d) other related activities. In the future, KA3 will make further efforts to promote the NAA applicability nationally and establish close international cooperation and relationships to seek out collaboration works for the upgrading and innovation of the technology with foreign counterparts.

For this special event, KA3 would like to introduce the current status of the utilization of the reactor neutron activation analysis in Korea and some recent papers on the application of NAA.

# Utilization and Facilities for a Neutron Activation Analysis at the HANARO Research Reactor in Korea

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#### Abstract

Since the commencement of the HANARO operation in 1995, some reactor systems have been gradually improved for a stable operation of the reactor. The experimental facilities for a neutron activation analysis (NAA) have been developed and installed to achieve the best measurement capability. That is, the automatic and manual modes of a pneumatic transfer system (PTS) were installed at three irradiation holes and tested to setup the optimum irradiation condition. In addition, gamma-ray measurement systems have been used for the analyses of samples by a quality control system. As a result, they have been effectively used during the past ten years. A device for a prompt gamma neutron activation analysis (PGAA) became available from 2002 for boron analysis in Boron Neutron Capture Therapy (BNCT) and other light elements in solid materials. Recently, a ko-standardization method was applied for more quantitative analysis. The examples of applications and international laboratory accreditation (KOLAS-ILAC) of NAA are also described. In addition, a delayed neutron activation analysis (DNAA) system will be installed by the end of this year.

Keywords: HANARO, Neutron Activation Analysis, Prompt Gamma Activation Analysis, Delayed Neutron Activation Analysis, Quality Control System, International Accreditation Scheme

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#### 1. Introduction

HANARO was designed as a powerful multipurpose neutron source that can provide a high neutron flux for various applications. As a nationwide neutron research facility, HANARO is now successfully utilized in various fields including neutron beam research, fuel and material tests, radioisotope production, neutron activation analysis, and neutron transmutation doping, etc. [1-2].

More detailed information is available at the HANARO home page for users. (http://hanaro.kaeri.re.kr; hanaro4u.kaeri.re.kr)

After the construction of the HANARO research reactor in 1995, experimental facilities have been

installed for a NAA. Generally, a NAA system consists of an irradiation facility such as a pneumatic transfer tube, vertical and horizontal irradiation tubes and a neutron beam guide for a PGAA, a gamma-ray measurement system and some NAA software for a gamma spectrum analysis and calculation of a component, chemical laboratories for a sample preparation or radiochemical separation, etc.

The prompt gamma neutron activation analysis (PGAA) facility has been under design from 1999 and it was installed in 2002. Detailed information is available from the published papers [3-7] and elsewhere in this journal.

Delayed neutron activation analysis (DNAA) is well known as a powerful method to determine fissile

elements such as U and Pu in suspicious samples. DNAA system has been designed in collaboration with the National Nuclear Management & Control Agency of Korea and it is additionally to be installed in the PTS by the end of 2005.

This paper is written to introduce the current status of the utilization and facilities for a neutron activation analysis at the HANARO research reactor and the applied research and development by using a NAA in Korea during the past ten years.

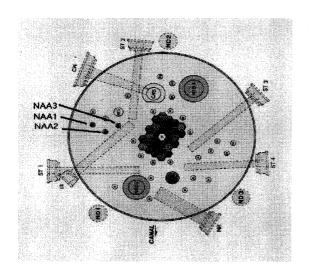


Fig. 1. Irradiation Holes for NAA in HANARO.

#### 2. Irradiation Facilities

A pneumatic transfer system (PTS) is one of the facilities to be used in an irradiation of a target material for an instrumental neutron activation analysis (INAA) in a research reactor. As for the irradiation facility, the automatic (PTS #2) and manual modes (PTS #1) of the pneumatic transfer systems were installed at three vertical irradiation holes within the reflector of HANARO in 1997. The positions for each irradiation hole (NAA #1-3) are shown in Fig. 1. The neutron of the NAA #1 irradiation hole is thermalized well. The irradiation tube of the NAA #2 irradiation hole was lined with Cd for an epithermal neutron activation analysis. Fig. 2 represents a schematic diagram of the PTS. PTS is

a simple shuttle system and operated by a pressurized nitrogen gas and a manual and automatic controller. The transfer time of a rabbit from a loader to an irradiation hole is 9 seconds, and when a rabbit is withdrawn, it takes about 4 seconds in the case of PTS #1.

Unfortunately, the Cd-lined irradiation tube for the epithermal NAA became unavailable in 2002 due to a depletion and contamination of Cd. One of the PTS (PTS #3) was remodeled in 2004 and the new designed PTS #1 and PTS #2 with a DNAA system will be reinstalled to improve the accuracy of the transfer time and the safety of its operation at the end of 2005.

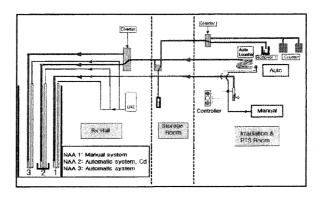


Fig. 2. Schematic Diagram of PTS in HANARO.

Characteristics of the NAA #1-3 irradiation holes in the operation of 30 MW<sub>th</sub> power are summarized in Table 1. Thermal, epithermal and fast neutron fluxes are measured by using high purity metal monitors like Au, Zr, Co, Ni, etc. The temperature on the irradiation position of the PE rabbit has to be limited to less than 80 °C because the melting point of the PE is about 120 °C. The temperature of the irradiation site was measured with the irradiation time using the thermo-label for the inside and surface of the rabbit with and without a sample during an irradiation. The measured temperature was in the range of 50 to 70 °C. For the safety of an irradiation, the optimum irradiation time was estimated as 4 hours. The Cd-

ratio,  $R_{Cd}$ , using Au-198 was determined by the a comparison of the activities created from a bare and Cd-covered monitor irradiation. The thermal neutron flux of NAA #1 is  $3.0 \times 10^{13}$  n·cm<sup>-2</sup>·s<sup>-1</sup> and the ratio of the thermal to epithermal neutron is more than 1000. The cadmium ratio was about 80.

Table 1. Characteristics of NAA #1, #3 Irradiation Holes (30 MW).

Items	Specific Feature
NAA #1 Neutron Flux, Thermal Epithermal Fast Cd-ratio of Au-198 Temperature Max. Irradiation Time	3.0 x $10^{13}$ n/cm <sup>2</sup> .sec 2.8 x $10^{10}$ n/cm <sup>2</sup> .sec 3.9 x $10^{10}$ n/cm <sup>2</sup> .sec 80 $50 \sim 70$ °C $\sim 4$ hrs
NAA #3 Neutron Flux, Thermal Epithermal Fast Cd-ratio of Au-198	1.3 x 10 <sup>14</sup> n/cm <sup>2</sup> .sec 1.5 x 10 <sup>11</sup> n/cm <sup>2</sup> .sec 9.1 x 10 <sup>10</sup> n/cm <sup>2</sup> .sec

A rabbit and sample vials were home-made by a high density polyethylene (HDPE) with a consideration of the impurity level as well as a physical stability. Fig. 4 shows the rabbits and vials used for a sample irradiation.

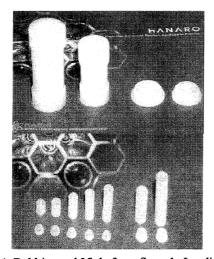


Fig. 4. Rabbits and Vials for a Sample Irradiation

The capacities of two difference rabbits with PTS are 35 mL and 60 mL, respectively. In addition, there are seven kinds of vials which have volumes from 0.5 mL to 10 mL. These vials can be more effectively used by a combination of them for many samples at a time.

#### 3. Measurement System

#### · 3.1 Gamma-Ray Measurement System

The measurements were carried out using a calibrated gamma-ray spectrometer (HpGe detector, GEM/GMX series and 919A MCB, Gamma Vision software, EG&G ORTEC). Energy and efficiency calibrations were done using multi-nuclide reference sources (Isotope Products Lab., ML 7500 series, 0.118" active diameter, disc type) traceable to NIST (National Institute of Standards and Technology, U.S.A). Since 1995, the number of advanced gammaray spectrometers has been increased according to the increasing demands. NAA Software for a data acquisition and quantification of an elemental concentration has also been upgraded from a DOS version to a Windows version. Among them, two sets of gamma-ray spectrometer with an automatic sample changer have been operated especially for the detection of medium and long lived nuclides. These systems which allow for the carrying out of an automatic counting up to 40 samples were designed and manufactured. An example of the gamma-ray measurement systems with an auto-sample changer is shown in Fig. 3.

#### 3.2 Data Acquisition and Calculation

From a report file of a measured spectrum (Gamma Vision, EG&G ORTEC), the elemental concentrations in the samples were absolutely determined by a software program available in the Windows environment, so called POWER-NAA,

which was developed for a fast and accurate calculation.

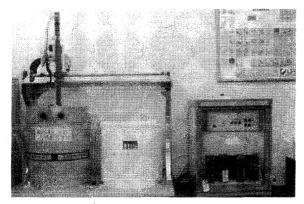


Fig. 3. Gamma-ray Spectrometer with Automatic Sample Changer.

The data for the operation of this program consists of the detector efficiency, nuclear data, count rates of the interesting gamma-ray energy and the thermal neutron flux. The determination of the elemental concentration using POWER NAA is based on a radioactivity creation formula. Fig. 5 represents the main display of the POWER-NAA program.



Fig. 5 POWER-NAA Program

#### 4. ko-Standardization

Recent advances and applications of the  $k_0$ -NAA technique have been published elsewhere by other authors.[8] In our laboratory, a feasibility study was

carried out for the application of  $k_0$ -NAA and the determination of the corresponding  $k_0$ -parameters of a well-thermalized NAA#1 irradiation hole at the HANARO research reactor and the results were reported in a previous work.[9-10] A comparison of the analytical data determined by three  $k_0$ -NAA programs, KAYZERO-SOLCOI, ADVNAA,  $k_0$ -DALAT, is carried out using airborne particulate matter collected from an urban site and the certified reference material of the air filter to evaluate the accuracy as well as the convenience and simplicity of the analysis. This year, the applicability of  $k_0$ -NAA to the NAA #3 irradiation hole was implemented and the neutron spectrum and  $k_0$ parameters are presented in Table 2. These  $k_0$ -NAA software and  $k_0$ -parameters for the NAA #1 and #3 irradiation holes of the HANARO were applied to the analysis of environmental and biological samples.

Table 2. Neutron spectrum and  $k_0$ -parameters of NAA #3 irradiation hole of HANARO.

Parameters	Mean	S.D.
α	0.1545	0.0524 (33.9%)
f	91.0	32.1 (35.3%)
$\mathbf{f_F}$	1.633	0.303 (18.5%)
Tn (°C)	18.24	5.70 (31.2%)
Asp(Au)/εp	4.151 × 10 <sup>13</sup>	4.255 × 10 <sup>11</sup> (1.0%)
Фth (n.cm <sup>-2</sup> .s <sup>-1</sup> )	1.262 × 10 <sup>14</sup>	$2.892 \times 10^{12} (2.3\%)$
Фері (n.cm <sup>-2</sup> .s <sup>-1</sup> )	1.495 × 10 <sup>12</sup>	4.691 × 10 <sup>11</sup> (31.4%)
Φf (n.cm <sup>-2</sup> .s <sup>-1</sup> )	9.057 × 10 <sup>11</sup>	1.697 × 10 <sup>11</sup> (18.7%)

#### 5. Applications

Instrumental Neutron Activation Analysis (INAA) has been one of the most important methods for a reliable and accurate, and more or less sensitive determination of a number of elemental constituents in many complex-matrix materials and has rapidly extended its application in various scientific and

technological areas. NAA has been applied to the trace, minor and major component analysis of nuclear, geological, biological, environmental and high purity materials, certified reference material (CRM), forensic samples, archaeological materials, and various polymers for the standardization and the validation of the method as well as research and development in Korea [10-23]. The results from the utilization of a NAA and PGAA are presented in more than 50 papers and technical reports every year, and the highlights are published in the HANARO Newsletter.

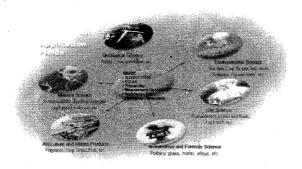


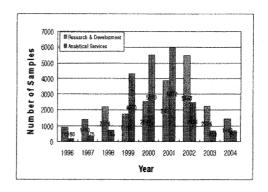
Fig. 7. Applications of NAA in Korea.

Annual statistics of the NAA utilization are presented in Fig. 6. More than sixty NAA users in Korea are registered as a member of the peer group of the NAA in HANARO and Korean Association of Neutron Activation Analysis.

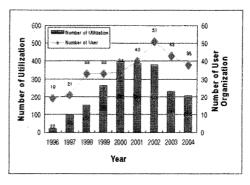
Recently, an analytical quality system of the NAA has been established as an international accreditation laboratory (KOLAS-ILAC) according to ISO/IEC 17025 technical and management requirements and a mutual recognition agreement (MRA).

NAA laboratory has carried out the international cooperation for the IAEA/RCA, FNCA programs to assess and compare air pollution levels in strategically chosen areas by the use of nuclear and complementary analytical techniques to identify and apportion critical air pollution sources and to

accumulate accurate air pollution data for a future work on trans-boundary transport of airborne particulate matter and collaboration projects between nations and international organizations.



#### (a) Annual trend of samples



#### (b) Annual trend of users

Fig. 6. Annual Statistics of Utilization in Neutron Activation Analysis

#### 6. Conclusion

During the past ten years, the NAA facilities in HANARO have been successfully utilized for the purpose of basic and applied researches. Nowadays, the irradiation facilities and measurement systems are being amended for a further effective use and a much safer operation in accordance with national demands. The utilization of the NAA is promoted in a variety of fields and by both internal and external users. International cooperation programs and collaboration works will be enhanced.

#### **Acknowledgements**

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### PGAA Facility in the HANARO Research Reactor of Korea

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#### Abstract

The prompt gamma-ray activation analysis (PGAA) facility was initially installed on the ST1 beam port of the HANARO for a BNCT study from 1998 to 2001, and it was reinstalled in 2003. The neutron flux is  $2.5 \times 10^8$  ncm<sup>-2</sup>s<sup>-1</sup> at 30 MW and the beam size at the sample position is  $2\times2$ cm<sup>2</sup>. The detection of the gamma-ray spectrum is a Compton suppression system which is composed of HPGe and BGO/NaI(TI) detectors. The functional information of the PGAA system is described including the specifications of the system, the calibrations, the sensitivity and detection limits and the measurement uncertainty.

Keywords: HANARO Research Reactor, Prompt Gamma-ray Activation Analysis, Compton suppression, Boron,

Hydrogen, Element Analysis

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#### 1. Introduction

In Korea, the prompt gamma-ray neutron activation analysis (PGAA) system was firstly installed and operated at the TRIGA Mark-III research reactor, Seoul, KAERI in the 1980's.[1-2] At the HANARO research reactor, a new PGAA system was initially installed on a platform located at the exit of the 4 m long ST1 tangential beam port in 2001 and modified to obtain a high quality neutron flux and a low background in 2003.[3-8] The thermal neutron beam is available by using pyrolytic graphite (PG) crystals which are used to extract the thermal beam by the method of a Bragg diffraction with the Bragg angle set at 45 degree. The diffracted beam is moved vertically 1.5m from the lower crystal to the sample position.

The fields of the present utilization are the boron analysis of tissue and cancer for the BNCT, DNA cell

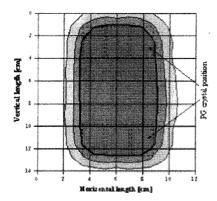
for nuclear medicine, the elements of boron and hydrogen in standard reference materials, biological and metal samples, environment samples and explosive materials and so on. The analysis of other light elements of sulfur, phosphorus, carbon, nitrogen and silicon are in progress.

This paper is to describe the functional information of the PGAA system in detail.

#### 1.1 Beam Flux

The beam size of the ST1 horizontal beam tube is  $120^{\rm h} \times 60^{\rm w}$  mm<sup>2</sup>, the unperturbed beam occupying the central  $60^{\rm h} \times 60^{\rm w}$  mm<sup>2</sup> area passes to a polarized neutron spectrometer (PNS) as shown in Fig. 1. Each of the  $30^{\rm h} \times 60^{\rm w}$  mm<sup>2</sup> of the upper and lower sides are used for the PGAA. The size of the crystal is  $35^{\rm h} \times 22^{\rm w} \times 4^{\rm t}$  mm<sup>3</sup>, occupying a beam area of  $70^{\rm h} \times 66^{\rm w}$  mm<sup>2</sup>. The thermal neutron flux at the crystal position with a 2.5 mm interval from the central position to

the edge was measured by gold activation wires and it was about 10<sup>10</sup> n·cm<sup>-2</sup>s<sup>-1</sup>. Neutron flux distribution at the PG crystal position in the ST1 beam tube is shown in Fig. 2.



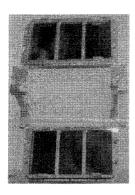


Fig. 1. Neutron flux distribution at PG position and front view of PG crystal setting.

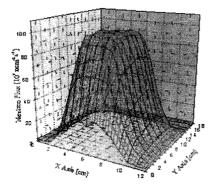


Fig. 2. Neutron flux distribution at the PG crystal position in ST1 beam tube.

#### 1.2 PGAA System

The neutron beam can be controlled by two kinds of beam shutters, one is placed at the ST1 tube as a main beam port and the other is to control the diffracted beam. The latter, PGAA beam shutter which is placed in the bottom lead frame to control the diffracted beam reduces the dose rates in the beam to negligible levels. Samples are inserted and removed using a manual operation, while the neutron beam is turned on, via a sample holder. The sample holder reliably positions the sample inside the lead wall, while keeping the user's body at a distance from the neutron beam.

The shielding materials used within the sample position are natural LiF tiles to avoid scattered neutrons towards the detector followed by 10 cm thick lead bricks to reduce the gamma-ray background. The gamma-ray detector is located at about a 25 cm distance from the sample and is placed on a perpendicular line with respect to the beam direction. To reduce the influences of the background by the scattered neutrons, enriched lithium tiles (<sup>6</sup>LiF) were attached in front of the HPGe detector in the direction of the gamma-ray emitted from the sample. The measured effective wavelength and velocity of the beam are 1.78±0.01Å and 2191±15 m/s, respectively. Flux uniformity of within 4 % is achieved in the central area of 1×1 cm<sup>2</sup> of the total beam cross section of 2×2 cm<sup>2</sup>. A detailed layout of the PGAA facility is shown in Fig. 3.

#### 2. Detection Assembly

In the measurement of the emitted prompt gamma-rays, the detection system was composed of a HP Ge detector (GEM-30190 EG&G ORTEC, relative efficiency 43 %) surrounded by eight bismuth-germanate (Bi<sub>4</sub>Ge<sub>3</sub>O<sub>12</sub>: BGO) detectors plus two NaI(Tl) scintillators (5.75HW6.30 BGO/NaI(Tl)/

(8) 1.5-x, BICRON) in order to reject the Compton-scattered photons.

The BGO detectors are covered with an aluminium case and shielded by 10 cm of lead to stop the gamma radiation appearing near the scintillation detector. This is surrounded with a 10cm boron sheet to shield the detector assembly from the thermal neutrons. A 10 cm thick borated polyethylene (BPE) and boron sheet were also added to reduce the variable background in the capture spectrum produced by the thermal neutrons interacting with the aluminium barrel, the lead shielding and the detector photomultiplier.

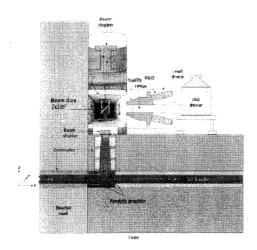


Fig. 3. Schematic layout of the PGAA system on the ST1 beam port at HANARO.

A HP Ge detector is placed in front of the sample position at 90 degree to the path of the diffracted neutron beam. The sample position is surrounded by shielding material comprised of a 2 cm natural LiF plate in a lead wall. This is considered to limit the unnecessary photons produced by neutrons that scatter from the sample and interact in the lead shielding surrounding the detector. The end of the HP Ge detector faced with the sample is shielded by 10 mm thick enriched LiF tiles. These shields serve to protect the HP Ge detector from a slow neutron

damage, and absorb the neutrons without the subsequent generation of capture gamma-rays.

The sample vial used is a thin sheet of  $26~\mu m$  thick and 10~mm dia. made of Teflon material. The background spectrum of Teflon is shown in Figure 4. The spectrum of Compton and a single mode for the standard reference material (NIST SRM, Spinach leaves) is shown in Figure 5 for comparison.

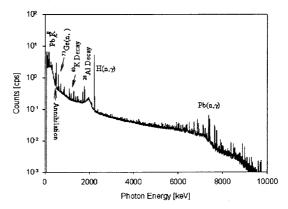


Fig 4. Background gamma-ray spectrum measured for an empty vial.

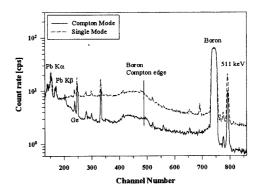


Fig. 5. Comparison of the spectrum emitted from NIST SRM (Spinach leaves) sample.

#### 3. Calibration

#### 3.1 Efficiency calibration

The full energy peak efficiency curve of the PGAA spectrometer was calibrated both by the conventional method using the standard radioactive sources and by measuring the prompt gamma-rays from the neutron capture reactions. A 43 % HP Ge detector

(EG&G ORTEC) connected to a PC based 16k MCA (919 MCB, EG&G ORTEC) is used for counting the prompt gamma-rays. The resolution of the detector is 2.2 keV at 1332 keV 60Co. The MCA has been calibrated in the region up to 11 MeV using the delayed gamma-rays from 152Eu and 60Co and the prompt gamma-rays from <sup>36</sup>Cl. In the low energy region of 60~1408 keV, <sup>133</sup>Ba, <sup>134</sup>Cs, <sup>137</sup>Cs, <sup>60</sup>Co, <sup>57</sup>Co, <sup>152</sup>Eu and <sup>241</sup>Am were used to generate the absolute full energy peak efficiency curve. A third polynomial was used for the energy calibration. In the high energy region of above 1 MeV, the sample for the measurement of the prompt gamma-rays was prepared from a mixture of ammonium chloride and melamine powder. The sample packed in Teflon (polytetrafluroethylene-PTFE) was irradiated for about 3 hours and the capture gamma-rays were accumulated. The absolute full energy peak efficiencies were determined for the low energy region (about up to 1.5 MeV) using an <sup>152</sup>Eu source and the relative efficiency plot for the energy region from 0.5 to about 11 MeV was obtained from the prompt gamma-ray spectrum of  $^{35}Cl(n,\gamma)$  and  $^{14}N(n,\gamma)$ . The relative efficiencies obtained from the chlorine spectrum were normalized to 152Eu efficiencies to obtain the absolute detection efficiencies of the gamma-rays. The typical efficiency values are in the order of 10<sup>-4</sup> to 10<sup>-5</sup>. The energy vs. efficiency curve is showed in Fig. 6. Full energy peak efficiency is determined by fitting the polynomials to the measured data; relative standard uncertainly is < 3% over the low energy region, and < 5% for the complete spectrum. Non-linearity of the spectrometer is determined in a similar manner by fitting a polynomial function to the observed data for the accurately obtained gamma-ray lines.

The sensitivity, detection limit and measurement uncertainty of the boron and hydrogen analysis by the PGAA system were estimated and they are summarized in Table 1. The sources of the uncertainties are mainly statistical errors, the detection efficiency, the background effect, etc. and the combined uncertainty was 3.75 % and 2.70 % for the Boron and Hydrogen analysis, respectively.

