VIII-II-1. Project Research

Project 7

K. Takamiya

Research Reactor Institute, Kyoto University

Objective: The collaborative research between KURRI and Korea Atomic Energy Research Institute (KAERI) started in 2006. Experiments by means of neutron activation analysis (NAA) and prompt gamma-ray activation analysis (PGA) have been carried out using the research reactor HANARO at KAERI in this project with the collaboration of KURRI and KAERI staffs. Because of the difference of the neutron flux between KUR and HANARO, the basic data for NAA and PGA experiments is important to perform effective analysis. After the test experiments performed in the early stage of this project, characteristics of those methods in using HANARO became clear. As the next stage of the project, analytical experiments for a variety of samples (biological, environmental, geochemical, industrial, and nanomaterials) should be carried out on the basis of the results of the test experiments.

Research Subjects: The common subject of this project is carrying out characteristic NAA or PGA experiments using HANARO which has higher neutron flux than KUR. The respective research subjects of eight research groups of this project are described as follows;

<u>P7-1</u>: Distribution of antimony and arsenic concentration in soil around smelter

<u>P7-2</u>: Electric conducting properties of doped carbon nanotubes

<u>P7-3</u>: Mineral change in human hair by milk intake

 $\underline{P7-4}$: Arsenic speciation in the extract of edible seaweed sample

<u>P7-5</u>: Neutron activation analysis for various elements in biological samples

<u>P7-6</u>: Quantification of ion-implanted arsenic in silicon by neutron activation analysis

<u>P7-7</u>: Prompt gamma-ray analysis of geochemical samples

<u>P7-8</u>: Development of gold-applied-paper neutron monitor

Results: Eight respective subjects described above have been studied in this project using HANARO. But, unfortunately, the research group of P7-5 had no chance to perform an experiment because of inconvenient schedules of the reactor operation and our visit to KAERI. The brief summaries of the subjects are as follows;

<u>P7-1</u>: Characteristics of antimony contamination in contradistinction to arsenic which is the homologous elements of antimony were investigated. Arsenic and antimony included in soil samples collected around an antimony smelter has been determined by NAA method.

And it is found that there is little correlation between concentrations of antimony and arsenic.

<u>P7-2</u>: The evaluation of the iodine contents by NAA was proved to be effective to evaluate the discrepancy of the nominal and the actual doping levels. And the experimental results indicate that iodine is one of the effective dopants to control the doping level of the SWNTs (single-walled nanotubes) even in the light doping region.

<u>P7-3</u>: The concentration of minerals in human hair samples assorted by dietary habit has been determined by NAA method using HANARO, and it was found that there are distinct differences between the mineral balances in the hairs with and without milk intakes.

<u>P7-4</u>: Arsenic compounds in the extract of hijiki were analyzed by NAA, ICP-AES and ICP-MS. As a result, the separation profiles for mixture solution of four standards and the extract of hijiki are obtained. There found As(III), MMA, As(V), and DMA peaks at the same elution volume, and two small peaks were found; after the peaks of As(V) and DMA. Unknown compounds causing the small peaks will be determined by further investigation in the future.

<u>P7-6</u>: INAA is applied to quantify arsenic as a dopant in silicon. It is found that INAA can quantify arsenic amounts of 10^{15} atoms/cm² with repeatability less than 0.78 %, and is adaptable to shallower dopants. INAA is also adaptable to evaluate smaller arsenic amounts, which is important to satisfy of the industrial demands.

<u>P7-7</u>: A performance of PGA system at HANARO has been evaluated in their previous study. As the next stage of the evaluation, geochemical standard reference materials were analyzed by k_0 method as well as conventional comparative method. As a result of evaluation, HANARO-PGA does not have a sufficient performance as an alternative facility of JRR3-PGA.

