JAERI-Conf
2004-010

PROCEEDINGS OF THE FNCA 2002 WORKSHOP ON THE UTILIZATION OF RESEARCH REACTORS JANUARY 13-17, 2003 JAKARTA & SERPONG, INDONESIA

(CONTRACT RESEARCH)

June 2004

Nuclear Technology and Education Center

日本原子力研究所
Japan Atomic Energy Research Institute

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編集兼発行　日本原子力研究所

The FNCA 2002 Work Shop on the Utilization of Research Reactors

January13-17, 2003 Jakarta & Serpong Indonesia

1. Opening January 13, Hotel Sahid Jaya, Jakarta

Opening Address by

Dr. S. Tamat (Local Organizing Committee, BATAN)
Mr. N. Tachikawa (MEXT)
Mr. I. Takeshita (JAERI)

Welcome Address by Dr. Soedyartomo Soentono
(Chairman, BATAN)

Group Photo

TCG Group

NAA Group

2. Plenary Session Hotel Sahid Jaya, Jakarta

General lecture

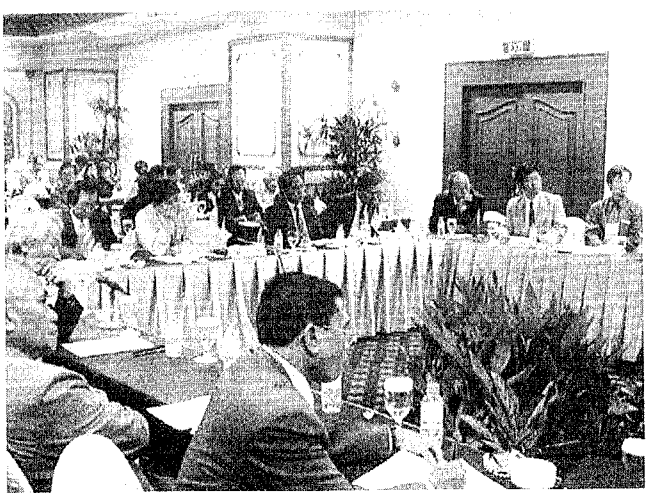
Dr. Soemewo (Pertamina Hospital, Indonesia)

Prof. Otoshi (Tohoku Univ. of C.S.S., Japan)

Research Reactor

Mr. Takeshita (JAERI, Japan)

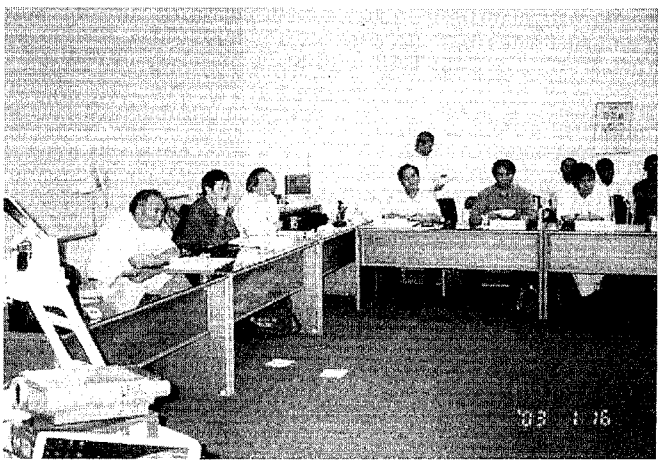
Mr. Charoen (OAP, Thailand)



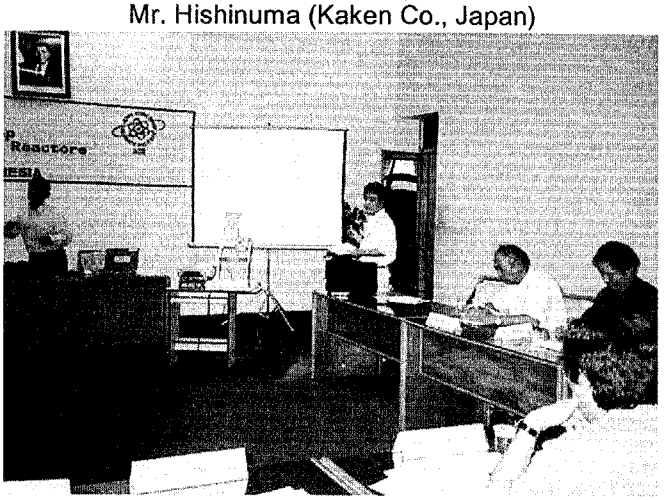
3. TCG Session January 14-16, PPR Building, BATAN, Serpong

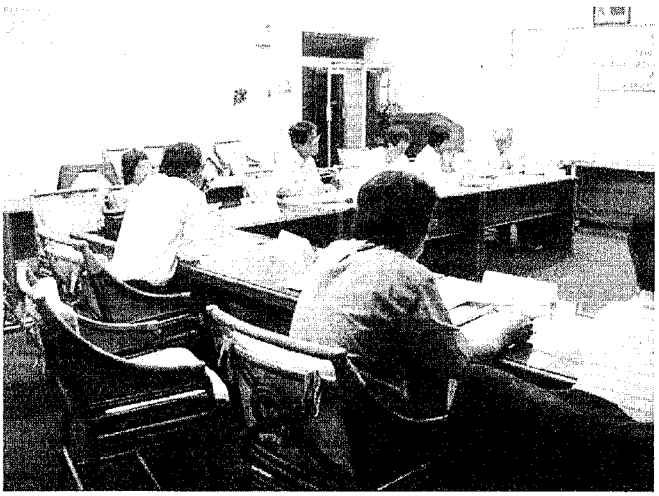
Ms. Sombrito (PNRI, The Philippines)

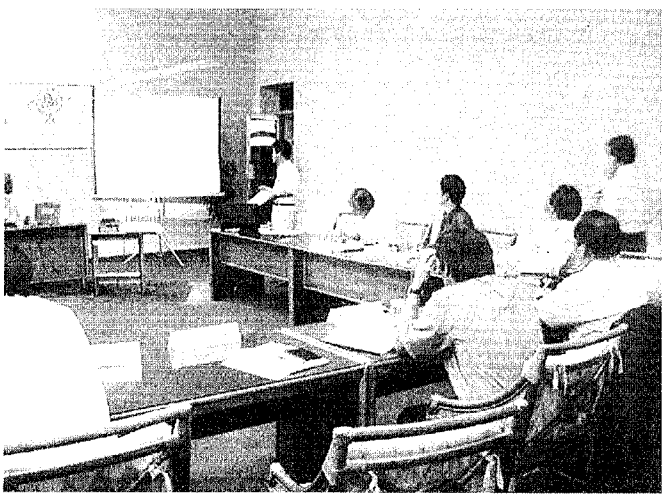
Prof. Le Van So (NRI, Vietnam)



Mr. Hishinuma (Kaken Co., Japan)

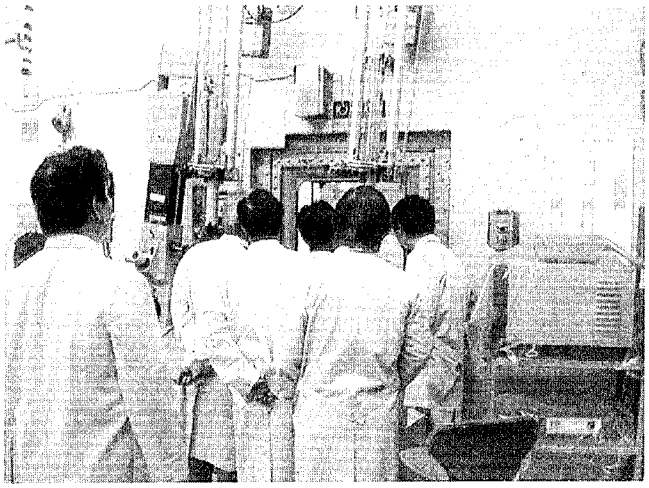




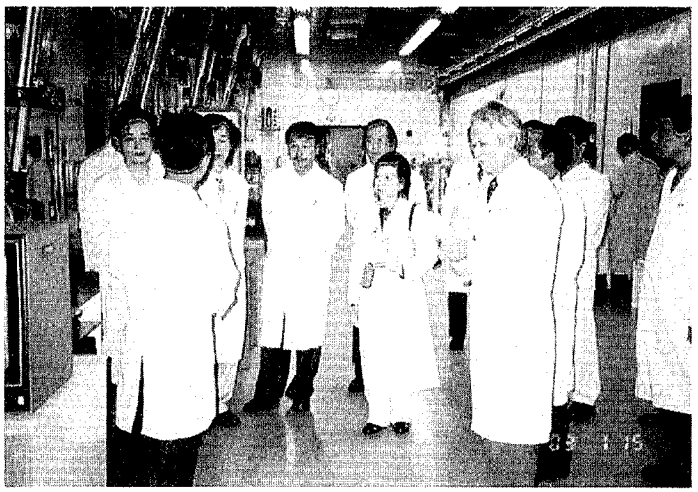


TCG Demonstration Experiment PPR BATAN

Hot Cell in the PPR Bldg

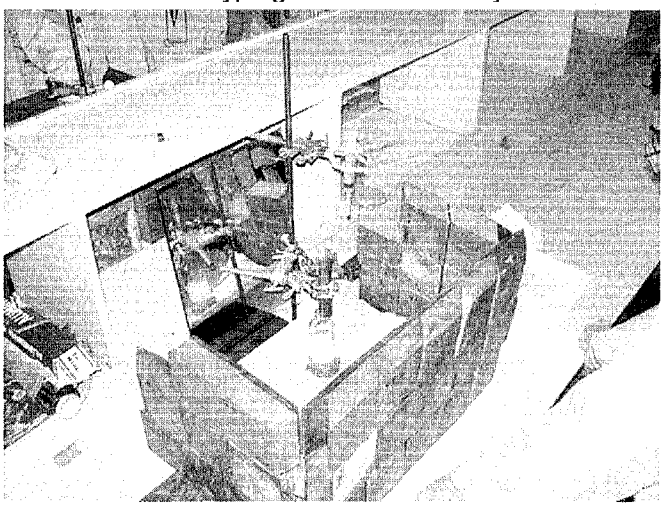


Dr. Machi and TCG members

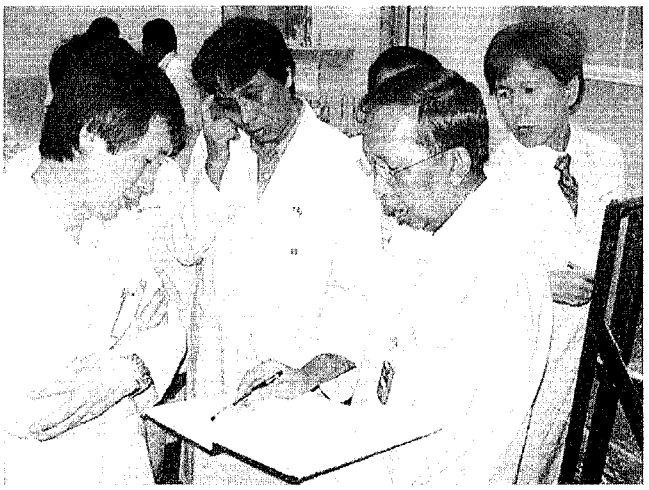


Laboratory for demonstration

PZC type generator assembly

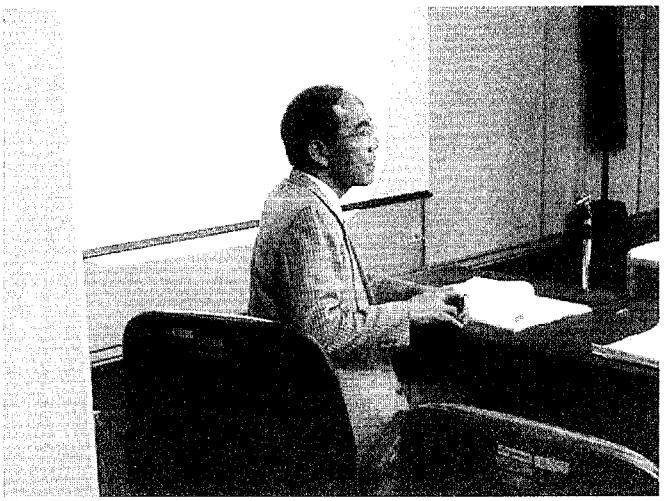


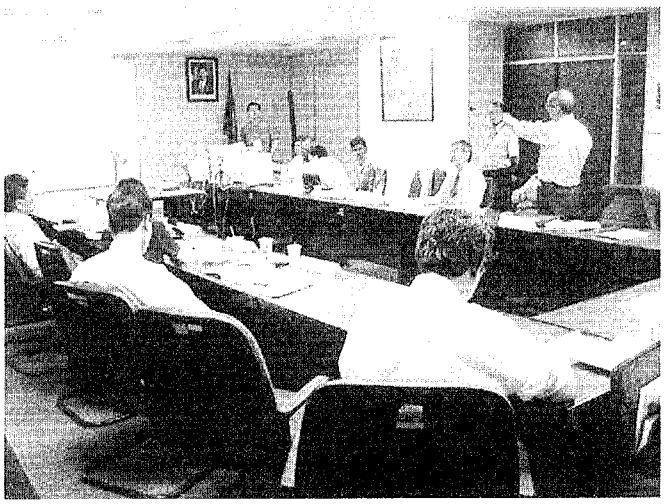
Discussion during the experiment



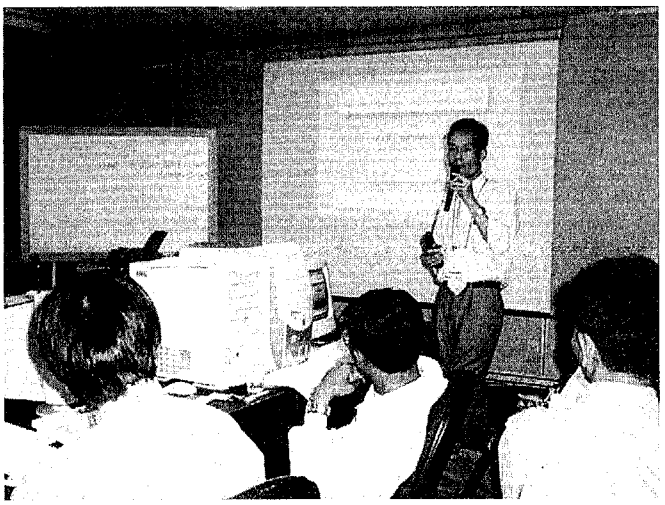
4. NAA Session January 14-16, PRSG Building, BATAN

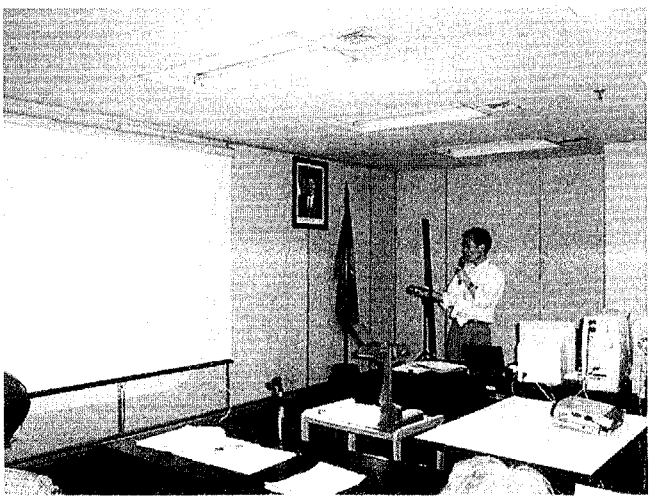
Prof. Ebihara (Tokyo Metropolitan Univ., Japan)





Mr. Ho Manh Dung (NRI, Vietnam)

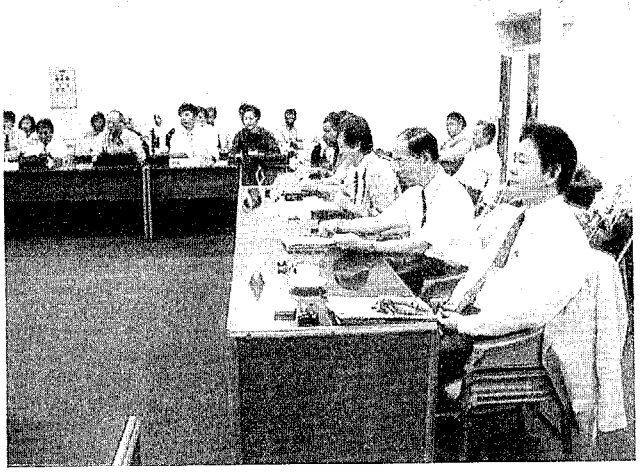




5. Closing Session

Round Table Discussion & Closing

Dr. Hastowo, Dr. Machi and Dr. Genka



Prof. Ebihara, Mr. Tachikawa (Japan)

Mr. Kuntoro, Dr. Mutalib (Indonesia)

Representatives

Prof. Ni, Mr. Yuan, Dr. Liu (China)

Dr. Chung, Mr. Han (Korea)

Dr. Hashim, Dr. Ab. Khalik (Malaysia)

Ms. Santos, Ms. Sombrito (The Philippines)

Mr. Charoen, Dr. Chueinta (Thailand)

Prof. Le, Mr. Ho (Vietnam)

JAERI－Conf 2004－010

Proceedings of the FNCA 2002 Workshop on the Utilization of Research Reactors

January 13 – 17, 2003

Jakarta & Serpong, Indonesia

(Contract Research)

Nuclear Technology and Education Center

(Tokai Site)

Japan Atomic Energy Research Institute

Tokai-mura, Naka-gun, Ibaraki-ken

(Received April 16, 2004)

The FNCA 2002 Workshop on the Utilization of Research Reactors, which is the eleventh workshop on the theme of research reactor utilization, was held in Jakarta and Serpong, Indonesia from January 13 to 17. This workshop was executed based on the agreement in the third Coordinator's Meeting of Forum for Nuclear Cooperation in Asia (FNCA) held in Tokyo, March 2002.

The workshop consists of two groups under the theme of the following fields; 1) Tc-99m Generator Technology, 2) Neutron Activation Analysis. The total number of participants for the workshop was 88 people from 8 countries; China, Indonesia, Korea, Malaysia, the Philippines, Thailand, Vietnam and Japan.

This report consists of 8 papers from the plenary session, 10 papers for Tc-99m Generator, 10 Papers for Neutron Activation Analysis and a summary report.

Keywords: Utilization, Research Reactor, Tc Generator, Neutron Activation Analysis, and FNCA

This Workshop was sponsored by the Ministry of Education, Culture, Sports, Science and Technology (MEXT) and the National Nuclear Energy Agency (BATAN), and the Japan Atomic Energy Research Institute (JAERI) had been assigned by MEXT as the responsible organization for holding the Workshop. This report includes the results carried out under the contract between JAERI and MEXT under the auspices of the special account law for electric power development promotion.

JAERI－Conf 2004－010

FNCA 2002 年度　研究炉利用ワークショップ　論文集

2003 年 1 月 13 日～17 日

ジャカルタ・スルポン　インドネシア

（受託調査）

日本原子力研究所

国際原子力総合技術センター

（2004 年 4 月 16 日受理）

研究炉利用ワークショップは、2002 年 3 月に東京で開催されたアジア原子力協力フォーラム第 3 回コーディネータ会合での合意に基づいて、文部科学省からの受託として 2003 年 1 月 13 日から 17 日まで、インドネシアのジャカルタ及びスルポンで開催されたものである。

Tc-99m ジェネレータ技術及び中性子放射化分析の 2 つのテーマについてワークショップを開催し、中国、インドネシア、韓国、マレーシア、フィリピン、タイ、ベトナム及び日本の 8 か国から 88 名が参加した。

本論文集は、8 編の全体会議からの報告、10 編の Tc-99m ジェネレータ分野の報告、10 編の中性子放射化分析分野の報告及び 1 編のサマリー報告を収録したものである。

本ワークショップは、文部科学省及びインドネシア原子力庁の共催として、文部科学省から原研が受託して実施したものである。この論文集は、電源開発促進対策特別会計法に基づき、文部科学省から受託して行なった調査の結果を含む。

日本原子力研究所(東海駐在)：〒319-1195 茨城県那珂郡東海村白方白根 2-4

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1. Workshop

Plenary Session

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1.1 NUCLEAR TECHNOLOGY FOR SUSTAINABLE DEVELOPMENT AND FNCA ACTIVITIES

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Abstract

Nuclear techniques have been contributing to sustainable development and human welfare through their applications in agriculture, health care, food supply, industry, water resources and environmental conservation. Nuclear techniques are more advantageous and/or complementary with other techniques to achieve goals. For many applications nuclear technique is more environmentally friendly because it does not need chemical agents to induce necessary reactions.

