VIII-II-1. Project Research

Project 8

PR8 Trace Elemental Analysis Using Research Reactor

K. Takamiya

Research Reactor Institute, Kyoto University

Objective: Neutron activation analysis (NAA) is one of the most sensitive and accurate method for elemental analysis. This method has been used in various research fields, such as biology, chemistry, material science, environmental science, and geochemistry, because it requires no sample preparation. And multi-elemental analysis can be performed in a single irradiation experiment. Many elements can be determined using this method, but it is difficult to analyze if a radioisotope which is produced by neutron induced reaction has too short or too long half-life. And if there exist too many elements in a sample matrix, the accuracy of gamma-ray spectrometry decrease because of the Compton effect and overlap of photo peaks. Various method has been developed to resolve these problem, for example, the prompt gamma-ray analysis (PGA) method can determine light elements which produce nuclei of very short half-lives by neutron capture reactions. In this method, samples are irradiated by neutron beam from such as Super Mirror neutron guide tube, and prompt gamma-rays are emitted from neutron-irradiated samples and detected by the detector. In this project research, various efforts to increase the precision of NAA method were planned. One side of the efforts is developing and installing new instruments for NAA method. PGA system and anti-Compton counting system have been developed at KURRI. The PGA system will be connected with Super Mirror guide tube which can supply high-quality thermal neutrons. The anti-Compton counting system will be installed at next to a pneumatic capsule transporting system to detect short-lived nuclei effectively. Another side of the efforts is developing and applying new NAA techniques.

Research Subjects: The common subject of this project is carrying out characteristic NAA or PGA experiments using research reactors such as KUR and HANARO. The respective subjects of the research groups of this project are described as follows;

P8-1: Neutron distribution measurements for high-precision NAA

P8-2: Development of prompt gamma-ray analysis apparatus in KURRI

P8-3: Antimony and Arsenic Concentration Distribution with Soil Particle Size

P8-4: Neutron activation analysis for short-lived products of Ag and Se in organisms

P8-5: Determination of chlorines in agricultural crops and soils by NAA method

P8-6: Study for concentration of elements in human hair

P8-7: Activation analysis of metals in hair from animals dosed natural medicines and health foods

Results: Because the restart of KUR operation delayed, some of subjects cannot be performed. But some have been carried out using HANARO. The brief summaries of the performed subjects are as follows;

P8-1: A new method of neutron flux measurements has been developed. The developed flux monitor was irradiated at HANARO in order to measure neutron distribution at the irradiation position. The neutron flux distributions were successfully measured to a precision of 5 mm range. Obtained results are consistent with the previous report in which Au-Al wire monitors were used as neutron monitors. The neutron flux at the irradiating position of pneumatic transporting system of KUR will be measured using the developed neutron monitor in the future.

P8-2: A PGA apparatus has been developed in KURRI from 2008. The electronics for detecting prompt gamma-rays with anti-coincidence technique has been checked in the present work. In order to check the whole system property, on-line experiments using reference standard samples will be carried out at thermal neutron beam line (B4) of KUR in 2010.

P8-3: Characteristics of antimony contamination in soils has been studied by NAA method. The soil samples collected around an antimony smelter were irradiated at HANARO. Concentrations of antimony and arsenic were determined by comparison method. Antimony concentration is on the decrease as the particle size of the soil becomes larger, and this tendency is the same as the previous results. Arsenic concentration distribution shows the same tendency as antimony. The distribution tendency of arsenic concentration with the soil particle size is comparable to elements of soil matrix such as iron, scandium and europium reported in their previous report.

P8-7: In the previous work using KUR, the correlation between intake of polyphenol and copper excretion in mice was observed. Copper in mouse hair was analyzed at HANARO to confirm the previous result. Copper was detected only in a specific group at 14th-day mouse. But, after enough time passed, copper was detected in all groups and the amount reached to equilibrium. Therefore, it was found that polyphenol accelerated the excretion speed of copper from mouse's inside of the body.

