1.48 Current Status of Neutron Activation Analysis in HANARO Research Reactor

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Summary

The facilities for neutron activation analysis in the HANARO (Hi-flux Advanced Neutron Application Research Reactor) are described and the main applications of NAA (Neutron Activation Analysis) are reviewed. The sample irradiation tube, automatic and manual pneumatic transfer system were installed at three irradiation holes of HANARO at the end of 1995. The performance of the NAA facility was examined to identify the characteristics of the tube transfer system, irradiation sites and custom-made polyethylene irradiation capsule. The available thermal neutron fluxes at irradiation sites are in the range of $3 \times 10^{13} \sim 1 \times 10^{14}$ $n/cm^2 \cdot s$ and cadmium ratios are in 15 ~ 250.

For an automatic sample changer for gamma-ray counting, a domestic product was designed and manufactured. An integrated computer program(Labview) to analyse the content was developed. In 2001, PGNAA (Prompt Gamma Neutron Activation Analysis) facility has been installed using a diffracted neutron beam of ST1.

NAA has been applied in the trace component analysis of nuclear, geological, biological, environmental and high purity materials, and various polymers for research and development. The improvement of analytical procedures and establishment of an analytical quality control and assurance system were studied. Applied research and development for the environment, industry and human health by NAA and its standardization were carried out. For the application of the KOLAS (Korea Laboratory Accreditation Scheme), evaluation of measurement uncertainty and proficiency testing of reference materials were performed. Also to verify the reliability and to validate analytical results, intercomparison studies between laboratories were carried out.

I. Facilities for Neutron Activation Analysis in HANARO

There are three irradiation holes(NAA 1-3) for Neutron Activation Analysis(NAA) in the HANARO research reactor as shown in Fig. 1. The PTS (pneumatic transfer system) has been mainly used for the sample irradiation of NAA and has been operated since the end of 1996. One of the irradiation holes (NAA 2) is equipped with a Cd lined tube for epithermal NAA. The PTS is operating on two modes : PTS #1 is handled manually as a simple shuttle system, and PTS #2 is controlled by a preset computer program; start from a sample loader via an automatic sample loader, diverter, two reactor core, radiation detector, delay stacker, two counters with and without window, finish to discharged receiver. The control program is written in Microsoft's Visual Basic for Windows which is an extension of Quick Basic and employs many of the structured features of C and other high level languages. Recently, software programs for the operation of PTS #2 were developed and upgraded. The air pressure system was changed to nitrogen gas to reduce the level of background radiation.

In 2001, PGNAA facility was installed so as to use neutrons by extracting diffracted neutron beams of ST1. It will be of use for analysis of not only B and Gd in patient's blood and tissue for BNCT, but also trace elements such as Cd, H, C, N, P, S in metals and alloys, semiconductors, geological samples, environmental samples, and biological samples.

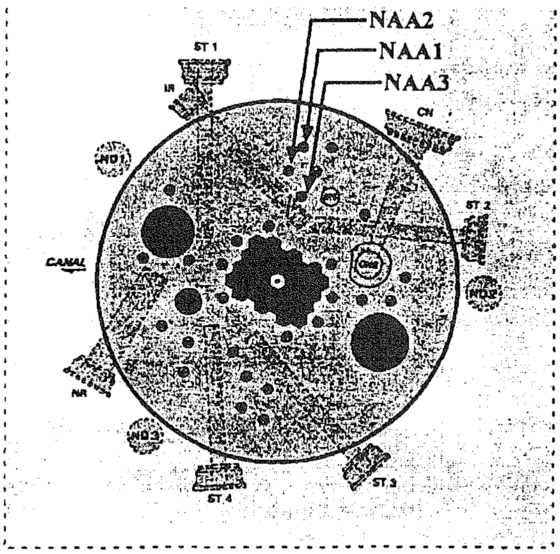

Fig. 1. Configuration of NAA Irradiation Holes in HANARO.