<u>P7-8</u>: Characteristics of the newly developed neutron monitor which is prepared using an ink-jet printer has been investigated by NAA method using HANARO. The uniformity and controllability of the preparing method has been examined. As a result, uniformity of monitoring material and reproducibility of the preparation method were confirmed. And it was found that the amount of monitoring material can be controlled precisely. The neutron flux at the neutron irradiation field such as inside of pneumatic capsule inserted into KUR will be measured using the developed neutron monitor in the future.

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Distribution of Antimony and Arsenic Concentration in Soil around Smelter

S. Fukutani, K. Takamiya, Y. Nakano, R. Okumura, J. H. Moon¹, S. H. Kim¹, T. Takahashi and A. Koyama

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

INTRODUCTION: 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 (Sb₂O₃) 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. In this study we attempt to analyze characteristics of antimony contamination in contradistinction to arsenic which is the homologous element of antimony.

EXPERIMENTS: The soil sample was collected around an antimony smelter where health hazards had been occurred in 1970's. The sampling was conducted with core samplers and the collected soil samples in the core samplers were divided into 10-12 depth direction fractions. Those were dried in air and sieved through a 2mm sieve. About 100 mg soil was sampled from each fraction and these soil samples were irradiated, using the neutron irradiation facility of NAA1 (thermal neutron flux: 3.9×10^{13} n/cm²/s) at HANARO (High-flux Advanced Neutron Application Reactor) in KAERI (Korea Atomic Energy Research Institute). Arsenic standards made from standard solution and BCR-176(city waste incineration ash, Commission of The European Communities) for antimony standard were irradiated together with soil samples. Irradiation time was 200 seconds for the each capsule, 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 in the soil samples at sampling site AA and AB are plotted in Fig. 1 with concentration as abscissa and depth as ordinate. Sampling site AA and AB are adjacent each other and considered to be about the same point. Background value of antimony in soil is reported a few mg / kg, on the contrary in Fig. 1, it is recognized that concentration values of antimony are several tens times larger than the



Fig. 1. Antimony concentration in the soil samples.



Fig. 2. Arsenic concentration in the soil samples.

background. It indicates that the antimony in those sampling area is loaded artificially. Additionally, the Fig. 1 tells that antimony loaded on the surface layer moved toward depth direction, and that the moving speed was gathered to be slowly from the antimony concentration gradient. Measurement results about arsenic at all sampling sites AA, AB and B are plotted in Fig. 2 with concentration as abscissa and depth as ordinate. By comparison with antimony of Fig.1, the concentration gradient of arsenic does not appear, and the concentration values were the nearly same as background. It became clear that there is little correlation between concentrations of antimony and arsenic.

REFERENCE:

[1] Industrial Rare Metals No.123; 2007 Annual Review, Arumu Publishing Co (in Japanese).

採択課題番号 20P7-1 土壌中有害元素の汚染経路の解明に関する研究 (京大・原子炉)福谷 哲、高宮幸一、高橋知之、小山昭夫

プロジェクト

PR7-2

S. Kitao, Y. Kobayashi, M. Seto, M. Saito¹, J. H. $Moon^2$, and S. H. Kim²

Research Reactor Institute, Kyoto University ¹Graduate School of Science, Kyoto University ²Korea Atomic Energy Research Institute

INTRODUCTION: Carbon nanotubes are graphitic tubules with a diameter of a few to a few tens of nanometers, which were discovered by Iijima in 1991 [1]. Since they have some unique physical properties, such as extremely-high electric and thermal conductivities, their investigations have been extensively carried out until today. However, even though it is important to improve and control their physical properties, the doping methods have not yet been well established. Since iodine is one of the effective dopants, we applied iodine doping to single-walled nanotubes (SWNTs) and investigated to reveal the effectiveness of the iodine doping using Mössbauer spectroscopy [2]. Since the evaluation of the doping level was not accurate by weight uptake, the doping level was evaluated by the neutron activation analysis (NAA), which is one of the most suitable methods to quantify the amount of specific element. The investigation on the comparison between open- and closed-end SWNTs for iodine doping was carried out in a certain doping levels and the doping level was confirmed by the NAA [3]. In this research, we tried to dope with iodine in the light doping regions to investigate the effectiveness of the doping method and the NAA evaluation of the doping levels. Open- and closed-end SWNTs were also compared to speculate the doped site of the iodine.