Acknowledgment: The author and colleagues of this project would like to thank J. H. Moon, G. M. Sun, S. H. Kim, and S. R. Baek (KAERI) for their operation of NAA and PGA systems and for their helpful cooperation. And we also thank Y. Nakano and R. Okumura (KURRI) for their technical support.

採択課題番号 21P8　　原子炉中性子を用いた微量元素分析　　プロジェクト

（京大・原子炉）高宮幸一

PR8-1 Neutron Distribution Measurements For High-Precision NAA

K. Takamiya, M. Takimoto, S. Shibata, R. Okumura, Y. Nakano, J. H. Moon[1] and S. H. Kim[1]

Research Reactor Institute, Kyoto University
[1] Korea Atomic Energy Research Institute

INTRODUCTION: A neutron flux is important parameter in neutron activation analysis, radioisotope production and other neutron irradiation experiments. In the case of neutron activation analysis, the analytical accuracy depends on the estimation of neutron flux at irradiations. In order to neglect a neutron flux parameter in the analytical determination, a standard comparison method is used. In this method, standard materials are irradiated with analytical samples at a time. But only one standard material is irradiated in many cases even through many analytical samples are inserted into an irradiation capsule. If neutron fluxes at an irradiating facility varies depending on the position, for example, vertical positions at an irradiation hole in a research reactor, difference of irradiating position between analytical samples and a standard material incites experimental errors in elemental determination. Therefore, distributions of neutron flux of irradiating facilities are essential parameters in such experiments.

A new preparation method of a neutron monitoring material using an ink-jet printer has been developed in the previous report [1]. In this method, gold solution is applied on a paper sheet using an ink-jet printer to prepare a neutron monitor material. The prepared monitor is thin enough to ignore the self-absorption in neutron irradiation and self-shielding effect in gamma-ray measurement and easy to handle. Because a target material of gold for neutron activation is applied on a paper sheet using ink-jet printer in this method, the position of the target material can be adjusted easily and correctly. Therefore, this method can be used as neutron distribution measurement. In the present work, measurements of neutron flux distribution have been tried using the monitoring material prepared as above.

EXPERIMENTS: As neutron monitors for neutron distribution measurements, two square paper pieces on which the gold solutions were applied in parallel stripes as shown in Fig. 1 were prepared. Height of each stripe was 5 mm, thus, the position resolution of this experiment is 5 mm. Measurements of axial neutron distribution at the position of an irradiating capsule in neutron irradiation holes of HANARO were tried. The stacked paper pieces were rolled up and inserted into the capsule. After the neutron irradiation at the irradiation holes, neutron monitors were separated each other. Measurements of gamma-ray spectra for the separated pieces were carried out using a Ge-detector.

RESULTS: Figure2 shows the activity variation of ^{198}Au which was produced in each monitor piece separated from the neutron monitor irradiated at PTS#1 and PTS#3 in HANARO. In this figure, the horizontal axis shows the center position of each monitor piece at the neutron irradiation. A trend or fluctuation of neutron fluence cannot be observed in the case of PTS#1, but there is a positive trend in PTS#3. This result is consistent with the result of a neutron flux measurement performed using Au-Al wire monitors previously [2].

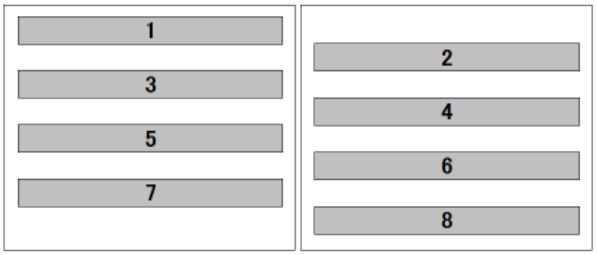

Fig. 1. Neutron monitors for a neutron distribution measurement at PTS#1 and PTS#3 in HANARO. Gold was applied in colored area.