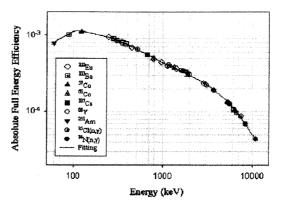


Fig. 6. Efficiency curve for the Compton suppression spectrometer of the PGAA of HANARO.

Table 1. The sensitivity, detection limit and uncertainties estimated in the boron and hydrogen analysis by PGAA.

	Boron	Hydrogen
Sensitivity (cps/mg)	1720	2.8
Detection Limit (μg/g)	0.002	6.8
Uncertainties due to	Un	cert (%)
- Statistical error	1.5	0.25
. Detection efficiency	2.0	2.30
- Mass (sample weight)	0.1	0.05
. Detection sensitivity	0.5	0.50
- Background subtraction	2.3	1.20
. high & low tails for hydrogen	1.5	0.50
	1	
Combined Uncertainty	3.75	2.70

#### 4. Application

Characteristics and testing of the PGAA facility were mainly carried out for the set-up of the system and an effective utilization and operation in 2003. In addition, the present PGAA facility is used for the boron and hydrogen analysis for the biological samples like spinach and pine needles, for a metal

sample like low alloy steel, for the BNCT and DNA like tissue and cancer cell, for an environment sample like monitoring of an air particulates and the second is the carbon, sulfur and nitrogen analysis related the explosives and the third is silicon and phosphorus related to sludge and health and so on.

Figure 7 shows the results of the analytical PGAA support in 2004. The major portion of the utilization is the light element analysis by our research and the remainders are university and medical researches and foreign usages.

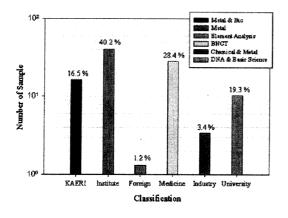


Fig. 7. Status of user supports in 2004.

#### 5. Conclusion

The present HANARO operation period is as follows: operating during three weeks and stop for on week. The PGAA beam fluctuation is below 2 %, which offers a stable beam state and is useful for micro element analysis. The affects of the background and interference coming from in and out side of the facility are low, therefore, the PGAA facility is useful for the analysis of light elements.

#### Acknowledgements

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### Duplicate Diet Study of Korean Geriatric Population by Neutron Activation Analysis

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#### **Abstract**

One day duplicate diet samples of 30 Korean geriatric population over 60 years old were collected. Samples were analyzed by NAA(Neutron Activation Analysis) after a sample preparation such as a homogenization and a freezedrying. SRM(Standards Reference Materials) from NIST were analyzed for a quality control of the analytical method. Analytical results were classified into two groups such as macro elements(Ca, K, Mg, Na) and minor elements(Cr, Fe, Mn, Se, Zn). Analytical results for a daily intake of the geriatric population in Korea showed that the macro and minor elements were lower than the RDA and literature except for Na. In conclusion, NAA is proved to be a sensitive analytical method useful for performing both a qualitative and a quantitative multi-elemental analysis of the macro and minor elements in a diet or biological materials.

Keywords: Duplicate Diet; Geriatric Population; Macro and Micro Elements, NAA

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#### 1. Introduction

Health status of a geriatric population can be affected more by malnutrition of micronutrients than the other age groups. Average span of life of the Korean geriatric population has become long and their population ratio is around 10% of the total population and expected to be 20% in 2020[1].

Minerals and micronutrients are very important components in diets[2]. Both their deficiency and excess may cause serious human health problems especially to the geriatric and pediatric population. The samples were collected from over 60 year old adults living in Seoul using the Duplicate Diet method[3]. Duplicate Diet method is most accurate method for estimating micronutrients intakes and an exact copy of the foods eaten and preferably weighed, to replicate the diet consumed by an individual.

Samples were analyzed by NAA(Neutron Activation Analysis) after a sampling and a sample preparation such as a homogenization[4], freeze-

drying, etc.[5,6]. and for a quality control of the analytical method the SRM(Standards Reference Materials)'s from NIST were analyzed.

#### 2. Experimental

#### 2.1 Sample Preparation

Immediately after the completion of the initial blending of the prepared one day Korean geriatric duplicate total diet in a blender with titanium blades, an analytical sub-sample representing about 20% of the total diet was withdrawn using a large plastic spoon and placed in a container for freeze-drying. Samples were initially frozen at -80°C for 3 hours and freeze-dried for 30 hours at the same temperature. Weight loss of the 18 samples after a freeze-drying is listed in Table 1. Freeze-dried samples were ground well to get a sample homogeneous for the analysis. Samples were stored in polyethylene vials at 2 - 4  $^{\circ}$ C for a short term storage and in a Teflon<sup>™</sup> vial at below -15°C for a long term storage to avoid an absorption or evaporation which may cause a change of the elemental concentrations.

#### 2.2 Neutron Activation Analysis

Samples were irradiated with thermal neutrons from the HANARO research reactor (20 MW) at the Korea Atomic Energy Research Institute. The Pneumatic Transfer System (PTS) was used for a sample irradiation. Samples were irradiated at the same geometrical position to minimize the geometry error. For a neutron flux monitoring, activation wires (Reactor Exp. Inc., R/X activation wires, 99.99% purity) such as Au, Cu, and Mn were used to monitor the thermal neutron flux ( $\Phi_t = 1.7 \times 10^{13} \text{ cm}^{-2} \cdot \text{s}^{-1}$ ). Samples were allowed to decay for an appropriate time to ensure satisfactory detection limits. Analytical condition is described in Table 2.

Table 1. Moisture Removal Rate by Freeze-Drying of Duplicate Diet.

Diving of Duplicate Diet.							
Sample	Wet	Dry	Moisture	Removal			
No.	(g)	(g)	(g)	(%)			
1	267.4	53.62	213.78	79.95			
2	176.11	22.58	153.53	87.18			
3	234.59	31.86	202.73	86.42			
4	217.9	37.91	179.99	82.6			
5	177.07	29.36	147.71	83.42			
6	185.05	34.38	150.67	81.42			
7	167	31.48	135.52	81.15			
8	142.79	31.56	111.23	77.9			
9	177.34	29.06	148.28	83.61			
10	174.43	39.52	134.91	77.34			
11	362.37	69.44	292.93	80.84			
12	287.81	35.17	252.64	87.78			
13	279.3	39.53	239.77	85.85			
14	300.51	53.02	247.49	82.36			
15	359.92	52.78	307.14	85.34			
16	419.84	66.72	353.12	84.11			
. 17	430.22	57.99	372.23	86.52			
18	353.81	67.61	286.2	80.89			
Mean				83.03			

Table 2. Analytical condition of Duplicate Diet by NAA

	by NAA	
D: :::-	Short-lived	Long-lived
Division	Nuclides	Nuclides
Sample	5 ~ 20 ···	100 ~ 200 mg
Weight	5~20 mg	100 ~ 300 mg
Irradiation	1 ~ 2 min	3 hr
Time	1 2 mm	3 III
Cooling	1st: $3^{\sim} 5 \min$	1st : 5 ~ 10 day
Time	2nd: 30~60 min	2nd: 14~21 day
Counting	1st: 300 sec	1st: 1000 sec
Time	2nd: 1000 sec	2nd: 4000 sec
Target	Co Ma Ma 7n	Cr, K, Na, Fe,
Element	Ca, Mg, Mn, Zn	Se, Zn

#### 2.3 Radioactivity Measurement

Gamma-ray spectra and the activity for the irradiated samples were obtained with the following

equipment. The crystal of the high purity germanium semi-conductor detector (EG&G ORTEC, GMX-25190P) was surrounded with a 10 cm thick lead shielding box (75x90x90 cm) with its inner wall lined with 0.1 cm Cd and Cu plate to reduce the influence of the external radiation. Gamma-ray interactions with a surrounding material were also considered. The detector is a coaxial type with a 25% relative efficiency and 1.9 keV resolution (FWHM) at 1332 keV (<sup>60</sup>Co) and a peak to Compton ratio of 45:1. The detector was connected to a personal computer and a 8K-multichannel analyzer (EG&G ORTEC, 919A MCB, Maestro-II) to complete the  $\gamma$ -ray spectrometer for an activity measurement and data analysis. Energy and efficiency calibrations were done with the radioactive multinuclide reference source (NEN Products Inc., NES-602, 1" diameter disc type) certified by NIST. The dead time of the detector was maintained at under 5% as much as possible. Decay and pile up correction were done automatically.

#### 2.4 Data Tratment

Concentrations of the elements were calculated with a new graphical programme, POWER NAA, coded with LabView version 5.0 for a rapid and simple data treatment after obtaining the  $\gamma$ -ray spectrum under the present analytical condition. The program is run with the counting condition, automatic identification of nuclides and their  $\gamma$ -ray energies, decay time correction for short half-life nuclides in order to calculate their activities, elemental concentrations, counting errors and detection limits. Elemental concentration was calculated from the peak area which is measured by the  $\gamma$ -ray activity along with the related nuclear data[7,8].

#### 3. Result and Discussion

Five biological reference materials such as NIST SRM 1548(total diet), NIST SRM 1567a(wheat flour),

NIST SRM 1547(peach leaves), NIST SRM 1566a(oyster tissue) and NIST SRM 1577b(bovine liver) were analyzed mainly by an instrumental neutron activation analysis for a quality control (Table 3-7). The elemental concentrations, relative standard deviations, and relative errors to the certified values were calculated. After the repetitive analysis(≥5 times), the relative standard deviations were within 10% except for Fe(12.4%) in SRM 1548, Rb(11.7%) and V(23.9%) in 1567a, Ce(14.4%) and La(15.7%) in 1547 and V(12.9%), Rb(15.8%) and Sc(10.8%) in 1566a. The relative errors to certified values are within 5% except for K(7.2%) and Zn(10.9%) in 1548, Mn(10.2%) and Rb(7.9%) in 1567a, Cl(13.2%), Fe(9.3%) and Rb(5.8%) in 1547, Cd(5.4%), Cl(7.6%), Co(5.7%), Cr(6.6%), Cu(7.2%), K(10.2%) and Zn(23.1%) in 1566a and Cu(8.7%), K(7.1%), Rb(8.6%) and Zn(9.4%) in 1577b. Analytical results of the elemental distribution in a Korean total diet and four Korean representative foodstuffs are listed in Table 8 - 12.

Table 3. Analytical Result of Total Diet, NIST SRM 1548 by NAA (unit : ppm)

		This W	'ork	Certified	Relative
Element	Nuclide -	Range	Mean (SD%)	Value (S.D.%)	Ептог (%)
Ca	Ca-49	1676~1765	1735 (0.08)	1740 (4)	031
Cl	C1-38	8210~8883	8460 (5.63)	8700 (5)	276
Fe	Fe-59	30.4~36.39	34.17 (12.39)	32.6 (11)	483
K	K- 42	5049~5958	5621 (1.28)	6060 (5)	724
Mg	Mg-27	521~568	551 (5.98)	556 (5)	095
Mn	Mn-56	50~5.4	53 (0.4)	52 (8)	094
Na	Na-24	6352~6605	6460 (2)	6250 (4)	336
Se	Se-75	0245~0.272	0257 (4.9)	0245 (2)	479
Zn	Zn-65	32.5~36.3	34.2 (7.89)	30.8 (4)	10.89
Al	A1-28	[32.6~377]	34.4 (10.39)	33	
Cs	Cs-134	[00137~0.015]	0014 (5.5)	0014	
Rb	Rb-86	[47~5.1]	49 (3.32)	48	

] : reference value

Table 4. Analytical Result of Wheat Flour, NIST SRM 1567a by NAA (unit : ppm)

		This Wo	ork	Certified Value	Relative
Element	Nuclide	D	Mann(SD0/)		Error
		Range	Mean(SD%)	(S.D.%)	(%)
Al	Al-28	53 ~ 6	5.7 (809)	5.7 (22)	03
Ca	Ca-49	178 ~ 200	188 (5.04)	191 (2)	147
Cu	Cu-66	20~23	21 (8397)	21 (10)	186
Fe	Fe-59	12.9 ~ 15.1	14.3 (0.69)	14.1 (4)	17
K	K-42	1239 ~ 1303	1271 (3.56)	1330 (2)	446
Mg	Mg-27	400~414	405 (2.15)	400 (2)	13
Mn	Mn-56	84 ~ 8.5	84 (0.18)	94 (5)	10.16
Na	Na-24	61 ~ 6.4	63 (1.35)	61 (13)	242
Rb	Rb-86	066~0.78	073 (11.69)	068 (4)	792
Se	Se-75	11 ~ 1.2	11 (3.26)	11 (18)	439
Zn	Zn-65	12.0 ~ 12.2	12.1 (1.11)	11.6(3)	471
Cl	Cl-38	[528 ~ 547]	535 (0.08)	565	
Co	Co-60	[00058~0.0062]	0006 (4.69)	0006	
V	V-52	[0013~0.018]	0015 (23.8)	0011	

[ ] : reference value

Table 5. Analytical Result of Peach Leaves, NIST SRM 1547 by NAA (unit : ppm)

El .	NT 11.1	This	Work	Certified Value	Relative
Element	Nuclide	Range	Mean(SD%)	(S.D.%)	Erro r(%)
Al	Al-28	124~ 129	249 (261)	249 (3)	005
Ba	Ba-139	14720~ 15210	126 (2.63)	124 (3)	18
Ca	Ca-49	242~253	14950 (0.95)	15600 (1)	417
Cl	Cl-38	306~322	312 (1.02)	360 (5)	13.21
Fe	Fe-59	234~ 242	238 (2.35)	218 (6)	925
K	K-42	23090~23190	23137 (0.12)	24300 (1)	479
Mg	Mg-27	4150~4450	4307 (2.79)	4320 (2)	031
Mn	Mn-56	93.7~96.7	95.1 (2.23)	98 (3)	295
Na	Na-24	22.4~23.4	22.8 (2.98)	24 (8)	515
Rb	Rb-86	18.9~22.1	20.9 (8.89)	19.7 (0)	584
Sr	Sr-87m	47.3~54.2	51.7 (9.38)	53 (8)	244
v	V-52	035~038	036 (606)	037 (8)	097
Zn	Zn-65	16.8~18.7	17.7 (2.64)	17.9 (2)	121
Br	Br-80	[10.5~120]	11.4 (2.37)	11	
Ce	Ce-141	[92~11.7]	10.8 (14.42)	10	
Co	Co-60	[0071~0.076]	0074 (3.92)	007	
Cr	Cr-51	[116~1.20]	118 (1.14)	1	
La	La-9	[944~11.96]	11.1 (15.71)	9	
Nd	Nd-147	[684~7.69]	723 (5.29)	7	
Sc	Sc-46	[0045~0.054]	005 (6.85)	004	
Sm	Sm-153	[114~1.22]	119 (3.87)	1	
Тb	Tb-160	[0105~0.111]	0107 (3.98)	· 01	
Th	Pa-233	[0049~0.055]	0051 (0.42)	005	

] : reference value

Table 6. Analytical Result of Oyster Tissue, NIST SRM 1566a by NAA (unit : ppm)

This Work Certified Rela					
Element	Nuclide	This	Work	Value	Relative Error
Biomoni	Traditad	Range	Mean(SD%)	(S.D.%)	(%)
Ag	Ag-110m	155 ~ 1.78	1.66 (451)	168 (9)	1.07
Al	Al-28	192.8 ~ 213.4	204.2 (2.42)	202.5 (6)	084
As	As-76	13.7 ~ 15.2	14.4 (3.63)	14 (9)	305
Ca	Ca-49	1857 ~ 1983	1936 (4.6)	1960 (10)	122
Cd	Cd-115	436~439	437 (0.58)	415 (9)	54
Cl	Cl-38	7501 ~ 7932	7656 (3.66)	8290 (9)	764
Co	Co-60	0596~0.611	0603 (1.8)	057 (19)	57
Cr	Cr-51	147~1.57	152 (4.92)	143 (32)	66
Cu	Cu-66	60.7 ~ 62.47	61.5 (1.931)	66.3 (6)	72
Fe	Fe-59	529 ~ 570	552 (3.5)	539 (6)	239
K	K-42	6720 ~ 7726	7223 (0.18)	7900 (6)	10.16
Mg	Mg-27	1139 ~ 1204	1165 (0.85)	1180 (14)	124
Mn	Mn-56	11.8 ~ 12.3	12.0 (2.77)	12.3 (12)	228
Na	Na-24	4047 ~ 4271	4185 (3.19)	4170 (3)	035
Se	Se-75	220~2.48	230 (8.12)	221 (11)	401
v	V-52	413 ~ 4.97	457 (12.91)	468 (3)	226
Zn	Zn-65	1004~1034	1022 (2.08)	830 (7)	23.09
Au	Au-198	[0.013 ~ 0017]	0015 (6.27)	001	
La	La-140	[029 ~ 0.30]	030 (0.91)	030	
Rb	Rb-86	[276~3.46]	311 (15.84)	3	
Sc	Sc-46	[0059~0.069]	0066 (10.79)	006	
Sm	Sm-153	[0058~0.061]	0059 (1.32)	006	

[ ] : reference value

Table 7. Analytical Result of Bovine Liver, NIST SRM 1577b by NAA (unit : ppm)

		This	Work	Certified	Relative
Element	Nuclide	Range	Mean(SD%)	Value	Error
		Kange	Mean(SD W)	(S.D.%)	(%)
Ca	Ca-49	113	113	116 (3)	25
Cl	Cl-38	2633 ~ 2685	2657 (0.85)	2780 (2)	442
Cu	Cu-66	173 ~ 175	174 (0.73)	160 (5)	873
Fe	Fe-59	179~197	188 (2.97)	184 (8)	221
K	K-42	9060 ~ 9572	9238 (3.92)	9940 (0)	707
Mg	Mg-27	594~658	620 (1.5)	601 (5)	312
Mn	Mn-56	98~10.4	10.1 (2.89)	10.5 (16)	341
NA	Na-24	2299 ~ 2391	2345 (1.42)	2420 (2)	311
Rb	Rb-86	13.8 ~ 16.4	14.9 (9.13)	13.7 (8)	859
S	S-37	8013 ~ 8235	8124 (1.932)	7850 (1)	349
Se	Se-75	066~0.76	072 (2.33)	073 (8)	15
Zn	Zn-65	130~146	139 (7.84)	127 (13)	937
Co	Co-60	[0.255 ~ 0273]	0262 (0.81)	025	
v	V-52	[0105~0.115]	011 (6.808)	0123	

] : reference value

Table 8 Analytical Result of Korean Total Diet by NAA

			•		
			Т	his Work	
Element	Nuclide	Half-life	_		Standard
			Range	Mean Value	Deviation
< ppm	level >				
Al	Al-28	228 min	16.07~1832	1755	1279
Br	Br-80 Br-82	17.68 min 35.299 hr	9.59~1062	9947	0583
Ca	Ca-49 Ca-47	8716 min 4536 day	1358 ~ 1496	1418	70.87
Cl	C1-38	37.24 min	10870 ~ 11780	11353	457.6
Fe	Fe-59	44.496 day	237~26.02	25.22	1317
I	I-128	24.99 min	159~2.01	18	03
K	K-42	12.36 hr	5394 ~ 5706	5579	1335
Mg	Mg-27	9462 min	8068 ~ 895.6	850.2	44.44
Mn	Mn-56	154.71 min	8464~8.778	8648	0164
Na	Na-24	14.659 hr	10080 ~ 10470	10298	1615
Rb	Rb-86	18.66 day	5427 ~ 5489	5457	0031
Sr	Sr-87m	167.7 min	53 ~ 5.803	5552	0356
Zn	Zn-65	244.1 day	31.57~33.74	32.48	1125
< ppb	level >				
As	As-76	26328 hr	492.7 ~ 6033	5264	5178
Co	Co-60	5271 yr	296~31.6	30.8	1058
Cr	Cr-51	27.704 day	348 ~ 4203	3931	3936
Cs	Cs-134	2062 yr	10.6~11.6	11.1	0707
Sc	Sc-46	83.83 day	32~3.6	3433	0208
Se	Se-75	119. <b>77</b> day	223 ~ 3284	2757	7453
Sm	Sm-153	46.704 hr	46.1 ~ 47.5	46.8	099
Th	Pa-233	27.0 day	238~2.82	26	03
U	Np-239	56.52 hr	31.45 ~ 38.55	35	50

# Table 9 Analytical Result of Korean Boiled Rice by NAA

		_		This Work		
Element Nuclide		Half-life	Range	Mean Value	Standard Deviation	
< ppm	level >					
Al	Al-28	228 min	0866~119	1028	0229	

Ca	Ca-49 Ca-47		8328 ~ 87.2	85.24	2772
Cl	Cl-38	37.24 min	297.9 ~ 330.5	316.1	16.63
Fe	Fe-59	44.496 day	2216~2.336	2276	00845
K	K-42	12.36 hr	757.4 <sup>~</sup> 785.9	774.9	12.3
Mg	Mg-27	9462 min	65.89 ~ 68.91	67.07	1613
Mn	Mn-56	154.71 min	3229~3.355	3272	00716
Na	Na-24	14.659 hr	108.8 ~ 111.7	110.4	1245
Zn	Zn-65	244.1 day	16~17.28	1663	0694
< pp1	level >				
As	As-76	26328 hr	103~142.2	129.8	18.09
Au	Au-198	64.644 hr	017~0.23	02	0044
Br	Br-80 Br-82	17.68 min 35.299 hr	645 ~ 742.1	678.5	43.85
Со	Co-60	5271 yr	464 <sup>~</sup> 55	5013	0441
I	I-128	24.99 min	77 ~ 120	100	30
Мо	Mo-101 Mo-99	14.60 min 65.945 hr	2681 ~ 384	3261	8195
Rb	Rb-86	1866 day	452~509.5	485.6	24.24
Sc	Sc-46	83.83 day	18 ~ 2.459	2174	0338
Sr	Sr-87m	167.7 min	77 ~ 91	840	10
Th	Pa-233	27.0 day	01~0.122	011	002
U	Np-239	56.52 hr	27~4.2	35	10

# Table 10 Analytical Result of Korean Kimch'i by NAA

		10 110	This Work				
Element Nuclide		Half-life	Range	Mean Value	Standard Deviation		
< ppm	level >						
Br	Br-80 Br-82	17.68 min 35.299 hr	1816~209.7	196.9	11.56		
Ca	Ca-49 Ca-47	8716 min 4536 day	4022 ~ 4480	4284	236.2		
Cl	C1-38	37.24 min	115900 ~ 120500	118433	2335		
Cr	Cr-51	27.704 day	1102~1.442	1295	0161		
Fe	Fe-59	44.496 day	49.23 ~ 54.77	51.38	2973		

К	K-42	12.36 hr	27320 ~ 30770	29535	1604
Mn	Mn-56	154.71 min	26.58 ~ 30.59	29.14	2221
Na	Na-24	14.659 hr	97790 ~ 104500	99843	3167
Rb	Rb-86	18.66 day	22.75 ~ 27.99	26.09	2469
Sr		167.7 min 64.84 day	2232~25.23	23.78	2058
Zn	Zn-65	244.1 day	42.51 ~ 46.47	44.67	1701
< ppl	b level >				
Co	Co-60	5271 yr	346.4 ~ 392	365.2	23.81
Cs	Cs-134	2062 yr	69.5 ~ 71.8	70.91	12.35
Sc	Sc-46,	83.83 day	30~33.84	3156	1748
Th	Pa-233	270 day	41 ~ 575	49	12