EXPERIMENTAL: The SWNTs used are the commercial "purified HiPco" tubes synthesized by the modified gas-phase processes. Open-end tubes are prepared by a heat treatment at 500°C for 10 min in air, while as-purchased sample contains almost closed-end tubes. The both samples were heated at 500°C for 10 min in vacuum to remove adsorbed molecules. Then, they were sealed in a T-shaped evacuated ampoule with a dried PdI_2 powder. By heating the PdI_2 -end of the ampoule, decomposed iodine was doped into SWNTs. The NAA was performed at High-flux Advanced Neutron Application Reactor (HANARO) in Korea Atomic Energy Research Institute (KAERI). The neutron irradiation was carried out for 1 min at the pneumatic transport system (PTS) #3 to evaluate I contents by

measuring 443 keV gamma-rays from ¹²⁸I with a half-life of 25 min. The irradiation for 30 min at the PTS #1 for Fe contents was carried out and the measurements were done a few weeks later to detect 1292 keV gamma-rays of ⁵⁹Fe with a half-life of 44.5 d. The measurement of the remaining Fe catalyst was used to confirm the quantities of the samples. The content of each element was calibrated by each standard sample.

RESULTS: Table I shows the observed contents of pristine SWNTs and the ones after iodine doping in the intended light doping regions. The fact that the iodine was clearly observed in the doped SWNT showed the iodine doping was successful even in the light doping regions. The evaluation of the iodine contents by NAA was proved to be effective to evaluate the discrepancy of the nominal and the actual doping levels. Although the precise control of the doping level was not achieved, the measured doping level was almost comparable to the nominal doping levels. This fact shows the control of the doping level is practically possible by this doping method. The closed- and open-end SWNTs are also compared and the obtained results were almost similar. This fact implies the doped iodine was mainly doped outside the tubes in this doping method. The experimental results indicate that iodine is one of the effective dopants to control the doping level of the SWNTs even in the light doping region. The result is useful for further improvement of the precise control of the electronic properties of the carbon nanotubes.

Table I. Measured iodine contents and doping levels x in CI_x .

Samples	Closed-end	Open-end
	µg∕ mg sample	µg/ mg sample
Not doped	0.007 ± 0.007	0.018±0.012
Indian domad	2 47 10 64	9 25 1 7
Ioume doped	5.47 ± 0.04	8.55±1.7
in nominal <i>x</i> =0.001	(measured	(<i>x</i> =0.0008)
	<i>x</i> =0.0003)	
Iodine doped	23.0±4.4	23.3±5.5
in nominal x=0.003	(<i>x</i> =0.0022)	(<i>x</i> =0.0022)

REFERENCES:

- [1] S. Iijima, Nature 354 (1991) 56.
- [2] S. Kitao, M. Seto, Y. Kobayashi, R. Haruki, S. Masubuchi, S. Kazama, H. Kataura, Y. Maniwa, S. Suzuki and Y. Achiba, *Hyperfine Interact.* (C) 5 (2002) 67.
- [3] S. Kitao, Y. Kobayashi and M. Seto, *Hyperfine Interact.*, 183 (2008) 179.

採択課題番号 20P7-2 ドープしたカーボンナノチューブの電気輸送特性 プロジェクト (京大・原子炉)北尾真司、小林康浩、瀬戸 誠(京大院・理)齋藤真器名、東谷口聡 S. Ohmori, K.Takamiya¹, Y. Nakano¹, J. H. Moon² and S. H. ${\rm Kim}^2$

Otsuma Women's University ¹Research Reactor Institute, Kyoto University ²Korea Atomic Energy Research Institute

INTRODUCTION: Quantities and qualities of minerals from ingested food seem to relate to the prevention and health damage. The human hair is a good archival recorder of mineral concentration in a human body. In the present work, changes of the concentration of mineral in human hair caused by milk intake were investigated. The concentration of minerals in human hair samples assorted by dietary habit was determined by neutron activation analysis (NAA) method using HANARO in KAERI.