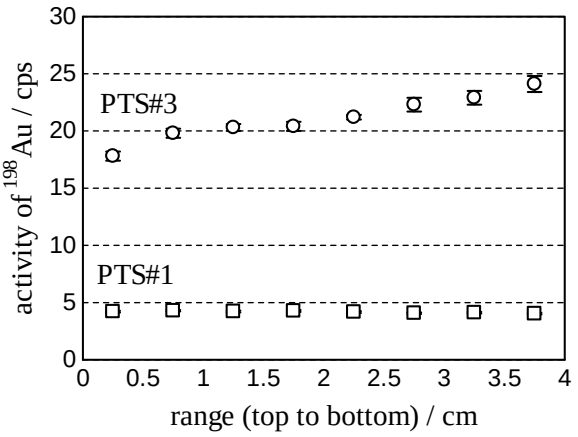

Fig. 2. The activity variation of ^{198}Au produced at PTS#1 and PTS#3 in HANARO.

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[2] Y. S. Chung, S. H. Kim, J. H. Moon, S. Y. Baek, Y. J. Kim, H. R. Kim, J. Radioanal. Nucl. Chem. **278** (2008) 707-712.

採択課題番号 21P8-1　中性子放射化により生成する短寿命核種の定量法の開発　　プロジェクト
（京大炉）高宮幸一、瀧本真己、柴田誠一、奥村良、中野幸弘（韓国原子力研究所）J.H.Moon, S.H.Kim

PR8-2 Development of Prompt Gammma-Ray Analysis Apparatus in KURRI

S. Sekimoto, K. Hirose[1], K. Takamiya, R. Okumura and Y. Kawabata

Research Reactor Institute, Kyoto University
[1]Research Center for Electron Photon Science, Tohoku University

INTRODUCTION: Prompt gamma-ray analysis (PGA) apparatus has been developed in KURRI from 2008. This development is promoted by Nuclear Energy Research Initiative, Research Reactor and Hot Laboratory Utilization program, which is titled as "Improvement of Utilization on KUR and HL in KURRI", under the project in Ministry of Education, Culture, Sports, Science and Technology. In PGA, it is necessary to observe the gamma rays emitted promptly from the samples through neutron induced reaction. The neutron irradiation for PGA in KURRI will be carried out at B4 reactor site of Kyoto University Reactor (KUR). The purpose of this project is to install the PGA in KURRI and to apply the total micro-element analysis system including the PGA and the short-lived nuclides measurement system, which has been developed under the same program, to the research on medical science, archeology, cosmo- and geo-science etc., in the future. In terms of the field of nuclear energy, this project is also directed toward the ideal model of small neutron source in university. In this report, the state of preparation for installation of the PGA system in KURRI is described.

PREPARATION: In 2008, the signal processing system for the anti-coincidence circuit was built with the NIM electronics. The anti-coincidence circuit consists of timing filter amplifier (TFA), constant fraction discriminator (CFD), gate and delay generator (GDG), Quad Logic (f-in/f-out) and time-to-amplitude convertor (TAC), as shown in Fig. 1. This circuit will follow the gamma-ray spectroscopic instrument which consists of an n-type high-purity germanium detector (Ge) and bismuth germinate scintillation detectors (BGO) with high voltage (HV), photo multiplier (PM) and preamplifier (pre-AMP). The signals from this circuit will be finally processed by pulse height analyzer (PHA).

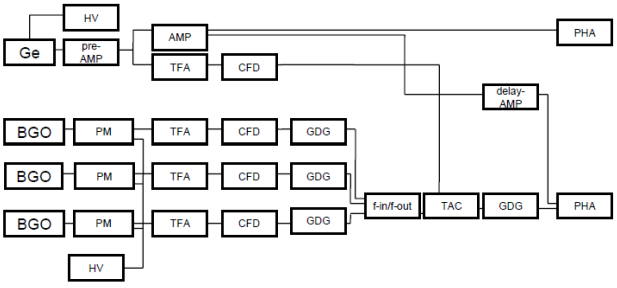

Fig. 1. Anti-coincidence system in KURRI-PGA.