# Table 11 Analytical Result of Mackerel by NAA

			This Work			
Element	Nuclide	Half-life	Range	Mea	ın Value	Standard Deviation
< ppm	level >					
Al	Al-28	228 min	12.6 ~ 158		1377	1767
As	As-76	26.328 hr	6.279 ~ 6924		6501	0367
Br	Br-80	17.68 min	1632 ~ 18.47		17.3	1013
	Br-82	35.299 hr				
Ca	Ca-49	8716 mir.	3448 ~ 3973		3675	269.6
	Ca-47	4536 day				
Cl	Cl-38	37.24 min	22980 ~ 24170	2	23747	665.2
Cr	Cr-51	27.704 day	0884~1371		1144	0211
Fe	Fe-59	44.496 day	45.31 ~ 49.44		47.64	1752
K	K-42	12.36 hr	11250~11790	1	11510	2232
Mg	Mg-27	9462 min	6489 ~ 693.6		675.3	23.42
Na	Na-24	14.659 hr	2183 ~ 2289		2230	5331
Rb	Rb-86	18.66 day	1058 ~ 1394		1166	0154
Se	Se-75	119.77 day	2913 ~ 3.099		2986	0099
Sr	Sr-87m	167.7 min	64~7.54		<b>7</b> 0	08

Zn	Zn-65	244.1 day	35.7 ~ 39.79	37.29	1885
< ppl	b level >				
Au	Au-198	64644 hr	12~17	145	0354
Со	Co-60	5271 yr	587~638	614.3	27.05
Cs	Cs-134	2062 yr	53.6 ~ 66.2	61.1	5466
Sc	Sc-46	83.83 day	32~3.8	35	0424
Th	Pa-233	27.0 day	085~1.07	096	015

Table 12 Analytical Result of Korean Mixed Pork and Beef by NAA

				This Work	
Element	Nuclide	Half-life	Range	Mean Value	Standard Deviation
< ppm	level >	, processor and a			
Al	Al-28	228 min	17.6~2139	195	268
Br	Br-80 Br-82	17.68 min 35.299 hr	1331 ~ 13.86	13.53	0239
Cd	Cd-115	53.472 hr	1545 ~ 1.579	1562	0024
Cl	C1-38	37.24 min	26020 ~ 26840	26343	4366
Fe	Fe-59	44.496 day	3978 ~ 46	43.38	2812
K	K-42	12.36 hr	11500 ~ 11930	11625	205
Mg	Mg-27	9462 min	602.9 ~ 674.1	635.2	36.05
Na	Na-24	14.659 hr	2110~2205	2162	4113
Rb	Rb-86	18.66 day	1863 ~ 21.63	20.8	1446
Zn	Zn-65	244.1 day	118.5 ~ 130.7	123.6	5832
< ppb	level >				
As	As-76	26328 hr	72.51 ~ 1068	85	15.71
Co	Co-60	5271 yr	13.6 ~ 17.3	15.35	174
Cr	Cr-51	27.704 day	336.8 ~ 682.5	515.4	143.2
Cs	Cs-134	2062 yr	34.6~46.2	40.2	5622
Hg	Hg-203	46.60 day	33.8 ~ 36.9	35.35	2192
I	I-128	24.99 min	150~250	200	70
Se	Se-75	119.77 day	448.7 ~ 606.1	527.3	79.06
Sm	Sm-153	46.704 hr	72~7.3	725	00707
Sr	Sr-87m	167.7 min	640 ~ <b>87</b> 0	<b>7</b> 60	150
Th	Pa-233	27.0 day	044~0.525	048	005

Daily intake of micro-elements by a Korean geriatric population is listed in Table 8. The data is compared with RDA values[9]. Ca intake is only 34% of the RDA value. Also some important micronutrients intakes are much lower than the RDA values such as Cr(24%), Fe(57%), Se(49% for male) and Zn(50%). However Na intake is 525% of the RDA values. High Na intake can be explained by their high consumption of salted food such as Kimchi which is a Korean representative foodstuff, salted soup, fish and etc.

Table 13. Comparison of Daily Diet Intake by

Korean Geriatric Population with RDA[9] and the

Literature Values[10].(Unit: mg/day)

Element	This Work	RDA	Literature	
Ca	407	1200	320~920	
K	1807	2000	2300~3900	
3.6	Male : 192	Male: 420	190 ~ 420	
Mg	Female: 245	Female: 320	190 420	
Na	2625	500	3500 ~ 6900	
Cr	0.048	0.2		
Fe	5.68	10	8.1 ~ 30	
Mn	4.2	5 .	2.2 ~ 8.6	
g -	Male: 0.034	Male: 0.07	0.034 ~ 0.133	
Se	Female : 0.044	Female: 0.05	0.034 0.133	
7	Male : 7.6	Male : 15	83 ~ 14	
Zn	Female: 6.0	Female: 12	8.3 ~ 14	

#### 4. Conclusion

Minerals and micronutrients are very important components in diets. Both their deficiency and excess may cause serious human health problems. The collected samples were prepared for over 60 year old adults in Seoul during a period of one day using the Duplicate Diet method. Duplicate Diet method is the most accurate method for estimating micronutrients intakes and an exact copy of the foods eaten and preferably weighed, to replicate the diet consumed by

an individual.

Analytical results can be classified into two groups such as macro elements(Ca, K, Mg, Na) and minor elements(Cr, Fe, Mn, Se, Zn). Analytical results for the daily intake of a geriatric population in Korea showed that except for Na every macro and minor element were lower than the RDA and IAEA study. This shows that the micronutrients intake of a Korean geriatric population is not well balanced and needs to be corrected or guided.

In conclusion, NAA is proved to be a sensitive analytical method useful for performing both a qualitative and a quantitative multi-elemental analysis of the macro and minor elements in a diet or biological materials.

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### A Provenance Study of Roof Tiles & Potteries using Neutron Activation Analysis from Katap-Ri, Korea

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#### Abstract

In Katap-ri, South Korea, potteries and roof tiles are excavated with the assumed raw materials. Using NAA, the provenance of these cultural remains was examined among the neighboring areas, and the correlation of the assumed raw materials with potteries and roof tiles was also examined. From the results of the statistics it is concluded that this cultural site is distinguished from others by several trace elements, Ba, Cr, Sc, Yb, Eu, and Rb with a 93.2 % accuracy. Also it turned out that the assumed raw materials weren't related to the potteries and roof tiles with a 96.3 % accuracy.

Keywords: potteries and roof tiles, neutron activation analysis, discriminant analysis

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#### 1. Introduction

This provenance study is aimed at figuring out the range regarding the distribution and transfer of ancient goods. Also, its purpose is to find direct proof of human activities in the production, distribution and exchange of goods. This study aims to find direct proof of the production, distribution and exchange of goods, and to understand the interrelations of those activities carried out by economical and social necessity within social communities[1].

Among the remains of Gatap-ri, Buyeo-eup, and Buyeo-gun (of Chungcheongnam-do), there are lands given to farmers by the government of Baekje, traditional homes, unidentified old facilities as well as architecture frames (estimated to be pottery kilns or attached buildings) of the Korea kingdom era.

This study will perform Neutron activation analysis (NAA) on the Gatap-ri remains and other

relics, and will discover the provenance of them on the basis of the experiment's result[2]. Also, we compare the clay samples collected and the pottery and roof tiles from Gatap-ri, Buyeo to establish the provenance.

#### 2. Experiment measure and statistical analysis

#### 2.1 Neutron activation analysis (NAA)

We performed a Neutron activation analysis on a total of 85 clay (or remains) samples collected from several places. There are 24 samples of pottery and roof tiles, 12 clay or soil material samples from Gatap-ri in Buyeo, 19 pottery samples from Anyeong-ri in Gongju, 20 pottery samples from Yongjang-ri and from ancient Wando temple sites as well as 12 samples from earthenware kiln remains from Cheongdo and 8 samples of roof tiles from the kiln sites of Cheongyang.

We examined the powder of preprocessed collected samples with the SRM at HANARO, an experimental reactor at the Korea Atomic Energy Research Institute for an hour. First, we removed the peak generated by a neutron interference during the gamma ray experiment and then selected 17 nucleuses (with high detection rate and large peak width) and used them in the analysis[3][4][5].

The analyzed elements that met those requirements are shown in Table 1.

Table 1. Analytical condition used for the analysis of archaeological samples

Irradiation	Neutron flux	Sample I	Irradiation	Cooling	Counting	Elements detected		
facility	(n/cm₂ · sec)	weight	time	time	time	Elements detected		
PTS	1.7 x 10 <sup>13</sup>	100mg	1 hr	5~10 days	2000s	Ca, K, Na, La, Sm		
PTS	1.7 x 10 <sup>13</sup>	100mg	l hr	15~20 days	4000s	Ba, Ce, Co, Cs, Cr, Eu Fe, Hf, Rb, Sc, Lu, Yb		

#### 2.2 Statistics analysis

The discovery of a remains' provenance from micro elements passed through NAA can be done by a discriminant analysis. This method is generally used for two purposes: the first one is a descriptive discriminant analysis done by analyzing the independent variables which are the best means to distinguish the remains with micro particles; the second is a predictive discriminant analysis which is used to forecast which remains are included in which relic site when there is not enough relevant information on the remains. Discriminant analysis verification is conducted to decide how meaningful the discriminant function is by using

the canonical discriminant function coefficient, Wilks' Lambda, eigenvalue and  $\chi 2$ .

General discriminant function is

$$Z_j = B_{0j} + \sum B_{ij} \cdot X_{ij}$$

Z means the discriminant point, B0 a constant, B a discriminant coefficient and X an independent

variable.

Statistically, meaningful discriminant function numbers equal to the collective number-1 or to the used predictive number, then take the smaller one among them. It is a general rule that the first or the second discriminant function is the most useful in distinguishing each group, and the other ones cannot provide key information for the classification.

#### 3. Results and Discussion

Clay, raw pottery or earthenware materials, are composed of Si, Al, Fe, Ca, K, Na, Mg. In the experiment, we don't use Ca, K, Na, Fe particles because their contents' distribution is widely overlapped and their distribution within the variable is larger than that in the fixed quantity[6]. The 21 sample's Ba value from the Yongjang-ri relic sites and from the ancient temple sites of Wando measured at ten times larger than the average, thus we replaced then with the average value since the unexpectedly large value seems unreasonable. [Co:Lu], [Ce:Sc], [La:Sm]and [Lu:Yb] elements are found to have a high fan shape and interrelation with each other given that their Pearson correlation coefficients show that the fan shape degree is higher than 0.6.

Discriminant analysis is carried out on the assumption that the provenances of the remains are all different. According to the analysis, Ba, Cr, Sc, Yb, Eu, and Rb are different according to the relics provenances and Ba is found to be the most important factor with 49.9% of  $\xi^2$ , the result of the analysis using Lambda. Table 2 represents the discriminant points from Fisher's linear discriminant functions and the values determine the relations between the relics' sites and the expected provenances. The accuracy rate of the function is 93.2%. Fig. 1 shows the dispersion based on the values of Fisher's linear discriminant functions 1 and 2.

On the assumption that the clay samples from

Gatap-ri of Buyeo are not raw pottery and roof tiles materials, we include the remains from the roof tile-kiln of Cheongyang and from Anyeong-ri in Gongju in our study. Sc, Ba, Ce, Rb and Eu can classify remains and its raw materials and Sc is the most important factor in dividing the remains and the materials, responsible for 52.7% of the classification rate. Table 3 represents the relations between the remains and the raw materials on the basis of the discriminant points of Fisher's linear discriminant functions. The accuracy rate of the discriminant functions in the classification work is 93.6%. Fig2 is a dispersion of the remains and their materials based on the values of Fisher's linear discriminant function 1, 2.

Table 2. Relations between each cultural site and predicted classification group

		•					
(%)	Site	Chungdo	Chungyang	Jindo 8	Anyung-Ri	Gatap-Ri	Total
	Chungdo	91.7	0	8.3	0	0	100
Original	Chungyan	. 0	87.5	0	0	12.5	100
Group Member	Jindo & Wando	0	0	100	0	0	100
-Ship	Anyung-Ri	0	5.0	0	90	5.0	100
	Gatap-Ri	0	0	0	7.1	92.9	100

Table 3. Relations between each cultural site containing soil samples and predicted

classification group

		Pridi	cted Group	Membershi	Р	Total
(%)	Site	Chunguaga	nungyang Anyung-Ri		Gatap-Ri	
		Chungyang.	Allyulig-Al	Garab-LT	(soil sample)	100
Original	Chungyang	100	0	0 .	0	100
Group	Anyung-Ri	0	100	0	0	100
Member	Gatap-Ri	0	7.1	85.7	7.2	100
<b></b>	Gatap-Ri	0	0	0	100	100
-Ship	(soil sample)	-	U	U	100	100

Fig. 1. Discriminant analysis result of each cultural sites: Anyung-Ri at Kongju, Yongjansung at Jindo and Bubhwasa at Wando, pottery site at Chungdo, roof tiles site at Chungyang, and Gatap-Ri at Buyeo. The possibility of each sites being seperated is of 93.2%.

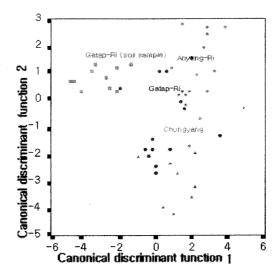
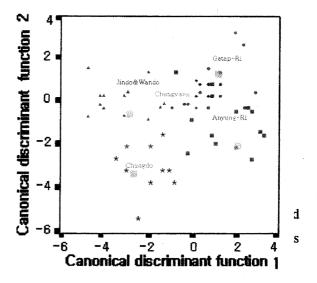


Fig. 2. Discriminant analysis result of each cultural sites containing soil samples: Gatap-Ri at Buyeo, soil samples of Gatap-Ri, Anyung-Ri at Kongju, roof tiles site at Chungyang. The possibility of each sites being seperated is of 96.3%.



#### 4. Conclusions

This provenance study is to compare pottery and roof tiles from Gatap-ri of Buyeo with other remains and to look into the relations between the clay material samples and pottery or roof tiles.

According to the result, the remains are distinguished from each other by Ba, Cr, Sc, Yb, Eu and Rb and the classification rate is as high as 96.3%. The result says that the clay samples collected from Gatap-ri of Buyeo are not the raw materials of pottery and roof tiles with a 96.3% probability. It means that

remains from Gatap-ri of Buyeo were made and used by the local residents, but the raw material, clay, was collected in other places and then transferred to Buyeo.

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## Quantitative Analysis of Environmental Hazardous Elements in Solid Polyethylene using XRF and INAA

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#### **Abstract**

Wavelength dispersive X-ray fluorescence spectrometry (WD-XRF) has been applied to the direct determination of hazardous elements in solid polyethylene (PE) materials. A series of six in-house PE reference materials was prepared by compounding some additives in to the basic resin. Determination of the elemental concentrations and the homogeneity of the elemental distribution for the references was analyzed by INAA. Emphasis was placed on the analytical performance in the analysis of the PE materials by WD-XRF. Satisfactory results and a good agreement were obtained for three elements (As, Cd and Hg). Calibration graphs of a very good linearity obtained by XRF for the three elements are presented. Finally, the performance of the technique in terms of a precision (0.1-6.6%) and accuracy (0.2-7.9%) was investigated.

Keywords: Hazardous Elements in PE, WD-XRF, INAA, Precision, Accuracy

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#### 1. Introduction

Polymers are one of the most important materials of manufactured products used in various fields. Their fields of application comprise of packaging, durables, electronics, vehicles and many other aspects of industrial and everyday life. A wide range of special properties of plastics is achieved by the incorporation of various additives to the different polymeric materials[1]. Many of these additives contain inorganic components: pigments, flame retardants, various stabilizers, catalysts, anti-oxidizing agents and minor impurities. These inorganic components are added in various

concentrations, depending on the required effect, usually from ppm to a few percent.

Therefore, there is a growing demand for accurate knowledge of the element concentration in polymers[2]. Moreover, new EU directives restricting the levels of certain additives in end-of-life vehicles (ELV), waste electrical and electronic equipment (WEEE) and The Restriction of Hazardous Substances in Electrical and Electronic Equipment (ROHS). The directives, of which will affect the use of cadmium, lead, mercury and hexavalent chromium as well as brominated flame retardants.

The demand for accurate knowledge of the elemental concentration of additives in plastics has led to the development of numerous spectroscopic techniques for their determination. These techniques can be classified into two groups. (a) The first group includes such as an atomic absorption spectrometry (AAS)[3], inductively coupled plasma atomic emission spectrometry (ICP-AES) and inductively coupled plasma mass spectrometry (ICP-MS)[4], which usually necessitate a sample dissolution prior to analysis[5]. With acids, the main limitation is the necessity to work at high temperatures, which can result in the loss of volatile species. (b) The second group includes X-ray fluorescence (XRF)[6] and instrumental neutron activation analysis (INAA)[7], which allow for the direct characterization of a solid without a major pre-treatment. However, these advantages are currently restrained by the difficulties of performing a calibration, because solid certified reference materials are not readly available for polymers. In consequence, the most usual approach consists of the use of polymer samples, which have been characterized by an alternative method.

The objective of this work is to provide a non-destructive and fast analysis of hazardous elements in polymer materials, especially in PE, that avoids the sample preparation of a polymeric matrix. In-house reference materials were prepared by compounding some elements in to the PE resin. Determination of the elemental concentrations and the homogeneity of the elemental distribution for the working references was analyzed by INAA. Finally, the analytical performance of WD-XRF in terms of a linearity, precision and accuracy was investigated.

#### 2. Experimental

2.1 Preparation of Polyethylene Reference Materials

The polymer matrix studied in this work was the HDPE (high density polyethylene), which is commonly used for household appliances manufactured with an injection molding process.

Three industrial PE additives were used, allowing for the incorporation of three elements: As, Cd and Hg. The chemical structures of all the incorporated elements were in a sulfide form (As<sub>2</sub>S<sub>3</sub>, CdS, HgS). In this work, it had been attempted to produce inhouse reference materials of an elemental concentration ranging from 10<sup>0</sup> to 10<sup>4</sup> ppm. An important point was the production of compound substances which are blended additives into a polymer matrix.

At the start, we mixed three initial mixtures of each additive with a PE matrix using a henschel mixer, respectively. Mixed proportion of the PE matrix with the additives is 10:1. Then initial compounds of a pellet type (2 mm in diameter) with elemental concentrations of about  $10^4$  ppm were made by an extruding process in a hot compounding extruder at  $170^{\circ}$ C. For the purpose of increasing the homogeneity, extruding processes had been performed 3 times for each compound.

According to the additives, each produced initial compound was mixed together with the same proportion and used as blended additives to make more diluted PE reference materials. The blended additives of the pellet type were compounded with the PE matrix of a granule type in the same proportion and the same extruding procedure as that of the initial compound. As a result of the procedure, a second compound pellet, which is diluted 10 times more than the initial compound, was produced. Through the sequential and repeated procedure, compound pellets of an elemental concentration ranging from 10<sup>4</sup> to 10<sup>0</sup> ppm had been made. Also,

for the purpose of reducing the concentration interval between the compound pellet, two additional compound pellets were produced which were mixed with the pellet of a 10<sup>2</sup> elemental concentration and that of a 10<sup>1</sup> with a 2:1 and 1:2 ratio. Finally, each produced compound pellet according to the elemental concentration was converted to a disk type reference material (30 mm in diameter and 3 mm thick) for the analysis by an injection molding. Fig. 1 presents the production scheme for the PE reference material with the concentration gradient.

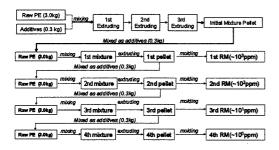


Fig. 1 The Production Scheme for PE RM with Concentration Gradient.

#### 2.2 Elemental Concentrations by INAA

Determination of the elemental concentrations and the homogeneity of the PE references was analyzed by instrumental neutron activation analysis (INAA). INAA is currently a method of choice for a trace analysis in environmental matrices, 8,9 including the characterization and certification of standard reference materials (SRMs) or certified reference materials (CRMs). 10 Each of the three samples according to each concentration level of the disk-type reference materials that had been made by an injection molding were selected. The samples were cut off to several fragments. Accurately weighed samples (about 300mg) were cleaned with 10% nitric

acid and dried for 2 hours in a drying oven at 50°C to remove the excess water.

For an irradiation, the samples that were sealed in small polyethylene capsules were placed in rabbits with an Au wire (Au/Al alloy wire: 0.1274% Au) for monitoring the neutron flux. Long time irradiation for a determination of the As, Cd and Hg were performed at the NAA #1 irradiation hole in the HANARO research reactor of the Korea Atomic Energy Research Institute and irradiated for 60 min. The maximum neutron flux attainable for a long time irradiation is  $3.19 \times 10^{13}$  n s<sup>-1</sup> cm<sup>-2</sup>. All the samples were counted by HPGe-gamma-ray detectors (Eg&G ORTEC). For long time irradiations, the decay times were in the range from 2 to 7 days. The counting time obtained for the irradiations was 2,000 sec. Evaluation and deconvolution of the peak activities were carried out using a Gamma Vision and a NAA software from Eg&G ORTEC. A Labview based computer program by using the nuclear properties of the elements and activity creation formula performed the calculations of the concentrations

#### 2.3 XRF Analysis

The elemental analysis of the PE reference samples was carried out with WD- XRF. All the measurements were carried out using a wavelength-dispersive sequential X-ray spectrometer (PW2404, PANalytical B.V., Almelo, The Netherlands) at Seoul Center, Korea Basic Science Institute (Seoul, South Korea), equipped with a side-window rhodium target 4 kW X-ray tube.

#### 3. Results and Discussion

3.1 Elemental Concentrations of the PE Reference Materials by INAA The concentrations of the incorporated elements of the reference materials determined by INAA are presented in Table 1. The analytical precision is related to counting the statistical errors (counting error Er=(N+2B)<sup>1/2</sup>/N, N is the net peak area of the nuclide of interest, B is the background area under the peak). The counting error is less than 1% for As, and between the 0.3-8.7% for Hg. For Cd, the counting error is better than 10% between 10<sup>4</sup>-10<sup>1</sup> ppm concentration level, but the counting error is 15% at less than a 10 ppm level because of the low S/N ratio, due to the low activation rate.

The homogeneity can be expressed as the relative standard deviation (RSD) of the analyzed samples. Each of the three samples of different concentration levels were analyzed. The samples were fragments of the selected disk type reference materials. The disks were selected by a pre-measurement of the X-ray signals from ten disks using XRF analysis. The method of selection was the best choice of the three disks, which radiated fluorescence X-ray signals from the interested elements of almost the highest, middle and lowest in ten samples. The RSD values are between 0.2-4.9% for the PE RMs in which the concentration level is higher than 10<sup>2</sup> ppm. But for the PE RMs with the concentrations lower than 100 ppm, the RSD values are between 1.5-8.6%.