METHODS: Hair was collected throughout a boarder of women's university. The numbers of hair samples are 32 persons that before they ingest and after they have been continued to ingest 500 mL of milk a day for six months and the number of hair samples are 2 persons that they cannot ingest milk. Hair was taken and washed by procedure recommended by IAEA arrange. The hair specimen was irradiated for 1 or 30 min. in the pneumatic tube irradiation facility of the HANARO in KAERI. Gamma-ray spectrometry of short-, middle- and longlived nuclides was carried out with a 4096 channel pulse height analyzer equipped with a 50 cc Ge-detector. Durations of irradiation cooling and measurement was combined appropriately. Concentrations were calculated in comparison with a value obtained from authentic samples. Standard materials of orchard leaves (NBS) and human hair (NIES) were irradiated together produced standard. And produced standard was recognized. Then, 16 elements in hair were quantitatively detected.

Tab.	1	Mineral	concentrations	
			in milk	(100g)

Mineral	contration
Na	55 mg
к	140
Ca	130
Mg	13
Fe	0.1
Zn	0.44
Cu	0.005

RESULTS and DISCUSSION: From the obtained γ -ray spectra, 16 elements were determined in the present experiments. Tab.1 showed mineral concentrations in us used milk. Fig. 1 and 2 show mineral concentrations standardized by normal adult female hair ^[1, 2]. The value given by concentration difference of elements between the hair ingest milk and the control, divided by standard deviation (σ) of control value is plotted against each element, setting ± 2 as normal range. Then we had comparative studies between the administrated mineral quality and the mineral balance of hair. As shown in Fig.1 and 2, changes of the mineral balances in hair were recognized compared with normal value, mineral concentration in hair by milk intake and hair ingest no milk.



Fig.2 Mineral change in hair by ingesting no milk

REFERENCES:

[1] S. Ohmori, et al., J. Radioanal. Chem. 63 (1981) 269-282.

[2] S. Ohmori, Tech. Environment, 15 (1986) 537-543.

採択課題番号 20P7-3 毛髪含有元素濃度の基礎的・応用的研究 (大妻女子大・社会情報)大森佐與子(京大・原子炉)中野幸廣 M. Fukushima, N. Muramatsu, K. Adachi and Y. Nakano¹

Department of Science and Engineering, Ishinomaki Senshu University ¹Research Reactor Institute, Kyoto University

INTRODUCTION: Arsenic is found widely in biology, geology, natural waters and so on. There are many arsenic compounds in natural, and it is known that inorganic compounds have higher toxicity comparing to organic compounds. Seaweeds have rather high levels of arsenic. It is important to analyze arsenic compounds in edible seaweeds. In this study, arsenic compounds in the extract of hijiki were analyzed by neutron activation analysis (NAA), inductively coupled plasma spectrometry (ICP-AES) and ICP-MS.

EXPERIMENTS:

(1) Chemicals

Standard solutions of As(V), monomethyl arsonic acid (MMA), and dimethyl arsenic acid (DMA) were prepared from disodium hydrogenarsenate heptahydrate, MMA, and DMA, respectively by solving appropriate amount to distilled water. Standard arsenic solution for atomic absorption spectrometry was used as As(III) standard.