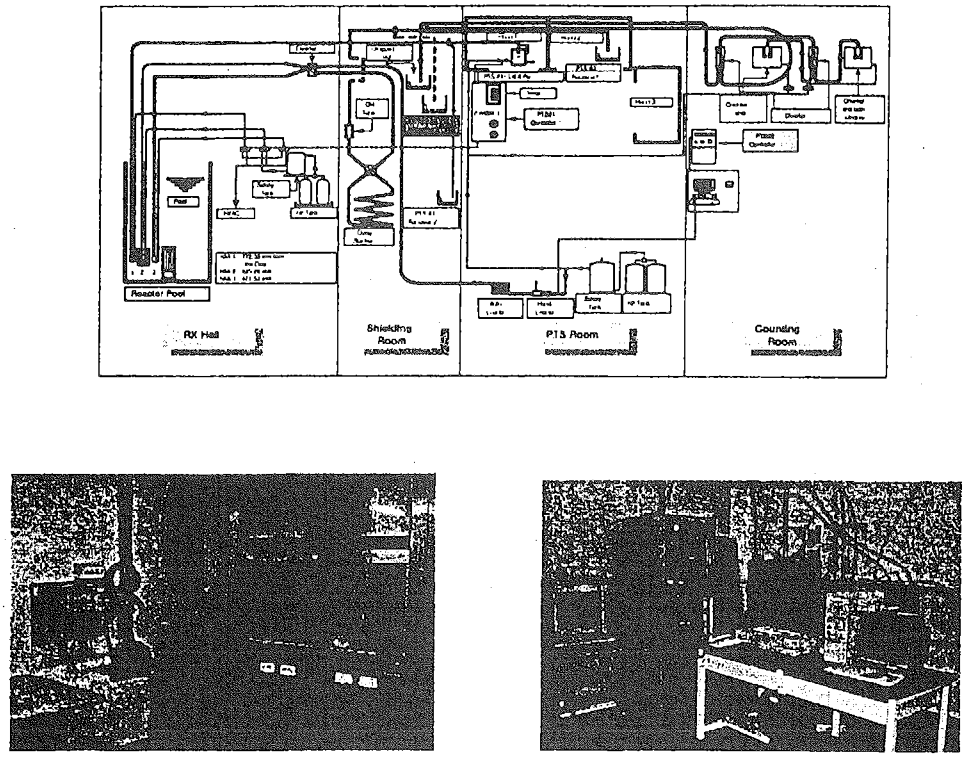

Fig. 2. Irradiation Facility(PTS) for Neutron Activation Analysis in HANARO.

The available neutron fluxes and cadmium ratios at irradiation sites are in the range of 3 x 10^{13} ~ 1 x 10^{14} n/cm^2 · s and presented in Table 1.

Table 1. Thermal, Epithermal and Fast Neutron Flux of NAA Irradiation Holes and Cadmium Ratios at 24MW Thermal Power.

Irradiation Hole	Neutron flux, n/cm^{2}sec. (24MW)			Cadmium Ratio(Au)
	Thermal, Φ_{th}	Epithermal, Φ_{epi}	Fast, Φ_f	
NAA 1	2.81±0.01 x 10^{13}	2.23±0.13 x 10^{10}	4.01±0.21 x 10^{10}	205
NAA2(Cd)	5.65±0.05 x 10^{13}	2.08±0.06 x 10^{11}	2.53±0.06 x 10^{11}	35
NAA 3	1.01±0.15 x 10^{14}	7.74±0.19 x 10^{11}	8.61±0.41 x 10^{11}	13

As shown in Fig. 3, two kinds of irradiation capsules(rabbit) were manufactured using a high density polyethylene material produced by the domestic industry. Also five kinds of inner sample capsules and two kinds of outer capsules made of pure high density polyethylene were manufactured. Some stability and physical properties such as heat-resistance, radiation-resistance and mechanical characteristics were checked to estimate a suitable irradiation condition. The physical properties of the polyethylene capsule are as follow: melting index(20.7), density(0.95), and melting point(131.6℃). Trace elements in the polyethylene rabbit and various polyethylene materials were analyzed by INAA and ICP.