This paper also illustrates successful applications of nuclear techniques and activities of the regional nuclear cooperation in Asia, FNCA (Forum for Nuclear Cooperation in Asia) to achieve common goals with limited resources.

1. RADIATION TECHNOLOGY FOR ENVIRONMENTAL PROTECTION

(1) Cleaning flue gases by electron beams

The emission of SO_2 and NO_x into the atmosphere from coal and oil burning power plants and industrial plants is one of the major sources of environmental pollution. These pollutants cause "acid rain" damaging forests, lakes, and even historical buildings.

Innovative technology using electron beams to simultaneously remove SO_2 and NO_x by irradiation was first developed in Japan and further studied by research groups in, Germany, Poland, China and Bulgaria. The flue gas is exposed to electron beams of 5~10kGy while it passes through an irradiation chamber. A small fraction of gaseous ammonia is injected into the chamber. As a consequence of reactions induced by radiation, SO_2 and NO_x are converted into a mixture of ammonium sulphate and nitrate particulates which can be separated from cleaned gases by standard techniques of electrostatic precipitators. These by-products are used as a fertilizer for agriculture.

The advantages of this technology over conventional processes for treating flue gases are:

- It is the only process to simultaneously remove SO_2 (90%) and NO_x (85%).
- The by-product of the process can be used as agricultural fertilizer.
- The process does not require large amounts of water.
- It can meet the stringent requirements for removal efficiency of SO_2 and NO_x.

After extensive studies using pilot scale plants in Japan and Poland with promising results in terms of technology and process costs, the industrial demonstration plant to clean flue gases of 270,000 Nm^3/hour from a power station in Poland has started operation in December 2000 with the IAEA assistance. Also in China an industrial demonstration plant to remove SO_2 in flue gas from a coal burning power station has been in operation for 3 years and the 2nd plant is under construction. In Japan a demonstration plant to clean heavy oil burning flue gas of power station of 220MWe is about to start operation in Chubu Power Co. (photo. 1).

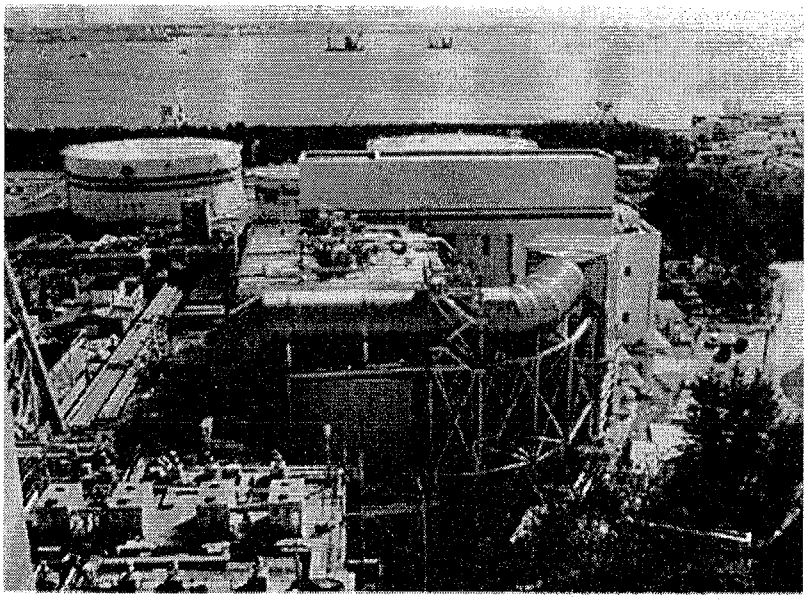

Photo1: Industrial plant to clean flue gases from heavy oil burning power station using electron beams by the Chubu Power Co. in Japan

Economic feasibility studies of this electron beam process by the IAEA and Japanese experts have shown that this technology is more cost effective than the conventional process in terms of both initial investment and operational cost. It should be noted that the conventional process, which involves the combination of catalytic reduction of NO_x and neutralization of SO_2 by lime-stone process, produces gypsum as a by-product, which is not useful in some countries. By contrast, the by-product of the electron beam process is a valuable fertilizer.

(2) Removing dioxin from incineration plants by electron beams

In 2001 the Japan Atomic Energy Research Institute (JAERI) has started operation of pilot plant to remove dioxin from flue gas of municipal incineration plant. In February 2002, it was announced that 90% of dioxin can be removed from the gas at 15kGy. An accelerator of 0.3 MeV•40mA is used to treat flue gas 1,000 N m^3/h from the incineration plant of city of Takasaki containing 1-5 ng/ m^3 of dioxin.

(3) Cleaning industrial waste water

In Korea a commercial company has been extensively studying treatment of wastewater from dye factories. An electron accelerator of 1.0 MeV•40mA is used to clean wastewater of 1,000m^3/day. Based on positive results of feasibility study, an industrial scale plant of 10,000 m^3/day has been proposed by EB Tech. Co. to the Korean Government and IAEA for financial support. The Brazilian Nuclear Energy Research Institute has been also working for cleaning wastewater and flue gases.

2. ENVIRONMENTALLY FRIENDLY AND PRODUCTIVE AGRICULTURE

Securing food for large population is an important challenge. Nuclear applications encompass mutation breeding, soil fertility and crop production, animal production, food safety, insect pest control, which contribute to sustainable agriculture and food supply.

(1) Plant mutation breeding

The radiation-induced changes in DNA sequences have resulted improved mutant crops demonstrating 1) disease resistance, 2) early maturity, 3) drought tolerance, and 4) better yield. Over the past 60 years, 1,800 new mutant plant varieties induced by radiation have been officially released and are now growing on millions of hectares of land.

In Japan a mutant variety of a pear was developed showing an excellent resistance against "black spot disease". Therefore, amount of pesticide has been decreased to one fifth, which brings about environmental benefit and economical profit. At the FAO/IAEA Laboratories mutant varieties of banana which are more resistant to plant disease (Fuzarium) and higher yield are being developed by mutation in combination with tissue culture techniques.

Examples of significant economical achievements of mutant varieties are: the better quality rice in China, the high yield cotton in Pakistan, the better quality barley for beer production in Central Europe, and disease resistant pear in Japan.

The FNCA is implementing the project on "Mutation Breeding of Drought Resistant Soybeans and Sorghum" and "Mutation Breeding on Insect Resistant Orchid".

(2) Food irradiation for better use of foods

Irradiation reduces food spoilage, improves food hygiene and extends shelf life. Radiation can kill insect pest of fruits and vegetable for quarantine regulation, replacing methyl bromide fumigation which deplete the ozone layer and will be phased out in the near future.

In the United States, food derived pathogenic bacteria such as Salmonella, E.Coli, Listeria, Campylobacter, Vibrios, Trichinella and other parasites claim estimated 5,000

lives annually and between 24 and 81 million cases of diarrhea of various kinds. Food irradiation is increasingly recognized by health authorities as a means of countering this health problem. In 2002, more than 2000 supermarkets in the USA are selling irradiated ground beef for hamburgers. (Photo 2)

Photo. 2 Irradiated ground beef for hamburgers sold in more than 2000 supermarkets in USA

More than seventy thousands tons of species have been on the commercial market in 35 countries. In Vietnam with the assistance of IAEA, an irradiator of 400kCi Co-60 recently started operation being fully utilized for irradiation of frozen shrimp and other foods. In China about 100,000 tons of foods such as garlic, dried vegetables, and spices are irradiated per year.

(3) Sterile insect technique (SIT) using radiation for control of insect pests replacing pesticide

Conservative estimates indicate that insect pests reduce world food production by 25% to 35%, in spite of enormous amounts of pesticide applications of about US$ 25 billion annually. This heavy reliance on pesticides brings about serious environmental pollution. To replace insecticide a nuclear technique, so-called sterile insect technique (SIT), has been successfully used against major insect pests in several countries.

Radiation can sexually sterilize insects. When mass-reared and sterilized male insects are released into the wild population, the female insects mate with sterilized males not to produce offspring. The Substantial reduction of fertile matings causes a fall in populations that eventually leads to eradication. The SIT has proved highly effective against several key insect pests such as, fruit flies, new world screwworm fly, tsetse flies, and lepidoptera. The SIT has been used successfully to eradicate or control the medfly in several countries such as Mexico, Guatemala, U.S.A., Chile, Argentina and Peru with substantial economic and environmental benefits. In Okinawa of Japan the melon fly has been eradicated in 1990 producing large economic benefits. Photo 3 shows specially designed radiation

facility to irradiate pupae at homogeneous dose.

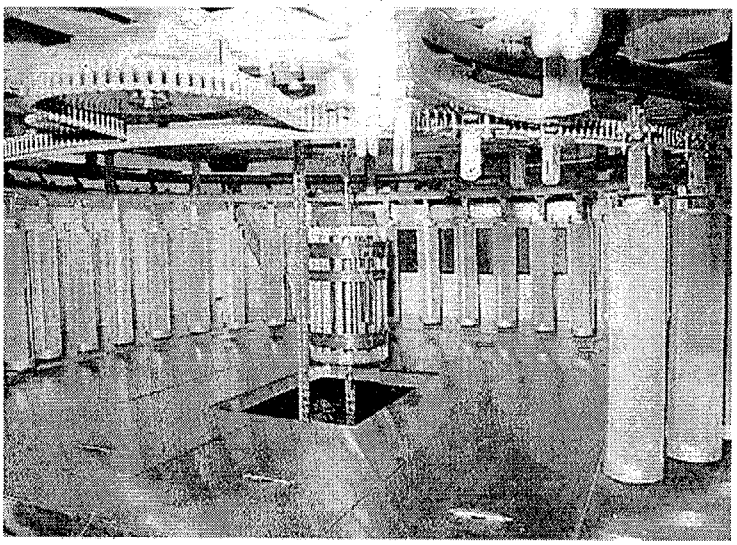

Photo 3 Radiation facility of pupae of melon fly using Co-60 in Okinawa, Japan

Pilot SIT projects to suppress rather than eradiate fruit fly pest as an insecticide replacement have been implemented in the Philippines, Thailand, and the border of Israel and Jordan to have confirmed the feasibility of SIT.

The IAEA has achieved the eradication of the tsetse fly from Zanzibar in Tanzania. Tsetse flies transmit trypanosomiasis, a disease which debilitates animals and causes sleeping sickness in humans. This disease is a major limiting factor in agriculture development for many African countries. A larger project of tsetse fly control and eradication is in progress in Ethiopia by the government and the IAEA.

The economic benefits from SIT are remarkable. In Mexico an annual cost of SIT operation is about $10 million to protect fruits and vegetable export markets of over $1 billion/year.

3. IMPROVED INDUSTRIAL PROCESSES AND UNIQUE PRODUCTS

Applications of radioisotopes and radiation in modern industry is of great importance for process improvements and product quality control bringing about energy and material saving. The major areas of application of isotopes and radiation in industry are as follows:

- Radiation processing for manufacturing
- Non-destructive testing by radiography for quality control
- Nucleonic control systems for process control and optimization
- Tracer technology for process optimization and trouble-shooting

This chapter focuses on the application of radiation processing which rapidly being adopted in developing countries.

(1) Heat and chemical resistant polymers produced by radiation cross-linking

Radiation can induce desired chemical reactions without using catalyst at room temperature. One of the advantages of radiation processing is savings in energy consumption. Some polymers cross-linked by radiation can be tailored to shrink when heated. This material is widely used for packaging, wire insulation and corrosion protection of pipeline welds. Wire industries are extensively using electron-beam to produce heat resistant insulation of wire.

Radiation cross-linking of polymers such as polyethylene, polyvinyl chloride and rubber improves their thermal resistance, chemical resistance and mechanical strength. Examples of commercial products manufactured by radiation cross-linking are shown in Table I.

Table I. Commercially produced cross-linked polymers

Products	Applications
Cross-linked polyethylene and PVC	Wire insulation resistant to heat and chemicals, pipes for heating systems
Foamed polyethylene	Insulation, packing, floating materials
Heat shrinkable tubings and sheets	Food packaging, insulation, protection against corrosion
Cross-linked rubber sheets	Automobile tires (high quality), roof protection sheets (weather resistant)
Cross-linked polyurethane (Japan)	Cable insulation for antilock brake sensor
Cross-linked nylon (Japan)	Automobile parts resistant to heat and chemicals
Super heat resistant SiC fibre (Japan)	Metal and ceramic composites, semi-commercial plant
Vulcanized natural rubber latex (Malaysia)	Surgical gloves, condoms

Cross-linked heat resistant polymer insulating wire is used in the automobile industry, telecommunications, and the aerospace industry and in home electrical appliances. In the automobile tire industry, radiation processing is used to cross-link rubber molecules to improve physical properties for the production of radial tires, in Japan, France, USA, Brazil, Korea, and recently in Indonesia.

(2) Surface coating without using organic solvents

Another environmentally friendly application of radiation processing is the curing of surface coatings of different products, such as wood panels, adhesive tapes, and surface coats for printing, floppy discs, and decorative steel plates. A major advantage of electron beam curing is that no organic solvent vapour is emitted into the environment by evaporation. The excellent quality of surface cured by electron beam has been proven. More than 400 low cost and low energy electron accelerators for surface coating are used through the world.

(3) New applications

New applications of radiation processing of polymers are show in Table II.

Table II. New Polymers by radiation in horizon

(1)	Hydrogel crosslinked by radiation being used for wound dressing
(2)	Adding values on natural polymers such as carrageenan, chitosane, alginates and silk
(3)	Crosslinking of PTFE at high temperature to improve mechanical properties such as abrasion resistance
(4)	Absorbent production by radiation grafting for selective absorption of uranium for its recovery from sea water

(4) Industrial radiation sources

Accelerators and Co-60 sources are major radiation sources for industrial applications. There are approximately 200 Co-60 gamma irradiators and 1,500 electron beam accelerators used mainly for industrial purposes throughout the world. Co-60 sources are commonly used for sterilization of medical products and foods, while electron accelerators for industrial processing.

4. RADIOTHERAPY OF CANCER

(1) Teletherapy

The teletherapy has tended towards the use of linear accelerators generating electrons and X-ray up to 25MeV, capable of reaching deep-seated tumours. About 2000 Co-60 machine and 6000 accelerators are in use worldwide.

New technique challenging cancer is particle therapy using proton and heavy ion which can avoid side effect by sharply focusing beam in tumor and give efficient damage to it. Heavy particle beam can be focused in area and depth and have high and biological

effects. Treatment by heavy ion beam such as C-14 has shown excellent recovering rate in about 1,300 clinical tests demonstrating benefits for patients of bone, liver and lung cancer at the National Institute of Radiological Sciences in Japan. In Japan there are 5 proton and 2 heavy -ion facilities for radiotherapy in operation.

(2) Brachytherapy using micro source

By using micro-size (1mm in diameter) Ir-192 sources with high specific activity, radiation source can be inserted into tumors and treatment time have been reduced to only 10-20 minutes instead of 10-20 hours. Procedures for insertion are easier and new sites such as bronchi of the lungs, bile ducts and small heart vessels are accessible.

There is large difference in number of teletherapy machines by the regions. In developing countries number of machines is in the range of 0.1 to 0.7 per million population while that in developed countries is in the range of 3.9 to 8.2.

5. REGIONAL COOPERATION IN ASIA FOR NUCLEAR APPLICATIONS

Asian region has large population and needs increasing amounts of energy, water and foods. Poverty is still common problem to be urgently solved. In this context the regional cooperation has been carried out through IAEA/RCA and FNCA (Forum for Nuclear Cooperation in Asia) to contribute the enhancement of the sustainable development.

(1) The FNCA has synergy with RCA/IAEA

For FNCA current activities are: 1)mutation breeding, 2)research reactor application (Tc-99m generator by PZC method, NAA for environmental protection, SANS for polymers), 3)radiotherapy for cervix uterine cancer, 4)waste management, 5)safety culture for research reactor, 6)nuclear public information, 7)low energy electron beam application, 8)bio-fertilizer application, 9)human resource development.

The FNCA aims to achieve tangible outcomes which have socio-economic impact meeting needs of society. Partnership is an important policy of the FNCA to sustain cooperation.

(2) Scientist exchange programme in Asia funded by Japan

Building R&D capacity is an important infrastructure for development, which needs human resource development (HRD). To enhance HRD the international cooperation can play significant role to be integrated with national training and education programme.

The Government of Japan (MEXT) has been implementing the "Scientist exchange programme in Asia for Nuclear Science and Technology" for 16 years. More than 1200 scientists from developing countries have been invited to Japan gaining research and technology experience.

6. Conclusion

The application of radiation and isotopes has been extended in developing countries
to contribute for the improvement of agriculture, industry, medical care and environmental protection.

Nuclear technique such as radiation application can provide environmentally friendly processes, which should be more widely used in the future.

The FNCA, the regional cooperation mechanism contributing to the promotion of nuclear application, should be further enhanced in the coming yesrs.

1.2 Neutron Activation Analysis as Analytical Tool of Environmental Issue

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Abstract

Neutron activation analysis (NAA) is applicable to the sample of wide range of research fields, such as material science, biology, geochemistry and so on. However, respecting the advantages of NAA, a sample with small amounts or a precious sample is the most suitable samples for NAA, because NAA is capable of trace analysis and non-destructive determination. In this paper, among these fields, NAA of atmospheric particulate matter (PM) sample is discussed emphasized on the use of obtained data as an analytical tool of environmental issue.

Concentration of PM in air is usually very low, and it is not easy to get vast amount of sample even using a high volume air sampling devise. Therefore, high sensitive NAA is suitable to determine elements in PM samples. Main components of PM is crust oriented silicate, and so on in rural/remote area, and carbonic materials and heavy metals are concentrated in PM in urban area, because of automobile exhaust and other anthropogenic emission source. Elemental pattern of PM reflects a condition of air around the monitoring site.

Trends of air pollution can be traced by periodical monitoring of PM by NAA method. Elemental concentrations in air change by season. For example, crustal elements increase in dry season, and sea salts components increase their concentration when wind direction from sea is dominant. Elements that emitted from anthropogenic sources are mainly contained in fine portion of PM, and increase their concentration during winter season, when emission from heating system is high and air is stable.

For further analysis and understanding of environmental issues, indicator elements for various emission sources, and elemental concentration ratios of some environmental samples and source portion assignment techniques are useful.

Introduction

NAA is known as one of the most sensitive analytical method for multi-elements. In these years, even rapid development of comparative analytical methods like ICP/MS (Inductively Coupled Plasma/Mass Spectrometry) and ICP/AES (Inductively Coupled Plasma/Atomic Emission Spectrometry), NAA has still its power. For instance, Japan Association of Activation Analysis, established in 1995 consists more than 180 research members of various fields and publishes journal twice a year. In its latest journal[1], more than 10 newly published theses on activation analysis including agricultural science, biology, geochemistry, cosmochemistry, and environmental science were introduced. Use of NAA is not rapidly increasing, but the use of NAA contributes to the development of various fields of sciences.

As reported in the 2001 Workshop on the Utilization of Research Reactors, basic advantage of NAA to a small amount sample or a precious sample like geological samples in space is clear[2], because NAA is capable of trace analysis and non-destructive determination. In this workshop, researchers of East Asian countries reported their activities on activation analysis on mainly for environmental sample like PM, river water and sediment samples. After the intensive discussion, the NAA group of the workshop decided PM sample as a common sample for NAA in 2002.