(2) Separation of arsenic compounds

Separation was done by the modification of the Yalcin's way [1]. Strong cation exchange resin (SCX, 50-100 mesh, H⁺ form, Bio-Rad Laboratories Co.) and strong anion exchange cartridge (SAC, Cl⁻ form, Waters) were used for separating As(III), As(V), MMA, and DMA each other. SAC was changed to OH- form by conditioning before use. For the separation condition of four arsenic species, SAC was connected onto SCX column, and the solution of As(III), As(V), MMA, and DMA mixture was loaded on SAC. As (III) was remained in mobile phase. After washing with distilled water, SCX and SAC were separated each other, MMA was recovered by 0.1 M acetic acid from SAC, then As(V) was recovered by 1M HCl from SAC. DMA was recovered by 2 M nitric acid from SCX. When mixture of As (III), As (V), MMA, and DMA (100 µg As, each) were separated by the way explained above, all the yield obtained for four compounds by AAS were 100%.

(3) Extraction of arsenic compounds from hijiki

For extracting arsenic compounds from seaweeds, 20 ml of solvent (distilled water : methanol = 1 : 0, 3 : 1,1 : 1, 1 : 3) was heated with 1 g of hijiki at 55°C for 1~6 hours. The extract was filtered with 0.45 µm membrane filter, then filled up till 25 ml. Thus obtained 5 ml of sample solution was loaded on the SAC connected to SCX, and washed with 10 ml of distilled water. MMA was separated with 6 ml of 0.1 M acetic acid from SAC, then As (V) was separated with 15 ml of 1M HCl from SAC. DMA was separated by 25 ml of 2M nitric acid from SCX. At each step, each 2 ml fraction of eluate was collected . Then 1 ml of each fraction was put into small plastic vial, and was dried up under the lamp. (4) NAA

Three vials were sealed in a plastic vial for irradiation, and was irradiated for 1 minute at #1 in HANARO. After 6 - 30 hours decay, gamma counting was done for each vial for 5 - 10 minutes.

RESULTS: The separation profiles for mixture of four standards and the extract of hijiki are shown in Fig. 1. As for the mixture of four standards, the yield for each standard was 40, 92, 47, and 210% for As (III), As (V), MMA, and DMA, respectively. Then the loss of each As compounds by the heating before the irradiation was examined. As a result, after heating standard solution for dryness the loss of each compound was 50%, 10%, 10%, and 10%, for As (III), As (V), MMA, and DMA, respectively. As for high yield of 210% for DMA, it might have caused by the high levels of Na-24 in the fraction after the irradiation. Since NaOH solution was used for conditioning of the SCX, Na⁺ on the resin was exchanged by H⁺ when 2M nitric acid was used for eluting DMA. This problem was solved by using KOH solution for conditioning the SCX. For further work, irradiation of solid phase without elution should be investigated for avoiding the loss of As by heating. As for the separation of hijiki extract, As (III), MMA, As (V), and DMA peaks were found at same elution volume. There were two small peaks were found; after the peaks of As (V) and DMA. By investigating further more As species, these compounds would be detected.



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REFERENCE:

[1]S. Yalcin, X. Chris Le, J. Environ. Monit., 3 (2001) 81-85.

採択課題番号 20P7-4 食用海草抽出液中のヒ素化学種同定 (石専大・理工)福島美智子、村松なつみ、安達奏恵(京大・原子炉)中野幸廣

PR7-5 Quantification of Ion-implanted Arsenic in Silicon by Neutron Activation Analysis

T. Takatsuka, K. Hirata, Y. Kobayashi, A. Kinomura¹, Y. Nakano², K. Takamiya², J. H. Moon ³ and S. H. Kim ³

National Metrology Institute of Japan, National Institute of Advanced Industrial Science and Technology ¹Research Institute of Instrumentation Frontier, National Institute of Advanced Industrial Science and Technology ²Research Reactor Institute, Kyoto University ³Komp Atomic Energy Research Institute

³ Korea Atomic Energy Research Institute

INTRODUCTION: For the high performance of silicon semiconductor devices, dopants are ion–implanted at lower energy to make shallower electric junctions. Dopant concentration should be controlled in 4 % precision at present and in 2 % after the year 2010 as indicated in the international technology roadmap for semiconductors (ITRS) 2007 [1]. Several techniques, such as secondary ion mass spectrometry (SIMS) and Rutherford backscattering spectrometry (RBS), are widely used for characterizing the depth profiles of dopant concentration. In order to quantify the dopant concentration by these techniques, reference materials are required for calibration.