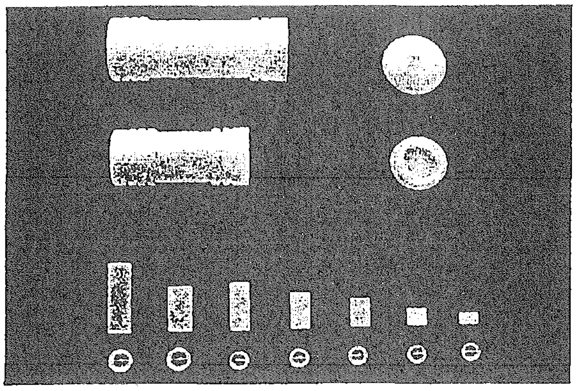

Fig. 3. Irradiation Capsule(PE) for PTS

For an automatic sample changer for gamma-ray counting, a domestic product was designed and manufactured. An integrated computer program(Labview) for the calculation of content was developed. Fig. 4 showed the gamma-ray spectrometric system.

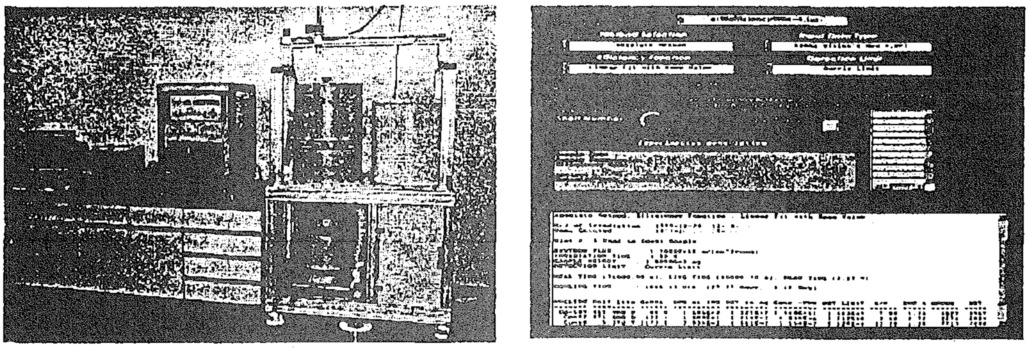

Fig. 4. Gamma-ray Spectrometric System for NAA

II. Application of Neutron Activation Analysis in HANARO

The research and development of NAA in KAERI has been applied to industry, agriculture, geology, environmental pollution, biology, archaeology, forensic science, and many other fields in Korea. Now we are using an advanced research reactor, HANARO. There are two major NAA groups: the KAERI (Korea Atomic Energy Research Institute) and the KIGAM (Korea Institute of Geology, Mining and Materials) as well as many users at universities and other organizations. In the future, we will extend the applied field of NAA and increase the number of users. Fig. 5 and Table 2 summarize trend in the utilization of NAA in HANARO.

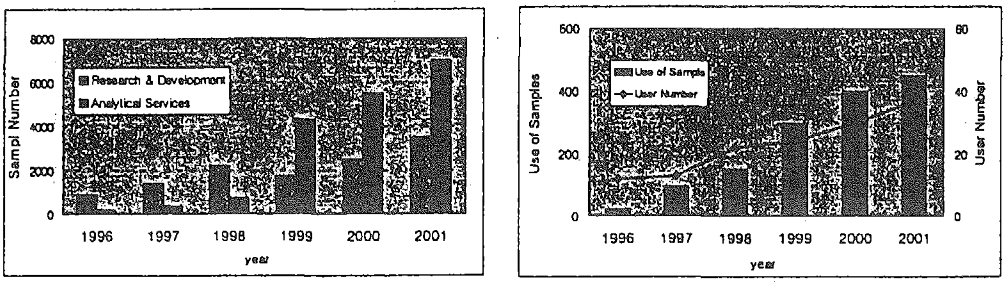

Fig. 5. The Utilization of NAA in HANARO.