Table 1 Elemental Concentrations of PE Reference

Materials by INAA (unit: ppm)

Sample	As(n=3)	Cd(n=3)	Hg(n=3)
PE-01	1753 ± 62	2305 ± 29	2256 ± 40
PE-02	199.3 ± 1.4	269.7 ± 7.6	$285.6 \pm 0.6$
PE-03	$130.0 \pm 1.3$	$145.0 \pm 0.7$	$185.0 \pm 9.8$
PE-04	$70.30 \pm 4.84$	91.1 ± 8.36	$110.0 \pm 9.12$
PE-05	$23.60 \pm 0.35$	$31.6 \pm 1.70$	$34.92 \pm 1.26$
PE-06	$1.54 \pm 0.05$	$1.83 \pm 0.12$	$2.50 \pm 0.23$

#### 3.2 Elemental Concentrations of PE Reference Materials by XRF Analysis

X-Y graphs were drawn for the purpose of a comparison of the results obtained for As, Cd and Hg, respectively, by two independent determinations on our six in-house PE materials. On the x axis we quote the results provided by the NAA, XRF analysis (net intensity) were plotted on the y axis. From these figures we can confirm that a very good linearity between the XRF signal and the elemental concentration measured with the INAA analysis was observed for As, Cd and Hg (R > 0.9987). Calculated slope, standard deviation of the slope and the correlation coefficient (R) are listed in Table 2. In the case of the slope and the standard deviations of a regression, there are slight differences with the concentration range. The slope is steeper in the lower concentration range than the higher concentration range. Also, the values of the standard deviation of regression are much better in the lower concentration range. This result was caused by the inter-element interferences with the XRF measurements.

Table 2 Calculated Slope, Standard Deviation of Slope and Correlation Coefficient

Element	Concentration slope	SD of	Correlation coefficient	
			regression	(R)
As	0-2000	0.5190	13.50	0.9994
	0-250	0.6824	3.140	0.9987
Cd	0-2500	0.0042	0.102	0.9997
	0-300	0.0051	0.014	0.9997
Hg	0-2500	0.1863	4.020	0.9997
	0-300	0.2194	1.382	0.9989

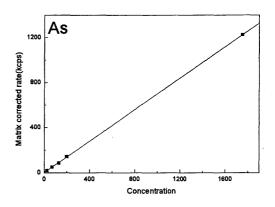
The six PE reference materials were used to obtain calibration graphs by XRF. XRF analysis was performed by a PANalytical SuperQ package. A Classic Model is used for the calculations of the

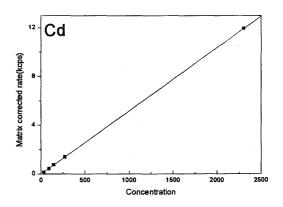
alpha' correction coefficients for matrix effects (inter-element influences) from the fundamental physical values such as an absorption and a secondary fluorescence. The matrix corrected calibration graph is shown in Fig. 2. Calculated correlation coefficient (R), intercept and standard deviation of the slope are listed in Table 3. Very nice linearity (R=1) and intercept signals made it possible to calibrate the technique for other numerous elements.

Both the repeatability and the accuracy of the measurement with PE RMs by XRF were evaluated. The analyzed and calculated values are listed in Table 4. The repeatability was estimated by calculating the relative standard deviation (RSD) between the mean values obtained from three consecutive measurements for each sample. Experimentally, we obtained relative standard deviations in the range 0.08 to 6.62% for elemental concentrations levels above 20ppm with WD-XRF. But in a lower concentration level (< 10ppm), the RSD values of all the elements exceed 10%.

Table 3. Calculated Correlation Coefficient (R), Intercept and Standard Deviation of Slope

Element	Correlation coefficient (R)	Intercept (kcps)	SD of regression
As	1.0000	0.0001	3.0666
Cd	1.0000	0.0002	0.0160
Hg	1.0000	0.0004	1.4435





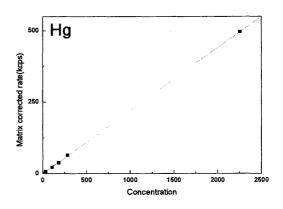


Fig. 2 Matrix Corrected Calibration Graph

The definition of an accuracy is the degree of agreement between the result of a measurement and the true value of the measured one. In this paper, the accuracy was evaluated by calculating the error between the measured mean value (n=3) by XRF and the reference value by INAA for each sample. We obtained a relative error (% error) ranging from 0.2

to 7.9% for elemental concentrations levels above 20ppm. But at a lower concentration level (< 10ppm), the % error values of Cd and Hg were above 50%. In the concentration levels over 20ppm, this good repeatability and accuracy made it possible to analyze the satisfactory confidence intervals on the calibration graphs.

Table 4 Precision and Accuracy of PE RM

Reference Material		PE-01	PE-02	PE-03	PE-04	PE-05	PE-06	
Precisio n	As	mean	1749	204.3	123.9	72.2	24.7	1.5
		SD	2.9	0.3	0.3	0.2	0.1	0.3
		RSD	0.2	0.2	0.2	0.2	0.4	22.0
	Cd	mean	2354	274.4	153.4	92.4	34.0	5.1
		SD	4.6	0.4	2.7	2.0	2.3	0.8
		RSD	0.2	0.2	1.8	2.2	6.6	15.1
	Hg	mean	2262	286.6	172.7	102.8	33.5	1.2
		SD	1.9	0.9	1.1	0.3	0.6	0.4
		RSD	0.1	0.3	0.7	0.3	1.9	33.9
	As	NAA	1753	199.3	130.0	70.3	23.6	1.5
		error	4.0	5.0	6,1	1.9	1.2	0,0
		% error	0.2	2.5	4.7	2.7	4.9	0.0
Асс <b>итас</b> У	Cd	NAA	2305	269.7	145.0	91.1	31.6	1.8
		error	49.0	4.7	8.4	1.3	2.5	3,4
		% error	2.1	1.8	5.8	1.4	7.9	189.3
	Hg	NAA	2256	286.0	185.0	110.0	34.9	2.5
		error	6.0	0.6	12.4	7.2	1.4	1.4
		% error	0.3	0.2	6.7	6.5	4.0	53.6

These values are attributable to both the good stability of the X-ray signal and the fine homogeneity of the PE samples produced. But, in the concentration levels below 20ppm, both values were increased rapidly except for the %error of Arsenic. The high RSD values and %error were caused by a signal unstableness. The fluctuation of the signals shows the low homogeneity of the incorporated additives in the sample bulk or the very low S/N ratio according to the lower limit of detection.

In this experiment, we used a sample cup of 27mm diameter for the XRF measurement. In other words, irradiation area of X-ray is circles of 27mm diameter. Also, the sample cup was rotated during the measurement by a sample spinner. Therefore, we drew a conclusion that the cause of the signal unstableness was attributed to the low S/N ratio rather than the sample homogeneity.

#### 4. Conclusions

The solid PE reference material production protocol suggested in this work seemed to be appropriate for the production of solid reference materials of other polymeric materials. Even if the present limitation is still the lack of commercially available certified polymer reference materials, it has been shown that such reference materials can be produced relatively easily at the laboratory level. Satisfactory agreement was obtained between the XRF signal and the elemental concentration measured with the INAA analysis of As, Cd and Hg. The results presented in this work suggest that X-ray fluorescence spectrometry is a suitable technique for a qualitative, semi-quantitative and quantitative determination of elements in plastics. It has been demonstrated that a satisfactory performance over a 10 ppm concentration level can be achieved. It thus seems very interesting to determine elements at a trace level and at higher concentrations without any sample preparation or dilution. The results demonstrate that this technique allows for a fast, nondestructive and reliable quantitative analysis in so far as reference materials are available, and the matrix matching of the sample and standard is verified.

#### Acknowledgements

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## Trace Elements of Fine Ambient Particulate in Korea by using an Instrumental Neutron Activation Analysis

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#### Abstract

In this study, using instrumental neutron activation analysis we undertook the measurement of about 30 elements in airborne PM 2.5 samples at an urban area of Daejeon city located in the southwestern region of Korea. An inspection of our measurement data indicated that the distribution patterns of the elements were clearly distinguished among the elements with their concentrations ranging over four orders of magnitude. The means for Lu and Dy were found to be the lowest at values of 0.01 and 0.04 ng m<sup>-3</sup>, while those for K and Fe showed the highest values of 671 and 653 ng m<sup>-3</sup>, respectively. The results of the FA analysis indicate that there are no more than six factors with a statistical significance, which appeared to play roles in regulating the elemental concentration levels in the area. These six factors can cover as much as 82.6 % of the total variance. Enrichment factor analysis supports an explicit interpretation of the results found by this factor analysis.

Keywords: Instrumental Neutron Activation Analysis, Air Pollution, PM 2.5, Factor Analysis, Enrichment **Factor Analysis** 

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#### 1. Introduction

In this study, using INAA we undertook the measurement of about 30 elements in airborne PM 2.5 samples that were collected from a roadside sampling station at a moderately polluted urban area of Daejeon city. Based on our measurement data, we attempted to elucidate the elemental distribution characteristics of the fine ambient particles. We also examined the inter-element relationships and the factors affecting the source processes through the application of diverse statistical approaches.

#### 2. Experimental

The sampling site is located several kilometers from the heavy-traffic intersection of the four-lane Honam highway and the Yusung hot spring region. In front of the sampling site there are two main roads with ten-lanes crossing each other. Hence, if one considers the geographic location of our study site on a real scale, the site can represent one of the areas that is affected most significantly by traffic-related pollution sources in the Daejeon city.

For the collection of airborne particulates, a PM 2.5 mini-volume air sampler (version 4.2, Airmetrics, USA) with a polycarbonate filter (φ 47mm, 0.4μm pore size, nuclepore) was used. 59 samples were collected from Dec. 2000 to May 2001. The sampler was placed about 4m above the ground. The air flow was adjusted to the rate of 5 l min<sup>-1</sup> at the beginning of a sampling, and the system ran for 48 hours. Total volume of air sampled through a filter was computed using a flow recorder from the start to the end of a sampling. Whenever the cleaning of the particle separator took place once a week, the flow rate was adjusted by a flow calibrator (MNF: 1022, Airmetrics, USA). The collected samples were prestored for 24 hours in a controlled atmosphere ( $20\Box$ , 50% relative humidity) and transferred into polyethylene vials after a weighing in the same conditions. Electrostatic charges are controlled by a <sup>210</sup>Po radioactive source.

Collected samples were irradiated with thermal neutrons using the Pneumatic Transfer System (PTS,  $\Phi_{\rm t} = 1.75 \times 10^{13} \, {\rm ncm}^{-2} {\rm s}^{-1}, \, R_{\rm cd} \sim 250)$  at the HANARO research reactor at the Korea Atomic Energy Research Institute. Samples were irradiated at the same position to minimize geometric errors. The measurements were carried out using a high purity Ge detector with a relative efficiency of 25%. This analytical system has a 1.9-keV resolution (FWHM) at 1332 keV of 60Co with the peak to Compton ratio of 45:1. The detector was connected to a personal computer and an 8k multichannel analyzer (EG&G ORTEC, 919A MCB). The calibration of the energy and efficiency was performed using multi-nuclide reference sources (NEN Products Inc., NES-602, 1" diameter disc type) that are certified by the NIST (National Institute of Standards and Technology, U.S.A.). A Gamma Vision software (EG&G ORTEC) was used for such purposes as: (1) energy and efficiency calibrations, (2) the acquisition of gamma spectra, and (3) peak analysis.

#### 3. Results and Discussion

#### 3.1 Analytical quality control

The optimum analytical conditions for various conditions including neutron flux, operating irradiation facilities, irradiation time, cooling time, counting time, etc were pre-estimated following the procedures established from previous studies [1,2]. In our previous study [2], the standard reference material (SRM: NIST, the National Institute of Standards and Technology, U.S.A., SRM 1648, urban particulate matter) was used for an analytical quality control. The concentrations of 31 elements were compared statistically with the certified or recommended values. The relative errors of Al, Ba, Br, Cl, Co, Cs, K, Mn, Na, Rb, Th, Ti, and V fell below 5%, while those of As, Fe, and Zn were less than 10%. The relative errors of Ce, Cu, Hf, I, La, Sb, and Sc were within 20%.

#### 3.2 Mass concentrations of PM 2.5 and trace element

An inspection of our measurement data indicated that the distribution patterns of the elements were clearly distinguished among the elements with their concentrations ranging over four orders of magnitude. The means for Lu and Dy were found to be the lowest at values of 0.01 and 0.04 ng m<sup>-3</sup>, while those for K and Fe showed the highest values of 671 and 653 ng m<sup>-3</sup>, respectively. To make a comparison of the data on a similar scale, the mean concentrations for each element were also plotted on a logarithmic

scale. Based on a simple comparison of the magnitude, the data sets can be grouped into five different categories: 1)  $< 10^{-1}$  ng m<sup>-3</sup>: Lu, Dy, and Hf; 2)  $< 10^{0}$  ng m<sup>-3</sup>: Sc, Th, Cs, Co, and La; 3)  $< 10^{1}$  ng m<sup>-3</sup>: Ce, Se, V, Rb, I, and As; 4)  $< 10^{2}$  ng m<sup>-3</sup>: Cu, Sb, Mn, Ba, Ti, Br, and Zn; and 5)  $< 10^{3}$  ng m<sup>-3</sup>: Mg, Ca, Cl, Na, Al, Fe, and K.

Though our measurements were made over a relatively short period of time, the PM 2.5 clearly showed the higher mean concentration of 43.6 µg m<sup>-3</sup> during spring than 30.2 µg m<sup>-3</sup> during winter with the range of 11.7 to 133 µg m<sup>-3</sup>. This enhanced PM 2.5 level during the spring season can be caused by the frequent intrusion of Asian dusts from China. In fact, this kind of intrusion occurred on Jan. 2, March 3 to 7, March 20 to 25, April 10 to 12, April 24 to 26, and April 28 in 2001. During the intrusion of Asian dusts the mean PM 2.5 level was 78.8 µg m<sup>-3</sup>, while that during without an intrusion was 31.8 µg m<sup>-3</sup>, respectively. Both major elements (like Al, Ca, Fe, K, Mn, and Ti) and minor elements (like Co, Dy, Hf, La, Lu, Sc, and Th) of crustal origin also showed much higher concentration levels even in the fine ambient particles during the period of the intrusion.

#### 3.3 Factor analysis

As a means to assess the major sources affecting elemental distributions in the study area, we conducted a varimax rotation factor analysis on our measurement data [3]. The results of the factor analysis indicate that there are no more than six factors (with statistical significance) which appear to be significant in regulating the elemental concentration levels in the area.

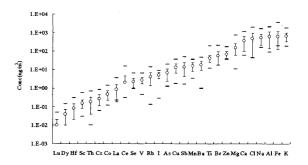


Fig. 1. A logarithmic plot of elemental concentration distributions observed from this study. Empty circles and error bars represent the mean and standard error values for each element, respectively. The thick marks are used to show both minimum and maximum concentrations for each specific element investigated during this study.

These six factors can cover as much as 82.6% of the total variance. The first factor was dominated by both many major/minor crustal elements and PM 2.5. It can explain the 44.2% of a variance. This factor indicates the contribution of resuspended soil and road dust. The elements originating from the burning of coal and fuel oils (like As, Se, and Zn) appeared to be the most important components of the second factor with an additional 9.9% of a variance. The third factor, while comprising 7.9% of a variance, was characterized by vehicular emissions - the term applied to both gasoline and diesel emissions. The fourth factor with an additional 7.8% of a variance was dominated by the elements of Ce and La. This factor means the 3-way catalytic converter of vehicles. The fifth and sixth factors were affected mostly by sea salt of Na and Cl and the copper smelting of Cu and I, respectively.

In fact, the results of the EF analysis are in line with the results found by this factor analysis. The dominant fraction of the first factor (such as Ca, Fe, K, Mn, Ti, and many rare earth elements) had consistently low enrichment factors (EF < 10). Most of them are found to be of a crustal origin. Most of the elements of an anthropogenic origin (like Sb, Se, Br, I, As, Zn, and Cu) were enriched with their EF values greater than 50. Thus, the results of the EF analysis support the idea that there are a number of source processes (including vehicle-related emissions) which control the metal concentration levels in the study area.

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#### **Applications of PGAA at HANARO Research Reactor**

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#### **Abstract**

In connection with the application of the PGAA in Korea, the concentrations of boron and hydrogen in metal and biological samples has been determined by using the PGAA facility at the 30MW HANARO Research Reactor, KAERI, for a few years now. The analytical quality control is implemented by using some standards and certified reference materials. BNCT application and the measurement of the ratio and contents for C, N and S in explosives compound are investigated. The silicon study on an air monitoring filter is processing.

Keywords: HANARO Research Reactor, Prompt Gamma-ray Activation Analysis, Boron, Hydrogen,

Element Analysis, BNCT

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#### 1. Introduction

Prompt gamma neutron activation analysis (PGAA) is useful as a nondestructive on-line measurement method for an elemental analysis. In this analytical method, the neutrons and gamma-rays are highly penetrating for most of the materials, a measurement is performed non-destructively and in removing problems of an incomplete dissolution, contamination, and loss [1]. Therefore, the PGAA method, which requires no special sample preparation as a non-destructive method is suggested as an appropriate analytical method for the analysis of light elements in a solid material[2]. Generally, because hydrogen and boron have a low atomic number as well as their nuclear properties, it is difficult to determine by neutron activation analysis, atomic absorption spectrometry, particle induced X-ray emission spectrometry or by X-ray fluorescence spectrometry, and also the chemical methods are not suitable for the determination of these materials at low levels [3].

Boron is known as an essential trace element for the growth of plants. It is in charge of the uracil formation and normal growth and its deficiency as well as toxicity threatens a crop productivity in many areas [4]. It has been reported that boron has effects upon the metabolism of calcium, phosphorus and magnesium in animals [5-6]. Boron is present in animal tissue in low concentrations of about a few mg/kg and is probably an essential micronutrient for humans. Its deficiency in plants may result in a reduced growth, yield loss, and even death, depending on the severity of the deficiency. In contradiction to this, an excess of boron in food products and drinking water is toxic to plants and animals, which is also considered harmful to ones health [7]. Boron occurs as an impurity in steels, a major component in glass, a thermalizing agent in nuclear reactor materials such as the boron carbide of the control rods, a detecting system for neutrons, a nutrient element in biological and environmental samples and a dopant in silicon wafers and semiconductor devices [8]. Boron is also used as a source for the short range alpha particles in a cancer treatment using boron neutron capture therapy (BNCT) [9-10].

Hydrogen content in various materials has a key role in a material characterization. For example, a labile trace element of the hydrogen content in metals causes an embrittlement, expands the contamination of heavy metal in soils and its deficiencies in plants causes growth damage. The presence of trace element amounts of hydrogen in metals and other materials is known to alter the material properties even at mg/kg levels. Although hydrogen in many materials is important and bound as moisture at percentage or sub-percentage levels, PGAA is suitable for the detection of hydrogen, both at high levels and at lower levels, in a wide variety of materials [11-12].

The purpose of the present paper is to introduce the measurement results of boron and hydrogen obtained from the PGAA system at the HANARO research reactor, KAERI.

#### 2. Experimental

#### 2.1 Measurement with PGAA

The sample is irradiated in a beam size of 2×2 cm<sup>2</sup> with a neutron flux of about 2.5×10<sup>8</sup> n·cm<sup>-2</sup>·s<sup>-1</sup>. The detector system consists of a high-purity Ge detector surrounded by eight BGO and two NaI(Tl) scintillators as an annulus type to reject the Compton scattered photons. A 43 % HPGe detector (EG&G Ortec, USA) connected to a PC based 16k MCA (919 MCB, EG&G Ortec, USA) is used for the counting of the prompt gamma rays. A detector is placed face-wise front of the sample position at 90° to the path of the diffracted neutron beam. The sample is usually positioned 25 cm from the face of the detector [13]. A sample for an irradiation is

suspended in a neutron beam by means of Teflon strings at a 45° angle to both the detector and the neutron beam. The variations in the neutron flux are monitored by a periodic irradiation of a Ti foil, which has a simple gamma-ray spectrum and its 1382 keV gamma ray was used.

#### 2.2 Sample Preparation

In the biological sample, the sample type for the boron analysis is prepared with powder. The powdered samples were dried using an oven at 30°C for two hours and then a cooling at room temperature. The obtained sample was put into a polytetrafluoro-ethylene (PTFE) vial with a lid and stored in a desiccator. To check the moisture content, the sample was weighed before and after a drying. The moisture content of the biological sample was less than 1.5 %. The standard reference materials (SRM) of the biological samples used, are the following; the peach leaves (NIST SRM 1547), the apple leaves (NIST SRM 1515), the spinach leaves (NIST SRM 1570a), the tomato leaves (NIST SRM 1573a), the total diet (NIST SRM 1548), the typical diet (NIST SRM 1548a), the oyster tissue (NIST SRM 1566b), and the corn bran (NIST SRM 8433) [14].

In the BNCT study, to obtain the accumulation rate in each organ such as skin cancer, blood, spleen, liver, kidney and the brain, a standard solution of boron was administered by i.p. injection with a dose of 750 and 75 mg/kg body weight in mice [15]. The B16-F10 melanoma cells exponentially growing in Dulbecco's modified Eagle's medium (DMEM) supplemented with 100 U penicillin/mL, 100 μg streptomycin/mL and 10% fetal calf serum were inoculated (1×10<sup>5</sup> cells) into the back of 6-week-old male C57BL mice. About 12 days later, the tumors reached a suitable size for the experiments. The aqueous suspension of BPA was prepared at a concentration of 100 mg/mL. A dose of 750 mg/kg body weight of BPA was administered to mice

intraperitoneally and the mice were sacrificed three hours later. BSH was dissolved in a physiological saline at concentrations of 6 mg/ml, and a dose of 75 mg/kg body weight was injected via a tail vein and the mice were sacrificed one hour later. In a combination of BPA and BSH, the BSH was injected two hours after the BPA administration and the mice were sacrificed one hour later. Many biological samples which contained a tumor were excised for the measurements of the <sup>10</sup>B-concentrations.

The sample preparation for the hydrogen analysis in alloy metal is as follows. The low alloy steel material is made using the mold. The mold was used for making the standard sample size with the upper part of 410×410 mm and the lower part of 300×300 mm of the inverse lozenge-shaped. The preparation procedures of reference materials are as follows; each ingredient was input into the 1000 kg vacuum induction furnace for a melting, next a casting and an ingot cutting that is the top and bottom with a band saw, forging after reheating at 1050 °C for one hour, homogenizing treatment at 900 °C for four hours, billet rolling and a repeat homogenizing, cutting of the appropriate size for the reference material which can be machined to a chip form, bar and disc form, and then storage. These designed chemical compositions for the reference materials of low alloy steel are mainly fluorine of 97.61~97.66 m/m%, and about 1.5 m/m% among the remainder is carbon, manganese, phosphorus, silicon and sulfur as main elements and the other elements are below 1 m/m%. The metal sample of the granule type had a circularity of one layer in the sheet made by KRISS [16]. The low alloy steel samples used are attached with one layer of a compressed paper sheet 8 mm in diameter. A granule of a sample has a size of 0.5 mm, which had a rhombic with intervals of 3 mm on paper sheet for reducing the neutron self-shielding and scattering and the gamma-ray attenuation. The SRM was also prepared by the same method. The

sample granule within the paper sheet is wrapped in a thin Teflon sheet for preventing a drop. The blank made with paper, which was wrapped with a thin Teflon sheet, is irradiated for confirming the background and interference peaks appearing from the paper.