Certified reference materials (CRMs) of shallow arsenic implants in silicon are now under development at the National Metrology Institute of Japan (NMIJ). To quantify the amount of ion-implanted arsenic atoms, one of the potential measurement techniques is Instrumental Neutron Activation Analysis (INAA) since INAA is a simple method without matrix-effects and pre-chemical treatments. In this study, INAA is applied to quantify arsenic in silicon.

EXPERIMENTS: Arsenic ions were implanted into Si(001) wafers at an energy of 10 keV and doses of A: 3 $\times 10^{15}$ /cm² and B: 3 $\times 10^{14}$ /cm². The wafers were diced into 15×15 mm squares pieces by dicing saw. The samples were cleaned in isopropyl alcohol with ultrasonic and rinsed in ultrapure water, and then cleaned ultrasonically in high-purity nitric acid followed by a rinse in ultrapure water. After cleaning, all samples were dried for a day in a clean bench. As standards for calibration of arsenic amount, a working standard solution was prepared by diluting SRM 3103a (NIST) gravimetrically. The arsenic standard solution was deposited from a polyethylene pipette onto filter papers, and the deposited solution was accurately weighted to calculate the arsenic content. All samples and standards were heat-sealed in clean polyethylene envelopes and stacked in an irradiation container, and then neutron irradiation was performed for 20 min at 3.9×10^{13} n cm⁻² s⁻¹ of thermal fluence rate in HANARO.

RESULTS: The largest peak at 559 keV of gamma ray from ⁷⁶As was integrated for each spectrum to calibrate the arsenic amount. Table 1 shows the results for three samples of A1, A2, and B1. Arsenic amounts were averaged from the results of two measurements for each sample. Area densities were calculated from the arsenic amounts, areas of sample surfaces, and arsenic atomic mass [2]. The mean arsenic density for A1 and A2 was 3.19×10^{15} /cm² and the relative standard deviation for four measurements was 0.78 %. Concerning sample B1 with lower arsenic density, the calibrated result was 3.00 $\times 10^{14}$ /cm² from two measurements, which shows good agreement with the nominal implantation dose. It shows that INAA has a possibility to quantify arsenic amounts when the area density is as low as 10^{14} /cm².

Table 1. Calibrated results of arsenic amounts in silicon. Area densities were calculated from area and arsenic atomic mass.

	As amount [ng]	area [cm ²]	area density [atoms/cm ²]
A1	899	2.28	3.18×10^{15}
A2	909	2.27	3.22×10^{15}
B1	83.4	2.23	3.00×10^{14}

SUMMARY: It is found that INAA can quantify arsenic amounts of 10^{15} atoms/cm² with repeatability less than 0.78 %, and is adaptable to shallower dopants. The evaluation of uncertainties is now under way. INAA is also adaptable to evaluate smaller arsenic amounts, which is important to satisfy the industrial demands for reference materials for calibrating the implanted dose of arsenic at shallow junctions.

REFERENCES:

- International Technology Roadmap for Semiconductors 2007.
- [2] ATOMIC WEIGHTS OF THE ELEMENTS 2005, IUPAC TECHNICAL REPORT (2006).