In this paper, NAA method applied to PM sample is discussed focused on its obtained data as an analytical tool of environmental issue.

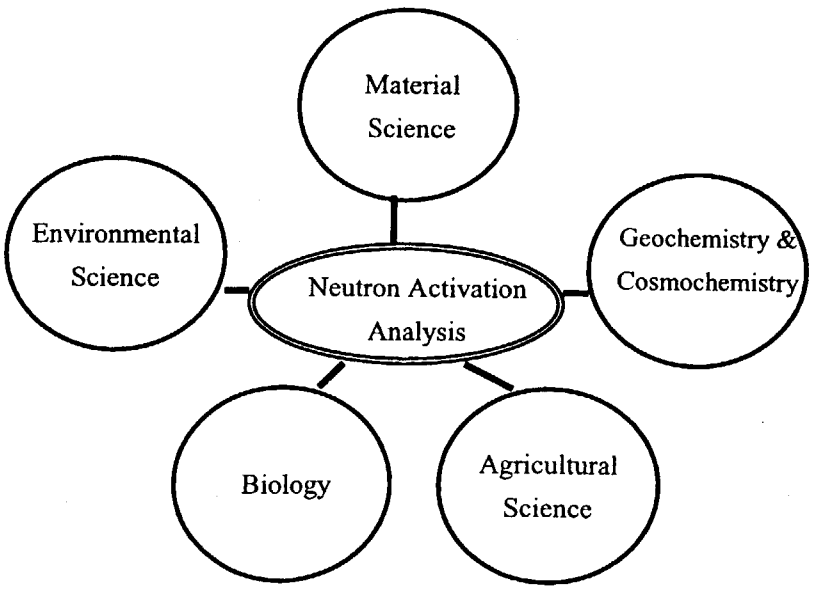

Fig.1 Example of applicable research field of neutron activation analysis

PM and NAA

-Elemental pattern

The National Air Sampling Network (NASN) of Japan employed a procedure for nondestructive instrumental neutron activation analysis (INAA) of PM sample for up to 30 elements[3]. The method could perform excellent sensitivity for multi-elements by combination of high density neutron fluxes of the atomic reactor and high resolution semiconductor detectors without chemical treatment of the sample. Example of analytical scheme of INAA is shown in Table1. Other important elements, such as Pb, Cd and so on which has relatively low analytical sensitivity of INAA were determined by X-ray fluorescence method.

Atmospheric concentration of PM and element are quite varied among sites in large/industrialized cities, sites in small/rural cities, and sites located in remote area. As shown in Fig.2, elements such a Cr, Fe, and Ni, which has anthropogenic emission sources in Kawasaki (typical industrialized city) was higher concentration than that of Sapporo (rural) and Nopporo (remote).

Table1 Example of analytical scheme of PM by INAA

Half life of nuclides	Elements	Neutron Irradiation	Cooling time	Measuring time
Short	Al, Br, Ca, Cl, Cu, Mn, Ti, V	1-3 min	3-4 min	300-500 sec
Medium	As, K, La, Na, Sb, Sm, W	5-6 hours	2-3 days	1,000 sec
Long	Ag, Ba, Ce, Co, Cr, Cs, Fe, Hf, Lu, Sc, Se, Th, Zn	5-6 hours	2-3 weeks	4,000 sec

-Trends and Seasonal variation

Trends of air pollution can be traced by periodical monitoring of PM by NAA method. In a case of Japanese NASN, some toxic elements emitted from anthropogenic sources in large industrialized city decreased in 1980's, because of improvement of emission sources and fuel conversion from coal to other energy sources. However, in some small and medium size cities, such kind of trend couldn't be seen, and elemental concentrations are stable and low (Fig.3).

Not so much systematically as seasonal pattern of CO_2 concentration in air, elemental concentrations in air also change by season. For example, concentrations of crustal elements increase in dry season and when encountering to sand storm far

from continents. On the other hand, sea salt components increase their concentration particularly a site near the coast when wind direction from sea is dominant. Elements which emitted from anthropogenic sources are mainly contained in fine portion of PM. These elements sometimes behave like gaseous pollutants and increase their concentration during winter season, when emission from heating system is high and air is stable.

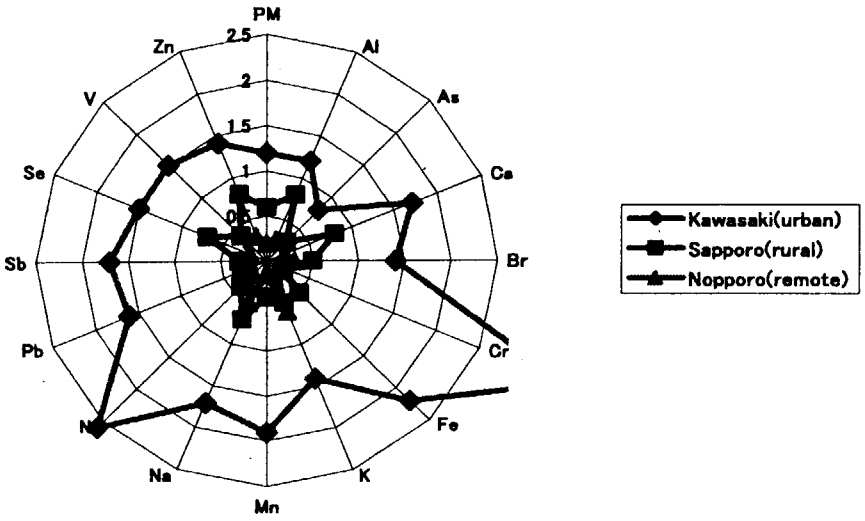

Fig.2 Elemental pattern (relative average concentrations) in several NASN sites (1996)

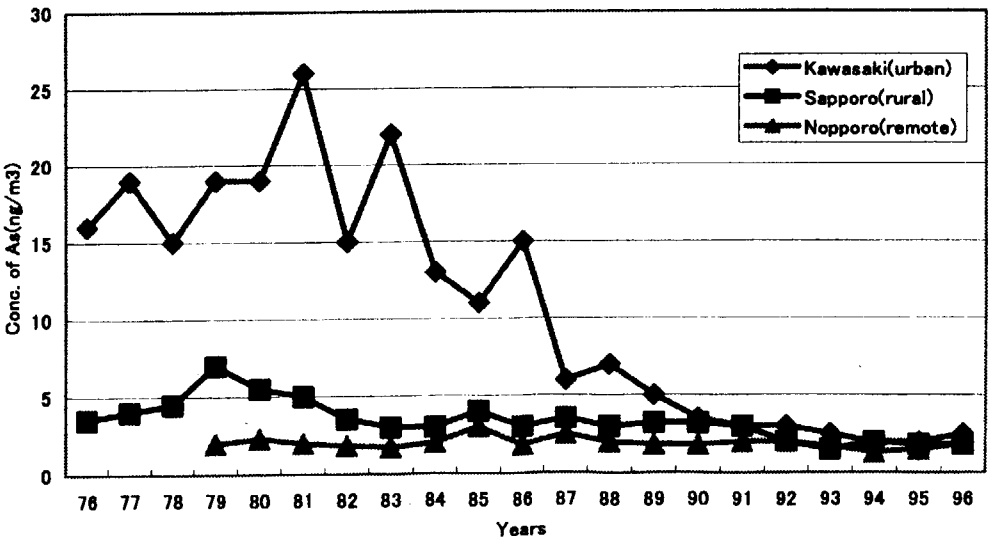

Fig.3. Annual average concentration of As in several NASN sites (1976-1996)

-Indicator elements

In Fig.4, a monthly variation of Total Suspended Particulate (TSP) concentration in Beijing was shown from Dec.1988 through Feb.1990, ranging from 170 to 420 microgram/m^3 [4]. Higher concentrations were generally observed in spring and winter. A monthly variation of Al, also plotted in Fig.4 showed a good agreement with that of TSP and higher correlations were found for TSP with other crustal elements. Then the influence of soil dust on the TSP concentration was discussed using Al as an indicator element of soil, with a hypothesis that all ambient Al originates from ground soil.

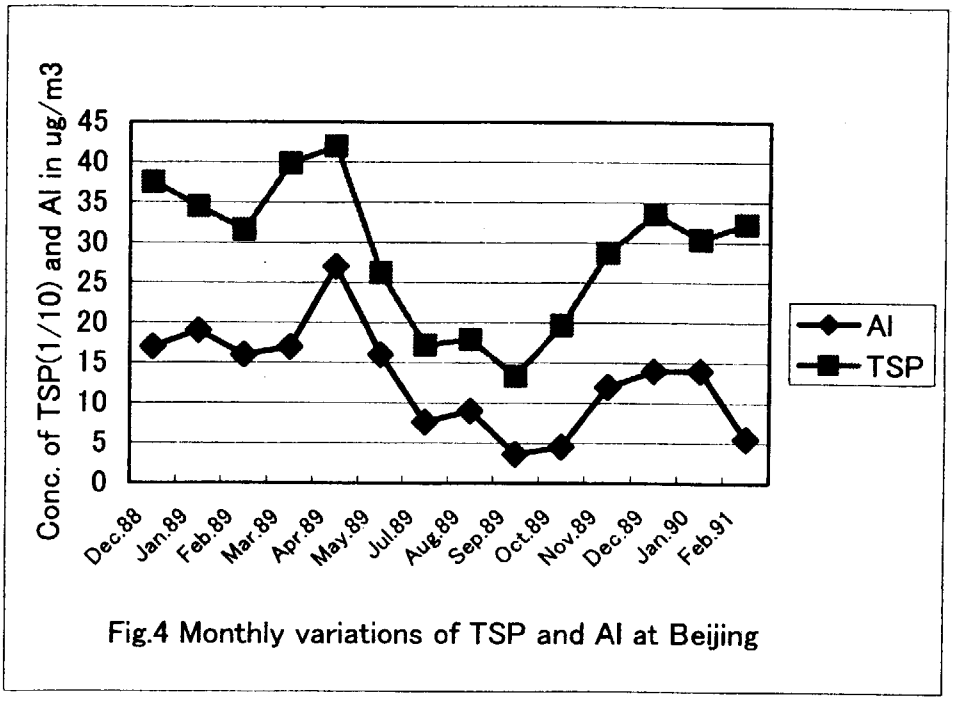

Fig.4 Monthly variations of TSP and Al at Beijing

One of the advantages of NAA is high detection sensitivity of rare earth elements. In the catalytic cracking process for the production of high octane gasoline, zeolite is used as a catalyst, and some zeolite contains considerable amounts of rare earth. Olmetz and Gordon[5] found that the La/Sm concentration ratio in the fine portion of atmospheric particles is higher than in the Earth's crust (La/Sm =5). They suggested the possibility of using the elemental concentration ratio of La/Sm as an indicator of oil industrial or automobile exhaust emissions. As shown in Fig.5 and Fig.6, the moving averages of La/Sm concentration ratios and Pb concentration for four years in Seoul indicates the increase of La/Sm and relationship with Pb concentration (r^2= 0.606)[6]. La/Sm concentration ratio seem to increase with the use of unleaded gasoline.

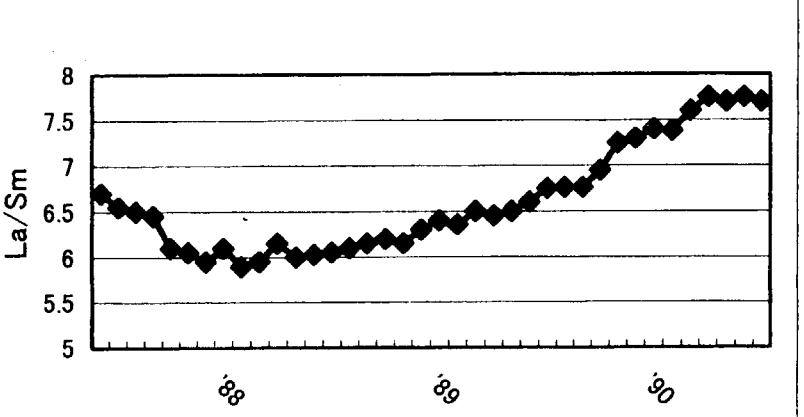

Fig5. Change of La/Sm ratio in Seoul

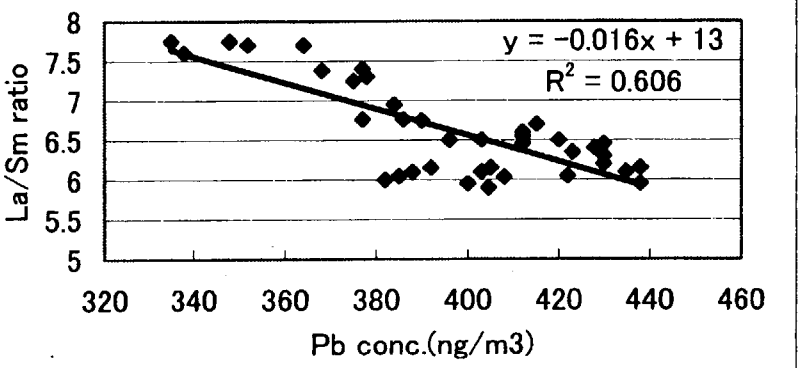

Fig.6 La/Sm ratio and Pb conc. in Seoul

-Source portion assignment

The Chemical Mass Balance (CMB) method has long been used for the emission source apportionment. This method is applied to the data obtained at monitoring site by following equation.

$Ci = Mj \cdot Xij \cdot \alpha ij$

Ci : Atmosperic concentration of element i (ng/m^3)

Mj: Atmosperic concentration of PM at receptor site released from emission source j (ng/m^3)

Xij : concentration of element i in a source emission j (g/g)

α ij: Coefficient of fractionation of element i (=1)

Contributions of each emission source are calculated by solving simultaneous equations set up by indicator elements for each emission source. Hashimoto et al. estimated contribution of 6 emission sources (soil, fuel-oil combustion, gasoline automobile, iron & steel industry, incinerated dust, and marine aerosol) in Seoul[7]. Hien et al. applied principal component factor analysis (PCFA) to evaluate aerosol source of Ho Chi Minh City[8]. These sources include vehicle emissions (Br and Zn), coal burning (Se), industrial process (Ce, Co, Cr, Pb and Sb), road dust (Al, Ti and V), soil dust (Fe and Th), biomass burning (K), marine aerosols (Na and Cl) and mineral fly ash (Sc and La). These source portion assignment study combined with size fractionated PM collection bring us more informative data for consideration of future mitigation measures.

Conclusion

Advantage of NAA and its application to the environmental issue is discussed. Among several comparative analytical methods, NAA is still useful particularly for small sized and precious samples. Various kinds of information can be obtained from the analytical results of multi-element of PM by NAA. It consists of situation around the site and the contribution of natural and anthropogenic emission sources at the site.

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1.3 The Current Status of Utilization of Research Reactors in China

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Abstract

Seminars on utilization of research reactors were held to enhance experience exchanging among institutes and universities in China. The status of CARR project is briefly described. The progress in BNCT program in China is introduced.

A brief overview on utilization of research reactors in China has been given in [1]. The main research reactors in China conduct most of utilization on research reactor although many of them are facing the aging problems.

The newly designed and under constructed research reactor, China advanced Research Reactor (CARR), is expected to take over the utilization tasks for the aged reactors in CIAE. In order to achieve this aim, we have to make our efforts in preparedness on utilization of research reactor, including all resources in technology, manpower, instrumentation, and budget, so as to make the reactor to be used in an effective way once it is completed.

In this paper, four aspects will be dressed about the related status of utilization of research reactors in China. They are: the status of CARR construction, the intention utilization on CARR, exchange on utilization of research reactors in China and the recent progress in BNCT program in China.

1. Status of CARR project

Having finished the reviewing work of the preliminary safety analysis report and environmental impact statement, the first construction license for the CARR project has been granted and the construction work with its first concrete pouring started on August 26, 2002. According to the schedule, the project will be completed within a total duration of 52 months.

Up to now, the irradiation test of single small plates of U_3Si_2-Al fuel and

then irradiation test of a fuel assembly have been finished in the MIR reactor of the Research Institute of Atomic Reactors, Russia. The metallographic inspection work is under way now. The results obtained are satisfactory and show that the performance of the CARR fuel design can meet the requirements of the conditions in CARR.

Some important evaluation and verification tests, such as the integrated reactor core flow-induced vibrations, the critical heat flux, flow distribution and flushing tests of the fuel assembly, critical velocity of the coolant are being conducted or under plan.

Now, seeking the vendors of a variety of devices and materials, the bidding for the design and fabrication of some subsystems is processing in due course.

Worth of mentioning is that the operation mode of the emergency ventilation for some severe accidents has been changed in the CARR design according to the safety review by the regulatory body. During an accident with radioactivity release, the emergency ventilation system will be completely switched off. The radioactivity is being held in the confinement building for a proper period of time. During this period, no venting flow via the stack and only a very low release of radioactivity corresponding to the leakage rate of the confinement building, say, 2.5 vol.% per day, is adopted. So, the design of very low leakage rate of the confinement building is the key point.

It is expected that the civilization construction work will be finished this year, 2003, and in 2004 and 2005 installation job will be completed then from the late 2005 to year 2006, the commissioning work will be conducted, and the power operation will start at the end of 2006.

2. Utilizations intended on CARR

In order to make the CARR to be effectively used once it is completed, many jobs have to be conducted in advance. A special symposium was organized and chaired by the president of CIAE last September. There are 17 papers, involving fundamental research, utilization of nuclear energy, radioisotopes production, science and technology in variety of fields, are presented on the meeting. Almost all utilizations are involved, such as neutron scattering research in life science and material science, preparing and their

application technology of transuranic elements, the application in health physics, development of new and high performance fuel, radioisotopes production including high purity ^{125}I, fission ^{99}Mo, ^{238}Pu and NTD silicon, nuclear data measurement for long life nuclides, on line isotope separating (ISOL), etc. It is emphasized that these utilizations on CARR should be better conducted through domestic and international collaboration and cooperation.

3. Enhancing experience exchanging in China

On aware of the importance of experience exchanging to promote the further utilization of limited research reactors resources, we organized a domestic symposium this year composed of experts from variety of Institutions or universities over China, which runs research reactor or nuclear facilities. They get together to present and discuss what they are doing and going to do in the area of utilization of research reactors. We found that the symposium is valuable for participants in their research work by the information and experience exchanging. So, such kinds of meetings or activities are asked to be periodically held later on by the participants. Some relevant organization to promote the experience exchange in utilization of research reactors will be set up in China.

4. Recent Progress in BNCT Program

Having passed 10 years of pacing up and down, the development of BNCT program in China rises again in Beijing, now headed by Dr. Zhou Yongmao, member of the Chinese Academy Engineering (CAE). The aim of the BNCT program is on the real treatment. The China's neurosurgery has got good foundation and relevant facilities. With a full figure of brain cancer patients in China, our first starting point for developing BNCT program is set to the treatment for brain glioblastoma at superficial position following the regularity of success or failure with long period international BNCT research, especially, drawing the achievements of clinical trial got by Japanese experts. The irradiation facility is a miniature neutron source reactor, which possesses "user affinity safety nature". The characteristics of its inherent safety have been fully reflected and verified in many such miniature reactors used for neutron

activation analyses in China. The irradiation facility under design provides two neutron beams, one thermal neutron and the other epithermal one. The later one can be available for the treatment for deep position brain cancer patients. The power of the facility is about 30 kW and scheduled to be placed in the basement of Tiantan Hospital of Beijing. Now the design of this facility is underway by the BNCT Research Group CIAE with the scheme comparation calculation, while the calculation of tiny-dose distribution in brain is performed by the Institute of Application Physics and Compute Math and the dose imitation tests are conducted by the China Institute of Radiation Protection. With the Safety limited value to ensure health, the thermal neutron flux at the exit position of thermal neutron beam is about 1×10^{9} n/cm^{2}·sec, while the epithermal neutron flux at the exit point of epithermal neutron beam 5×10^{8} n/cm^{2}·sec. The new funding pattern of state support combined with nongovernmental people is adopted for this program development, if the fund arrives in due time, this facility will be erected and puts into running in the year of 2004-2005.