The prepared explosives sample of the chemical compound was put into a Teflon wrap with a lid and stored in a desiccator. To check the moisture content. the sample was dried before and after a weighing. The moisture content of the sample was less than 0.5 % from the weight difference after a drying. The weight of the samples were measured three times per each sample. The chemicals used are the following; melamine (MEL, C<sub>3</sub>H<sub>6</sub>N<sub>6</sub>, Aldrich, 99%), 4-nitrophenol (NP, C<sub>6</sub>H<sub>5</sub>NO<sub>3</sub>, Fluka, 97%), paminobenzoic acid (ABA, C<sub>7</sub>H<sub>7</sub>NO<sub>2</sub>, Fisher, 98%), nitrobenzene (NB, C<sub>6</sub>H<sub>5</sub>NO<sub>2</sub>, Osaka, 99%), triethyl phosphate (TP,  $C_6H_{15}O_4P$ , Yakuri, 99%), polyethylene (PE, (-CH<sub>2</sub>CH<sub>2</sub>-)<sub>n</sub>, Aldrich), sodium nitrite (NaNO2, Junsei, 99%), sodium nitrate (NaNO<sub>3</sub>, Kanto, 99.95%), and sodium carbonate (Na<sub>2</sub>CO<sub>3</sub>, Aldrich, 99.95%). Imitators of ethylene glycol dinitrate (EGDN,  $C_2H_4N_2O_6$ and pentaerythritol tetranitrate (PETN, C<sub>5</sub>H<sub>8</sub>N<sub>4</sub>O<sub>12</sub>) reproducing the same relative concentrations of carbon, hydrogen, nitrogen, and oxygen as in the real ones which were prepared from a mixture of PE, NaNO<sub>2</sub>, NaNO<sub>3</sub> and NaCO<sub>3</sub>.

#### 3. Analysis

#### 3.1 Analysis of Boron

The boron peak can be overlapped with the prompt gamma rays from <sup>24</sup>Na (472 keV). This contamination came from the NaI(TI) scintillator making up parts of the Compton-suppressed detector. The statistical fluctuation of the boron peak concealed the sodium peak in the case of a high boron content in the samples. A minor contribution results from the LiF tiles used as a neutron shielding material. In this case, the 478 keV peak from the

lithium  $[^6Li(n,\gamma)^7Li]$  reaction was not broadened. The most important background sources are from the sodium impurities in the detector, oxygen and nitrogen in the air, aluminum in the sample holder and fluorine from the Teflon holding the samples. The prompt gamma-ray spectrum in Fig. 1 shows the Doppler-broadened boron peak (478 keV), as well as the strong hydrogen peak (2223 keV) and the single- and double-escape peaks. Several peaks corresponded to the neutron capture reactions of carbon and fluorine. In Fig. 2, the boron peak was overlapped with the prompt gamma-rays from <sup>24</sup>Na with 472 keV. The results are either known from the literature or can be determined using the standards experimentally. These intensity ratios can be used to subtract the appropriate counts from the combined boron plus the sodium peak in the 468-487 keV region. The effect of an interference from lithium can also appear, but in the high boron concentration, the peak shape of lithium appeared weakly in the boron region and the count ratio was below 1.5 % within the boron broaden peak area. In the low boron concentration below 5 mg/kg, the lithium peak of the 477.6 keV has no effect on the boron peak [17]. The analysis results were reconsidered about the effect where the count ratio of lithium in the broadened peak area was about 3~5 %. After a background subtraction, the broaden boron peak shape is showing as in Fig. 3.

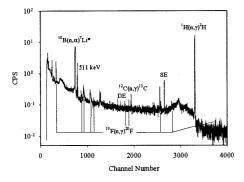


Fig. 1. Partial prompt gamma-ray spectrum of the peach leaves measured at the PGAA facility.

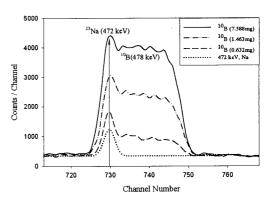


Fig. 2. Part of the prompt gamma-ray spectra near the boron peak for the various boron concentrations.

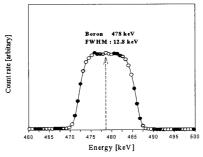


Fig. 3. Doppler-broadened prompt boron peak at 478 keV obtained from the thermal neutron irradiation of a boron (boric acid) sample. The circles in the figure are the measured data points and the solid line is the fit.

#### 3.2 BNCT Application

The boron concentrations in tumors, tissues, blood and cultured cells were estimated from the calibration curves obtained by using standard samples for the BNCT study. Boronophenylalanine (BPA) was used for the boron neutron capture therapy of brain tumors in both experimental animals and humans and/or sodium borocaptate (Na<sub>2</sub>B<sub>12</sub>H<sub>11</sub>SH; BSH). BPA is well known as a boron compound for malignant melanoma and BSH for malignant glioma. When tumor cells accumulate these compounds and receive thermal neutrons, the tumors could be destroyed efficiently because the  $^{10}$ B(n, $\alpha$ ) $^{7}$ Li reaction releases a particle and a recoiling  $^{7}$ Li ion with an average total kinetic energy of 2.34 MeV. These particles have the characteristics

of high linear energy transfer (LET) radiations with large biological effects. However, their ranges are very limited; namely, they have a path length of approximately one cell diameter ( $10~\mu m$ ). Since this average path length is probably the size of a tumor cell of  $10\text{-}14~\mu m$ , it is possible to selectively destroy a tumor without affecting the surrounding tissues. Therefore, a concentration of  $10\text{-}35~\mu g$  boron per gram for a tumor which is equivalent to  $10^8\text{-}10^9$  atoms of  $^{10}B$  per cell, is required to destroy the tumor. This concentration range is due to the location of the boron compound at or inside the tumor cells. The boron contents of the animal tissues are very important for BNCT and especially in the areas of tumors.

The <sup>10</sup>B concentrations in the tumor and other tissues after a BPA and a BSH administration were measured as shown in Fig. 4. In the boron concentration in the BAP, BSH and a combination of BPA and BSH, the value of the kidney was about five to seven times higher than the other tissues. Generally, in the present measurement of the mouse tissues, the boron concentration ratio of the tumor for each tissue in the BPA appeared to be about 1~4 times higher, but the ratio of a part of the tissue in the BSH was high.

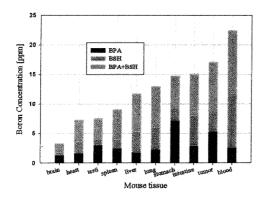


Fig. 4. Comparison of the boron concentration in the BPA, BSH and a combination of BPA and BSH.

#### 3.3 Analysis of Hydrogen

Neutron captured by hydrogen in the shielding materials and in the atmosphere gives rise to hydrogen background gamma rays in the PGAA facilities. The magnitude of this background is dependent upon the matrix of the sample and the surrounding component. To confirm the background effect for the determination of hydrogen, the measurement of a blank spectrum is necessary prior to a sample measurement. The count rate of the hydrogen peak arising from the off-neutron beam was estimated to be about 0.07±0.01 cps corresponding to approximately 13 µg of hydrogen. The count rate for the interference peaks without the blank appeared at 0.20±0.01 cps in the measurement for 10.000 sec.

The interference peaks by the measurement condition with a sample position as a system matrix and the background level in the hydrogen energy region as a sample matrix were investigated periodically. The interference peaks which appear from 2220~2222 keV and 2224~2225 keV give rise to an influence for a hydrogen analysis. The main background regions appear from the tail of a high and low energy region near the hydrogen peak area. Another possible source of a background arises from the Compton scattering of high energy gamma-rays. This results from a decrease in the signal to noise ratio by raising the continuum baseline under the hydrogen peak. Because of the complexity of the prompt gamma-ray spectra, spectral interferences often become a major source of uncertainty in the PGAA measurements.

As shown in Fig. 5, interferences for the hydrogen peak region appear with a few peaks. The background energies are 2221 keV, 2223.3 keV, 2224.4 keV and 2226.8 keV, where the remainder of the peaks except for 2222 and 2225 keV have an extremely small peak area. The total count rate in the hydrogen gamma energy region for a blank is about 0.43 cps and the percentage of the count rate per energy for the above four energies is 6 %, 21 %, 45 % and 28 % respectively. Neutron capture by hydrogen in the atmosphere and shielding materials

gives rise to about 0.2 counts per second of a gamma radiation. The count rate for the blank in the hydrogen energy region was about two times higher than that for the air condition of the on-beam stage. Before the sample is measured, the fluctuation of the count rate for the hydrogen peak was checked using a blank and the variation of the peak area appeared as below 1 % for a 10,000 sec measurement as shown in Fig 6.

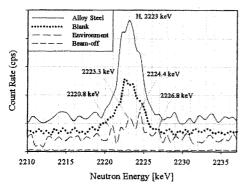


Fig 5. The comparison of the sample and blank spectrum obtained from measurement of the low alloy steel. A few peak weakly appeared give to influence at the hydrogen peak in spectrum.

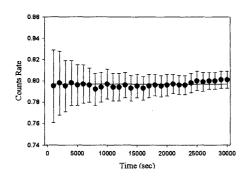


Fig. 6. The Variation Rate of Peak Area on the Hydrogen Energy Region.

#### 4. Results and Discussion

#### 4.1 Analysis of Boron

#### 4.1.1 Biological

The combined uncertainties were in the range of 3.86 to 4.0 % for the boron analysis in the relevant materials. The main sources of the uncertainties are due to statistical errors ( $0.20 \sim 0.34$  %), the detection efficiency (2.8 %), the background subtraction of 472 keV Na ( $2.4 \sim 2.65$  %), and error sources

including some other corrections. The analytical results of NIST SRMs are summarized in Table 1. The relative errors of the measured values are  $1.0 \sim 3.0$  % for the apple leaves, tomato leaves, spinach leaves and peach leaves, 13 % for the typical diet, and about 40 % for the total diet, oyster tissue, and corn bran. The analysis of boron using the present PGAA facility is in good agreement with the certified values in above a 10 mg/kg concentration, but in below a 5 mg/kg concentration, the measured values are higher than the certified values. Fig. 7 shows the Z-score from the comparison between the experimental and certified values in terms of the ratio of the experiment to certified values.

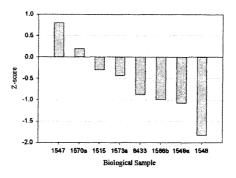


Fig 7. Comparison of the analytical results for the biological NIST SRM.

Table 1. Comparison between the measured values and certified values for the NIST SRM samples.

Sample	Present	Certified	Others	
(NIST SRM)	(mg/kg)	(mg/kg)	(mg/kg)	
Peach (1547)	28.2±1.0	29.0±2.0	27.6±2.80 11(a)	
			26.2±1.00 <sup>12(e)</sup>	
Apple (1515)	27.3±1.0	27.0±2.0	28.6±3.70 12(e)	
Spinach (1570a)	37.3±1.5	37.6±1.0	30.1±3.50 <sup>13(b)</sup>	
Total Diet (1548)	3.6±0.6	2.50	-	
Typical Diet	4.7±0.1	4.16±0.1	4.22±0.04 <sup>14(c)</sup>	
(1548a)			<u> </u>	
Oyster Tissue	4.0±0.2	2.80±1.2	-	
(1566b)				
Tomato (1573a)	33.9±1.2	33.3±0.7	32.4±4.00 13(b)	
Corn Bran	3.9±0.4	2.80±1.2	2.06±0.06 15(d)	
(8433)			4.50±1.40 12(e)	

- (a) PGAA Method, (b) PGAA  $k_0$ -Method,
- (c) PGAA  $k_0$ -Method, average value of  $4.15\pm0.02$
- (cold neutron) and 4.28±0.07(thermal neutron),
- (d) ICP-MS Method, (e) ICP-MS and ICP-OES, average value, where corn bran 8433 is RM.

#### 4.1.2 BNCT

first samples **BPA** were given intraperitoneally at a dose of 750 mg/kg body weight and the distribution in the various tissues was measured three hours after an administration. The BPA <sup>10</sup>B concentrations in the blood, liver, kidney and tumor were 2.58, 1.77, 6.26 and 5.32 mg/kg, respectively. The tumor/ blood, tumor/lung, tumor /heart, tumor/liver, tumor/ spleen, tumor/kidney, tumor/stomach, tumor/testis, tumor/brain and tumor/intestine <sup>10</sup>B ratios were 2.06, 2.31, 3.26, 3.01, 2.17, 0.85, 1.77, 0.73, 4.09 and 1.82, respectively.

The second samples were given BSH at a dose of 75 mg/kg body weight, the <sup>10</sup>B concentrations in the blood, liver, kidney and tumor were 8.94, 3.61, 16.23 and 3.54 mg/kg, respectively (Table 2). The tumor/blood, tumor/lung, tumor/heart, tumor/liver, tumor/spleen, tumor/kidney, tumor/ testis, tumor/ stomach, tumor/brain and tumor /intestine <sup>10</sup>B ratios were 0.40, 0.93, 1.74, 0.98, 2.24, 0.22, 3.28, 1.65, 6.56 and 0.69, respectively. In the case of a combination of BPA and BSH, the BSH at a dose of 75 mg/kg body weight was injected two hours after the BPA administration at a dose of 750 mg/kg body weight. The measured boron concentrations in the blood, liver, kidney and tumor were 10.91, 6.35, 17.05 and 8.24 mg/kg, respectively

Boronophenylalanine (BPA) was administered at a dose of 750 mg/kg body weight intraperitoneally. The <sup>10</sup>B concentration in the tumor was 5.32 mg/kg and the boron concentration ratio of the brain to tumor was 1.30. In the case of the borocaptate sodium (BSH) at a dose of 75 mg/kg body weight, the <sup>10</sup>B concentration in the tumor was 3.54 mg/kg and the boron concentration ratio of the brain to tumor was 0.54.

Table 2. <sup>10</sup>B concentration of each tissue after administration.

#### 4.2 Analysis of Hydrogen

#### 4.2.1 Metal and Biological

The hydrogen concentration of the SRM sample was determined from the measured count rate of the hydrogen peak, sample mass and hydrogen sensitivity. The results are summarized in Table 3 together with the certified values. The relative errors of the measured values are about 3.5 % for a high hydrogen concentration such as SRM 2453, about 6 % for the 60 mg/kg concentration such as the titanium-base alloy. The analysis of the hydrogen content using the present PGAA facility is in good agreement with the certified value of the NIST SRM.

Table 3. Comparison with the Present and Certified Values for the Biological and Metal Samples [In the A±B, B is S.D].

Reference Samples	Hydrogen [mg/	RE (%)	
NIST SRM	Certified	Present	` ´
1566b (Oyster Tissue)	$7.20 \pm 0.40$	$6.50 \pm 0.5$	9.7
1632C (Coal)	5.11 ± 0.12	4.55 ± 0.7	11
173C (Titanium Alloy)	60	56.6 ± 1.5	5.7
2453 (H in Titanium)	114	109.5 ±12	4.0

#### 4.2.2 Low Alloy Steel

In the determination of hydrogen as shown in Table 4, the content deviation appeared at about 3 % for the SRM. In the case of each RM sample, the content of the hydrogen had the same results. The spectrum for the low alloy steel is shown in Fig. 8. The ratio of the hydrogen count in the low alloy steel is not high, but the measured data appeared high because the used blank is compressed with a few paper filters which were adhered to the sample.

	Boron Concentration (mg/kg) (mean ± SD)					
Dose of BPA and BSH	blood	lung	heart	Liver	spleen	kidney
BPA 750 mg/kg/b.wt	2.58±0.21	2.30±0.25	1.63±0.18	1.77±0.18	2.45±0.26	6.26±0.51
BSH 75 mg/kg/b.wt	8.94±1.33	3.81±0.51	2.04±0.33	3.61±0.37	1.58±0.43	16.23±2.15
BPA 750 mg/kg+BSH 75 mg/kg	10.91±0.51	6.89±0.55	3.66±0.30	6.35±0.33	5.03±0.28	17.05±0.51
	Boron Concentration (mg/kg) (mean ± SD)					
Dose of BPA and BSH	testis	stomach	brain	intestine	tumor	
BPA 750 mg/kg/b.wt	3.01±0.32	7.25±0.66	1.30±0.20	2.92±0.28	5.32±0.43	]
BSH 75 mg/kg/b.wt	1.08±0.43	2.14±0.30	0.54±0.10	5.14±0.54	3.54±0.44	
BPA 750 mg/kg+BSH 75mg/kg	3.45±0.15	5.42±0.50	1.48±0.14	7.04±0.42	8.24±0.57	

The sample of below 0.5 mg had an influence on interference peaks that gave a fluctuation of more the 8 % to the hydrogen net peak. In the high mass above several mg, the net area by the interference peaks was small, the fluctuation of the net peak area nearly did not show the influences.

Table 4. The comparison with the SRM and the RM for the row alloy steel, which is obtained from each calibration function for hydrogen content.

Sample	Present (mg/kg)	Other (mg/kg)**
NIST SRM 364	5.19±0.24	< 5
RM-01*	1.14±0.12	<1
RM-02*	0.91±0.14	44
RM-03*	0.98±0.10	"
RM-04*	1.11±0.08	44
RM-05*	1.03±0.08	۲,
Ave. RM	1.03±0.10	

<sup>(\*)</sup> The low alloy steel is made for using as laboratory standard material from KRISS. The sample number is to show voluntary selected sample among made reference sample

(\*\*) The value obtained from ICP/OES method

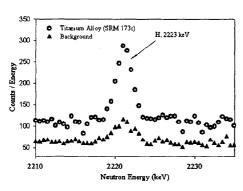


Fig. 8. The Comparison of the hydrogen peak from the real sample spectrum for the low alloy steel.

The reference value of hydrogen for the NIST SRM 364 sample was about 5 mg/kg, but the measured value was a little higher than the estimated value. The measured values for the alloy steel materials show a range of about one mg/kg, but the hydrogen concentrations of these samples have not yet been definately reported using other method. The combined uncertainty was about 3.4 % for the

hydrogen analysis. The statistical error where counting uncertainties arise from observing the radiation emitted in a nuclear decay, which shows some degree of statistical fluctuation, appeared at 2.2 % in the present study. The sources of the other uncertainties are mainly neutron flux variation for a 10,000 sec measurement (0.5 %), the spectral interference for a low sample weight (1.2 %).

#### 4.2.3 Explosives

In the present measurement for the chemicals, the count rate for 10 mg of powder type sample is about 5~6 cps and the blank is 0.33 cps. The DSPEC spectrum is well known to be the same as the single mode but the count rate was lower than the single mode as shown in Fig 9. The count rate of the single mode relative to the Compton mode for the hydrogen region appears at about 92 % and in the case of the DSPEC it is about 88 % for the Compton mode.

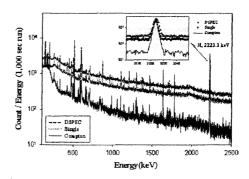


Fig. 9. Comparison of the count rate for Compton, single and DSPEC mode.

The net count rate for the chemicals is obtained from the Compton mode and shown in Table 5. The ratio of net count of carbon, 1261.8 keV and nitrogen, 1884.8 keV to hydrogen, 2223.3 keV were similar with each sample and the carbon net count of 1261.8 keV appeared in the range of 0.11 % to 0.13 % for the hydrogen net area and the nitrogen net count of 1884.8 keV is in the range of 0.33 % to 0.35 %. The explosives show that the nitrogen content in these materials appears at about three

times the carbon content, and the detection study for the explosives is shown to be useful for the prompt gamma activation analysis.

Table 5. Comparison of the ratio of counts for the carbon and nitrogen.

Sample	Composition	Weight (mg)	C/H (%)*	N/H (%)*
TP	C <sub>6</sub> H <sub>15</sub> O <sub>4</sub> P	121.36	0.12	- (/8)
ABA	C <sub>7</sub> H <sub>7</sub> NO <sub>2</sub>	101.55	0.11	0.34
Melamine	C <sub>3</sub> H <sub>6</sub> N <sub>6</sub>	100.81	0.12	0.33
NB	C <sub>6</sub> H <sub>5</sub> NO <sub>2</sub>	107.88	0.12	0.34
NP	C <sub>6</sub> H <sub>5</sub> NO <sub>3</sub>	101.93	0.11	0.33
A	C <sub>3</sub> H <sub>6</sub> N <sub>3</sub> O <sub>6</sub>	99.79	0.13	0.35
EGDN	C <sub>2</sub> H <sub>4</sub> N <sub>2</sub> O <sub>6</sub>	100.01	0.13	0.35
PETN	C <sub>5</sub> H <sub>8</sub> N <sub>4</sub> O <sub>12</sub>	99.21	0.13	0.34

<sup>\*)</sup> Gamma energies of Carbon 1261.8 keV, nitrogen 1884.8 keV and hydrogen 2223.3 keV.

#### 5. Conclusion

The degree of spectral interference peaks as a matrix effect in the sample was established for an accurate peak analysis, and then the content for the boron and hydrogen was assured with the certified reference materials (NIST SRM). In the present measurement, the measured values for the SRM under the stable beam flux which has a below 2% beam fluctuation are in good agreement with the certified value.

One of the important things about the basic and clinical studies on the BNCT is the precise and accurate measurement of the boron concentrations in biological samples. Boron must accumulate as much as possible at the tumors and cancer cell. Thus, we must develop the analysis technology for measuring the boron concentrations by using a PGAA and create good boron compounds from an accumulation respect.

The metal sample with no influence of the moistures received the affects of the interference peaks, which appeared as a count deviation from the background subtraction. The measurement results of

the low alloy steel for the hydrogen content could be confirmed within the range of a standard deviation of the five reference materials, and the total average value also showed an adjacent value with the estimate to another independent analysis method (ICP/OES).

Until now, the HANARO PGAA facility has been used for analyzing boron, hydrogen, carbon and nitrogen in a biological and metal sample. In the analysis of those samples, the biological samples of a powdered type are influenced by the moisture but metal samples of a granule type were influenced by the system matrix. On the whole, the analysis of a concentration for the light elements like boron, hydrogen, carbon, nitrogen and silicon using the HANARO PGAA facility shows good results to a few mg/kg but the concentration in the below one mg/kg appeared higher than the standard reference materials because of the influence of the background and interferences. Therefore, for an accuracy and precision analysis to below one mg/kg, the cold neutron PGAA facility will be useful for the lower concentration analysis on the light elements.

#### Acknowledgements

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## 研究会報告

8<sup>th</sup> International Conference on Nuclear Analytical Methods in the Life Sciences (NAMLS8) [2005年4月17日~22日]

東京農工大学留学生センター 川端 良子

2005年4月17日から22日まで、ブラジルのリオデジャネイロで、8th International Conference on Nuclear Analytical Methods in the Life Sciences (NAMLS8) が、開催された。参加者は総勢 197名で、発表者は年々増加しており、参加者全員が同じ会場で発表するという従来の方針を見直さなければならなかったとのことであった。また、多くの人が会議場のホテルに宿泊していたこともあり、非常に親睦的な会議であった。そのため、初めての私も、いろいろな場所で、議論に参加することができた。

個々の発表は、放射化分析のみならず、いろいろな分析方法を用いたものであり、比較検討を 行っている研究者も多くみられた。また、スタンダードの比較検討を行っている発表もみられ、 生物試料におけるスタンダードの難しさも議論されていた。また、生物試料を扱う上での、前処 理の問題も議論になっていた。

私の専門である中央アジア関係の研究発表は、順天堂大学の千葉百子先生だけで、他の国からの参加者による発表がなく残念であった.しかし、水環境汚染については、いろいろな国からの発表があり、国々によって違いはあるが、安全な飲料水確保が世界的な問題であることを再認識した.