採択課題番号 20P7-6 中性子放射化分析を用いた珪素中イオン注入ひ素の定量分析 プロジェクト (京大・原子炉)中野幸廣(産総研・計測標準)小林慶規、平田浩一、高塚登志子 (産総研・計測フロンティア)木野村淳

Prompt Gamma-Ray Analysis of Geochemical Samples

Y. Oura, B.S. Wee, M. Ebihara, J. H. Moon¹ and G. M. Sun¹

Graduate School of Science and Engineering, Tokyo Metropolitan University ¹Korea Atomic Energy Research Institute

INTRODUCTION: Domestic practical prompt gamma ray analysis (PGA) facility for determination of multielemental concentrations for geochemical and cosmochemical samples is only PGA system in JRR-3, JAEA. Thus, alternative facility for PGA is strongly required, especially during the intermission of reactor due to a periodic check or a failure. PGA system at HANARO, KAERI is an important candidate for such facility in Asia. Therefore we evaluated a performance of PGA system at HANARO ourselves in 2006 and 2007. In 2008, we tried to analysis geochemical standard reference materials by k_0 method as well as conventinal comparative method.

EXPERIMENTS: Powder samples (JB-1, JSO-2, JZn-1, and JCu-1) in FEP bag were set on Teflon dish fixed at a center of neutron beam. These were irradiated by thermal neutrons for about 2 hours and induced prompt gamma rays were detected during irradiation under anticompton mode. Since Teflon dish was fixed at a center of neutron beam, reproducibility of sample position was higher than the experiment in 2007. Peak area was calculated using HypermetPC software. Kayzero factor values used for determination in this study were cited from [1] and a relative detector efficiency function was supplied by Dr. Sun, KAERI.

RESULTS: Comparing with PGA system at JRR-3 (JRR3-PGA), significant disadvantage of PGA system at HANARO (HANARO-PGA) is a higher background (BKG) count [2]. Although we had found about 1/3 times decrease of BKG count rate in experiment 2007, BKG count rate in this experiment became higher and almost same condition in the experiment 2006. Thus, contribution of BKG count rate was corrected for determination of B, Na, Al, Fe, Ni and Cu,.

Several inconsistencies between determination values obtained at HANARO-PGA and JRR3-PGA were observed in experiment 2007 [2]: for example, Si concentrations by HANARO-PGA were systematically higher. In this study same samples were analyzed again. Most elemental concentrations for all samples are consistent with recommended or reference values within $\pm 10\%$, though some elemental concentrations were inconsistent. Most of large different concentration values were found to be caused by low count rates and/or larger contribution of BKG count. No systematic deviation was observed.

In contrast to comparison method, analytical results by k₀ method were not good. Significant inconsistent cases are Mn/Si, Fe/Si, and Sm/Si concentration ratios. These ratios for all samples were systematically higher, though Si, Fe, and Sm concentration values obtained by comparison method were good, as shown in Table 1. Kayzero factor values for PGA determined at different facilities are known to be consistent for elements whose neutron capture cross section comply with 1/v law. Although neutron spectrum at the sample position in HANARO-PGA is characteristic, that is, there are several peaks at the energy region of under 1 eV, the k₀ factors cited in ref. [1] are supposed to be applicable to HANARO-PGA. In fact, it was found that observed S/Na ratio for Na₂SO₄ reagent sample were same as a theoretical value within its error. For Sm, non-1/v element, k₀ factors [3] obtained at HANARO-PGA was used instead of ref. [1]. However, results did not improve. The cause of this deviation was still not clear. Same samples will be analyzed at JRR3-PGA both comparison and k₀ methods in order to assist in solving this unclear situation.

Unfortunately, HANARO-PGA does not have a sufficient performance as an alternative facility of JRR3-PGA. In near future, we expect that a new PGA facility using guided neutron beam at HANARO will have a excellent features.

Table1. Si and Fe concentration by comparison method and Fe/Si concentration ratio by k_0 method.