5. Conclusion

Although many research reactors in China are now facing the aging problems, the utilization of research reactors is still subjected to pay great attention to.

Once the new designed and constructed research reactor CARR is erected, its utilization will certainly be prosperous.

The research and development on BNCT program in China has been paid great attention and highly concerned recent years.

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1.4 Operation and Utilization of Indonesian Research Reactors

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ABSTRACT

For supporting the R&D in nuclear science and technology and its application, BATAN own and operate three research reactors namely, TRIGA-2000 , KARTINI and RSG-GAS having thermal power of 2 MW, 100 kW and 30 MW respectively. The main features , operation and utilization progress of the reactors are described in this report.

1. INTRODUCTION

National Nuclear Energy Agency Indonesia, BATAN owned three research reactors located in three separate region as main tools for conducting the R&D in nuclear science and technology. TRIGA-2000 reactor with the power of 2 MW is operated since 1964 by the Center for Research and Development of Nuclear Technique, P3TKN-BATAN in Bandung, West Jawa. The reactor is originally TRIGA MARK II type, 250 kW and then up graded to 1000kW by replacing the new core and lastly up graded to 2000 kW in the year of 2000.

KARTINI reactor with the power of 100 KW is operated since 1979 by Center for Research and Development of Advanced Technology, P3TM-BATAN located at Yogyakarta. The reactor was built by using the ex- TRIGA MARK-II Bandung. While the Multipurpose Reactor G.A. Siwabessy (RSG-GAS) with the power of 30 MW is operated by Center for Development of Research Reactor Technology, at Serpong Center, Banten Province. All these three reactors have their own specific mission in supporting nation goal of National Nuclear Energy Agency.

2. TRIGA-2000 RESEARCH REACTOR

The reactor was just up-graded and formally inaugurated by the President of Republic of Indonesia in June 2000. Detail information on this reactor is displayed as the followings.

Reactor Name	TRIGA-2000, BANDUNG
Location	Bandung
Owner	National Nuclear Energy Agency
Operator	Center for Research and Development of Nuclear Technique.
Construction started	*01/01/1961*
First criticality	*10/19/1964*
Upgrading started	*01/04/1996*
First Criticality	*24/06/2000*
Status	Operational
Reactor Type	Triga Mark II
Fuel	UZrH
Steady State Power	*2000 kW*
Moderator	H2O, ZrH
Coolant	Light Water
Reflector	Graphite, H_2O.

The reactor is routinely operated by mode of 2 weeks operation and two weeks off. The reactor is mainly applied for radioisotope production, pneumatics systems for NAA, beam tube for science and material study and training. The reactor also serve as a back up of the RSG-GAS reactor whenever RSG-GAS reactor is not available for isotope production.

3. KARTINI RESEARCH REACTOR

The Kartini research reactor is now operating at 100 kW, using up-graded instrumentation and control. Detail information on this reactor is as the followings.

Reactor name	KARTINI
Location	Yogyakarta
Owner	National Nuclear Energy Agency
Operator	Center for Research and Development of Advance Tchnology
Construction started	*04/01/1975*
First criticality	*01/25/1979*
Status	Operational
Reactor Type	Triga Mark II
Steady State Power	100 kW
Fuel	UZrH
Moderator	H_2O, ZrH
Coolant	Light Water
Reflector	Graphite

The reactor is routinely operating at 100 kW, 5 days a week mainly for neutron activation analysis of environmental samples, biological samples as well as ore samples (uranium and thorium) by using delayed neutron counting system and neutron radiography. In the mean time, preparation activities for up-rating the reactor to 250 kW is being conducted.

4. RSG-GAS REACTOR

The reactor core has completely replaced its oxide fuel with silicide, U_3Si_2 Al and preserved its nominal power of 30 MW and average flux of 2×10^{14} n/cm^2 sec. Detail information of RSG-GAS is described as the followings.

Reactor Name	RSG-GAS Reactor
Location	Serpong, Tangerang, Banten
Owner	National Nuclear Energy Agency
Operator	Center for Development of Research Reactor Tecnology
Construction started	01/01/1983
First criticality	*07/29/1987*
Status	Operational
Reactor Type	Pool, multipurpose reactor
Steady State Power	30 MW
Fuel	MTR type, U_3Si_2-Al matrix
Moderator /Coolant	Light Water
Reflector	Be, H_2O

The fuel conversion was started at 36^{th} core by inserting 2 standard U_3Si_2-Al element into the core and followed by 5 standard elements and 1 control element for rest of the cycles until 45^{th} core. In order to fulfill the requirement of Nuclear Control Board for reactor modification some measurements were done to test the performance of the new fuel core. The test included rod drop test, criticality, control rod calibration, low power kinetics parameters, power calorimetric calibration and neutron flux measurement, xenon reactivity measurements, moderator temperature reactivity coefficient, and radiation exposure data as well as operation test at nominal power of 30 MW. The transient test namely loss of force flow was carried out also. The operation test of reactor up to 30 MW showed that all operation parameters are mostly the same as that of the oxide fuel and meet the operation and safety limits. The silicide core configuration is illustrated in Figure 1.

RSG-GAS reactor is operated for about 600 MWD per cycle. Upon optimization among fuel availability, user requirements and efficiency, annual reactor operation program was set-up as illustrated in Figure 2 and 3.

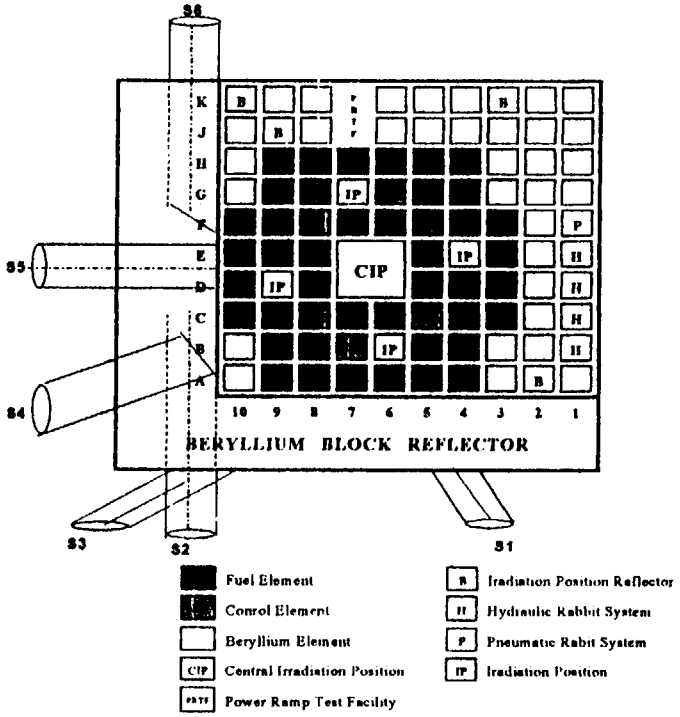

Figure1. RSG-GAS core configuration

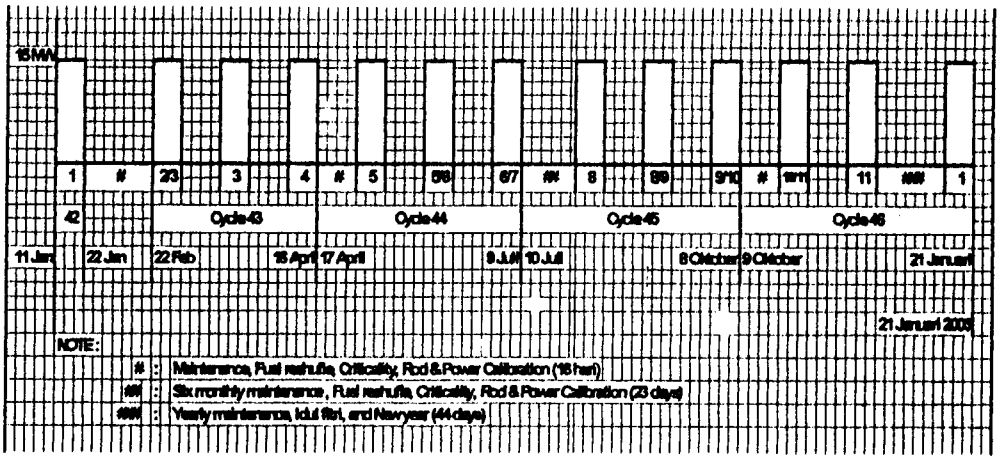

Figure 2. Yearly operation schedule of the RSG-GAS reactor.

The reactor is operated at power level of 15 MW for 4 cycles a year. Each cycle is divided in three phases of operation. Each operation runs for 12 days and shutdown time between two operations is 16 days. By taking into account a couple of weeks to prepare core experiments and preventive maintenance, one cycle lasts about three months.

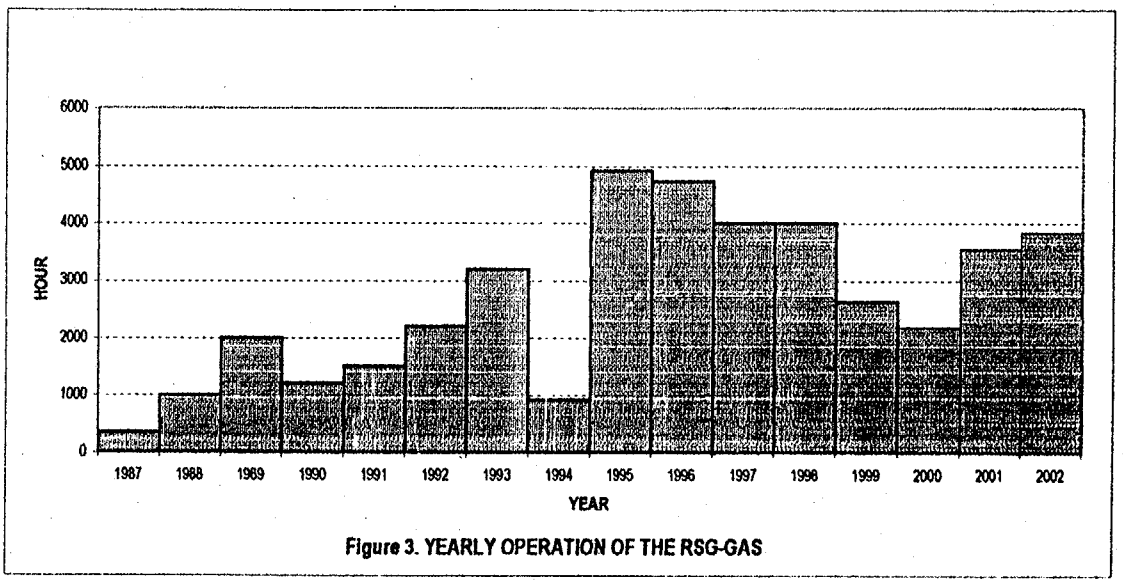

Figure 3. YEARLY OPERATION OF THE RSG-GAS

The RSG-GAS is mainly applied for radioisotope production, research and development of nuclear material and science as well as training and education. The number of samples were irradiated using RSG-GAS can be shown at Figure 4.

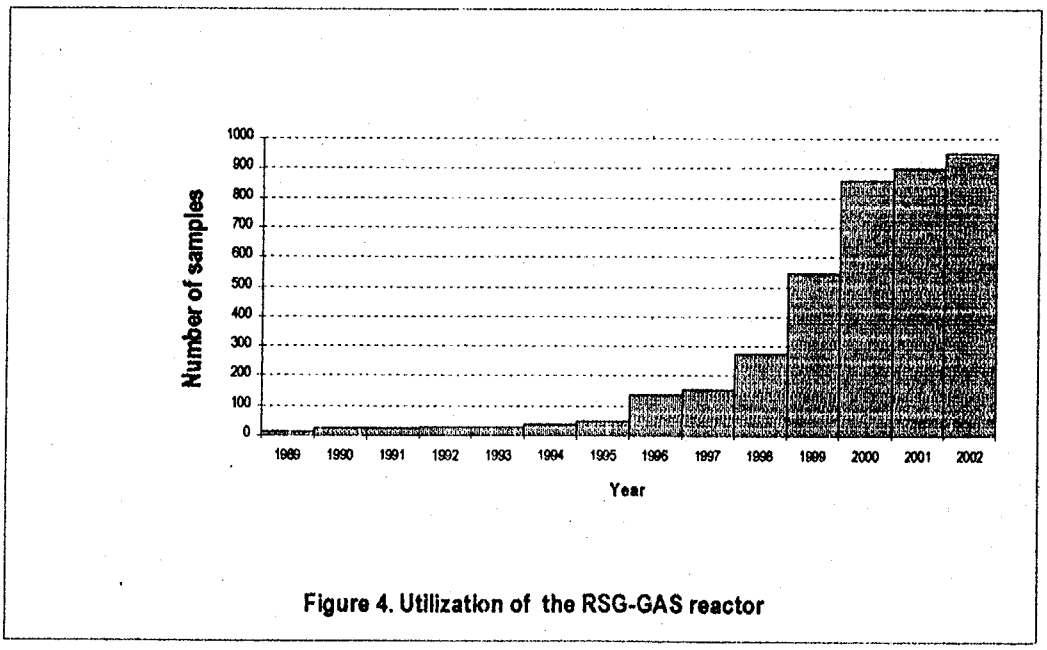

Figure 4. Utilization of the RSG-GAS reactor

Radioisotopes Molybdenum-99, Irridum-191, Iodium-131 etc. are routinely produced as presented in Table 1. The first production of iodine-125 from xenon gas was successfully performed in November 2002 at the xenon loop facility at beam tube no.1. The isotope has been delivered to India and China.

Tabel 1. : Routine irradiation for radioisotope production at RSG-GAS reactor per cycle.

No.	Target	Products	Weight, g/batch	# Batch	Activity, Ci/batch	Total Activity, Ci/cycle	Total Activity, Ci/ year
1.	U-235	Mo-99	2.2	12	88.5	1062	4248
2.	Ir	Ir-192	8	3	1750	5250	21000
3.	TeO_2	I-131	100	10	12	120	480
4.	S	P-32	20	2	2.2	4.4	17.64
5.	Sm_2O_3	Sm-153	0.03	5	0.02	0.1	0.4
6.	ReO_3	Re-186	0.03	2	0.015	0.03	0.12

For research and development has also provided with the power ramp test facility, neutron radiography, rabbit system for NAA, 5 beam tubes for science and material study. Lastly, for training and education, almost 30 under graduate and graduate students used this facility for research besides routine training of operator and maintenance group.

5. FUTURE PLAN

A basic objective of nuclear activities is to obtain an optimum use of three research reactor which are operated by BATAN for Scientific and Technological research and development as well as application. Therefore, it is important to find an appropriate system to stimulate active participation of the private sector in R&D activities. Collaboration research among researchers from government organization, universities, and business might be one way to promote R&D activities that lead to higher productivity.

In order to guarantee the continuity of supply of market demand , the operation of TRIGA-2000 will increase its isotope production in order to be able to back up the operation of RSG-GAS.

Application for NAA for the three reactor will be increased for serving the public demand in the area of environment, geology, and health.

Due to the aging problem and an obsolete technology of some I & C component activities to modify and refurbish the system was performed. Modification on computer system for RSG- GAS reactor monitoring is in progress.

6. CLOSING REMARK

All three reactors are routinely operated to serve radioisotope production, neutron activation analysis and beam experiments. From the utilization point of view, these reactor are still under utilized compared to the available capacity of the irradiation facilities Indeed, BATAN should actively seeking a new user and application and promote them through university and industry.

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1.5 Country Report: Utilization of MINT's Research Reactor

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Abstract

MINT has only one research reactor, i.e. TRIGA MKII reactor, equipped with various neutron irradiation facilities such as rotary rack and rabbit system. Apart from counting facilities for NAA work, other facilities available for the respective studies include facilities for neutron radiography and SANS. At Present most of reactor operation time has been utilized for samples irradiation related to the NAA application. Majority of the samples are from MINT analytical chemistry laboratory where I and my colleagues work, and the rest of the samples are from local universities. We do provide analytical chemistry services for other government departments as well as private companies.

In order to improve the reactor utilization, the management of MINT has formed Reactor Interest Group (RIG) at the national level in 2002, which embraces members from various institutions in this country. To support the RIG activities, MINT provides seed funding to finance various activities for the reactor utilization, which include financing project to make use of SANS, neutron radiography and radioisotopes production (mainly for tracer studies carried out by MINT's tracer group) facilities, and funding for basic study in BNCT.

1.0 INTRODUCTION

Reaktor TRIGA PUSPATI (RTP) is the one and only research reactor in Malaysia. It come into operation in 1982 and reached its first criticality on 28 Jun 1982. RTP is a pool type reactor, designed to allow greater flexibility in experimental works in the core region. It uses demineralized light water as moderator to dissipate heat, high purity graphite as reflector and uranium-235 as nuclear fuel.

The average neutron flux of RTP operated at maximum power of 1 MW is 1.2×10^{12} $cm^{-2} s^{-1}$. Using a mixture of Am/Be as neutron source and four control rod to control the fission reaction inside the reactor core, RTP produces free neutrons with energies ranging up to 10 MeV.

These free neutrons produced in the reactor core are used in many research related activities such as NAA, radioisotope production, neutron radiography and SANS. Irradiation facilities available in the reactor core include central thimble (CT) located at the center of reactor core, dry tube (DT), isotope production system (IPS), cadmium cover irradiation tube, two triangle irradiation positions and hexagonal irradiation positions.

2.0 STATUS OF REACTOR UTILIZATION AND FACILITIES.

Throughout the year 2002 the reactor has been in good condition and ready to be operated. The reactor however was closed for three months in August until November for annual maintenance. Generally, the reactor was operated based on user request typically 6 hours daily for 4 working days a week. Most of the time the reactor is used for irradiation of samples for NAA work. However, a 12-hours operation time has been accomplished several times for the production of radioisotopes such as samarium-153, phosphorus-32 and iridium-191 (iridium glass). Status of related facilities around the reactor are as follows:

2.1 Small Angle Neutron Scattering (SANS)

The specification of the SANS instrument available at one of the beam port of the reactor is given in Table 1.

Table 1: Specifications of the SANS facility at reactor TRIGA MINT.

Parameter	**Specifications**
Beam tube	Radial piercing beam port
Monochromator	Three (double layer) set of ZYB highly Oriented Pyrolitic Graphite (HOPG) Crystal
Incident wavelength	0.5 nm
Wavelength resolution	5.2%
Source to sample distance	1-4 meter
Beam size at specimen	12 to 50 mm
Q range covered	$0.0008 < Q < 0.036$ nm
Maximum flux	1.7×10^{4} n cm^{-2} s^{-1}
Detector	PSD (128 x 128 pixeles; 0.5 x 0.5 cm element dim., approx.)

Even though the facility is ready to be used since 1997, the system needs to be upgraded for instances to include automated beam stopper, liquid nitrogen top-up facility and better neutron scattering software. Despite the setback the system has been utilized to study several surfactant based colloidal samples including:

i. Samples which exhibit transition from worm micelle to spherical micelle through doping process (by using surfactant and block copolymer) on worm micelle system,
ii. Samples which show camera phase to micro emulsion phase transition in a ternary system.
iii. Samples which have symmetrical transition of cubic phase for ternary system.
iv. Samples of liquid crystal surfactant mixed with kaolinite clay to study microstructure variation.

Latest information I received showing that the upgrading of SANS facility is starting and expected to complete end of 2003.

2.2 Neutron Radiography

The first facility namely NUR 1 was built as a test facility at the beamport #1 which is one of the radial beamports. It was a temporary facility for obtaining and verification of data as well as calculation for a better facility. The experience and data acquired were the used to construct a permanent facility for neutron radiography named NUR 2. The construction of the facility was commenced in 1985. Both transfer and direct methods of NR can be done at the facility. At present a radiography image is recorded using films.

The facility will undergo further development beginning of 2003. Example of related project to be done is the development of a new thermal neutron radiography facility that will include the follow up task as follows:

a) Beam port characterization, design, instrumentation, fabrication and testing.
b) Neutron collimator, shielding and design
c) Development of imaging system
d) Determination of neutron beam quality and evaluation of image quality.