日本からは、千葉先生のほかに、首都大学東京の海老原充先生、金沢大学の小村和久先生が、参加されていた。しかし、中国と比べ日本人の参加はすくなく、特に、中国の若手研究者がたくさん参加していたことにパワーの差を感じた。

初めての南アメリカであったが、会議スケジュールが密で、半日のリオデジャネイロ見学しか 自由時間がなかったが、有名なコルコバードの丘に登り、キリスト像をみることができ、良い記 念になった。また、フェアウェルパーティーでは、リオのカーニバルを彷彿させるカーニバルダ ンサーの舞台があり、皆さん一緒に踊るなどとても、盛り上がっていた。

## 2005FNCA 研究炉利用ワークショップ [2005年8月8日~12日]

―アジアの環境問題に貢献する放射化分析―

東北公益文科大学 大歳恒彦

平成17年8月8日から12日まで、「2005FNCA(アジア原子力協力、Forum for Nuclear Cooperation in Asia)研究炉利用ワークショップ」が、マレイシアのクアラルンプールで開催された。今年1月のタイにおける前回の 2004 ワークショップ(「放射化分析」No.18 で大浦さんが報告)に続いての開催となった。放射化分析グループには中国、インドネシア、韓国、フィリピ



2005 FNCA研究炉利用ワークショップ参加者 (開会式)

ン、タイ、ベトナムおよび主催国のマレイシアから参加があり、日本からは海老原先生(首都大学東京)、大浦先生(同)および大歳の3名が参加した。ホスト役のマレイシア原子力庁の強力なサポートによって、有意義な会合となった今回のワークショップの概要について報告する。

#### 1. ワークショップの内容

ワークショップでは、初日、開会式に続いて全体会議があり、その後、それぞれのグループに 分かれての討論が行われた。

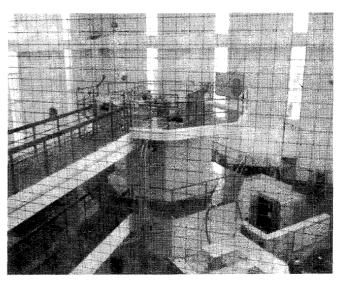
放射化分析グループでは、従来は前回のワークショップから1年間のデータやk<sub>0</sub>法の進捗状況についての報告や検討を行ってきたが、本ワークショップにおいては、放射化分析が各国においてどの程度、環境試料に適用されているかの現状レポートと、今後、FNCAの放射化分析の活動を各国の環境改善に向けて反映させるためにはどのような新しいプロジェクトが期待されるかというプロポーザルが提出された。

参加国のうち、現在は原子炉が停止しているフィリピンを除いて、環境試料の放射化分析が、個別のプロジェクトなどによって非常に精力的に行われていることがわかった。主な試料の種類としては、大気浮遊粒子、海洋試料などである。また、韓国、インドネシア、タイのように、現在ko整備段階にあるところを含め、ほとんどの機関でko法による放射化分析を実用レベルに引き上げてきており、このワークショップの成果のひとつとして認められる。参加国の多くは開発途上国であり、途上国の環境問題の究明などに放射化分析法が大いに貢献しているものと推察される。一方、現在、環境当局との連携が十分密接に行われているかどうかという点については、必ずしも満足がいくような状況ではなく、恒常的な国家環境モニタリング体制の一部に組み込まれるような放射化分析の位置づけが今後の課題となろう。一般的に、開発途上国では環境当局の力が弱く、十分な予算の裏付けが得られない場合が多いなかで、優先的に放射化分析法が取り上げられるかどうかが問題となるが、一方で、地方の環境測定機関が脆弱である開発途上国では、放射化分析による中央試験所的なシステムが威力を発揮するものと考えられ、参加国の原子力研究機関の活躍が期待される。今後のプロジェクト提案でも、各国ともに大気浮遊粒子及び海洋試料への放射化分析の適用が大半を占め、大気汚染及び海洋汚染への強い関心がうかがえる。大気汚染では、多元素分析のパターンから、発生源の寄与率推定を目的としたプロジェクトなど

#### も提案された。

#### 2. マレイシア原子力庁の見学

マレイシア原子力庁(Malaysian Institute for Nuclear Technology Research, MINT)の保有する TRIGA 炉は新首都プトラジャヤの郊外、クアラルンプールからもバスで約 1 時間の距離に立地する。1982 年に臨界に達し、熱出力 1 0 0 k w、平均中性子束密度  $1.2 \times 10^{12} \text{cm}^{-2} \text{s}^{-1}$  の性能を有する。毎週 4 0 時間程度の運転を行っている。よく整理整頓された施設内部は全体に気持ちがよい印象であり、途上国にいることを忘れてしまうほどである。放射化分析の施設もよく整備されており、気送管照射システムに隣接する測定室に設置された  $5 \sim 6$  台の Ge 半



マレイシア原子カ庁の TRIGA 炉

導体検出器が同時に稼働することができるようになっている。

#### 3. 開発途上国の環境事情

今回のワークショップ期間中に、会場のクアラルンプールでは、お隣にあるインドネシアのスマトラ島の森林火災による大気汚染(ヘイズ、靄の意味)が警戒レベルにまで達し、空港などが一時閉鎖になるとともに、学校も休校になり、われわれも戸外に出ると息苦しいような二、三日を過ごした。通りにはマスクを売る人が急に増え、町行く人々も不安げな表情であった。幸い、最終日の施設見学の日には状況も改善して青空がのぞいたが、このような状況は数年おきに起こ



森林火災による大気汚染を伝える新聞記事

っているようだ。森林火災は自国には責任のない越境大気汚染であるが、マレイシア自体も、急速な工業化に伴う生活レベルや収入の向上と引き替えに、深刻な大気・水質の汚染に直面している。バスで少し移動すると、大きな工業団地などを通りかかり、おなじみの日本企業の看板に遭遇する。我々の生活とも決して無縁とはいえない途上国の環境に接する時、複雑な心境となる。このような環境問題の解決に、放射化分析が少しでも役立てばと思う。

前回のタイの大浦さんの報告にもあるように、 東南アジアのスパイスのきいた料理はとても魅力 的だ。マレイシアはマレイ系の他、中国系、インド系などからなる多民族国家であり、料理についても、多様な味が楽しめる。筆者が好きなのは、清潔なホテルでいただく食事よりも、町中で庶民が入るレストランの野性味のある味で、おどろくほど安く、また、現地の人がどんな生活をしているのかがよくわかる。

最後に、今回のワークショップに参加させていただいた事について、海老原先生、大浦先生ならびに事務局の皆様方に心より感謝申し上げたい。

2005 日本放射化学会年会・第 49 回放射化学討論会 [2005 年 9 月 28 日 ~30 日]

武蔵工業大学工学部 鈴木章悟

標記学会が 2005 年 2005 年 9 月 28 日(水)~30 日(金)の会期で金沢市観光会館を会場にして、金沢大学・中西 孝先生を委員長に開催されました。兼六園・金沢城の近くにある会場で、日差しが痛いくらいの好天に恵まれ、高い高い秋空が金沢城の上にひろがる風景は印象的でした。

特別講演3件、学会賞・奨励賞受賞講演4件、口頭発表52件、ポスター発表85件の合計144件の講演がありました。また討論会翌日には飛騨市神岡町にあるニュートリノ研究施設への見学会が行われました。

放射化分析分科会は 30 日 12:10 より開かれた。第1部では、金沢大学・小村和久先生が「極低レベル放射能測定と放射化分析-環境中性子、宇宙線及び原爆中性子による放射化-」と題して講演されました。極低レベルの放射能による放射化について大変興味深いお話を聞くことができました。第2部では、「放射化分析研究会の総会」を兼ねて、拡大幹事会報告、研究用原子炉の現況報告と共同利用の将来、MPGA の開発状況と今後の計画について等、放射化分析に関する様々な情報交換が行われた。

放射化分析に関連した発表は、例年に比べて少なく、次の9件であったが、ポスター発表では 活発な討論がなされていた。

#### 口頭発表

k<sub>0</sub>標準化法を用いた中性子放射化分析 養殖マガキ軟体部の中性子放射化分析

(首都大)大浦泰嗣 他 (石巻専修大理工)福島美智子 他

#### ポスター発表

1964年福岡降下物における微量元素並びにセシウム 137 の分布と挙動(武蔵工大)北原照央 他 鉄製錬におけるウラン及びトリウム同位体の分配 (武蔵工大)中原将登 他

多重ガンマ線放射化分析法を用いた宇宙・地球科学研究

(武蔵工大)中原将登 他 (原研)初川雄一 他

即発ガンマ線ドップラー広がり法によるシリカゲル細孔のキャラクタリゼーション

(大同工大)酒井陽一 他

即発γ線分析装置における収斂中性子ビーム利用

(原研東海)松江秀明 他

リョウブの葉に含まれるコバルト分布の解明 高純度鉄中の不純物 Mn 測定

#### 第49回放射化学討論会に参加して

宮本 ユタカ

今年もまた、放射化学討論会の季節となった。今年の開催地はテレビの旅番組で頻繁に紹介される観光地の金沢である。10 月 17 日から中国・北京で開催される APSORC2005 へ参加するために今年の討論会参加者は激減するのではないかと危惧されていたようであったが、300 名を超える参加者で会は盛況に終わり、心配は杞憂だったようである。口頭発表用に 2 会場、ポスター発表用に 1 フロアと程よい広さであったように思う。特にポスター会場は見たり、通り抜けたりするのに不便を感じなかった。プログラムをご覧になった方はよくお分かりかと思うが、放射化分析を使った研究発表の件数は以前に比べてかなり少なかった。発表件数と放射化分析の利用者数の指標になっているとは限らないが少し寂しい印象を受けた。その一方、京都大学原子炉実験所(KUR)から韓国 KAERI の原子炉 HANARO との協力研究の話題が出るなど、今後の放射化分析の動向として気になる新たな展開も見え始めている。個人的な意見としては、KUR だけでなく日本全体と韓国、更にはアジア全体に発展した放射化分析のコミュニティーが出来れば良いと思う。

今回の討論会の特別講演では、「カムランドが切り開くニュートリノ科学の世界」と題して、東 北大学の鈴木厚人先生からカムランドの整備やそこで得られた観測データの紹介をして頂いた。 その講演の中で放射化分析として興味深かったのは、液体シンチレータの極微量分析についてで ある。ニュートリノの検出器である液体シンチレータは低エネルギー・ニュートリノ反応現象の 検出を可能にするため、4ºK や U および Th 系列核種の放射能を極力低く抑えなければならない ことは誰でも想像できるであろう。先生達は、精製したシンチレータが検出器として用いるのに 十分な純度に達しているかを確認するため、K, U および Th 濃度を中性子放射化分析(NAA)を用 いて極微量分析したというのである。蒸発乾固させた試料を米国  $MITR(\phi_{th}:5\times10^{13}~cm^{-2}s^{-1})$ で中 性子照射し、<sup>40</sup>K: <2.4×10<sup>-15</sup> g / g, <sup>232</sup>Th: <5.5×10<sup>-15</sup> g / g, <sup>238</sup>U : <8×10<sup>-15</sup> g / g の上限値(信 頼限界 90%)を得たとのことである。講演では結果のみを紹介して分析方法などの詳細には触れ なかったが、文献検索をして関連する論文[1]を見つけたのでその概要を紹介しておく。照射に用 いた液体シンチレータは約 450g であり、それを蒸発乾固させて石英管に封入した。封入した石 英管は 2001 年 1 月から 2002 年 7 月まで照射した後、カリフォルニア工科大学で開封してアセト ンと硝酸を使って内容物を洗い出した。その洗液を集めて Eichrom 社製の TRU 樹脂カラムに通 した。そのカラムをそのまま Ge 検出器で測定して樹脂上に吸着した U と Th の定量核種である <sup>239</sup>Np と <sup>233</sup>Pa の放射能を求めた。また、溶出液を Ge 検出器で測定して K の定量核種 <sup>42</sup>K の放射 能を求めた。誘導放射能から濃度への換算方法は論文に述べられていないが、試料と同時に既知 量の Na, K, Cr, Zn, Br, Sr, Sn, Sb, La, Au, Th, U の塩を照射して熱中性子束および熱外中性子 リョウブの葉に含まれるコバルト分布の解明 高純度鉄中の不純物 Mn 測定

#### 第49回放射化学討論会に参加して

宮本 ユタカ

今年もまた、放射化学討論会の季節となった。今年の開催地はテレビの旅番組で頻繁に紹介される観光地の金沢である。10 月 17 日から中国・北京で開催される APSORC2005 へ参加するために今年の討論会参加者は激減するのではないかと危惧されていたようであったが、300 名を超える参加者で会は盛況に終わり、心配は杞憂だったようである。口頭発表用に 2 会場、ポスター発表用に 1 フロアと程よい広さであったように思う。特にポスター会場は見たり、通り抜けたりするのに不便を感じなかった。プログラムをご覧になった方はよくお分かりかと思うが、放射化分析を使った研究発表の件数は以前に比べてかなり少なかった。発表件数と放射化分析の利用者数の指標になっているとは限らないが少し寂しい印象を受けた。その一方、京都大学原子炉実験所(KUR)から韓国 KAERI の原子炉 HANARO との協力研究の話題が出るなど、今後の放射化分析の動向として気になる新たな展開も見え始めている。個人的な意見としては、KUR だけでなく日本全体と韓国、更にはアジア全体に発展した放射化分析のコミュニティーが出来れば良いと思う。

今回の討論会の特別講演では、「カムランドが切り開くニュートリノ科学の世界」と題して、東 北大学の鈴木厚人先生からカムランドの整備やそこで得られた観測データの紹介をして頂いた。 その講演の中で放射化分析として興味深かったのは、液体シンチレータの極微量分析についてで ある。ニュートリノの検出器である液体シンチレータは低エネルギー・ニュートリノ反応現象の 検出を可能にするため、4ºK や U および Th 系列核種の放射能を極力低く抑えなければならない ことは誰でも想像できるであろう。先生達は、精製したシンチレータが検出器として用いるのに 十分な純度に達しているかを確認するため、K, U および Th 濃度を中性子放射化分析(NAA)を用 いて極微量分析したというのである。蒸発乾固させた試料を米国  $MITR(\phi_{th}:5\times10^{13}~cm^{-2}s^{-1})$ で中 性子照射し、<sup>40</sup>K: <2.4×10<sup>-15</sup> g / g, <sup>232</sup>Th: <5.5×10<sup>-15</sup> g / g, <sup>238</sup>U : <8×10<sup>-15</sup> g / g の上限値(信 頼限界 90%)を得たとのことである。講演では結果のみを紹介して分析方法などの詳細には触れ なかったが、文献検索をして関連する論文[1]を見つけたのでその概要を紹介しておく。照射に用 いた液体シンチレータは約 450g であり、それを蒸発乾固させて石英管に封入した。封入した石 英管は 2001 年 1 月から 2002 年 7 月まで照射した後、カリフォルニア工科大学で開封してアセト ンと硝酸を使って内容物を洗い出した。その洗液を集めて Eichrom 社製の TRU 樹脂カラムに通 した。そのカラムをそのまま Ge 検出器で測定して樹脂上に吸着した U と Th の定量核種である <sup>239</sup>Np と <sup>233</sup>Pa の放射能を求めた。また、溶出液を Ge 検出器で測定して K の定量核種 <sup>42</sup>K の放射 能を求めた。誘導放射能から濃度への換算方法は論文に述べられていないが、試料と同時に既知 量の Na, K, Cr, Zn, Br, Sr, Sn, Sb, La, Au, Th, U の塩を照射して熱中性子束および熱外中性子 束の校正を行ったと書いている。

講演の中で、鈴木先生は「この試料の照射は日本で行えなかったので米国にお願いした。」と話されており、このようなすばらしい研究に日本の研究炉が関与できなかったのは非常に残念だと思った。

[1] "Novel technique for ultra-sensitive determination of trace elements in organic scintillators", Z.Djurcic et al., Nucl. Instr. and Meth. in Phys. Res. A, 507 680-690 (2003).

## 動向

KAERI の研究用原子炉 HANARO と京都大学原子炉実験所との協力研究の紹介

京都大学原子炉実験所 高宮幸一

#### はじめに

京都大学研究用原子炉(KUR)は、2005 年2月から約1年半から2年間にわたり、新燃料の準備のために運転を一時休止する予定である。もちろんこの間、KUR を用いて行われてきた中性子放射化分析やトレーサー製造といった共同利用も休止せざるを得ない。しかし長年にわたり継続され、さまざまな重要な研究成果を生み出してきた中性子を用いた研究を、たとえ1~2年間とはいえ休止させることはできない。そこで京都大学原子炉実験所では、休止期間中にも中性子を用いた研究を継続するため、また新たな研究分野の開拓のため、韓国原子力研究所にある研究用原子炉 HANARO を用いた協力研究体制の準備を進めている。本稿では研究用原子炉 HANARO の照射施設を紹介するとともに、進行中の協力研究の方向性について説明する。

#### KAERI と HANARO について

韓国原子力研究所(KAERI)は韓国のほぼ地理的中心に位置するデジョン(大田; Daejeon)にあり、研究用原子炉である HANARO を所有する原子力関連の研究機関である。日本からのアクセスは、まず航空機によりインチョン(仁川)国際空港へ向かい、空港からデジョンまでは高速バスでおよそ3時間である。デジョンは再開発が進行中であり、中心部には近代的な高層ビルが並ぶが、郊外は森林や畑地も多くのんびりした雰囲気が漂っており、KAERIはこの郊外にある。最近ではKAERIの付近にも建設中の高層マンションがみられるようになり、周囲の環境は随分と変化しつつある。このような環境にある KAERI の敷地の一番奥まったところに研究用原子炉である HANARO は設置されている。KUR のように炉室とホットラボのような実験施設が研究者の居室から離れておらず、ひとつの建屋に原子炉、実験室、そしてスタッフの居室が同居している。そのためか、居室にいても原子炉施設や実験室が非常に身近に感じられる。

HANARO はオープンプール型の軽水炉で、最大熱出力は 30 MW である。つまり現在の KUR(5 MW)の 6 倍の熱出力をもつ。休止後再開する際の KUR の出力は 1 MW(医療照射時のみ 5 MW)であるので、 1 桁以上中性子フラックスの大きな中性子実験施設である。HANARO に直結した実験装置には、KUR と同様の圧気輸送管照射設備や即発ガンマ線分析装置などの照射孔があるが、以下では中性子放射化分析に関係する圧気輸送管照射設備などについて詳細に紹介する。

#### 圧気輸送管照射設備と周辺設備

圧気輸送管照射には NAA1,2,3 と呼ばれる 3 つの照射孔が用いられており、それぞれ  $3.9\times10^{13}$ ,  $9.4\times10^{13}$ ,  $1.6\times10^{14}$   $n_{th}/cm^2/s$  の中性子束による照射ができる。 KUR の 5 MW 運転時での Pn-1,2,3 の中性子束は、それぞれ  $1.93\times10^{13}$ ,  $2.75\times10^{13}$ ,  $2.43\times10^{13}$   $n_{th}/cm^2/s$  であり、照射可能

な中性子束は現在の  $2\sim6$  倍(1 MW 運転時と比較すると  $10\sim30$  倍)となる。NAA1 は同位体製造にも用いられるが、NAA2 および NAA3 は中性子放射化分析のみに利用されている。これらと接続された照射設備が 3 種類あり、それぞれ PTS#1,#2,#3 と呼ばれている。

PTS#1 は NAA1 に接続されており、試料の輸送と取り出しを行い、あらかじめ設定された時間での中性子照射ができる。PTS#2 と PTS#3 はそれぞれ NAA2 と NAA3 の両方に接続され、照射管の途中に分岐装置が設置されており、照射に用いる照射孔を選択することができる。PTS#2 と PTS#3 では試料の自動照射装置が利用可能で、複数の試料キャプセルを自動的に順次照射することができる。PTS#1 と PTS#2 では試料の取り出し場所が 2 箇所用意されており、放射能が弱いものは直接フードに、強いものは遮蔽体に囲まれた格納容器に取り出すことができる。また PTS#3 は照射管の末端が Ge 検出器に接続されており、照射後すぐに自動的に $\gamma$ 線の測定を開始することができる(PTS#3 は今秋から遅発中性子放射化分析(DNAA)を主目的とした装置となる)。

照射の方法は KUR などの一般的な圧気輸送管照射装置と同様で、ポリエチレン製のキャプセルに試料を封入し、圧縮ガス(HANARO では窒素ガス)を用いてキャプセルを照射孔に搬送し、予定照射時間の経過後、実験室にキャプセルが戻ってくる。照射に用いるキャプセルは円筒状で、その大きさは内径 24.3 mm、長さ 75.3 mm であり、充填できる試料の重量は梱包材などを含めた総量で 30 g(試料重量は  $10\sim15$  g)までとなっている。また照射時間は4時間までと制限されているが、安全性が HANARO 側に認められた場合、より長時間の照射ができる可能性がある。

 $\gamma$ 線スペクトルの計測は照射装置室に隣接する測定室で行うことができ、Ge 検出器(20~35%) が 5 台設置されている。うち 2 台は 40 個までの試料の自動測定が可能で、長寿命核種の測定に用いられている。HANARO の放射化分析グループでは、これらの装置を用いて主に K0 法による INAA を行っている。照射装置室や測定室に隣接して、放射性物質の取り扱いが可能な化学実験室もあるが、RNAA はほとんど行われていない(化学操作を行うことができないわけではない)。

HANARO では中性子放射化分析による元素分析サービスも行っている。試料数や定量する元素数によって価格が設定されており、韓国内の研究機関や企業はもとより、海外の研究機関からも分析の依頼を受けており、40以上のユーザーグループが利用している。ここ数年間では、研究開発のための照射が1000~6000件/年、分析依頼照射が2000~9000件/年となっている。

#### 京都大学原子炉実験所との協力研究

京都大学原子炉実験所では、KUR の一時休止している期間、つまり 2006 年 4 月から2年間、放射化分析および中性子ラジオグラフィを主とした協力研究を HANARO において行う予定である。この協力研究では、京都大学原子炉実験所の HANARO 利用プロジェクト研究として日本の研究グループから実験テーマを公募し、テーマの採択された研究グループと共同でプロジェクト研究を行うことになる。この際、京都大学原子炉実験所が窓口となり、照射実験の段取りや旅費の支給を行う予定である。しかし予算に限りがあるので、協力研究の採択数はこれまでの KUR 利用の場合より縮小される。また、本協力研究の趣旨は上述の元素分析サービスの窓口として機能するのではなく、あくまで HANARO を利用した新たな放射化分析研究の開拓を目的としているため、単なる照射利用というよりはチャレンジングな研究テーマが歓迎されることになる。現在、研究プロジェクトの公募内容や協力研究開始後の手続きの方法について準備中であるが、10

月末には研究公募を開始する予定である。また本協力研究は KUR の再開後においても継続して 行いたいと考えているため、再開後は KUR と HANARO のお互いの利点を生かすことにより、 放射化分析の適用範囲のさらなる拡張が期待できる。

#### 最後に

以上が HANARO の放射化分析実験設備の紹介と京都大学原子炉実験所との協力研究の紹介であるが、HANARO の非常に強力な中性子束と整備された照射設備を利用すれば、KUR の圧気輸送管による照射では定量できなかったり、長期照射を行わねば定量できなかった極微量元素についても新たに分析のターゲットとすることが可能となり、放射化分析の適用範囲を拡大することができる。しかし高中性子束であるため、KUR の Pn-1 などで行われているような短時間照射での短寿命核種の定量については照射に多少の工夫が必要となるであろう。来年 4 月から開始予定の協力研究では、HANARO を利用した放射化分析の特徴をわれわれが理解し、どのような研究を展開できるのかを熟考した上で実施すれば、KUR の利用だけでは不可能であった新たな放射化分析の可能性を切り開くことができるであろう。放射化分析を行っている、またこれから行おうとしている研究者の皆様には、10 月末に開始予定の HANARO 利用実験の公募において、新たな研究の拡がりが期待される多くのテーマの提案をお願いしたい。