In 2009, n-type high-purity germanium detector (CANBERRA GR3019), multi channel analyzer (MCA, SEIKO EG&G, 7600-000) including HV (7600-300) and PHA (7600-510), and spectroscopy amplifier and gated integrator (ORTEC 673) were installed for the PGA system. The lead and LiF shieldings from background gamma rays and scattered neutrons, respectively, were also prepared for the gamma-ray spectroscopic instrument.

SIGNAL TEST: Using some electronics and detectors described above, output signals of following two kinds of circuits were checked by an oscilloscope. Figure 2 shows the output signal from BGO processed through PM, Pre-AMP. and TFA. Figure 3 shows that processed through PM, f-in/f-out and TFA.

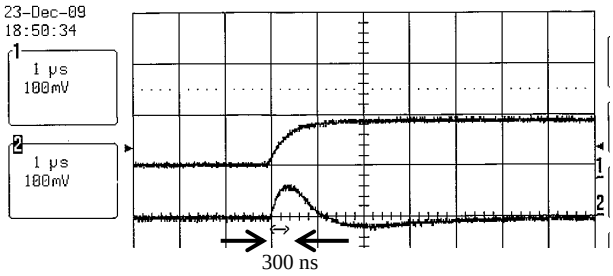

Fig. 2. Signal from the following circuit.
(BGO→PM→Pre-AMP.(Ortec 113)→TFA)

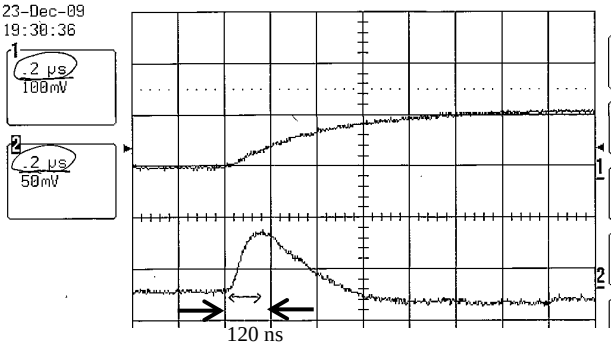

Fig. 3. Signal from the following circuit.
(BGO→PM→f-in/f-out→TFA)

It was confirmed that the signal from BGO in the circuit shown in Fig. 3 was able to be processed faster than that in Fig. 2. Therefore, the circuit shown in Fig. 3 will be adopted for the electronics of the PGA system in KURRI.

FUTURE PLAN: In 2010, on-line experiments using reference standard samples will be carried out at B4 reactor site of KUR.

採択課題番号 21P8-2 京大炉における即発ガンマ線分析装置の開発 プロジェクト
（京大炉）関本 俊、高宮幸一、奥村良、川端祐司

PR8-3 **Antimony and Arsenic Concentration Distribution with Soil Particle Size**

S. Fukutani, K. Takamiya, Y. Nakano, R. Okumura, T. Takahashi and A. Koyama

Research Reactor Institute, Kyoto University

INTRODUCTION: In this study we attempt to analyze characteristics of antimony contamination in soils around a antimony smelter in contradistinction to arsenic which is the homologous element of antimony. Antimony (Sb) is one of natural occurring trace elements in soil, but it is being identified as a pollutant by increasing its usage. Flame retardants, paints and pigments are important industrial products, and 13,279 t of antimony trioxide (Sb_2O_3) was in demand at 2006 in Japan [1]. It is pointed out that these important products, flame retardants, paints and pigments, are end up in the garbage and loaded in the environment. Antimony is categorized as items to be monitored in Japan, and the guideline values of antimony are under 0.02mg/L and 0.015mg/L for the public water quality and the tap water quality, respectively. As just described, antimony is needed to be studied about its behavior in the environment because of its large amount of usage and the strict guideline values.