2.3 Neutron Activation Analysis

The NAA is an analytical chemistry technique and the Analytical Chemistry Laboratory of MINT operates the facility along with other complementary non-nuclear analytical technique such as ICP-MS, ion chromatography, GCMS and AAS. Neutron Activation Analysis plays an important role in the analysis of trace inorganic chemical in various types of samples for various applications in research as well as services. Examples of research undertaken with significant contribution from NAA technique include regional IAEA projects on air pollution trend in the region and marine environmental pollution. The Analytical Chemistry Laboratory also received samples from other organizations in the country for the elemental analysis. The non-destructive nature of NAA technique enables the analyst to deal with material, which is difficult to make up into solution such as industrial sludge, oil sludge, minerals and rubber products. The NAA can also be used to validate analytical data generated by other analytical techniques.

2.4 Radioisotope Production

Radioisotope from elements such as Mo, I, Ho, Au, Ir, Br, P, and Zn can be produced in the RTP. Irradiation facilities for producing radioisotope available in RTP are Dry Tube (DT), Isotope Production System (IPS), Rotary Rack (RR) and Central Thimble (CT). The radioisotope production laboratory is equipped with hot-cells and glove-boxes for handling various radioisotopes and for labeling compounds and complexes.

In nuclear medicine, radioisotopes such as I-131, Sm-153 and Ho-166 are used as radiotheraphy agents. Due to low neutron fluxes some of these radioisotopes ceased to be produced in the RTP facilities.

For radiotracer application, radioisotopes such as Au-198, Ir-192, Br-82, P-32, Fe-59 and Zn-65 are produced using RTP irradiation facilities. These radioisotopes are introduced as labeling agents into systems such as pipe ways, river, underground water channel and air to mark the trajectories of these systems.

2.5 Education and Training

The RTP has also used for education and training for the following field of study:

a) Reactor physics and engineering
b) Reactor operation and maintenance
c) Reactor instrumentations
d) Reactor utilization

Reactor physics and engineering cover areas such as the study of reactor behaviour, e.g. Neutron reactivity insertion, neutron fluxes measurements, reactivity coefficients of reactors, criticality, transport and diffusion of neutrons, reactor kinetics and thermal hydraulics. It also covers areas in theoretical computations through the use of computer codes for estimating physical reactor parameters and thermal hydraulic properties of the reactor.

Reactor operation modes include the start-up and shutdown procedure, steady-state and square wave as well as core excess procedures. Reactor maintenance includes reactor power calibration using doubling time method and inspections and service of reactor components including the mechanical, electrical, electronics and instruments of the reactor.

RTP is available to provide undergraduate as well as postgraduate students hands-on experience in their field of studies, especially in reactor physics and engineering and reactor operation training. The research reactor normally used for basic training of reactor operators before advancing to a higher and more advance level.

Even though there are many areas of R&D regarding the reactor utilization that has the potential to be exploited, this is not fully utilized. As it is now, current reactor utilization is low – require a lot of commitments in terms of human resources and funding to propagate the R&D research reactor related activities.

3.0 REACTOR INTEREST GROUP (RIG)

With the objective of arresting the decline of interest in the utilization of RTP, MINT has initiated the formation of RIG in early 2000 and was formally launched at national level on 25 Jun 2002. The group comprises members representing stakeholders and/or those who have interest in the reactor utilization. The formation of RIG will provide platform for the members to channel ideas and suggestions to the management of MINT to improve the usage of the reactor and upgrade its system.

In the initial stage, the RIG activity members from universities academic staffs and research institutes in Malaysia have been invited in the RIG seminar held on 25 Jun 2002 with the objective of identifying research activities to be carried out. The RIG workshop was later held in Penang on 27-30 Aug. 2002 to detail out the proposed research program. The proposed research program to be implemented in the near future funded by the Ministry of Science, Technology and Environment include:

a) The application of nuclear and complementary analytical technique to health related issue comprising the following research activities: Essential and toxic metals in food of an average Malaysian Dietary intake, Characterization of air-borne particulate particles in the assessment of air quality, Drinking water quality and its health impact, Geochemical effects on ground water quality for use in public drinking.
b) The application of isotope and tracer, comprising the following research programs: predicting soil erosion using radioisotope technique and sediment transport study at Sungai Kemaman estuary.
c) In-core development facilities comprising the following research activities: Development of neutron flux mapping display and TRIGA fuel management and modeling.
d) Utilization of neutron beam for medical and industrial application (auxiliary/out core activities) comprising of the following research activities: Development of (thermal) new Neutron radiography facility, enhancement of SANS to include beam port characterization, design, instrumentation, fabrication and testing, Neutron collimator, shielding and design, development of imaging system and determination of neutron beam quality, feasibility study on the possibility of developing Prompt gamma NAA at RTP, and study on Characteristic of neutron from RTP thermal column as part of design and feasibility study for BNCT.

4.0 CONCLUSION

At present, the reactor and facilities except for NAA are under utilized. The current declining of reactor utilization will be overcame with the commitment from all party concerned including the researchers involved from MINT and other research institutions in the country as well as MINT's management and the Government of Malaysia.

THE 2002 WORKSHOP ON THE UTILIZATION
OF RESEARCH REACTORS
January 13-17, 2003
Serpong, Indonesia

COUNTRY REPORT

1.6 STATUS OF RESEARCH REACTOR UTILIZATION AND OTHER RELATED ACTIVITIES

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ABSTRACT

The report covers two parts; the first is on the progress of the cooperative projects planned for 2002 under the FNCA and the other part on the activities related to the PRR1, Philippine Research Reactor. In the 2001 Workshop at Beijing, the Country agreed to participate in the three areas for collaboration. A brief reports on these three projects are included. The Country representatives during this 2002 Workshop will do a more detailed presentation on Radioisotope Production (TcG) and Neutron Activation Analysis projects. The second part of this report deals with the issues/concerns impeding the rehabilitation of PRR1*. In January 2002, the Institute created the PRR1 Strategic Plan Committee to look deeply into these issues and concerns. The results of the Committee's work are discussed.

* "A Rehabilitation Concept for the Core Container and Associated Structures of the PRR1", L.S. Leopando presented to the IAEA/RCA Regional Seminar on Aging Management of Research Reactors, Mumbai, India 4-6 December 2002

Research Reactor Utilization and the FNCA Projects

In the 2001 Beijing Workshop the Philippines agreed to participate in the three areas of research reactor utilization projects. Despite the inherent constraints due to the unavailability of the neutron facility work, have been done that contributed to the objectives of the cooperative activities that mutually satisfies the local needs and hopefully that of the other participating countries.

1. Neutron Scattering(Neutron Beam Applications)

The Philippine's proposal on the small angle scattering of carrageenan had been discussed in the 2001 Beijing Workshop. Carrageenan, a natural polymer produced from red seaweed, has various types of isomers, such as kappa(κ), iota(ι) and lambda(λ), and other. They differ in the location of the OH groups and/or the number of the $-SO_4$ groups. Carrageenan is one of the important natural products of the Philippines. Various R & D on the applications of the carrageenan have been supported by the government however very few and limited studies on understanding the basic characteristics of the material have been done. The proposal was adopted as one project for collaboration with Prof. M. Shibayama of Tokyo University and his colleagues. Two studies were proposed and were evaluated by Japanese polymer scientists.

a. Structural investigation of gamma irradiated carrageenan
The structure-properties of gamma ray irradiated carrageenan can be investigated from various points.
   i. structure-mesh size, local ad global structure, heterogeneities, sol-gel transition
   ii. properties-mechanical properties, swelling behavior, hysteresis, phase behavior, thermal properties

As suggested by the evaluators, we have concentrated on the kappa carrageenan. Philippine refined κ-carrageenan was further purified by re-precipitation with alcohol, converted into pure salt form, dialyzed and lyophilized. Samples were irradiated using Co-60 at 5 and 100KGy at ambient temperature. Molecular weight, FTIR, XRD and XRF of the unirradiated and gamma irradiated samples are obtained. These samples will be used in the SANS experiments.
Complimentary experiments on the film gel using angular dependent total reflection X-ray fluorescence spectrometry is being prepared using local.facility.

b. Structural investigation of carrageenan/Polyvinyl pyrrolidone (PVP) blends
Samples with different amount of kappa carrageenan(KC) and PVP in water were prepared and irradiated at a dose of 5-50KGy. SANS experiments are proposed to study (a) cross-linked PVP (b) grafted KC to PVP and (c) KC trapped as semi-interpenetrating network.. Prof. M. Shibayama had commented and gave suggestions on the proposed experiments.

Documents and procedures for conducting experiments using the SANS set up of Tokyo University at Tokai was also provided by him. Arrangement for one of the polymer chemist from PNRI to join the group of Prof. Shibayama to conduct the

SANS experiments this 2003 is being made. Meanwhile, other sample characterization complementing SANS data needed to fully understand and explain the structural change will be continued at PNRI.

2. Neutron Activation Analysis

PNRI is implementing a project entitled "Nuclear Analytical Techniques for Air Quality Management" in support of The **Clean Air Act,** a legislation of the congress of the Philippines to address air pollution problems and related environmental concerns. NAA is one of the nuclear analytical techniques to be employed for the analysis of the air filters, together with the x-ray fluorescence methods and PIXE currently being used. Filters from air samplers strategically located various part of Metro Manila have been collected for more than five years already. These filters were mostly analyzed by XRF and some by PIXE. NAA of few samples by a staff while on OJT abroad was also conducted. Since NAA is one of the utilization area during the operational years of the reactor a number of researchers have experience on this technique but whose knowledge of the Ko method is minimal. It is desired to use NAA to compliment the XRF technique and at the same time evaluate their performance through intercomparison. To realize this, a mechanism to access irradiation/NAA facility outside through the IAEA/RCA project on Sharing of Research Reactor Resources or/and under the FNCA will be explored. Details of this will be discussed by the country representative at Workshop.

3. STATUS OF THE APPLICATION OF REACTOR PRODUCED RADIOISOTOPES

Reactor produced radioisotopes are continued to be used in the bio-medical, industrial, agricultural field and in various areas of research in the Country. Due to the unavailability of a neutron source, the need for these radioisotopes are met through importation. Some of these widely used radioisotopes are; ^{131}I, ^{99m}Tc, ^{201}Tl, ^{32}P, ^{3}H, ^{192}Ir, ^{35}S, ^{55}Fe, etc. High activity sources such ^{60}Co, ^{137}Cs, ^{192}Ir, ^{90}Sr, etc. present in nuclear medicine, NDT and nucleonic instrumentation facilities are are used and found even in the Visayas and Mindanao.

R & D on the ^{99m}Tc generator had been conducted since the early 90's. A gel type column was developed which had shown satisfactory Molybdenum absorptive capacity. For the 2002 the performance of the gel-type polyzirconium compound (PZC) column material developed by Kaken Co. in Japan in cooperation with Japan Atomic Energy Research Institute (JAERI) is being tested under the framework of the FNCA project on Research Reactor Utilization. A separate report on this will be presented by the Country representative.

STRATEGIC PLAN FOR PRR-1: REHABILITATION/UTILIZATION

A rehabilitation concept for the core container and associated structure of the Philippine Research Reactor (PRR-1) was made when it was decided to continue the repair and operate the reactor. The assistance of the IAEA was sought to give advice and

evaluation on the proposed rehabilitation. Two experts were dispatched by the Agency who made a review and evaluation of the conceptual design of the core tank and decay tank. In addition, the experts recommended the following:

1. Preparation of a strategic plan for PRR-1 utilization to provide rationale for its rehabilitation prior to any reactor system modification.
2. The establishment of an adequate organization with substantial financial and human resources that will take responsibility for the rehabilitation. The organization and function of this group must comply with Agency Guide(SS 35-G2)
3. Establishment of a legal standing for the Nuclear Regulations, Licensing and Safety Division (NRLSD) to enforce regulatory functions related to PRR-1 rehabilitation. NRLSD is one of the technical divisions under PNRI.

The Institute acted on the recommendation and the Director created the PRR-1 Strategic Plan Committee. The members are senior staff of the three technical divisions who had involvement with PRR-1 as user, operator or regulator. They are also the most experienced and knowledgeable people in PNRI as far as PRR-1 is concerned. Some retired Division and Section chiefs served as advisers. The Committee's task has 2 phases:

1. Determine whether the rehabilitation and operation of PRR-1 will be sustainable
2. Preparation of strategic plan for implementation

The committee met regularly for six months for Phase 1. In this period the following were conducted:

1. Review and evaluation of the present status and condition of PRR-1
2. Identification of the regulatory requirement that will be necessary to assure safety and public acceptance of operation of PRR-1
3. Identification of real needs that will provide justification for the rehabilitation and will provide long term sustainability of its operations
4. Identification and quantification of resources needed to assure sustainable operation. The resource include those for rehabilitation, operation, to create and operate the utilization facilities and operate the regulatory infrastructure
5. Identification of sources for financial and manpower needs

PRELIMINARY CONCLUSIONS/OUTPUT(Phase 1)

1. Committee was unanimous that there were national needs that only a research reactor could satisfy
2. Administrative Policy placing PRR-1 and other nuclear facilities of PNRI under regulatory control by the NRLSD to ensure safety conditions as recommended in the relevant parts of the IAEA code of practice.
3. Identified needs that PRR-1 could serve

4. At least US$12 M would be needed to complete repair(reactor to operate at 3 MW from the original 1 MW before TRIGA conversion)
5. Repair must be complete in 5 years

These were presented to the Director and the senior management staff who will have to act on specific items and finally made decision on its recommendation to the Department of Science and Technology which supervises PNRI.

1.7 RESEARCH REACTOR AND ITS APPLICATION IN THAILAND

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ABSTRACT

The first Thai Research Reactor (TRR-1) was established in 1961. TRR-1 had been operated with power of 1 MW from 1962 to 1975 and was shut down for modification during 1975 to 1977. The Thai Research Reactor1/Modification1 (TRR-1/M1) is a multipurpose reactor with nominal power of 2 MW. Since 1977 TRR-1/M1 has been operated and utilized for various applications such as neutron activation analysis, radioisotope production, gem irradiation, neutron radiography and research works. To expand and promote the utilization of research reactor, the new 10 MW Reseach Reactor will be established in the Ongkarak Nuclear Research Center (ONRC) project and the project will be finished in the near future.

Introduction

The first Thai Research Reactor (TRR-1) was established in 1961. The TRR-1 was a swimming pool type reactor with power of 1 MW and plate typed high enriched uranium (U_3O_8-Al) fuel. The reactor was cooled and moderated by light water. The TRR-1 had been operated from 1962 to 1975 and was shut down to modified to TRR-1/M1. The reactor core and control system were disassembled and replaced by TRIGA mark III. The TRR-1/M1 was a multipurpose reactor with nominal power of 2 MW. with low enriched uranium (UZrH) fuel. The initial criticality of TRR-1/M1 was reached on November 7, 1977. At the beginning the whole core was composed of 8.5% wt. of 20% enricled U-235 fuel elements. Later in 1980, some fuel elements were replaced by 20% wt. of 20% enriched U-235, resulting in mixed core operation unitil now.

The TRR-1/M1 has been routinely operated to serve the users in goernment organizations and private sectors. The main applications of TRR-1/M1 are neutron activation analysis, radioisotope production, gem irradiation, neutron radiography and other research works.

Thai research Reactor-1/Modification 1 (TRR-1/M1)

Characteristic of TRR-1/M1

Reactor Type	TRIGA Mark III
Maximum Steady-State Power Level	2000 kW (thermal)
Maximum Pulse	2.1% δk/k ($3.00)
Fuel Element Design	
Fuel-moderator material	U-$ZrH_{1.6}$[a]
Uranium content	8.5 wt-%
Uranium enrichment	20% U-235
Shape	Cylindrical
Length of fuel	38 cm (15 in.) overall
Diameter Of fuel	3.63 cm (1.43 in.) OD
Cladding material	Type 304 SS
Cladding thickness	0.051 cm (0.020 in.)
Number of Fuel Elements	100
Excess Reacitivity, max	6.3% δk/k (cold, clean)
Number of Control Rods	
Transient	1
Regulating	1
Shim	2
Safety	1
Total Reactivity Worth of rods	10.12% δk/k
Reactor Cooling	Natural convection of pool water

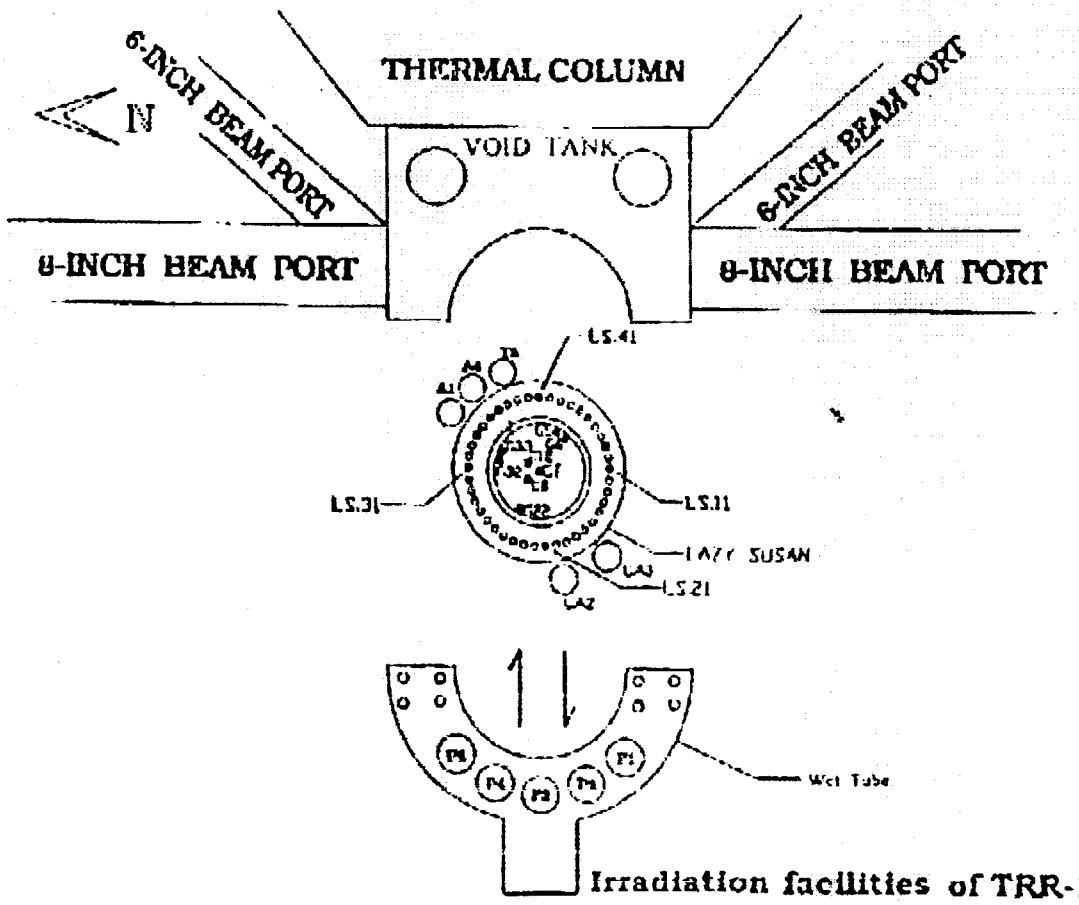
Operating Schedule of TRR-1/M1

Nowadays, TRR-1/M1 is operated at the power of 1.2 MW, 3 days per week with total 34 hours per week. The reactor is shut down 2 months every year for yearly maintenance.