		,	
	Comparison method		k ₀ method
	Si, %	Fe, %	Fe/Si
JB-1	25.5 ± 0.8	5.58 ± 0.45	0.313 ± 0.019
	24.5^{*}	6.29^{*}	0.257^{*}
JSO-2	19.3 ± 0.7	6.76 ± 0.7	0.500 ± 0.048
	19.9^{*}	6.81*	0.342^{*}
JZn-1	23.8 ± 1.3	11.9 ± 0.8	0.713 ± 0.046
	20.5^{*}	11.8^*	0.576^{*}
JCu-1	14.3 ± 0.6	15.2 ± 1.1	1.52 ± 0.08
	13.6^{*}	17.5^{*}	1.29^{*}

recommended or reference value

REFERENCES:

 IAEA "Database of Prompt Gamma Rays from Slow Neutron Captutre for Elemental Analysis" (2007).
Y. Oura *et al.*, KURRI Progress Report 2007, p162.
G.M. Sun *et al.*, J. Radioanal. Nucl. Chem., **256** (2003) 541-552.

採択課題番号 20P7-7 地球化学的試料用ならびに環境試料用標準試料の 即発ガンマ線分析 (首都大院・理工)大浦泰嗣、Wee Boon Siong、海老原充、竹田光世 プロジェクト

PR7-7

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

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

INTRODUCTION: The neutron flux is one of the most important parameters in neutron irradiating experiments such neutron activation analysis (NAA), prompt gamma-ray activation analysis (PGA) and radioisotope production. In NAA experiments, parameter of neutron flux irradiated to samples is not so important in irradiating analytical samples with standard samples. But, if standard and analytical samples placed separately in the irradiating field each other, the correction for the neutron flux is necessary. Then the information about neutron flux is required for almost all the neutron irradiation experiments. Metallic wires are often applied for measurement of neutron flux because metallic wires are easy to locate them at the neutron irradiation fields such as inside of pneumatic capsules. But self-absorption effect causes inhomogeneous activation between the surface and center of the wire in neutron irradiation. And another thing, self-shielding effect causes broadening of photoelectric peaks in gamma-ray spectroscopy. These effects trigger experimental error. On the other hand, there is less influence of those effects in using metallic foils. But fixing foils at the irradiation fields and treating them is not easy because they are fragile. In this work, the new method for preparing neutron-monitor materials using a common ink-jet printer has been developed for the purpose of neutron monitoring at various neutron irradiation fields. As like this preparation method, the fabrication method of standard radioactive surface source using ink-jet printer has been developed ^[1], and the uniformity and positional resolution of this method was confirmed. In this report, the uniformity and controllability of the preparing method has been examined in detail by NAA method.

EXPERIMENTS: In this preparation method, a gold solution was applied onto a paper sheet using an ink-jet printer because an ink-jet printer can apply pigment to a print sheet uniformly. In order to confirm the uniformity of applied gold, the amount of gold on the print sheet was determined by NAA method using HANARO. The five paper fragments to which gold solution was applied with the same concentration at different positions of the same paper sheet were irradiated by neutrons with a gold standard sample. After the irradiation, the gamma-rays from activated samples were measured by a Ge-detector to determine the amount of gold in the irradiated samples. And using the same NAA method, the correlation between the amount of the applied gold and the input parameter of the pigment concentration for ink-jet printer was examined.

RESULTS: The amounts of gold contained in five samples are shown in Fig. 1. It is found that the variation of the applied amount is small enough. The uniformity of applied gold on a paper sheet and the reproducibility of the preparation method were confirmed. The correlation between the preset (configured) pigment concentration and the observed amount of applied gold is shown in Fig. 2. A linear correlation between them is not found in this figure, but there found the secondary correlation. This result represent that the amount of applied gold can be controlled by using the obtained correlation as calibration parameters.



Fig. 1. Variation of the amounts of gold contained in five samples which are prepared with the same printing parameters.



Fig. 2. Correlation between printing concentration and the amount of applied gold.

REFERENCE:

[1] Y. Sato, Y. Hino, T. Yamada and M. Matsumoto, Applied Radiation and Isotopes, **60** (2004) 543-546.

採択課題番号 20P7-8 地球外物質及び地球物質の中性子放射化分析 プロジェクト (京大・原子炉)高宮幸一、柴田誠一(日大・文理)小林貴之(京大院・工)関本 俊、曹 順美 (京大・工)瀧本真己