EXPERIMENTS: The soil sample was collected around an antimony smelter where health hazards had been occurred in 1970's. The collected soil was dried in air and sieved through a 2mm sieve. This pretreatment soil was then sieved through 2000, 1000, 850, 425, 250, 106, 75, 44μm sieves, and 9 fractions of the soil sample were prepared. About 100 mg soil was sampled from each fraction. Prepared soil samples were irradiated by neutron at HANARO (High-flux Advanced Neutron Application Reactor) in KAERI (Korea Atomic Energy Research Institute). The facility of neutron irradiation was NAA1 (thermal neutron flux: 3.9×10^{13} n/cm^2/s). SRM 2711a (Montana Soil, supplied by NIST) was used for standards of antimony and arsenic quantitative analysis. Certified values of antimony and arsenic concentration are 23.8 ± 1.4 and 107 ± 5 mg / kg, respectively. Irradiation time was 130 seconds, and after about one day for cooling, γ-ray spectrometry was conducted. Concentrations of antimony and arsenic were calculated by using comparison method.

RESULTS: Measurement results about antimony and arsenic in the soils are plotted in Fig. 1and Fig. 2, respectively. logarithmic approximate curves are also drawn in the Fig. 1 and Fig. 2. Fig.1 shows that antimony concentration in the soil is on the decrease as the particle size of the soil becomes larger, and this tendency is the same as

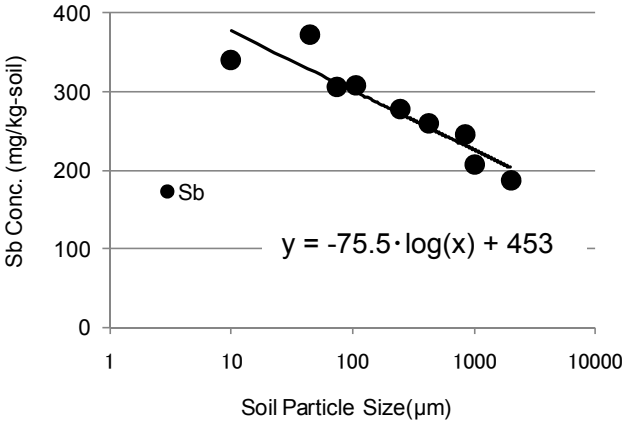

Fig. 1. Antimony concentration distribution.

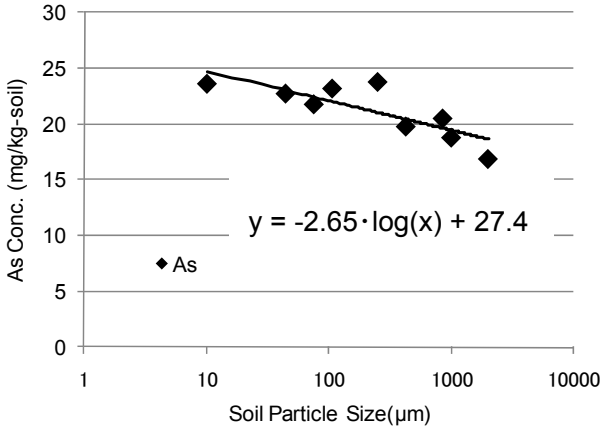

Fig. 2. Arsenic concentration distribution.

we reported before [2]. In Fig. 2, arsenic concentration distribution shows the same tendency as antimony, but judging from the slope values of the approximate curves, changes of arsenic concentration are small. The distribution tendency of arsenic concentration with the soil particle size is comparable to elements of soil matrix such as iron, scandium and europium we reported before [2]. Further studies are necessary to elucidate details.