Operating Schedule :

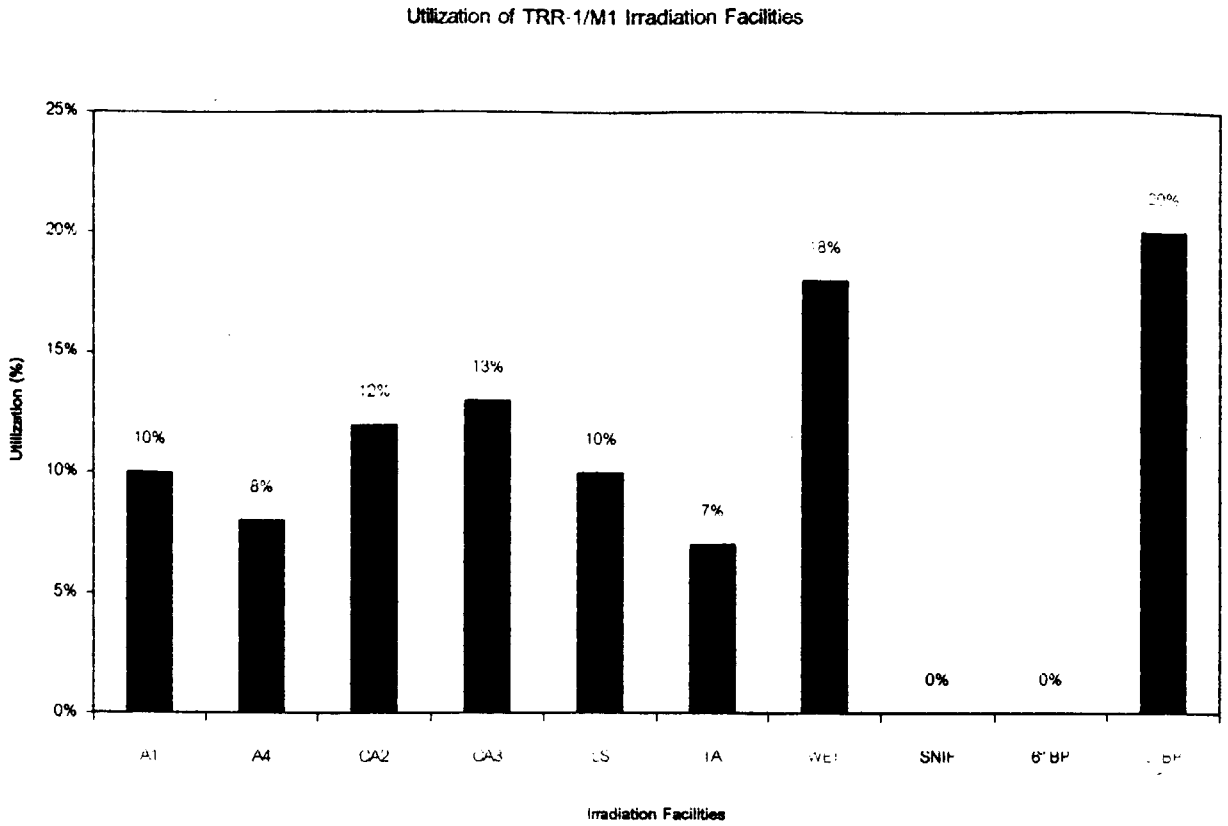
Monday	:	weekly maintenance
Tuesday	:	reserve for special experiment
Wednesday	:	Operate at 1.2 MW for 12 hours
Thursday	:	Operate at 1.2 MW for 12 hours
Friday	:	Operate at 1.2 MW for 10 hours

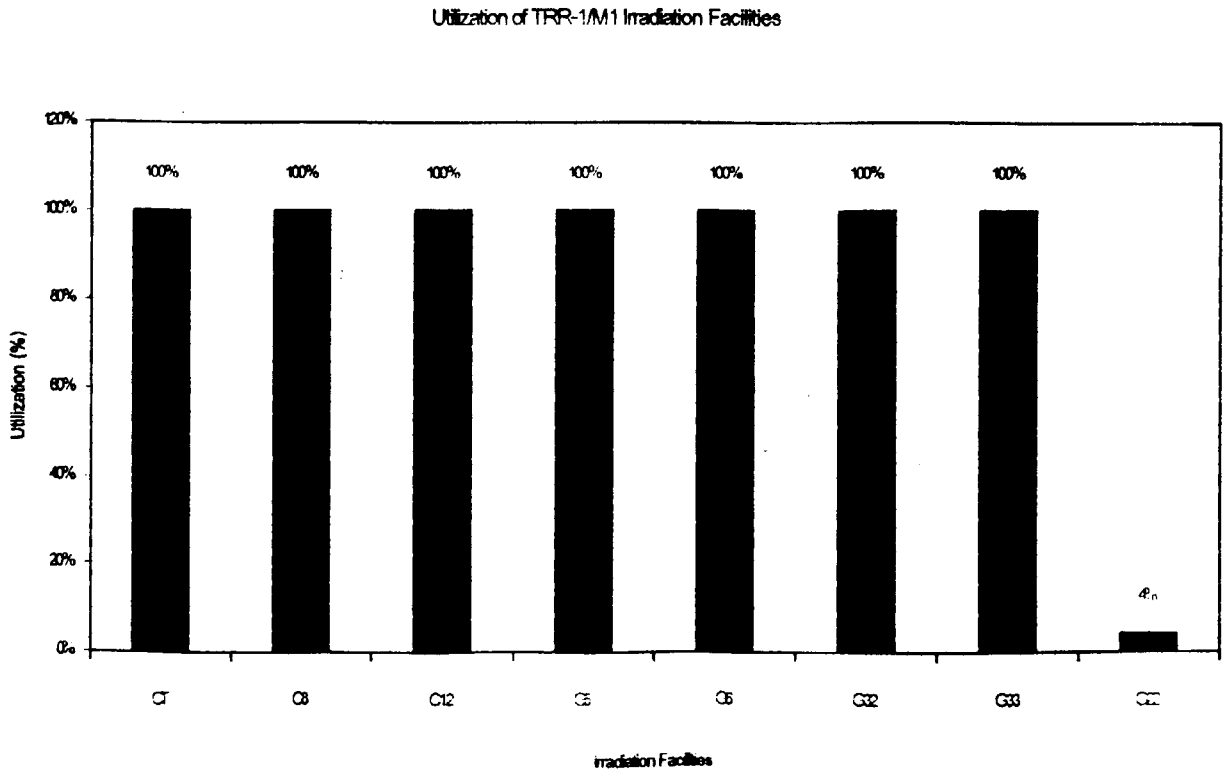
Irradiation Facilities of TRR-1/M1



Irradiation facilities of TRR-1/M1

Irradiation Tube	No. of Tube	Neutron flux N/cm^2sec (at 1.2 MW)	Purpose of Utilization
CT	1	3.0×10^{13}	Radioisotope Production
C8	1	2.4×10^{13}	Radioisotope Production
C12	1	2.5×10^{13}	Radioisotope Production
G5	1	1.1×10^{13}	Radioisotope Production
G6	1	1.0×10^{13}	Radioisotope Production
G32	1	0.9×10^{13}	Radioisotope Production
G33	1	1.2×10^{13}	Radioisotope Production
G22	1	7.5×10^{12}	NAA
Rotary Specimen Rack(Lazy Susan)	41	3.5×10^{12}	NAA
A1	1	8.7×10^{11}	NAA
A4	1	8.7×10^{11}	NAA
CA2	1	1.1×10^{10}(epithermal)	NAA
CA3	1	1.0×10^{10}(epithermal)	NAA
TA	1		NAA
Wet Tube	13	$\approx 1.0 \times 10^{11}$	Gem Irradiation
SNIF	1		NAA
6" Beam Port	2	$\approx 1.0 \times 10^{6}$	PGNAA
8" Beam Port	2	$\approx 1.0 \times 10^{6}$	Neutron Scattering, Neutron Radiography


Utilization of TRR-1/M1 Irradiation Facilities
25%
20%
15%
10%
5%
0%
Utilization (%)
10%
8%
12%
13%
10%
7%
18%
0%
0%
Irradiation Facilities


Utilization of TRR-1/M1 Irradiation Facilities
120%
100%
80%
60%
40%
20%
Utilization (%)
100%
100%
100%
100%
100%
100%
100%
Irradiation Facilities

Reactor Utilization

1. Neutron Activation Analysis
2. Radioisotope Production: I-131, Sm-153, P-32 etc.
3. Gem irradiation
4. Nuclear Physics and Reactor Engineering experiments
   - Neutron Radiography
   - Neutron Spectrometer Test
   - Heat generator Measurement in Irradiation Tube
   - Flux Measurement
   - Neutron Radiography Corputed Topography-film Scanning Technique
5. Reactor Operator Training

Users

1. Physics Division
2. Isotope Production Division
3. Chemistry Division
4. Waste Management Division
5. Radiation Measurement Division
6. Department of Mineral Resources
7. Universities: Chulalongkorn University, Naresuan University, Rajabhat Institute etc.
8. Private Companies

Future Plan

The new 10 MW Research Reactor will be established in the Ongkarak Nuclear Research Center (ONRC) Project.

The reactor system shall be a multipurpose, pool type reactor, cooled and moderated by light water, reflected by beryllium and heavy water (D_2O),that will use low-enriched uranium (LEU) fuel. This uranium erbium zirconium hydride fueled TRIGA reactor shall have a steady state thermal power rating of 10 MW, and shall be capable of generating a neutron flux suitable for the following functions:

- Beam experiments: Neutron Diffraction (HRPD), Neutron Radiography (NR), and Prompt Gamma Neutron Activation Analysis (PGNAA.)
- Radioisotope production for medical, industrial and agricultural uses.
- Neutron transmutation doping (silicon.)
- Applied research and technology development in the nuclear field.
- General training to master the fundamental principles of the reactor and its operation.
- Training in reactor physics (neutron physics, thermal hydraulics, reactor experiments, etc.)
- The capability for later modification to perform medical therapy for patients using the boron neutron capture (BNCT) technique, and/or small angle neutron scattering (SANS.)

The ONRC Project is in the process of submittal for construction permit of the reactor.

Conclusion

The Thai Research Reactor has been safety operated for 40 years. The TRR-1/M1 is still operating in good condition and can be routinely utilized for neutron activation analysis, radioisotope production, neutron radiography, gem irradiation and other experiments.

To expand and promote the utilization of Research reactor, the new 10 MW Research Reactor is planned in the ONRC project and expect to be finished in the near future.

1.8 CURRENT STATUS OF OPERATION AND UTILIZATION OF THE DALAT RESEARCH REACTOR

(Presented at the FNCA Workshop on the Utilization of Research Reactors, Jakarta, Indonesia, 13-17 January, 2003)

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ABSTRACT

The Dalat Nuclear Research Reactor (DNRR) is a 500 kW swimming pool type reactor used the Soviet WWR-SM fuel assembly with 36% enrichment of U-235. It was upgraded from the USA 250 kW TRIGA Mark-II reactor. The first criticality of the renovated reactor was in November 1983 and its regular operation at nominal power of 500 kW has been since March 1984.

The DNRR is operated mainly in continuous runs of 100 hrs, once every 4 weeks, for radioisotope production, neutron activation analyses and research purposes. The remaining time between two continuous runs is devoted to maintenance activities and also to short run for physics experiments and training purpose. From the first start-up to the end of December 2002, it totaled about 24,700 hrs of operation and the total energy released was 490 MWd.

After 10 years of operation with the core of 89-fuel assembly configuration, in April 1994, the first refueling work was done and the 100-fuel assembly configuration was set-up. The second fuel reloading was executed in March 2002. At present time, the working configuration of the reactor core consists of 104 fuel assemblies. This fuel reloading will ensure efficient exploitation of the reactor for about 3 years with 1200-1300 hrs per year at nominal power.

The current status of operation and utilization and some activities related to the reactor core management of the DNRR are presented and discussed in this paper.

I. REACTOR DESCRIPTION AND ITS OPERATION

The DNRR is a pool type reactor, moderated and cooled by light water. It was upgraded from the TRIGA Mark-II reactor built in early 1960's. First criticality of the renovated reactor was in November 1983 and since March 1984, its regular operation has been done. Main specifications of the DNRR are shown in Table 1.

Table 1. Reactor Specifications

Reactor type	Swimming pool, TRIGA Mark II, modified to Russian type of IVV-9
Nominal thermal power	500 kW, steady state
Coolant and moderator	Light water
Core cooling mechanism	Natural convection
Reflector	Beryllium and Graphite
Fuel type	WWR-SM, U-Al alloy, 36% enrichment
Number of control rods	7 (2 safety rods, 4 shim rods, 1 regulating rod)
Control rod material	B_4C for safety and shim rods, Stainless steel for automatic regulating rod
Neutron measuring channels	9 (6 CFC, 3 CIC)
Vertical irradiation channels	4 (neutron trap, 1 wet channel, 2 dry channels) and 40 holes at the rotary rack
Horizontal beam-ports	4 (1 tangential, 3 radial)
Thermal column	1
Spent fuel storage (temporary)	inside reactor building, next to the reactor shielding

From the first start-up of the reactor in March 1984 to the present time, the reactor has proved to be safe and reliable, as it has never suffered from any incident which significantly affected the environment, and annual operation schedules have been rigorously respected (see in Table 2 and Fig. 1).

Table 2. Operation data of the DNRR

Year	***Operation time (hrs)***	***Released Energy (MWd)***	***Accumulated released energy (MWd)***	***Number of scram***		
				Electric network failure	*Other reasons*	*Total*
1984	1120	18.6	18.6	11	17	28
1985	1771	31.7	50.3	5	8	13
1986	1387	26.9	77.2	6	4	10
1987	993	17.9	95.1	6	4	10
1988	1286	25.2	120.3	14	4	18
1989	1343	26.3	146.6	9	2	11
1990	1505	30.4	177.0	13	5	18
1991	1654	33.9	210.9	8	2	10
1992	1486	30.4	241.3	8	2	10
1993	966	17.7	259.0	1	2	3
1994	1302	26.8	285.8	11	1	12
1995	1351	27.9	313.7	9	2	11
1996	1370	28.1	341.8	7	3	10
1997	1206	24.7	366.5	9	1	10
1998	1203	24.9	391.4	2	4	6
1999	1215	25.1	416.5	9	4	13
2000	1113	22.8	439.3	8	0	8
2001	1228	27.5	466.8	4	2	6
2002	1220	24.0	490.8	1	3	4
Total	***24,719***		***490.8***	***141***	***70***	***211***

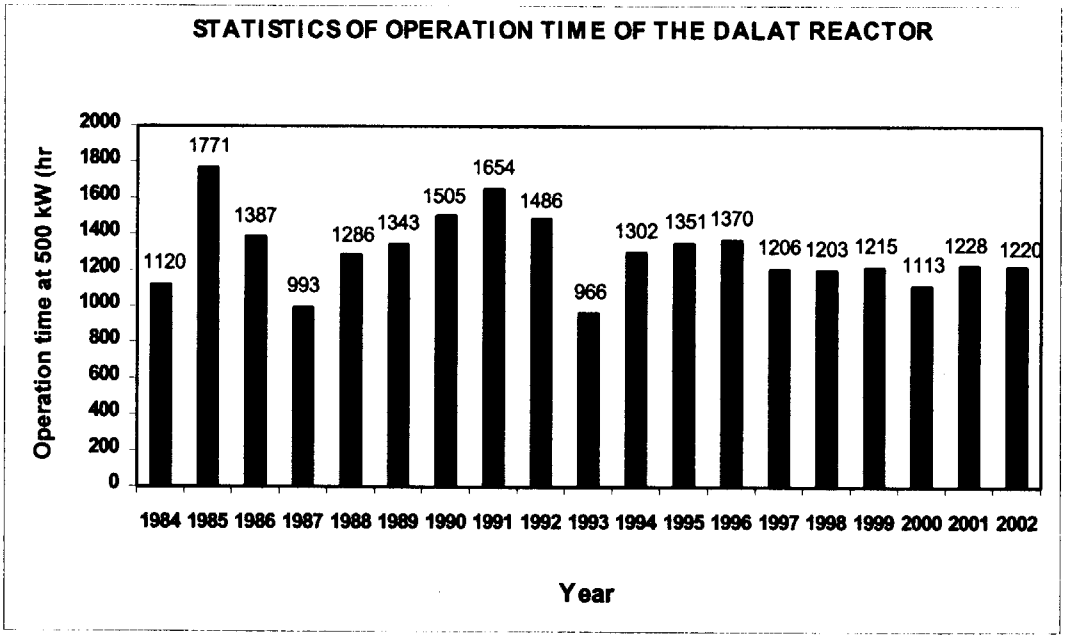

Fig. 1. Statistics of reactor operation time from 3/1984 to 12/2002

II. REACTOR MANAGEMENT

II.1. Ageing Management

In order to carry out the visual inspection of reactor aluminium tank and in-side components, an home-made high-resolution camera system has been designed and installed.

In the past years, using the designed camera system, some inspections have been done. It should be noted that the upper cylindrical shell prevented visual access to the major part of the reactor tank's surface, both because it covers the upper part of the tank and because its horizontal bottom plate prevents from easy immersion of video camera. Thus, only about 20% of the total reactor tank's surface could be visually inspected. All information about inspected surface has been recorded on videotape. Based on the results obtained, it could be noted that most of the pits seem to have initiated from mechanical defects (scratches on the aluminum surfaces during commissioning and re-commissioning time). However, these pits seem to be unplugged in most of the cases, which is in favor of stopping pitting propagation. Most of cracks like defects appears to be due to bad welding conditions during reactor constructing time.

It seem to us that, using an immersed video camera coupled to a control monitor and a video tape recorder with high contrast and resolution, is good enough for the visual inspection of the surface of reactor pool tank and in-side components.

II.2. Core Management

The fuel assemblies of the DNRR are of Soviet-designed standard type WWR-SM, enriched to 36% in U-235. Each fuel assembly composes of 3 concentric layers: 2 circular inner tubes and one hexagonal outer tube. The fuel layer with a thickness of 0.7 mm is wrapped between two-aluminum alloy cladding layers of 0.9-mm thickness.

After 10 years of operation with the core of 89-fuel assembly configuration, in April 1994, refueling work was done by adding 11 fresh fuel assemblies in the core periphery at

previous beryllium rod locations. The second refueling work was done in March 2002 by adding 4 fresh fuel assemblies more in the core periphery. At present, the reactor has been working with 104-fuel assembly configuration (Fig. 2).

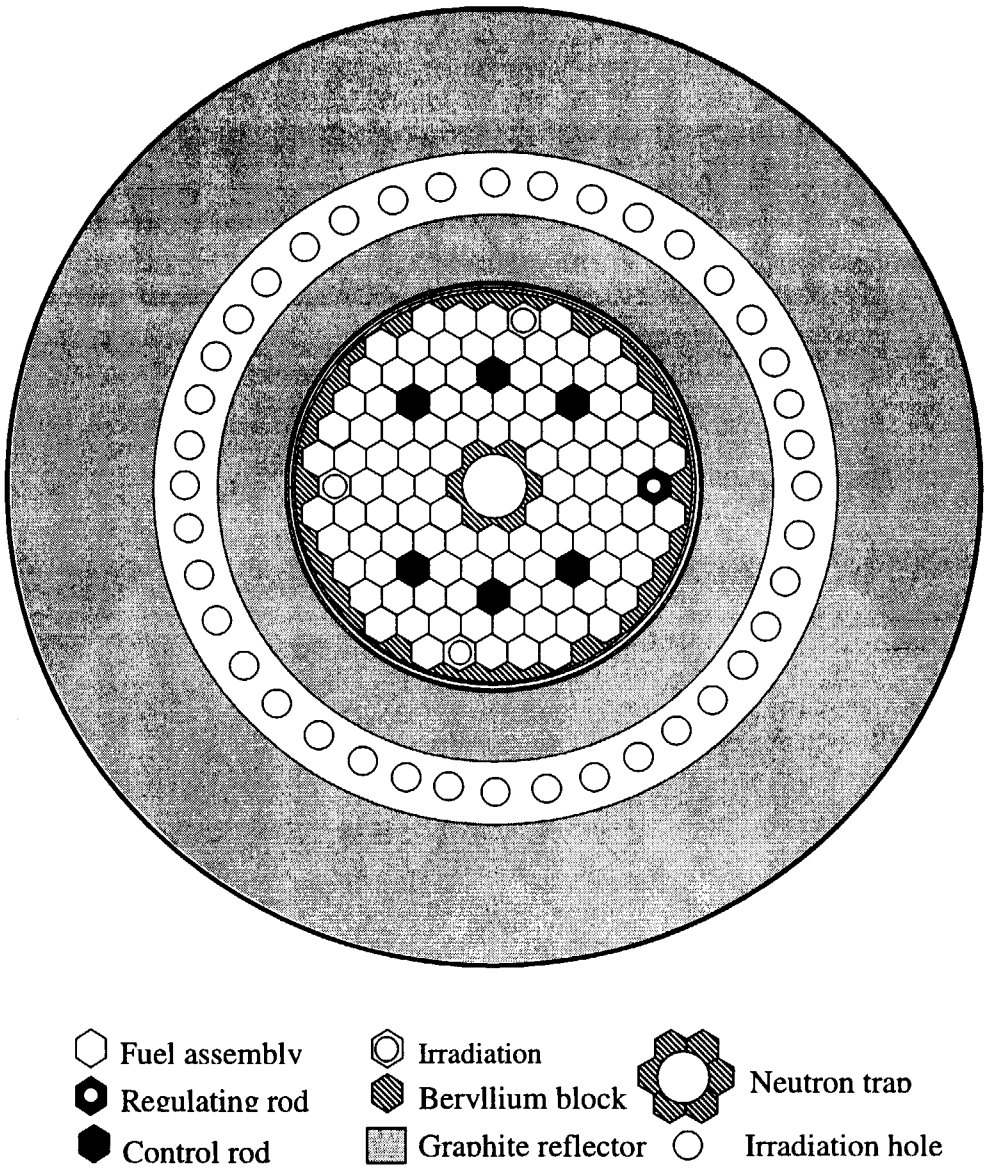

Figure 2. Cross-section view of the reactor core

In order to prepare a plan for the second reloading, a fuel management programme including calculation and experiment researches was implemented.