#### MTAA 12 日本開催決定

首都大学東京 都市教養学部 海老原充

かねてより次期 MTAA (Modern Trends in Activation Analysis) の開催に関して、本誌でご報告してきましたが、日本で開催されることが決定されました。MTAA は放射化分析に関する国際会議で、第 1 回が 1961 年に開催されています。最近では 1995 年に韓国・ソウルで第 9 回 (MTAA-9)、1999 年に米国・ワシントン DC 近郊で第 10 回 (MTAA-10)、そして 2003 年に英国・ギルフォードで第 11 回 (MTAA-11) が開催されています。次回、MTAA-12 を日本で開催する方向で検討し、招致プロポーザルを提出する予定であることを前号の「放射化分析」誌(放射化分析 No. 18、p. 89-91)でお知らせしました。ここではその後の経過を簡単にご説明し、結果(本稿のタイトルにありますが)をお知らせ致します。また、今後の予定についても簡単にふれたいと思います。

#### 「プロポーザル作成及び提出」

前号以降の経過ですが、まず日本開催の具体的プランを作成するにあたって、作業グループを つくりました、メンバーとして、できるだけ集まりやすいことや、これまでの MTAA (や関連し た放射化学関係の学会) に参加された経験を持っていることを考慮して、桝本和義 (KEK)、松尾 月末には研究公募を開始する予定である。また本協力研究は KUR の再開後においても継続して 行いたいと考えているため、再開後は KUR と HANARO のお互いの利点を生かすことにより、 放射化分析の適用範囲のさらなる拡張が期待できる。

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場所について、多少説明を加える必要があるかと思われます。これまで過去数回出席した MTAAを振り返ってみますと、韓国での MTAA-9 はホテルで開催され、全体的にかなり力が入っていました。MTAA-10 は米国での会議らしく(と私には思えるのですが)、場所も NIH の会議場(だと記憶しています)を使い、すべてにおいて実質本位と印象づけられる会の運営でした。MTAA-11 はある面ではそれにさらに輪をかけたようなところがありました。Banquet で挽回したかにみえたのですが、その帰りに見事に裏切られたこともあり、全体として、MTAA-12 がやりやすくなった、と感じたものです。以上の経験を参考にしたワーキンググループでの結論として、MTAA-12 では基本的には実質本位で行うのが良いのではないか、ということになりました。金銭的に実質本位に行うとなりますと、ホテルでの開催はあきらめ、大学施設を使うという選択が残ります。箱としては公共施設でも良いのでしょうが、発表の場を考えるとやはり大学の方が楽です。というようなことから、首都大学南大沢キャンパスで開催する、ということで合意しました。首都大学は本年 4 月に旧都立大他、都立の 2 大学、1 短大を統合して発足した大学ですが、それにあわせて一部施設を新設しました。また、手頃な講堂もあり、鉄道駅(京王電鉄相模原線、南大沢駅)から徒歩でアクセスできます。

開催年としては、最近の MTAA は 4年ごとに開催ざれてきましたので、次回 MTAA-12 は 2007 に開催することを想定されていました。開催時期は大学の夏休みで、気候がある程度落ち着いて来る 9 月中~下旬を考えました。

以上のことを踏まえてプロポーザルを作成し、締め切りの 7 月 1 日までに、MTAA の実施母体である国際放射化分析委員会 (International Committee of Activation Analysis; ICAA)の MTAA 開催場所選定委員会 (Site Selection Committee) 委員長である NIST の Dick Lindstrom 氏に提出しました。この提出した招致プロポーザルを資料 1 として、この原稿に添付します。なお、このプロポーザルには LOC のメンバーのリストが載せてありますが、記載の通り暫定的なものであり、それぞれの方にご相談なしに記載しました。事情をご理解頂き、御了承下さいますようお願い致します。

#### 「決定」

プロポーザルは日本以外に、インド、スロベニア、ポルトガルから提出されたことを知らされました。場所選定委員会でこれら 4 つのプロポールを回覧し、最終選考過程に移るかどうかの審査をおこないます。8月はじめに Lindstrom 氏から追加の情報を寄せるようにとの要請がすべての提案者にありました。日本の提案書にはすべて情報が盛られていましたが、念のために追加情報を提出しました(参考資料 2 として添付)。この段階で選定委員会としては 4 つの提案をすべて受理するという決定をし、最終選考が行われることになりました。最終選考は前出 ICAA の委員が投票で行います。今回の場合、受理された 4 つの提案書が各委員のもとに送られ、それをもとに 1 番から 4 番まで順位をつけて、Lindstrom 氏に知らせます。ICAA は現在 21 名のメンバーからなります。投票は 10 月初旬に締め切られ、10 月 19 日に投票の結果が電子メールで伝えられ

ました (参考資料 3). 丁度中国で開催されていたアジア・太平洋放射化学シンポジウム (APSORC) の期間中のことでした.

投票の結果の詳細は知らされていませんが、日本の次がインドで、スロベニア、ポルトガルの順でした。Lindstrom から投票結果に対する統計的な解析結果のメールがとどいでいるので、それも参考のために参考資料3にあわせて添付します。

#### 「今後の予定」

とりあえず、以上のような経緯で日本開催が決まりました。開催結果を受けてからはまだ具体的な活動は何もしていませんが、今後、早急に組織作りをして、準備に入りたいと考えています。 前号でも書きましたが、放射化分析研究会の総力を挙げて是非とも良い会にしたいと思います。 皆様のご協力をお願い致します。

資料1 (招致申請書)

#### Site proposal for MTAA-12

# 1. Name, affiliation, postal address, telephone number, fax number, and e-mail address of the proposed Conference Chair:

EBIHARA, Mitsuru, Professor

Department of Chemistry, Tokyo Metropolitan University

1-1 Minami-Ohsawa, Hachioji, Tokyo 192-0397, Japan

Phone: +81-426-77-2553

Fax: +81-426-77-2525

Email: Ebihara-mitsuru@c.metro-u.ac.jp

#### 2. Name and address of sponsoring organization:

Japan Association of Activation Analysis (JA<sup>3</sup>)

c/o Branch office of Advanced Studies in Atomic Energy, University of Tokyo Japan Atomic Energy Research Institute

Tokai, Ibaraki 319-1195, Japan

#### 3. Dates proposed for Conference:

Late September, 2007

#### 4. Location (venue, city, country) proposed:

Minami-Ohsawa campus (main campus) of Tokyo Metropolitan University (TMU)

The main campus of TMU is located 40km west to the downtown Tokyo and can be reached in 40 to 45

min from Shinjuku by train (Keio-line). Rapid and express trains run three times an hour for each (total 6 times) with local train being served three times an hour during daytime.

#### 5. Co-sponsoring organizations, institutions, or societies:

- (1) Tokyo Metropolitan University
- (2) Japan Society of Nuclear and Radiochemical Sciences
- (3) Japan Society for Analytical Chemistry
- (4) Chemical Society of Japan
- (5) Japanese Society for Neutron Science
- (6) Atomic Energy Society of Japan
- (7) Korea Association of Activation Analysis
- (All are pending.)

#### 6. Composition of Conference arrangements committee and staff:

#### (LOC-tentative)

Fukuoka, T (Rissho Univ.)

Fukushima, M. (Ishinomaki Senshu Univ.)

Hatsukawa, Y (JAERI)

Katayama, Y. (Human Environ. Univ.)

Kawano, M. (Ehime Univ.)

Kikawada, K. (Sophia Univ.)

Masumoto, K. (KEK)

Matsuo, M. (Univ. of Tokyo)

Matsue, H. (JAERI)

Minai, Y. (Musashi Univ.)

Miyamoto, Y. (JAERI)

Muramatsu, Y. (Gakushuin Univ.)

Okada, Y. (Musashi Inst. of Tech.)

Oura, Y. (TMU)

Shibata, S. (Kyoto Univ. Res. Reactor Inst.)

Sasajima, F. (JAERI)

Sawahata, H. (Univ. of Tokyo)

Yagi, H. (S. H. I. Examination & Inspection, Ltd.)

#### 7. Lecture room accommodation (location, seating capacity, and facilities):

(Opening ceremony)

University hall (small) with 500 seats, stage and screen/sound facilities

(Regular lecture)

Two classrooms with 150-200 seats and screen/sound facilities

(Meeting)

Several small rooms for formal and informal meetings

#### 8. Poster room accommodation (location, poster capacity, and facilities):

A classroom next to or close to lecture rooms. Additional room can be prepared in accordance with the number of poster presentations.

#### 9. Exhibition space (location, floor area, and facilities):

A classroom close to lecture/poster rooms can be used for exhibition.

#### 10. Residential accommodation (state where hotel, guest house, university residence, etc):

Distance from lecture and exhibition:

Dining facilities and room amenities:

Approximate daily cost including taxes (state whether single or double room, and whether breakfast is included):

(1) Keio Plaza Hotel Tama Center (two stops from Minami-Ohsawa by train; 5 min)

(s/106 rooms) 10,164yen; (t/117 rooms) 18,249yen; (w/24 rooms) 15,937yen (tax incl.)

Breakfast: 1,300yen

(2) Hashimoto City Hotel (two stops from Minami-Ohsawa by train; 5 min)

(s/63 rooms) 6,120yen; (t/9 rooms) 9,890yen; (w/9 rooms) 9,580yen (tax incl.)

Breakfast not served.

(3) Hashimoto Park Hotel (two stops from Minami-Ohsawa by train; 5 min)

(s/88 rooms) 6,300yen; (t/2 rooms) 10,500yen (tax incl.)

Breakfast not served.

(4) International House on campus

(s/15 rooms) 10,000yen/week; (t/5 rooms) 15,000yen/week (tax incl.)

There are utilities in rooms for preparing meals.

There are many convenience stores around stations and hotels. There also are fast food stores (for hamburgers and donuts) and coffee shops there.

#### 11. Nature and amount of possible financial assistance:

Toward the direct cost of conference activities such as facility rentals, staff salaries, postage, publications, etc.:

Supports from the Tokyo Metropolitan Government, scientific societies and government-related offices will be used for this category. Benefits from advertisements and exhibitions are also allocated to this category. At least, a total of 1,000,000yen is expected to be accumulated.

#### Towards the cost of travel, registration fee, etc. for selected participants:

Donation from companies and public organizations (including IAEA, hopefully) will be used for this category. Accommodation at the International House will also be allotted to this category. We expect to

#### 12. Local facilities for the preparation of abstract booklet and participant list:

There are several local printing offices which can prepare both abstract booklet and participant list.

#### 13. Transportation facilities:

#### Name and location of nearest international airport:

Tokyo International Airport (Narita)

#### Modes of transport, time, and approximate cost from airport to residential accommodations:

Most foreign Participants are supposed to stay at the area closed to TMU. Tama Center is the local center in this area and easily accessed from Shinjuku (a subcenter of Tokyo) by trains. There are several ways to come to Shinjuku and then to Tama Center. The followings are some easy ways;

(1) From Narita to Shinjuku: Train (Narita Express: 3110yen) or Limousine bus (3,000yen).

From Shinjuku to Tama Center: Train (330yen) (by Keio line and Odakyu line)

(2) From Narita to Tama Center: Limousine bus (3,330yen)

#### Modes and cost of transportation within the city:

Local train (Keio line); 150 yen (Minami-Ohsawa to either to Tama Center or to Hashimoto), 390 yen (Minami-Ohsawa to Shinjuku) and 330 yen (Tama Center to Shinjuku)

#### 14. Working language for Conference:

Conference policy regarding other languages for

Abstract:

Manuscript:

#### Oral and poster presentation:

English is the only language for these items.

#### 15. Banking, internet, and travel planning facilities on site and nearby:

For local banking: A bank is located near the Minami-Ohsawa station.

International banking: Several banks are located near the Tama-Center station.

Internet: It is planned that internet can be used on campus.

Travel planning facilities: There are several commercial travel offices at the Tama Center station.

# 16. Approximate registration fee, items included in the fee, and whether credit cards will be accepted in payment:

	General	Accompanying	Student
		persons	
	\$400	\$150	\$250
Abstract booklet	0		0
Participant list	0		0
Reception	O 1	0	0
Banquet	0	0	
Coffee/tea	0	0	. 0
Proceedings	0		0

# 17. Plan for the publication of the Proceedings (in print or CD-ROM or both), including the name of the journal or publisher and anticipated schedule for receipt of manuscripts, reviewing, and publication:

Following the established procedures, Journal of Radioanalytical and Nuclear Chemistry will be arranged for the journal. Manuscripts will be collected on site of MTAA. Editing including reviewing will be completed in 10 months and final manuscripts will be forwarded to the publisher within a year.

# 18. Plans for social program (e. g., welcome reception, banquet, provision for accompanying persons, and other events during and after the Conference):

Welcome reception: On campus in the Sunday evening. Beer and snack/fruit are served.

Banquet and excursion: Banquet will be planned, being coupled with an excursion, probably on Wednesday. Participants may choose one of two or three destinations for excursion, depending on their taste. In the evening, all participants will meet at the banquet place. All transportation will be served by chartered buses.

#### Programs for accompanying persons:

Japanese culture classes (Tea ceremony, Flower arranging, Kimono, etc.) on TMU campus.

Sanrio Puroland (Theme park) at Tama Center.

Tama Zoo accessed by monorail from Tama Center.

Kamakura (historical city; 90 min), Yokohama (harbor city; 70 min), Tokyo (downtown; 45 min) - all these destinations can be easily accessed by train. Minutes shown in parentheses are the time required for train from the Minami-Ohsawa station.

#### <u>Post-conference tours</u>:

We are planning several post conference tours including Science tour at the Tsukuba Science City and J-PARC (Japan Proton Accelerator Research Complex) tour at Tokai.

#### 18. Other relevant information:

(1) Registration fee will be paid by using major credit cards.

(2) The main campus of TMU, a conference site, is not located in the downtown, but is in the suburb of the Tokyo Metropolitan area. It takes 40-45 min by train. There are many train services from Shinjuku and Tama Center (a city center of the area where TMU is located) and Minami-Ohsawa (the nearest station to TMU). Most participants are supposed to stay either at Tama Center or Hashimoto (both are two stops away from Minami-Ohsawa). However, someone may stay at Shinjuku in downtown and commute to TMU every day. Don't worry about commuters' hell; your directions in the morning and evening are opposite to most commuters. You can easily find a seat to sit down.

(3) The area around TMU is healthy and safety.

(4) We are contacting with a commercial office for designing scientific conventions, which can help us in preparing conference itself and proceedings.

(5) We have just started to have a collaboration with the Korean Society of Activation Analysis (KA³) and will have a joint workshop/ meeting between JA³ (Japan Association of Activation Analysis) and KA³. Mitsuru Ebihara (the proposed chair; a representative of JA³) have contacted with a representative of KA³ in last April and received an informal agreement of a strong support by KA³ when MTAA would be taken place in Japan. Ebihara also discussed with Prof. Chai on the same issue and heard warm words for our holding MTAA in Japan. We are confident that MTAA12 held in Japan can be a good chance for Asian countries to be allied in promoting activation analysis not only in Asia but also in the world.

資料2 (招致申請書への追加資料)

August 29, 2005

#### Supplement to the proposal

Nature of the RA and CS surroundings (campus, city, resort...). Recreational and scenic possibilities for attendees and accompanying persons at the CS and RA at lunchtime, in the evening, and before and after the Conference.

As our CS is the campus of TMU and our university is located in the suburb of Tokyo, it is easy to go to either scenic countryside or downtown, Tokyo, depending upon attendees' and/or accompanying persons' choice. It takes about 40 min to Shinjuku as a subcenter of Tokyo, 60 min to Yokohama, a harbor city, 60 min to Takao (a public garden good for light hiking), and so on. An outlet mall is located just in front of our main entrance gate.

Choices of food available near the RA (vegetarian, Western, Chinese...); notable regional specialties. Does the RA cost include breakfast or other meals? Are there restaurants near the RA where groups of attendees might dine together?

There are several restaurants (Japanese, Chinese, Western,...) around the Minami-Ohsawa station, the nearest station to the campus. Vegetarian's dishes are also available at many restaurants. Unfortunately, I don't come up with notable regional specialties, but foreign visitor must enjoy Japanese food and hospitality. Groups of attendees can dine together here and there. In addition, several fast food restaurants are located near the station. Normally, RA doesn't include meals if not requested.

Means of transport from international airport to the RA and between the RA and the CS: public or chartered, points of interest passed, transit time, frequency, cost, walking distance to stations, difficulty for foreigners or the handicapped to master, and any other information a visitor should know.

Responses to most of the above inquiries are stated in the site proposal. There are several means of transport to RA. As efficiently works public transportation system from the airport at Narita to the downtown, Tokyo and within the metropolitan area, lent-a cars are not recommended. Direction boards at stations show English names along with Japanese. Hankul and Chinese are often seen.

#### Possible tours to NAA facilities or other sites of scientific interest.

We are planning tours to JAERI (Japan Atomic Energy Research Institute) and Tsukuba scientific city. At JAERI, a large facility of accelerator complex for generating neutrons (J-PARC) is now under construction and will be completed in 2007. In Tsukuba area, many scientific institutes are located.

What is included in the registration fee at different price points, and whether credit cards will be accepted in payment.

Information responding to these inquiries are stated in the site proposal. Major credit card will be accepted for payment.

#### Criteria for deciding how discounted fees (i.e., for students or certain countries) will be applied.

The content of applicants' abstract is highly valued. We will be making best effort to collect financial support allocable for supporting students and attendees from developing countries.

#### Mean high and low temperature and rainfall for the proposed conference period.

Late September is our proposed period. Normally, it is just beginning of the tourist season. Mean high and low temperature in Tokyo is around 27 and 18 Celsius. It is rather dry with few rain.

#### Visa and health requirements.

Visa is required for attendees from certain countries. Visitors from most European countries, US and most Asian countries are exempted from visa if their stay is within a limited period. Health requirements are not needed for normal cases.

#### Internet and printing availability at the CS and RA; mobile phone usability.

It is planned that internet can be used at CS. Many hotels have free internet service. Mobile phone is getting

more popular than regular phone (fixed phone). As a person in charge of the site proposal form Japan has

decided to become the last user of a mobile phone, the usability of mobile phone can't be directly

commented, but it must be excellent.

Links to local tourist offices and other relevant information on the WWW.

The following web sites may be helpful to get additional information. Unfortunately, a web site for our

university is under construction, because a new university consisting of three universities and a collage

founded by the Tokyo Metropolitan Government was reconstructed in April, 2005, mostly following the

former Tokyo Metropolitan University. The Japanese name was renewed, but the English name stay

unchanged.

Narita New Tokyo International Airport

http://www.narita-airport.jp/en/index.html

Tokyo Convention and Visitors Bureau

http://www.tcvb.or.jp/en/index\_en.htm

Airport Limousine (Narita to Downtown, Tokyo)

http://www.limousinebus.co.jp/e/

JR-East (Japan Railway-east)

http://www.jreast.co.jp/e/index.html

Narita Express (Narita to main stations in downtown, Tokyo, including Shinjuku)

http://www.jreast.co.jp/e/nex/index.html

Chemistry Department/Tokyo Metropolitan University (old version)

http://www.sci.metro-u.ac.jp/chem/English/\_notes/index\_files/TMU\_Chemistry.htm

(Please note that our university has just been renewed In April, 2005. A new home page is under

construction.)

Mitsuru Ebihara

(proposed conference chair)

資料3(開催地決定の通知と投票結果に関する統計的考察)

From: richard.lindstrom@nist.gov

**Subject: MTAA-12 location** 

Date: 2005年10月19日 21:16:43:JST

Dear Colleague,

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It is our pleasure to announce that the Twelfth International Conference on Modern Trends in Activation

Analysis will be held in Tokyo in the fall of 2007, chaired by Prof. Mitsuru Ebihara of Tokyo Metropolitan

University.

Four groups submitted detailed offers to host MTAA-12. In addition to TMU, these were from India (a joint

proposal from Bhabha Atomic Research Centre and the Saha Institute of Nuclear Physics), Slovenia (Jozef

Stefan Institute), and Portugal (Instituto Tecnológico e Nuclear). The proposals were voted upon by all

twenty-one members of the ICAA. Because all the offers were attractive, the voting was very close. Every

proposal received at least one vote as the best venue, and every one received at least one vote in fourth

position. India was second in the voting, followed by Slovenia and Portugal.

Thanks to all the members for their thoughtful considerations, and for returning your votes with due regard

for the lead time required to prepare a successful MTAA-12. We look forward to seeing you in Tokyo.

With best regards,

A. Chatt, President, IÇAA

Dick Lindstrom, Chair, Site selection subcommittee

\*\*\*\*\*\*

From: richard.lindstrom@nist.gov

**Subject: Voting statistics** 

Date: 2005年10月20日 6:34:37:JST

Dear Colleagues,

You may be interested in a few of my observations about the voting for MTAA-12. As you know, the final

ranking was decided by the sum of scores (best = 1 ... last choice = 4; winner has the lowest sum). The result

was Japan-India-Slovenia-Portugal.

Although all four proposals received at least one first vote and at least one last vote, the number of votes as

first choice also followed the sequence Japan-India-Slovenia-Portugal.

Out of 4! = 24 possible combinations of four ranks, twelve were picked by some ICAA members. The final

ranking of Japan-India-Slovenia-Portugal was the ranking chosen by the largest number (six) of members.

Four other combinations received two votes each, and seven combinations were chosen by only one member.

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I believe we can conclude that, despite the wide variety of opinion expressed in the voting, there is broad consistency in the choice that we have made. Thank you all for your input; the MTAA-12 site selection subcommittee is now dissolved.