Table 2. Summary of Utilization of NAA and Users.

Year / Items	Objects	Users
Research & Development	· Environmental Samples: Airborne Particulates · Biological Samples: Rice, Algae, Foodstuff, Hair · Rocks, Soils, Sediments · PTS Examination · Measurement of Neutron Flux, Cadimium Ratios · Data Intercomparison of Reference Materials · International CRP · Plastic Samples · Proficiency Test of Reference Materials	· Institutes (KAERI, KRISS, KIGAM, KORDI, etc.) · Industries (LG Corp., Samsung, EnTec, etc.) · Universities (Seoul, Yonsei, Kyungsan, Chungnam, etc.)
Analytical Services	· Impurities in Iodine · Flyash, Air Dust, Air Filter, Resin, Human Hair · Ground Water, Waste Water, Sludge, Sediments · Bentonite, Granite, Silicate, Rock, Soil, Sand · Dry Bid Sample · Reactor Coolent, Fuel Materials, Irradiation Test · Flux Monitors, Fission Track Analysis · HP SiC Plate & Tube · Impurities in RI Medicines · Tooth Sample Irradiation · Impurities in Iodine, Molybdenum, Nickel	· Institutes (KAERI, KRISS, KIGAM, KORDI, etc.) · Industries (LG Corp., Samsung Elect., KIA Motors,Samrim Corp., Sampung Industry, etc.) · Universities (Seoul, Yonsei, etc.)

Recently, basic and applied research for an analytical quality control system and trace component analysis of nuclear, geological, biological, environmental and high purity materials and various polymers have been carried out using a neutron activation analysis facility in HANARO. The major contents are summarized as follows:

1) Improvement of analytical procedures and establishment of an analytical quality control system.

With the analysis of six kinds of biological and environmental standard reference materials(NIST SRMs : Peach Leaves, Citrus Leaves, Total Diet, Bovine Liver, Oyster Tissue, Wheat Flour), the analytical error was identified(15 elements were within 10% of the relative error, and 20 elements were within 15% of the relative standard deviation). A radiochemical separation procedure(RNAA) for U and Th to improve analytical sensitivity(> 0.5 ng) was developed by using the co-precipitation and ion exchange resin methods, and the ENAA method for I, and Sr determination(sensitivity > 0.1 ppm) was also examined. Up to date, since the k_0-quantitative method which is accurate, convenient, and user-friendly, has been generalized, we are trying to implement the method to our system. Therefore, as a first step, k_0-parameters such as α and f were experimentally determined by the multi-monitor method. The measured values of α and f were 0.097 ± 0.021 and 1148 ± 95, respectively.

In connection with fundamental qualification for the application for the KOLAS, education and training for practical application, evaluation of measurement uncertainty and proficiency test reference materials were performed. Recently, our laboratory was accredited by KOLAS in September 2001.

To verify the reliability and to validate the analytical results, intercomparison studies between laboratories for twelve kinds of samples such as dust, soil, coal fly ash, sediment, algae, etc., were carried out. All results obtained were excellent and a typical result of urban dust was plotted in Fig. 6.

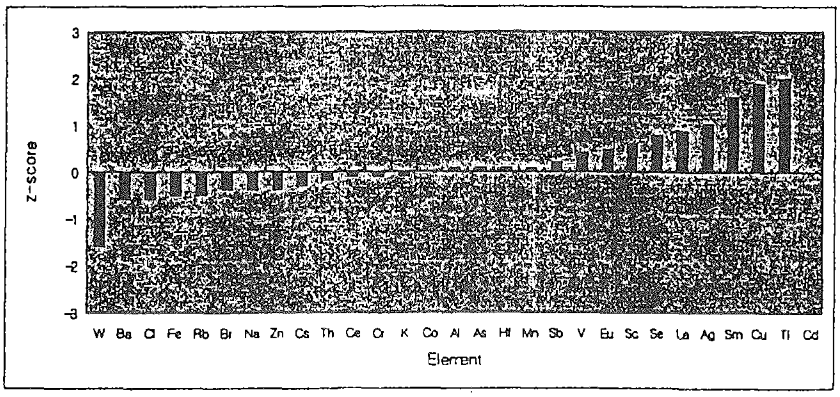

Fig. 6. Evaluation of z-score for the Elements Measured in Urban Dust.