The calculation of fuel burn-up and burn-up distribution for Dalat reactor has been carried out based on cell calculation program WIMS and two diffusion calculation programs HEXAGA and HEXNOD in two dimensional geometry. The codes of WIMS, HEXAGA, HEXNOD have been developed to solve the problem of in-core fuel management optimization basing on the perturbation theory and binary shuffle technique. The code can search an optimal fuel loading pattern with the lowest power peaking factor for any available set of fuel assemblies. To determine the number of fuel assemblies to be discharged at the

end of cycle, which satisfies with operational constraints, several different reloading schemes with the different number of discharged fuel elements are investigated.

For checking calculation results, some experiments have been done. For this purpose, experimental measurement of fuel burn-up for the reactor was realized by gamma scanning method to measure long live isotopes from fission products.

Gamma spectra, treated by a multi-channel analyzer can be used to evaluate the component of fission and activation products in fuel elements. Based on the analyzing the gamma spectra, we can define the burn-up of each fuel element. It is clear that the activity of long-lived fission product - Cs^{137} has concern with fuel burn-up, But because of the difficulty with the efficient calibration of the measurement system, the relative method is usually used. By relative method, ratios of Cs^{134}/Cs^{137}, Eu^{154}/Cs^{137}, Cs^{134}/Sb^{125} can be used for evaluating the fuel burn-up. In our case, we are interested in using Cs^{134}/Cs^{137} ratio because radioactivity of Cs^{137} can measured after several days of fuel cooling and Cs^{134} can be measured after one month of fuel cooling with the existing $70cm^3$ HPGe system.

Using calibrated curves of variation of Cs^{134} and Cs^{137} activity by fuel burn-up by PHDOSE program, the burn-up of each fuel element can be determined.

During last two years, the burn-up of 68 fuel assemblies was determined by experimental measurements.

Results obtained show that the difference between calculation data and experimental data is about 5%.

III. REACTOR UTILIZATION

III.1. Radioisotopes Production

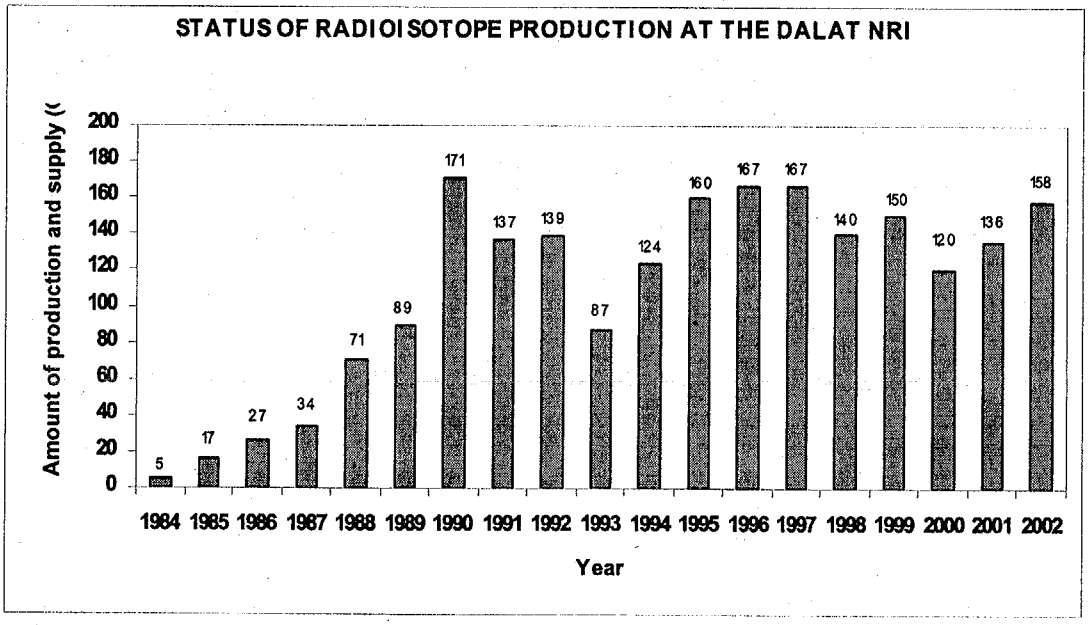

Fig. 3. Status of radioisotope production for medical use from 3/1984 to 12/2002

Radioisotopes and radiopharmaceuticals have been produced serving nuclear medicine departments and other users such as in industry, agriculture, hydrology, scientific research, etc. For medicine purpose our products are routinely delivered to more than twenty hospitals in the country, allowing them to diagnose and treat about 50 thousands of patients per year. The main products are P^{32} applicators, Tc^{99m} generator and its label, I^{131} solution/capsules and others *(Fig. 3).* It is totaled about 2,100 Ci of radioisotopes and radiopharmaceuticals have been produced and supplied to medical users so far, giving a yearly average of about 120 Ci.

III.2. Neutron Activation Analysis

In the nuclear analytical field, requests of many branches of the national economy for various types of samples have been answered *(Fig. 4).* Nuclear technique is of advantage in serving geology exploration, oil prospecting, agriculture, biology, etc. Environmental studies are carried out efficiently with nuclear and associated methods. These studies have led to following the radioactivity background at various locations inside the country. Environmental pollution studies especially at big cities and industrial centers have contributed to implementing the environment control mission of the Government. It is totaled about 41,500 samples have been irradiated at the reactor and analyzed so far, giving a yearly average of 2500 samples.

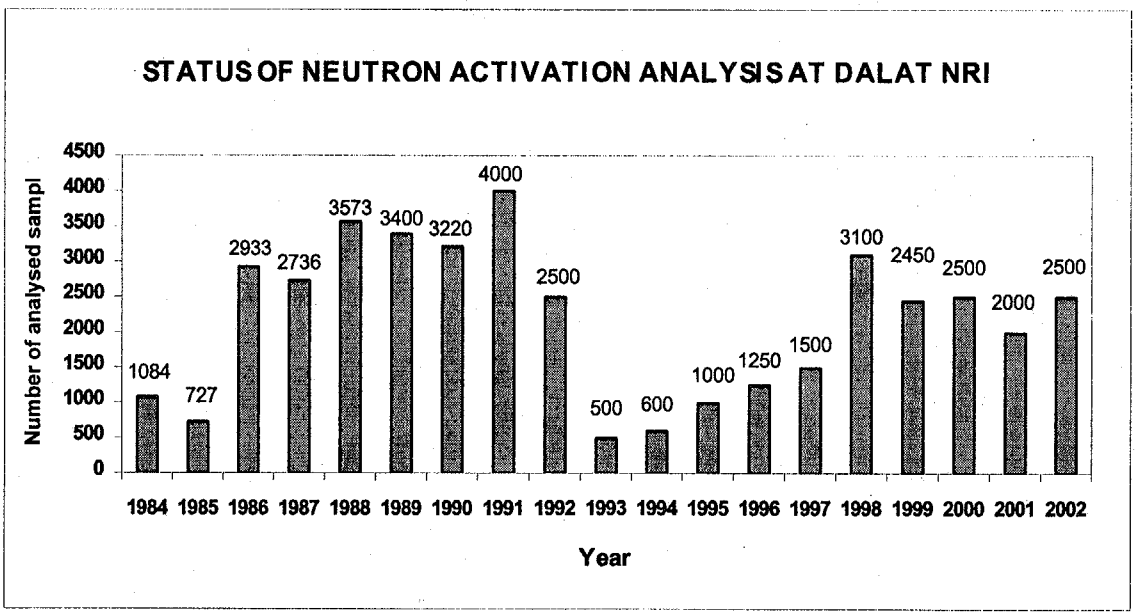

Fig. 4. Status of neutron activation analysis at DNRR from 1984 to 12/2002

III.3. Neutron Beam Researches

The reactor has four horizontal beam tubes, which provide beams of neutron and gamma radiation for a variety of experiments. They also provide irradiation facilities for large specimens up to 15cm in diameter in a region close to the reactor core. In configuration, three of the beam tubes are oriented radically with respect to the center of the core, and one beam tube is tangential to the outer edge of the core. Besides, there is a large thermal column with outside dimensions of 1.2m by 1.2m in cross section and 1.6m in length (Fig. 5).

Because lacking of the experimental facilities, during the last two years, at the Dalat reactor some activities related to neutron beam utilization have been carried out, such as PGNAA, nuclear data measurements, radiation protection study.

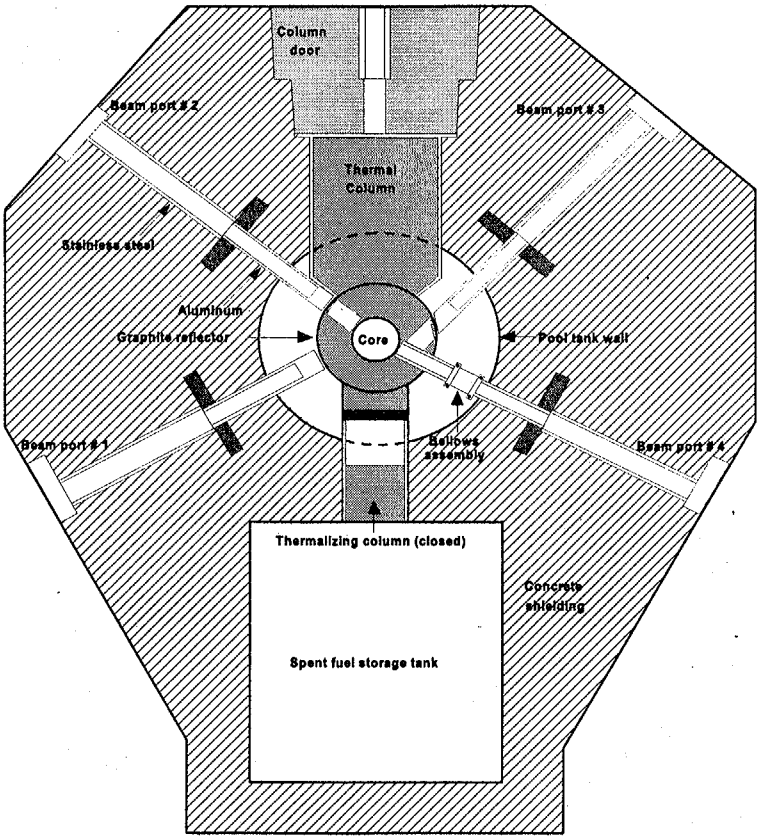

Fig. 5. Horizontal section view of the Dalat research reactor

IV. CONCLUSION

The Dalat reactor has been safely operated for 19 years. To achieve that, maintaining and upgrading the reactor technological facilities have been done with a high quality. The reactor physics and thermal hydraulics studies have also provided the important bases for safety evaluation and in-core fuel management to ensure its safe operation and effective exploitation.

Taking this opportunity, we would like to express our thanks to Japanese Government for co-ordination and financial support for this collaboration program.

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Workshop

Tc Generator

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1.9 The Performance of Gel Technetium-99m Generator

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Abstract

Technetium-99m, as one of the important radionuclides in nuclear medical science, has been widely used for diseases diagnosis in both developed and developing countries for many years. Technetium-99m can be obtained from both fission-type and gel-type Tc-99m generator. Fission-type generator was prepared by Molybdenum-99 separated from fission products of uranium-235 and gel-type was prepared by irradiating nature MoO_3 in reactor, and a series of chemical and physical processes.

This paper briefly describes the manufacturing technical process of gel-type Technetium-99 generator, including the preparation of target containing nature MoO_3, the target irradiation in reactor, gel preparation, gel filtration and drying、dried gel cracking、generator loading and activity calibration of generator. The performances of gel-type Technetium-99m generator, such as elution efficiency, elution profile, the pH, Mo breakthrough, Zirconium content, radiochemical purity, radionuclidic purity, sterility and pyrogencity of eluate, are also expatiated in detail.

Comparing with fission-type Technetium-99m generator, the defects of gel-type Technetium-99m generator are enumerated and their overcoming solutions are recommended in this paper.

Key words:
Gel Technetium-99 generator　Zirconium Molybdate　performance

1. Introduction

Tc-99m, which was widely used in hospital for diseases diagnosis, can be obtained by eluting Tc-99m generator. Nowadays, two types of Tc-99m generators are provided in the world market—fission-type Tc-99m generator and gel-type Tc-99m generator(differentiated by their source of its parent—Molybdenum-99). Because of the better performance of fission generator, such as high concentration of Tc-99m in eluate, narrow elution peak, high elution efficiency, light weight of shielding container, the fission generator is most frequently used in the world. However, because of the requirement of elaborate manufacturing facilities and the environment pollution problem, more scientists strive to develop another method of acquiring Mo-99 from neutron activation of nature or enriched Mo-98 and great achievements have been made. Especially, the elution efficiency was greatly promoted and elution peak profile was improved by improving the conditions of gel preparation and gel drying. NPIC Has solved the problem of quickly and semi-quantitively loading generator in 1986, and started to produce gel-type Tc-99m generator on large scale. The gel Tc-99m generator produced in NPIC occupies 30% ~40% of the Chinese market and fission Tc-99m generator produced in CIAE occupies 50%~60%. The rest market is occupied by imported fission Tc-99m generator.

2. The technical process of gel Technetium-99m generator preparation

Gel Tc-99m generator is produced by the following steps (Shows in fig.1) and the working environmental conditions are different from steps to steps.

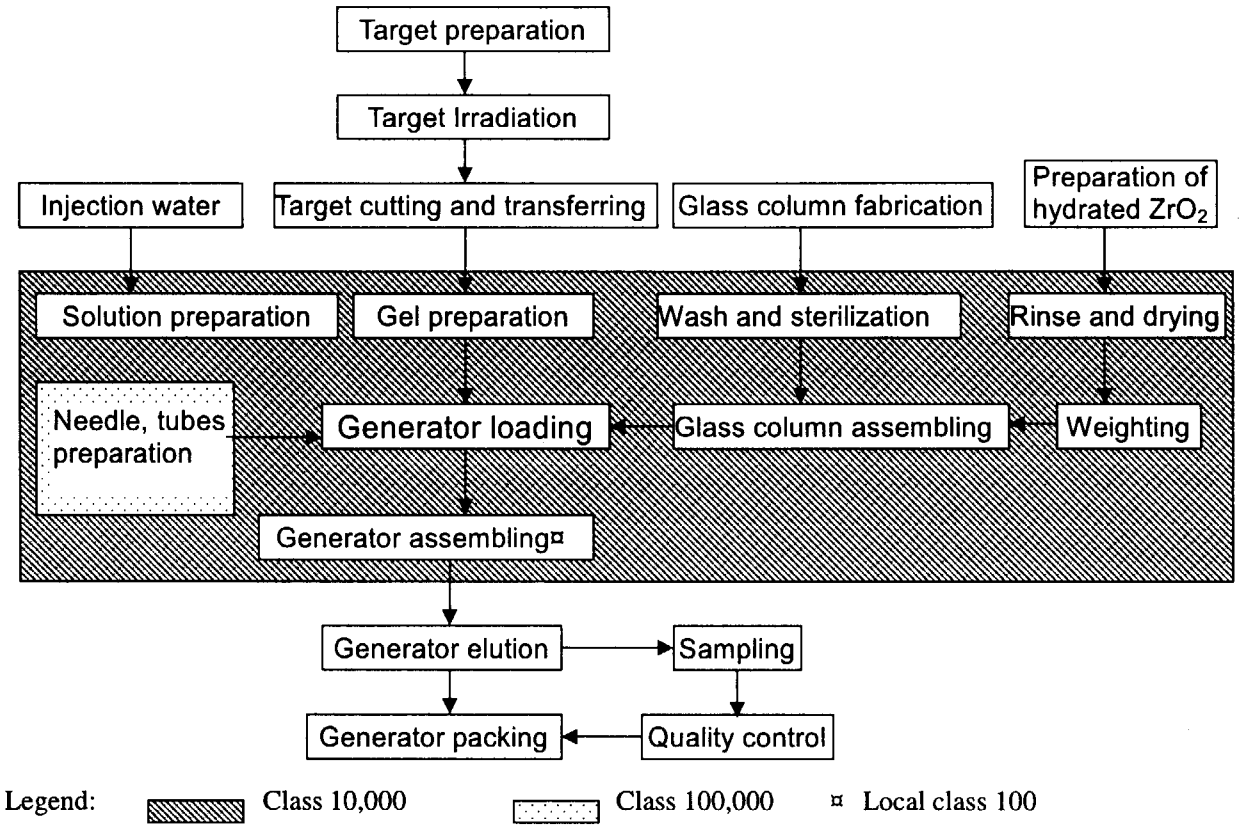

Fig.1 The procedure chart of Gel Tc-99m generator production

2.1 Target preparation and irradiation

High pure natural molybdenum trioxide powder is sealed in double capsules (quartz tube inside capsule and pure aluminum outside capsule) and is irradiated in MJTR (5MW) or HFETR (125MW). The activity of every generator depends on the irradiation time, irradiation position in reactor and loaded quantity of Zirconyl Molybdate gel in glass column. The quantity of MoO_3 is depended on the required generator number. Generally, the target is irradiated in MJTR for 9days, the average activity of one generator is approximate 1.0~1.5Ci, and irradiated in HFETR for 2~3day, the average activity of one generator is 1.5~2.2Ci.

2.2 Gel preparation

▼Transfer the quartz tubes containing irradiated MoO_3 to the chemical hot-cell and wash them with pure water and injection water respectively;

▼Crash the quartz tubes and dissolve the MoO_3 with NaOH solution to form $NaMoO_4$ solution;

▼Neutralize the above solution with 1mol/L HNO_3 solution to be pH≈4.5;

▼While stirring, add the above solution slowly to a stainless steel container containing $ZrOCl_2$ solution;

▼ While stirring, adjust the pH of above mixed solution to be pH≈4.5 with NaOH solution to form Zirconium Molybdate gel;

2.3 Gel filtrating, drying and dried gel disintegrating

▼Filtrate the gel solution by vacuuming and dry the filtrated gel at the temperature of 105±5℃ with running clean hot air for 7~8hours;

▼For easy loading of generator, the dried gel is transferred to a container containing saline to disintegrate the gel into uniform particles and the rinse the particles with saline to remove the fine particles.