Best regards,

Dick

# 施 設 便 り

## 原研施設利用総合共同研究

### 一東京大学大学院工学系研究科原子力専攻共同利用管理本部(大学開放研究室)-

1. 平成 18 年度 原研施設利用総合共同研究の課題公募は 10 月 31 日をもって締め切られました。応募された研究課題は来年 1 月に開催予定の原研施設利用総合共同研究委員会において課題採択の検討が行われ、その結果は 3 月下旬には応募された方々へ採択通知という形でご報告されます。

平成18年度の研究課題として応募された件数は下記のとおりです。

• 一般共同研究

86 件

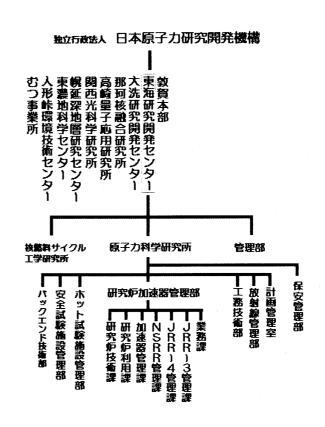
• 連携重点研究

6 件

協力研究(助成申請)

平成18年度より廃止予定

平成 17 年 10 月 1 日をもちまして日本 原子力研究所と核燃料サイクル開発機 構の原子力二法人が統合され、日本原 子力研究開発機構(略称は原子力機構、 JAEA) となりました。旧日本原子力研 究所東海研究所は日本原子力研究開発 機構東海研究開発センター原子力科学 研究所となり、旧核燃料サイクル機構 は日本原子力研究開発機構東海研究開 発センター核燃料サイクル工学研究所 と名称が変わりました。統合に伴い、 原子力機構は中期計画で原子炉や大型 研究施設をはじめとして様々な設備に 対しての"施設共用"という新しい形 態を掲げ、産学連携を旗印に大学関係 者をはじめとして民間企業など外部利



用者への利用を促進するように努める意思を表しています。この形態を遂行することにより利用者にとっては、原子力機構の施設を利用できる枠が広まることになるので多分野での利用が増加し、強いては原子力利用全体の活性化に繋がると期待されています。

3. 平成 18 年度からの京都大学原子炉実験所 (KUR) の休止に伴い、放射化分析や医療照射 (BNCT) の研究者にとっては原子力機構の有する研究用原子炉 JRR-3 と JRR-4 が国内で

は唯一の施設となりますので暫らくはこの二つの原子炉の利用増加が予想されます。また、 それに加えて上述した原子力機構の施設共用という新しい枠を使った産業界の利用も増加 すると考えられるので原子炉の限られた共同利用運転の枠の中ではマシンタイムの確保が 難しくなっていく事が十分に考えられます。

4. 本紙 No.18 でお知らせした通り、平成 17 年 4 月 1 日より東京大学原子力研究総合センターは東京大学大学院工学系研究科原子力専攻と改組され、原子力機構の有する施設の共同利用を管理する部署として共同利用管理本部(旧全国共同研究部門)が設置されました。大学開放研究室は、これからも利用者と原子力機構とのより一層強固な架け橋となって共同利用の管理・運営に努めて参ります。

(澤幡浩之)

## 京都大学原子炉寒鹸所

研究用原子炉(KUR)は、現在所有している高濃縮燃料での運転は、燃料を使い切ってしまうため、2006年2月までで終了します。しかし、2年ほどの運転休止の後、2008年には、新しい燃料(低濃縮)を購入し、定格出力1 MW で運転再開ができることになりました。現在は運転休止の期間が出来るだけ短くてすむよう更に努力しているところです。

運転再開後は、医療照射と放射化分析が共同利用研究の二本柱として位置づけられています。 定格出力は 1 MW と下がりますが、医療照射の際は特に 5 MW での運転となる予定です。放射化分析は通常 1 MW での利用となりますが、5 MW での照射を希望される場合は、医療照射の際に利用可能です。ただ、医療照射が終わり次第 1 MW に戻すことが予定されていますので、照射時間については注意が必要です。

KUR 運転休止期間中の対応として、特に韓国原子力研究所(KAERI)の原子炉(HANARO)を利用することが少しずつ具体化してきています。このことについては本誌別稿に紹介されていますのでそちらを参照下さい。

放射化分析関連の平成17年度下半期共同利用研究採択は次の一件です。

福島美智子 石巻専修大 養殖マガキの中性子放射化分析 平成 17 年度通年共同利用研究採択については前回発行の本誌を参照して下さい。

(柴田誠一)

# 東北大学原子核理学研究施設

東北大学大学院理学研究科原子核理学研究施設(核理研)では平成 17 年度後期において、以下のような課題が採択されています。放射性同位元素製造から核化学、放射線化学と多分野に利用可能です。もちろん放射化分析には中性子放射化分析では分析不可能な元素の定量も可能となりますので、ご利用ください。

は唯一の施設となりますので暫らくはこの二つの原子炉の利用増加が予想されます。また、 それに加えて上述した原子力機構の施設共用という新しい枠を使った産業界の利用も増加 すると考えられるので原子炉の限られた共同利用運転の枠の中ではマシンタイムの確保が 難しくなっていく事が十分に考えられます。

4. 本紙 No.18 でお知らせした通り、平成 17 年 4 月 1 日より東京大学原子力研究総合センターは東京大学大学院工学系研究科原子力専攻と改組され、原子力機構の有する施設の共同利用を管理する部署として共同利用管理本部(旧全国共同研究部門)が設置されました。大学開放研究室は、これからも利用者と原子力機構とのより一層強固な架け橋となって共同利用の管理・運営に努めて参ります。

(澤幡浩之)

## 京都大学原子炉寒鹸所

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2554	大槻勤	東北大核理研	標識化による金属内包フラーレン及びヘテロフラーレン
			の製造及びその応用研究
2555	関根勉	東北大理学研究科	長寿命放射性核種の環境中移行における基礎化学 反応研
			究
2556	山崎仁丈	東北大工学研究科	水素誘起空孔による拡散の促進
2557	中西孝	金沢大自然	制動放射線誘導放射性核種の生成量に関する実測値と計
			算値の比較
2558	鹿野弘二	函館高専	光量子放射化分析法による光通信用材料中の不純物評価
2559	桝本和義	KEK	金属及び半導体材料中の軽元素の定量のための分離・捕
			集法の開発
2560	大浦泰嗣	首都大都市教養	宇宙化学的試料および環境試料の光量子放射化分析
2561	鈴木達也	東工大原子炉研	固体抽出剤を用いた希土類元素とアクチノイド元素の分
			離に関する研究
2562	結城秀行	東北大核理研	電子ビームによるフリーラジカル生成の研究
			(大槻勤)

# 事務局より

### ◇平成17年度第1回放射化分析研究会幹事会議事録

1. 日時, 場所

2005年6月4日 13:00~

東大(旧)原総センタータンデム加速器実験施設会議室

2. 幹事の交代 (下線:出席)

選出された幹事: <u>海老原充</u>,河野公栄,木川田喜一,澤幡浩之,<u>宮本ユタカ</u> 留任幹事:大浦泰嗣,片山幸士,笹島文雄,桝本和義,松尾基之,八木宏親

3. 代表幹事の選任

代表幹事の選出について議論し,以下の点で合意した;

- (1) (選出方法) 年度初めの幹事会で互選(出席者の過半数)
- (2) (任期) 2年. ただし, 各年度初めの幹事会で確認する.
- (3) (再任) 再任は妨げない.

以上の合意に基づき、2005-2006年度の代表幹事として海老原を選出した.

### 4. 議事内容(報告)

(1) 会計 (河野)

出来るだけ早くメールによって監査報告を受け、その後、報告内容の可否をメールで問うことのプロセスを了承した。

(2) 会誌(大浦, 宮本)

今後の会誌の発行予定について、別紙をもとに意見交換した、議論の要点は以下の通り;

- ・毎号連続でなく、途中に別の特集を組むことを考える(弾力的に).
- ・訪問記的な記事も特集(海外施設)に組み込む.
- No. 20 を記念号とする.
- ・「ぶんせき」誌に最近掲載された岡田氏の進歩総説を転載,あるいは書き換えたものを掲載する可能性を探る.
- ・「考古学」特集を積極的に考える.
- ・京大原子炉実験所・専門研究会での発表の要旨等を掲載する.
- (3) ホームページ(笹島)

ホームページ関連の進捗状況について、別紙をもとに説明された.

(4) 冬の学校

京大原子炉専門研究会として 12/20-22 の間の 2 日間で開催の予定.次期申請について意見交換をし、中西(金沢大;学外)、柴田(学内)のお二人に申請をお願いする方向で、片山幹事に依頼した.

### 5. 議事内容(議題)

(1) 2005 年度会務分担

会誌:大浦, 宮本, 木川田, 八木

会計:澤幡 監査:河野

行事:松尾,澤幡,片山

HP: 桝本, 笹島, 大浦, 宮本, 澤幡

(2) MTAA の招致について

招致のための working group を作ることが提案され、了承された.メンバーは以下の通り: 海老原、大浦、桝本、松江、松尾(アイウエオ順)

(3) KA3 (放射化分析研究会) との連携について

この件について意見交換をし,以下の点で合意した;

- ・連携を前向きに考える.
- ・連携の方法として、共同研究、合同研究会開催を実行する方向で考える.
- ・合同研究会として、京大原子炉専門研究会の場を使えるかどうか、打診する.
- ・韓国側にも資金的な裏付けを積極的に探してもらう.
- (4) その他

例年通り、「理工学における放射線研究会」の場で拡大幹事会を開催する方向で検討する.

次回の(拡大)幹事会開催予定(可能性として)

- •7月6-8日(放射線研究会)
- ·9月28-30日(放射化学会)
- ·12月20-22日(京大原子炉実験所)

## ◇平成 17 年度 放射化分析研究会拡大幹事会議事録

開催日:平成17年9月29日(木)

開催場所:金沢市観光会館

参加者:海老原、河野、片山、八木、松尾、宮本、松江、大浦、桝本、高宮、澤幡

### 議事内容:

- 1. 会誌について (大浦)
  - ・ No.19 を 10 月中に発行予定。No. 19 からの「特集」は海外放射化分析施設の紹介で、今回は韓国原子力研究所を掲載する予定. 依頼した原稿はほぼ入手済み。また、「研究会報告」として HANARO2005、NAMLS 8、FNCA など、「動向」として「KAERI の研究用原子炉HANARO と京都大学原子炉実験所との協力研究の紹介」、「MTAA-12 招致活動状況」などが掲載予定。その他の内容は別添参照。
  - ・ 会員名簿の個人情報保護法への対応について議論されたが、会員が自ら通知した経緯があるので、掲載について問題があるか否かを知らせてもらって事務局が随時対応する事とした。 今回の討論会についての報告は天野先生か中西先生に海老原幹事からお願いする事とした。
  - ・ 次号、No.20 は記念号となるので、何か良い提案を出して頂きたいとの依頼があった。
- 2. 会計報告について
  - ・ 河野幹事の会計監査が行われ、適正に処理されている旨の報告がされた。

- ・ 所属や住所の不明な会員情報及び Mailing list における Host unknown のアドレス情報に ついては事務局が適宜対応する事となった。
- ・ 会費の未納については、会誌発送の際に請求文書 (3 年分前まで)を入れて対応する事とした。
- 3. MTAA-12 の準備について (海老原)
  - ・ 日本・インド・ポルトガル・スロベニアが参加を表明しており、今後 ICAA (国際放射化 分析委員会) 委員に資料が配布され、投票 (10 月 10 日締め切り) が行われる. 結果については中国で行われる APSORC までには決定する模様との報告があった。

### 4. 冬の学校 (片山)

- ・ 京都大学原子炉実験所専門研究会(放射化分析研究会「冬の学校」)が 12 月 21 日 (水)、 22 日 (木)に開催予定であるとの報告があった。また、韓国原子力研究所のメンバーも招致したいとの意見もあったが、時期的な問題があり現実には至らなかったとの報告があった。 然しながら、来年の研究会では是非実現させたいとの事も合わせて報告された。
- 5. KA-3 (韓国の放射化分析研究会) について (海老原)
  - ・ 韓国側は合同の研究会開催に非常に積極的である。問題は旅費の捻出であるが、来年には Joint して行いたいとの意向が示された。
- 6. 新機構の方針について(松江)
  - ・ 統合後の新法人、日本原子力研究開発機構の基本方針は平成 18 年 3 月末までは現行と大きく変わる事はないとの報告があった。4 月以降の状況については、詳細についてはまだ不詳な点が残されている。料金体系などは変更される可能があるが、組織については大きな変革は無いであろうとの報告があった。
- 7. Mailing list のドメインが変更について (澤幡)
  - ・ 新法人の発足に伴い、ネットワーク運用も変更される。そのため現有の JA-3 Mailing list が使用できなくなるとの報告があった。早急に他のドメインを取得する必要があり、対応策としては東大のドメインを取得する、或いは無料で運用できるプロバイダーを模索することが考えられた。12 月末には新たな Mailing list に切り換えなければならず、10 月中には打開策を決定する必要がある。
- 8. 新しいホームページが公開された。(桝本)
  - ・ 桝本幹事、笹島幹事によって、JA-3 のホームページがリニューアルされた。今後は、よりリアルタイムな情報提供をしていくため様々な情報提供、および収集が必要であり、広く情報の提供を呼びかけていく必要があるとの報告があった。

# ◇第49回放射化学討論会 放射化分析分科会報告

開催日時:平成17年9月30日(金) 12:10~14:00

開催場所:金沢市観光会館 B 会議室

参加者:約30名

世話人:金沢大院医系 天野良平、東大院総合 松尾基之

### 議事内容:

第1部

小村和久先生(金沢大学自然計測応用研究センター低レベル放射能実験施設)による「極低レベル放射能測定と放射化分析-環境中性子・宇宙線・原爆による放射化-」と題する講演が行われた。講演の内容は、放射化分析の基本的なことから始まり、天然の照射源である宇宙線や土壌、岩石中の核反応などの説明、或いはそれら低フラックスにて照射するターゲット(分析試料)への着目、更には、そのような生成核種の濃度が極めて低い放射化を行っていく際に必要不可欠である尾小屋地下測定施設の説明など、大変興味深いものであった。後半では、小村先生がこれまで手がけられてきた「60Co、152Eu による原爆中性子評価の実測値と計算値の不一致問題の解明」、「天然の152Eu 発見」、「JCO 臨界事故の環境影響評価」、「原爆線量評価」などの研究成果を説明された。最後に小村先生の宝とも言うべき「尾小屋地下測定施設」の将来計画の説明があり、大変有意義な講演であった。

### 第2部

第2部は「放射化分析研究会からのお知らせ」とそれに関連する質疑応答が行われた。 内容は下記の通りである。

- 1.「京都大学原子炉実験所と韓国原子力研究所 KAERI の協力研究」と題して、京大炉の高宮幸一先生から KUR の今後の動向及び韓国原子力研究所の研究炉 HANARO の共同利用の話題を頂いた。HANARO は高中性子束な原子炉でありほぼ一年中利用可能といった利点があるが、KAERI 側の検出器やスタッフの人数の不足や試料の返却(RI の国際輸送の問題)など、まだ解決すべき問題点が多いとの見解も報告された。また、2005 年 10 月から HANARO 利用研究のための公募が開始する予定であること、2006 年 4 月からは実際に協力研究を開始する予定であることなどが紹介された。
- 2. 澤幡幹事より「放射化分析研究会事務局から研究用原子炉の現状報告と共同利用の将来」と題する報告があった。その中で、2004年度の会計報告が行われ、出席者の了承が得られた。また、平成17年10月1日より日本原子力研究所と核燃料サイクル機構が統合されることに伴い、組織改変の話題が提供され、新たに発足する独立行政法人日本原子力研究開発機構(原子力機構と略称する)の組織図が発表された。それに補足して原子力機構における試験研究炉の役割として重要な研究施設・設備で、外部から利用ニーズが高い施設・設備については、利用支援体制を整備し、研究者や企業等に対する共用の促進を図る、との方針が打ち出されたとの報告があった。それを受けて、原子力専攻としては現行する共同利用の体制は大きく変わることがないとの報告があった。
- 3. 続いて、日本原子力研究所の大島真澄氏及び首都大学東京の海老原充先生より「多重即発ガンマ線分析の開発状況と今後の計画について」と題する報告があった。報告は JRR-3 における多重即発ガンマ線分析の現状と計画、及び J-PARC における今後の計画という二つのテーマに分かれており、前者では、多重即発ガンマ線分析装置にオートサンプルチャンジャーが装備されたこと、クローバー検出器が 3 台装備されたことが報告され、更には、文科省公募型特会事業「革新的原子力システム開発」によりクラスター検出器を 2 台、クローバー検出器の 4 台を整備する準備があることが報告された。また、現在のところ予定していた感度が得られないなどの問題点も報告された。また、今年度中に世界最高速のデータ収集系の開発も行うことが合わせて報告された。後半では、今後の開発計画と将来の利用についての提案がなされ、二法人統合後の組織的な対応が不安定であるなど問題点も報告された。最後に多重即発ガンマ線分析装置の全システム完

成までの期間として約3年はかかるとの報告があり、関係者に対してより一層の協力依頼があった。

次に 2 つ目のテーマである J-PARC における今後の計画が報告され、J-PARC の工事進捗状況 の説明と 2007 年後半のビーム入射に向けたタイムスケジュール、J-PARC 中性子核反応実験装置 の共同提案と今後の課題などが報告された。また、ビームライン実現に向けて「新規 JST 公募原子カシステム研究開発事業」への提案を行い、現在書類審査をパスした旨の報告があった。

4. その他として、大浦幹事より放射化分析研究会機関紙「放射化分析 No.19」の編集状況が報告された。No.19 は特集として「海外放射化分析施設-韓国原子力研究所-」が掲載され、研究会報告として HANARO2005、NAMLS 8、FNCA など、動向として「KAERI の研究用原子炉HANARO と京都大学原子炉実験所との協力研究の紹介」、「MTAA-12 招致活動状況」などが掲載され予定であることが紹介された。総じて韓国原子力研究所との協力関係など国際的な情報を紹介する内容になる模様であるとの報告があった。また、海老原代表幹事より MTAA12 の招致状況と準備活動の報告、片山幹事より京都大学原子炉実験所専門研究会(「京都大学原子炉実験所での放射化分析と今後の中性子利用分析」)が 12 月 21 日 (水)、22 日 (木)の両日に、放射化分析研究会「冬の学校」をかねて行われる予定であるとの報告があった。

### ◇放射化分析冬の学校(京大炉専門研究会)

京都大学原子炉実験所専門研究会のご案内 (京都大学原子炉実験所での放射化分析と今後の中性子利用分析)

京大原子炉は平成 18 年度より、現在の高濃縮ウランでの運転を中止し、約2年間の休止後再開が予定されています。

そこで、昨年度は、代谷所長より原子炉休止の事情や将来計画等を伺うとともに、京大炉を使った放射化分析の成果と原子炉が休止している期間の対応について 6 名の方にご講演をしていただきました。

再開後は運転が大幅に変更され、燃料も低濃縮へ、出力も医療照射時以外の通常利用は1MWの計画になっており、連続運転から各日運転になりそうです。

今後京大炉による放射化分析をどのように発展させていけばよいか、また、原研施設の利用や、 韓国の研究用原子炉ハナロの利用なども話題に取り上げる予定です。

今年度は下記の日程で研究会を開催いたしますので、多くの方のご参加をお待ちしております。 なお、この専門研究会を放射化分析研究会「冬の学校」の活動の一部といたします。 プログラム等、詳細が決まり次第ご案内を差し上げます。

日 時: 平成17年12月21日(水)13時-17時 12月22日(木)9時-12時30分

場 所:京都大学原子炉実験所事務棟大会議室 宿 泊:京都大学原子炉実験所 共同研究員宿舎

参加申込期限:11月25日(金) (事務手続き上、できるだけ早くお申し込みください)

申込先:

₹444-3505

愛知県岡崎市本宿町上三本松 6-2

人間環境大学

片山幸士

TEL/FAX: 0564-48-1014, E-mail: katayama@uhe.ac.jp

-----参加申込書 ------

氏名:

所属:

職名:級号俸:

連絡先:住所(郵便番号)

E-mail

TEL

FAX

自宅住所 (郵便番号):

参加日:12月 21日(水) 22日(木) (参加日を残してください) 宿泊: 12月 20日 21日 22日 (希望日を残してください)

問い合わせ先: 片山幸士

TEL/FAX: 0564-48-1014 E-mail: katayama@uhe.ac.jp

高田實彌

TEL: 0724-51-2441、FAX: 0724-51-2622 E-mail: takada@HL.rri.kyoto-u.ac.jp

## ◇会員名簿について

昨今,個人情報保護が話題になっています.(本来の趣旨よりも拡大解釈されているような気もしますが.)本会誌の末尾には毎号会員名簿を掲載しておりますが,氏名以外の登録事項の掲載を希望されない方がおられましたら,事務局までご連絡ください.次号から,希望される登録事項を削除いたします.

また、登録事項に変更等ありましたら速やかに事務局に連絡いただけますようお願いいたします.

# カレンダー

2005/12/15-20

2005 環太平洋国際化学会議

ホノルル (アメリカ合衆国)

http://www.csj.jp/learned-society/pacifichem/pacifichem2005top.html

http://www.pacifichem.org/

発表申し込み締切: 2005/4/13

参加申し込み締切: 2005/10 月末

2006/1/23-27

Internatinal Conference on Application of Radiotracers in Chemical, environmental and

Biological Scinces

India

http://netsrvrcs.saha.ernet.in/arcebs/index.html

発表申し込み締切: 2005/8/1

参加申し込み締切: 2005/12/1

2006/4/3-7

Seventh Methods and Applications of Radioanalytical Chemistry (MARC VII)

USA

http://www.min.uc.edu/nuclear/marc/

発表申し込み締切: 2005/12/15

参加申し込み締切:未公表

2006/4/23-28

15th Radiochemical Conference (RadChem2006)

Czech Republic

http://www.fjfi.cvut.cz/radchem/

発表申し込み締切: 2005/11/30

参加申し込み締切: 2005/12

2006/9/13-15

Environmental Radiochemical Analysis 10th International Symposium

Oxford, UK

http://www.rsc.org/era2006

発表申し込み締切: 2006/1/31

参加申し込み締切: 2006/7/31

### 本会誌の内容および投稿募集

放射化分析研究会の情報媒体は、定期刊行機関紙「放射化分析」、不定期の「放射化分析研究会ニュース」、および電子メールです。

会誌「放射化分析」は下記の記事で構成されます。

### 講座・特集・解説:

専門的な記事を毎号企画していきます。記事の内容についてのご希望や提案をお寄せください。

### 研究紹介:

放射化分析に関する研究、放射化分析を用いた研究を紹介していきます。 研究の当事者にまとまった内容を執筆していただくものと、秀れた論文を紹介するものとがあります。 特に紹介したい研究についてご意見をお寄せください。

#### Q&A:

会員から質問を受けて、回答やアドバイスを掲載します。どのような質問でも結備です。 回答者は幹事会が責任をもって会員の中から選定します。

### 施設•研究室紹介:

放射化分析を行うことができる施設については、放射化分析関係の共同利用、 共同研究が行われている施設で採択されている研究課題などを掲載します。 また放射化分析を行っている研究室の紹介を適宜行います。

### 談話室:

会員の気軽なおしゃべりの場です。要望、話題、新刊紹介、その他、どのような内容でも結構です。

#### 芸い書・

学生や着手研究者およびこれから放射化分析を手がけようとする方々の新鮮な声を反映させる欄です。

### 研究会報告・最近の動向等:

最近開催された研究会の報告です。放射化が取り扱われている研究は広範囲な分野に 顔を出している筈です。放射化が関係している研究課題が1件でもあれば、この欄に取り上げたいと思います。 会員の皆さんからの情報提供をお待ちしています。

### アナウンスメント

### 論文紹介(発表論文アブストラクト)

会員から届けられた論文別刷りを元に、発表された学術論文のタイトルとアプストラクトが随時掲載されます。 博士・修論文の要約も掲載します。

### 会員名簿

原稿は A4 横書き、40 字×40 行、上下マージン 25mm、左右マージン 20mm を標準として、 そのままオフセット印刷又はコピーして使える形に清書して送付して下さい。 編集の際にレイアウトを工夫させていただくこともありますので念のためファイルを添付して下さい。 (ファイルは MS-DOS のテキスト又は MS-WORD が最適ですが、Macintosh のファイルやワープロ専用機のファイルもかなりの程度処理できます)

### 電子メーリングシステム

会員及び幹事会の電子メールアドレスが、下記のエイリアスに登録されています。 これを用いて会員相互の交流・意見交換を促進していきます。

会員: JA3@kaihoken.nuclear.jp

幹事会: JA3-kanjikai@kaihoken.nuclear.jp

JA3@kaihoken.nuclear.jp に発信すれば、(電子メールアドレスが登録されている) 会員全体に配信されます。幹事会・事務局への連絡・要望等は JA3-kanjikai@kaihoken.nuclear.jp に発信して下さい。

メールアドレスを持っているけれど、事務局等からの電子メールを受け取ったことの無い会員は、アドレスがエイリアスに正しく登録されていない可能性があります。お心当たりの方は事務局宛にご連絡下さい。登録を修正します。

### 放射化分析研究会

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### 事 4

海老原充 河野公栄 木川田喜一 澤幡浩之 高田寶彌 豊田和弘 大浦泰嗣 片山幸士 笹島文雄 桝本和義 松尾基之 八木宏親