2) Applied research and development in the fields of the environment, industry and human health.

A study on the development of analytical methods and the standardization of a procedure for the analysis of airborne particulate matter were carried out using standard reference materials(NIST SRM 1648, NIES CRM-8) and laboratory intercomparison work. Air pollution monitoring was performed using a Gent stacked filter unit low volume sampler and polycarbonate membrane filter(coarse particle : 2.5～10 μm, fine particle : <2.5 μm). To compare air quality, airborne particulates with two particle sizes were collected from urban, rural and industrial areas from May 1997 to the present. The concentrations of impurities in the blank filters were estimated and used for correction. The concentrations of 25 heavy metals and trace elements, and the mass of airborne particulate matter($PM_{2.5}$/PM_{10}) and elemental black carbon were measured. From the measured data, a presumption of the source origin and a correlation pattern between PM and the elements were statistically treated with the calculation of the enrichment factor and correlation coefficient. The results of air monitoring are plotted in Figs. 7 and 8, respectively. In addition, a probabilistic risk analysis of the inhalation of airborne toxic metals in PM_{10} based on the concentration profile was performed in an industrial complex using the analytical data by NAA and ICP-MS.

Concerning applied research on human health and the environment using NAA of biological samples, five kinds of biological standard reference materials were analyzed and the measurement error of the analytical results was evaluated. Furthermore, a study of the daily intake of micro-nutrients by the geriatric population by NAA is now being carried out by the duplicate sampling method these days.

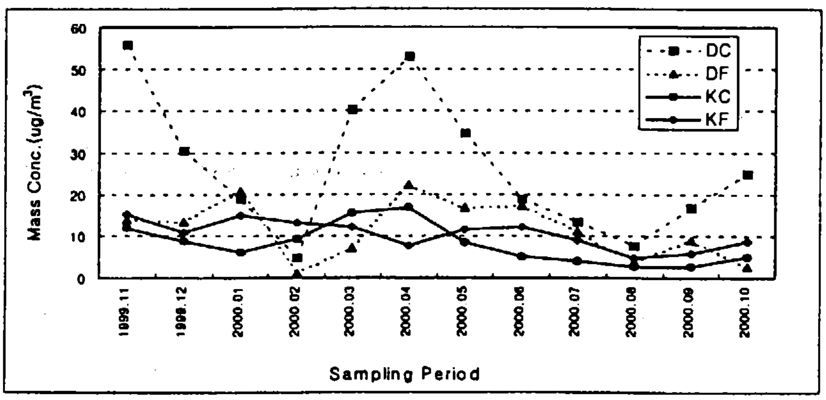

Fig. 7. Monthly Variation of Mass Concentration of Airborne Particulate Matter Collected in Urban and Industrial Areas('1999. 11 - '2000. 10)