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採択課題番号 21P8-3　土壌中微量環境負荷元素の汚染経路の解明に関する研究　　プロジェクト

（京大・原子炉）福谷　哲、高宮幸一、高橋知之、小山昭夫

PR8-4

Activation Analysis of Metals in Hair from Animals Dosed Natural Medicines and Health Foods

M. Kawase, S. Morimoto[1] and T. Saito[2]

Faculty of Bioscience, Nagahama Institute of Bio-science and Technology
[1]Department of Pharmacy, Osaka Ohtani University
[2]Radioisotope Research Center, Osaka University

INTRODUCTION: The plant is one of important raw material of the herbal medicine. Moreover, the effect of the herbal medicine is largely affected by the home and the quality of a used plant. The trace elements are considered to be related with the quality of herbal medicine, but the study about the trace elements in plant is not done widely. Therefore, the role of trace elements in herbal medicine is little clarified.
We confirmed that the polyphenol promotes the exhaust of Cu from mouse's inside of the body by the activation analysis at KUR, before. We analyzed mouse's hair again at HANARO to confirm a previous result, and the results are reported here.

EXPERIMENTS: The green tea extract was administrated to the C57BL/6 C constantly for 2 or 4 weeks. The used concentration of green tea extract was 0.2 g/l (L group) and 2.0 g/l (H group). The body hair was collected as a reaping analytical sample on the 0th day of the intake beginning and the 14th day and 24th day. The samples were subjected the activation analysis at HANARO after washing.

RESULTS: Observed gamma-ray energy spectra are shown in Fig. 1. Cu was detected in hair from H group for 2 weeks administrated mice (upper spectrum in Fig. 1). However, Cu was not found in that of L group (lower spectrum in Fig. 1). Cu was also not detected in both H and L group of 0th day mice. In both H and L group mice at 24th day, Cu was detected at the same level.

From these results it was found that polyphenol accelerated the excretion speed of Cu from mouse's inside of the body. Thus, Cu was detected only in H group mice hair at 14th day. But, after enough time passed, the excretion of Cu reached to equilibrium, and Cu was detected in hair of all samples.

To confirm this hypothesis, more experiments are required.

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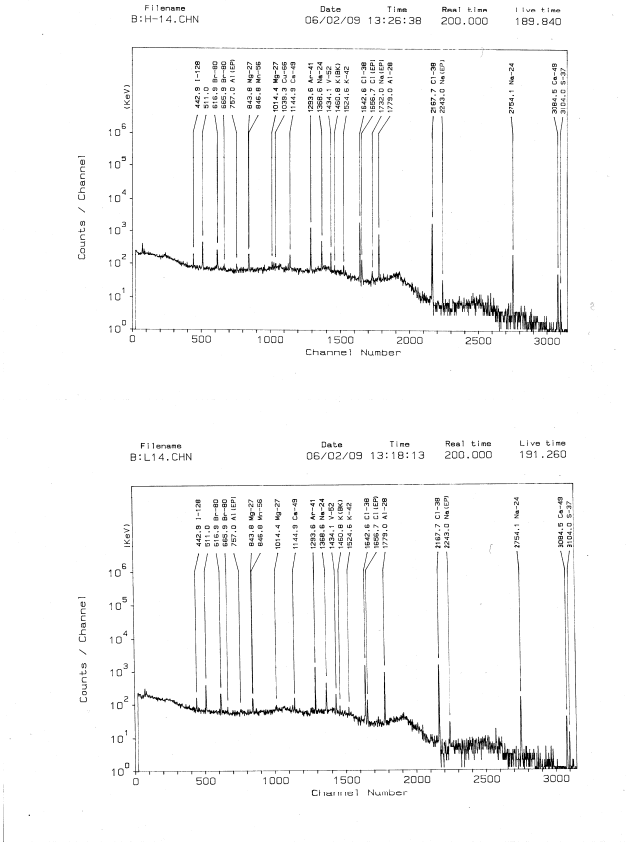

Fig. 1. Observed gamma-ray energy spectra
Spectrum of H group of mice at 14 th day is shown in upper spectrum.
Spectrum of L group of mice at 14 th day is shown in lower spectrum.

採択課題番号 21P8-7　植物由来医薬品および健康食品中の微量元素とこれを与えた動物体毛中の微量元素の放射化分析　プロジェクト

（長浜バイオ大・バイオサイエンス）川瀬雅也（大阪大谷大・薬）森本正太郎
（大阪大・RI センター）斎藤　直