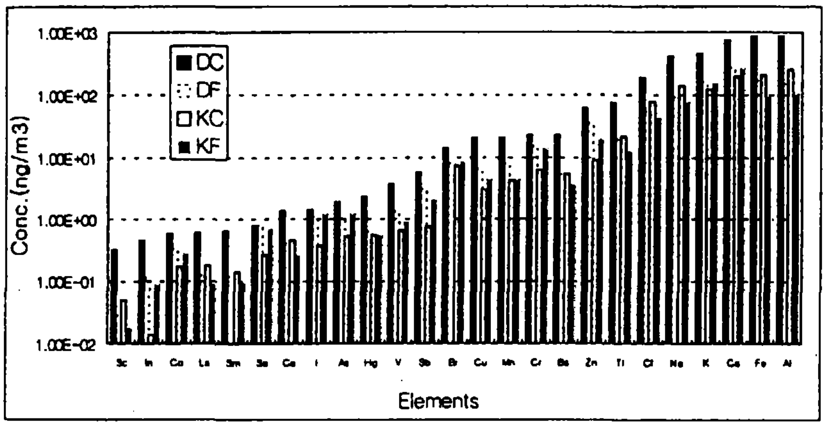

Fig. 8. Average Elemental Concentration of Airborne Particulate Matter Collected in Urban and Industrial Areas('1999. 11 - '2000. 10)

The amounts of trace elements in the air dust samples collected from inside a foundry working place were analyzed and the distribution of trace elements in the hair and blood of corresponding workers were measured to investigate the correlation. In addition, identification of the elemental distribution in healthy supplementary foodstuffs, comparison of the component elements of six kinds of herb medicines in Korea and China, the distribution of trace elements in the Korean total diet(Table 3) and representative foods were studied.

In connection with the analysis of trace elements in plastics(raw material and fabricated products) using NAA, identification of the results by NAA and comparison with those by the XRF and ICP methods, the optimum analytical conditions for NAA were determined, a new freezing-crushing technique for the preparation of a polymer sample was developed, analysis of inorganic components and additive agents in plastic materials(PE, PP) was carried out for the manufacture of irradiation rabbits and sample capsules, and the elution characteristics of toxic elements in plastic materials was also evaluated. As a geological application of NAA, 814 samples were taken from 6 regions in the Chungnam area and 12 elements including Ce, Yb, Th, Cr, etc., were analyzed by NAA. These data will be useful in identifying the characteristics and the correlation of geo-environmental and biological materials.

Table 3. Analytical Result of Korean Total Diet by NAA.

Element	Nuclide	Concentration of element		
		Range	Mean Value	Standard Deviation
		< ppm >		
Al	Al-28	16.1～18.3	17.6	1.28
Br	Br-80/Br-82	9.59～10.6	9.95	0.58
Ca	Ca-49/Ca-47	1358～1496	1418	70.9
Cl	Cl-38	10870～11780	11353	458
Fe	Fe-59	23.7～26	25.2	1.32
I	I-128	1.50～1.77	1.64	0.19
K	K-42	5394～5706	5579	134
Mg	Mg-27	807～896	850	44.4
Mn	Mn-56	8.46～8.78	8.65	0.16
Na	Na-24	10080～10470	10298	162
Rb	Rb-86	5.43～5.49	5.46	0.03
Sr	Sr-87m/Sr-85	5.3～5.80	5.55	0.36
Zn	Zn-65	31.57～33.74	32.48	1.13
		< ppb >		
As	As-76	493～603	526	51.8
Co	Co-60	29.6～31.6	30.8	1.06
Cr	Cr-51	348～420	393	39.4
Cs	Cs-134	10.6～11.6	11.1	0.71
Sc	Sc-46	3.2～3.6	3.43	0.21
Se	Se-75	223～328	276	74.5
Sm	Sm-153	46.1～47.5	46.8	0.99
Th	Pa-233	2.38～2.82	2.6	0.3
U	Np-239	31.5～38.6	35	5.0

IV. Future Plan

We will analyse airborne particulate as well as other environmental and biological samples for air and marine environmental pollution studies. The scope of this work is also included in a part of our long term nuclear research and development project related to intensive study on the application of NAA. Particularly, we are interested in the applications as well as the supporting system for analytical quality control of NAA involving the PGAA facility. To continuously improve the analytical accuracy and detection limit, we are making plans for the development of various irradiation facilities and equipment, and research of analytical techniques using the *k*o-standardization method. However, a successful application of international standards is essential to making the above project, i.e., standardization and harmonization of analytical techniques in sampling and sample preparation, analytical procedures and data evaluation. Collaboration with RCA member states, IAEA and other countries should be promoted.

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