2.4 Generator loading, assembling and leakage testing

▼Automatically loading the suitable size particles into glass columns with required volume of gel particles;

▼Connect the glass column with silicon pipes, elution needles and other components and put the loaded glass columns into lead shielding , plastic houses;

▼Take the leakage test by eluting the generator with saline.

2.5 Activity calibration of generator

▼After six hours, elute the generator with physiological saline, the activity of generator can be calculated by measuring the activity of eluate. The activity of gel Tc-99m generator is marked with the activity of Tc-99m in 12ml eluate eluted from generator.

3. The performance of gel-type Technetium-99m generator and their effect factors

Unlike Mo-99 in a fission-type Tc-99m generator, neutron activated Mo-99 in gel generator is chemically fixed by incorporating itself into insoluble zirconium molybdate gel.

The performance of gel Tc-99m generator includes appearance character, pH of eluate, elution efficiency, elution profile, radionuclidic purity, radiochemical purity, Zirconium content, Mo-99 break-through, biologic character and the weight of generator unit etc. Their requirement are listed as follows:

Column	ZrMo gel
Weight of gel loaded in one generator, g	5~10
Weight of hydrate ZrO_2 loaded in one column, g	1.5~2
Appearance character of eluate	Colorless and clear
Radioactivity of Mo-99, Ci	0.5~3.0
pH of eluate	4.0~7.0
Elution efficiency of Tc-99m, %	70~80
Peak width of elution profile, ml	5~9
Mo breakthrough, μ g/ml	<2
Zirconium content, μ g/ml	<2
^{99}Mo / ^{99m}Tc, %	<0.01
Biological test	Sterile and pyrogen-free
Useful life	2 weeks
Unit weight, kg	35

The performance of gel Tc-99m generator can be affected by the following factors:

1) The irradiation time and irradiation position of MoO_3 in reactor, quantity of gel particles loaded in generator affect the radioactivity of generator;
2) The gel preparation conditions (such as molar ratio of zirconium and molybdenum, pH of reaction mixed solution, etc.), drying temperature of gel, particle size of disintegrated gel, volume of gel

loaded in glass column and ingredient of disintegrated gel rinsing solution can significantly affect generator's elution performance (Efficiency and elution profile);

3) The pre-treatment method and loaded quantity of hydrated ZrO_2 in glass column affect the pH, Mo-99 breakthrough and Zr content of eluate;
4) The volume of saline used for disintegrated gel rinsing affects the radionuclidic purity and Mo-99 breakthrough.

4. The differences of gel Tc-99m generator performance comparing with that of fission Tc-99m generator

Both gel generator and fission generator are produced and supplied to Chinese market. The fission Tc-99m generator is produced by China Institute of Atomic Energy (CIAE) and gel Tc-99m generator is produced by Nuclear Power of China (NPIC). The following table presents the performance of the two type of Tc-99m generator

Table 1 Performance comparison of gel and fission Tc-99m generator

Item	Fission Tc-99m generator	Gel Tc-99m generator
Raw material	HEU/LEU, hard to get	MoO_3, easy to get
Capital cost	High	Low
Waste treatment	Difficult and high expense	Easy and low expense
Radioactivity of Mo-99	Loaded as required	High than required
Elution efficiency of Tc-99m	>90%	65~80%
Peak width of elution profile,	Narrow (~3ml)	Broad (6~8ml)
Appearance character	Colorless and clear	Colorless and clear
pH of eluate		4.0~7.0
Radionuclidic purity	^{131}I<0.005% ^{99}Mo<0.05 ^{103}Ru<0.005% $^{89}Sr<6x10^{-5}$ % $^{90}Sr<6x10^{-6}$ % a<$1x10^{-7}$ % Other ß、γ<0.01%	^{99}Mo / ^{99m}Tc, <0.01% Other radionuclides: No detected
Radiochemical purity, %	>99	99
Mo breakthrough, μ g/ml	—	<2
Zirconium content, μ g/ml	—	<2
Aluminum content, μ g/ml	<10	—
Biological test	Sterile and pyrogen-free	Sterile and pyrogen-free
Glass column volume	Smaller	Bigger
Weight of generator unit	Light	Heavy

5. Conclusion

The quality of pertechnetate eluted from gel generator has no significant difference compared with that of fission generator. All the specifications of pertechnetate can meet the requirements of pharmacopoeia. Comparing with the gel Tc-99m generator, the treatment of fission by-products of Uranium-235 is a big problem for fission Tc-99m generator.

Simultaneously, lower elution efficiency, broad elution profile, lower Tc-99m concentration in eluate, and heavy generator unit is the shortcoming of gel generator. The big advantage of gel generator is lower expense of raw material, easy treatment of waste.

The gel Tc-99m generator can be acceptable by the customers.

1.10 The Utilization and Performance of Poly Zirconium Chloride (PZC) as Chromatographic Column for ^{99m}Tc Generator

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ABSTRACT

We report on the performance test of column ^{99m}Tc generator utilizing poly zirconium chloride as an adsorbent. Natural molybdenum trioxide was irradiated for 2 hrs at a neutron flux of about 1.5 x 10^{12} n/cm^2/s. A total of fourteen generators have been prepared from the experiments. Yield of ^{99m}Tc has been 60 – 93 % and measured ^{99}Mo content in eluates is 0.20 – 0.44 %. The adsorption efficiency of ^{99}Mo on PZC column is between 85 - 92 %. The flow rates from the elution are relatively very poor and take about 5 minutes to finish the elution of 1 ml saline.

Introduction

Presently ^{99m}Tc is mainly obtained from generators based on column chromatography over alumina and using ^{99}Mo of very high specific activity obtained from fission or by high flux neutron activation of molybdenum trioxide (MoO_3). Sublimation, solvent extraction, zirconium molybdate gel and titanium molybdate techniques have been studied as alternatives methods to replace the utilization of fission molybdenum as a parent to generates ^{99m}Tc. The feasibility of using PZC as a column matrix has been explored and studied more details, particularly after the work of Tanase *et al.*(1997). Three batches of high performance adsorbent poly-zirconium chloride received from Department of Radiosotopes, Japan Atomic Research Institute was used to prepare the ^{99m}Tc generators. We report our finding on the adsorption of molybdenum by PZC column, elusion efficiency and molybdenum content in the eluates.

Experimental

2 g of MoO_3 was irradiated for 2 hr at a neutron flux of about 1.5 x 10^{12} n/cm^2/s. This produces about 10 mCi ^{99}Mo at the end of irradiation. After cooling, irradiated material was dissolved in 5 ml of 6 M NaOH and dilute with water to 25 ml. The mixture contains 53.4 mg Mo/ml solution.

5 ml was removed from the solution and pH of the solution was adjusted to 7 with 1 M HCl. The volume is adjusted to 10 ml with water. 1 g of PZC was added to the solution and keep at either 70 ^{0}C or 90 ^{0}C for 3 hours in a water bath. Percentage of ^{99}Mo adsorbed by PZC is then estimated by measuring the radioactivity of ^{99}Mo in the remaining solution after loading into column.

The PZC absorbing ^{99}Mo is packed into a glass column, with a glass filter of 0.20 μm at the bottom, of 8 mm inner diameter and 50 mm length. The column is then washed with 50 ml saline and taken this time as t = 0. Elution is done at the interval of 24 hr since last elution and for four consecutive days. Desorption rate of ^{99}Mo was estimated by measuring the total activity of eluate using CRC dose calibrator.

Results and Discussion

Adsorption of ^{99}Mo to PZC

Result in Table 1 shows that the adsorption rate of ^{99}Mo solution at pH 7 to PZC is between 80 – 92 % and the estimated error from the measurements is ±1%. Variation in the adsorption rate is probably due to the lost of radioactivity during loading of solution into column. Adsorption capacity may vary significantly on pH of the solution adjusted and maintained at 7. In our study, the measurement of pH is done using pH paper indicator and assumed to be accurate enough in the experiments. Work reported by Department of Radioisotope, JAERI on the High Performance Mo Absorbent PZC in the previous workshop suggested that changes from pH 7 to 8 of solution produces about 4 % reduction in the absorption capacity. Our results show that variation in the adsorption capacity is within ±10 % which suggests pH of moly solution shall be at 7 more accurate. Changes of temperature solution between 70 and 90 ^{0}C has no significant effect on the capacity of PZC to adsorb moly.

Table 1: Percentage adsorption of ^{99}Mo solution at pH 7 to Poly Zirconium Chloride.

Column	Adsorption (%)	Temperature of solution (^{0}C)
1	90	90
2	85	70
3	88	90
4	92	90
5	91	90
6	87	90
7	79	70
8	82	90
9	88	90
10	85	90
11	91	90
12	82	70
13	84	90
14	87	90
15	82	90

Elution Profile

Figures 1- 3 show the elution profile from PZC Tc generators prepared under the procedure described previously. Column number 1 was discarded due to some errors in the preparation. A total of 8 ml saline is used for every elution. Results from cumulative activity measured indicated that between 80 – 90 % of ^{99m}Tc has been removed from the column by the first 5 ml saline and typical example from the results of elution is Table 2. Elution in done by gravity in all cases.

Table 2: Elution activity for the first 5 ml saline cumulative activity from column 6 – 10.

Elution day	% removed ^{99m}Tc from Column no.				
	6	7	8	9	10
1	86	94	90	88	87
2	88	90	85	82	80
3	86	92	79	75	68
4	81	88	81	76	74

Molybdenum content in the eluates

Result in table 3 shows the average desorption rate for all generators is about 0.20%. In all cases, higher desorption rate is observed. This suggest that he use of small alumina column which act as a second column to minimize the molybdenum breakthrough is unavoidable and very substantial.

Table 3: The percentage average desorption of molybdenum in the eluted ^{99m}Tc solution from PZC column generators.

Column no.	Average desorption (%)
1	-
2	0.44
3	0.28
4	0.21
5	0.24
6	0.24
7	0.26
8	0.24
9	0.23
10	0.23
11	0.22
12	0.23
13	0.24
14	0.21
15	0.25

Conclusion

It is apparent that PZC based Tc generator can be prepared by low neutron flux irradiation of natural molybdenum trioxide powder. Elution profile for fourteen generators prepared show acceptable elution efficiency when compared with commercially available fission product Tc generators. Molybdenum content is higher than specified by British Pharmacopea, which is should be less than 0.1%. In this case, second alumina column is required to reduce the moly breakthrough. We need to further investigate elution pattern for a longer periods. Our work suggests that PZC can be prepared but for higher radioactivity preparation, a suitable handling technique, or probably suitable processing plant must be developed to reduce radiation exposure to operators..

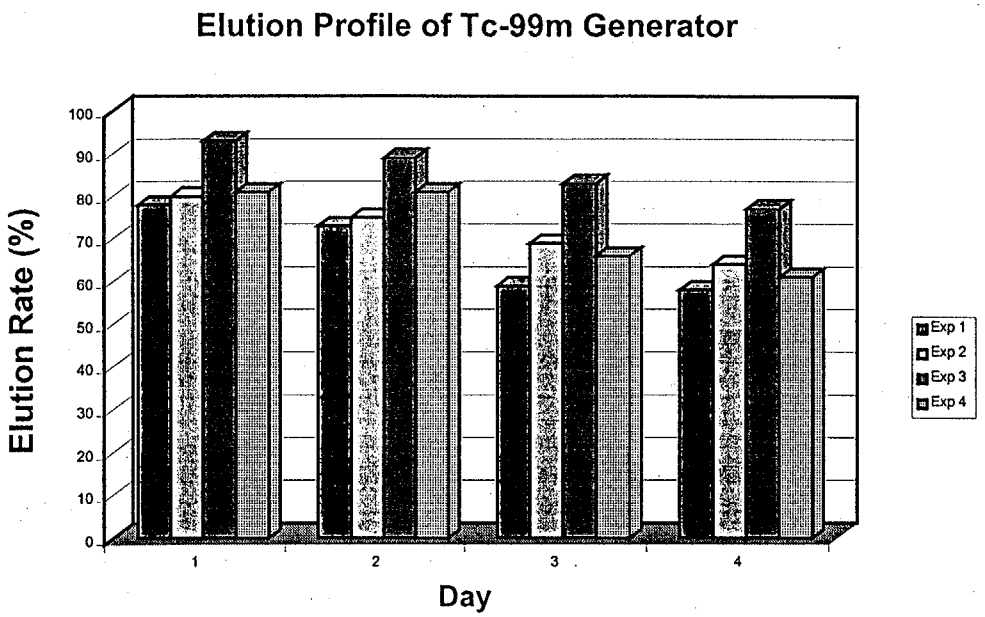

Figure 1: The elution profile of ^{99m}Tc PZC generator obtained from 4 days elution.

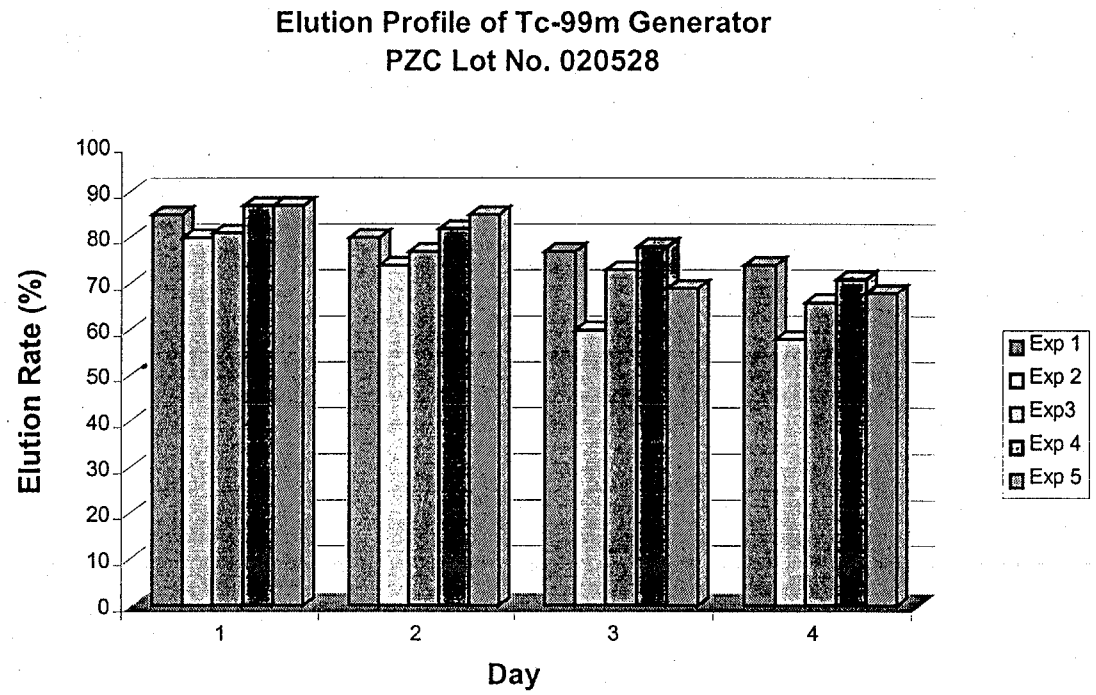

Figure 2: Elution profile of ^{99m}Tc PZC generators from lot No. 020528

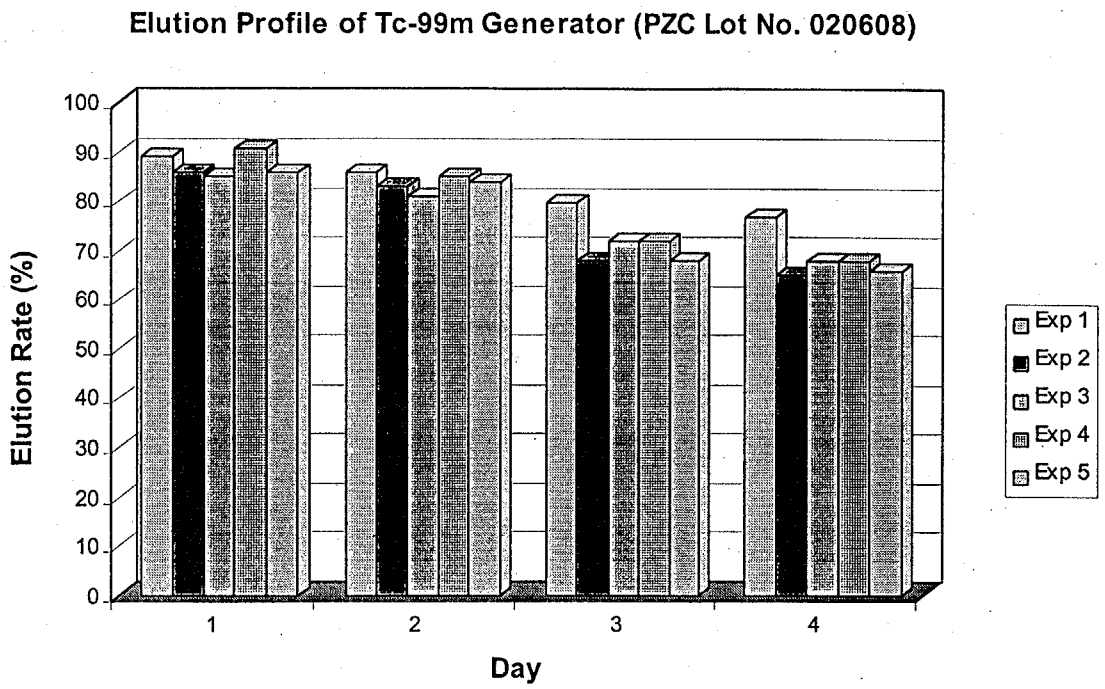

Figure 3: Elution profile of ^{99m}Tc PZC generators from lot No. 020628

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1.11 A Comparative Study on the Labelling of Radiopharmaceutical Kits with ^{99m}Tc Obtained from F.P. ^{99}Mo/^{99m}Tc Generator and PZC- Based ^{99}Mo/^{99m}Tc Generator

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ABSTRACT.

A Comparative Study on the Labelling of Radiopharmaceutical Kits with ^{99m}Tc Obtained from F.P. ^{99}Mo/^{99m}Tc Generator and PZC- Based ^{99}Mo/^{99m}Tc Generator.
Labelling comparison of radiopharmaceutical kit with ^{99m}Tc obtained from different generator may assist in the evaluation of a generator quality. Technetium-99m from ^{99}Mo/^{99m}Tc Generator based on fission product is commonly used in nuclear medicine all over the world. A ^{99}Mo/^{99m}Tc generator was prepared by reacting high activity ^{99}Mo (± 300 mCi) of neutron natural molybdenum with PZC materials. The generator resulted in high ^{99m}Tc yields and low ^{99}Mo breakthrough in the presence of a low concentration oxidizing agent ($NaOCl^-$). Technetium-99m obtained from both generators were labelled to pyrophosphate, HMPAO and MIBI kits, respectively. The chemical and biological properties of ^{99m}Tc obtained from both generators afford the similar radiochemical purity and biodistribution pattern.

Keywords : radiopharmaceutical, PZC, generator, ^{99m}Tc, kits

INTRODUCTION

Technetium-99m , the daughter of ^{99}Mo, is the most commonly used radioisotope in nuclear medicine. It is used in over nine million medical procedures annually in the US and comprises 80 % of all nuclear medicine procedures in the world. By the milking procedure, it is normally eluted as the daughter isotope of molybdenum-99 from ^{99}Mo/^{99m}Tc Generator. Molybdenum-99 is typically produced by the thermal neutron fission of ^{235}U, the targets are ussually from HEU or LEU. Molybdenum-99 also can be generated by neutron capture in targets of natural molybdenum or targets that have been enriched in ^{98}Mo, however, neutron capture produces ^{99}Mo with low specific activity compared than from fission product.

The development of low specific activity of ^{99}Mo produced by (n, γ) nuclear reaction for use in gel generators of zirconium or titanium molybdate encourages several countries to consider it as an alternative one even though it needs a rather complicated process. Recently, a new adsorbent named PZC (Poly Zirconium Compound) has been developed by Kaken Co., Japan, that have adsorbability on molybdate higher 100 times than alumina shown as a promising adsorbent in the (n, γ) ^{99}Mo-^{99m}Tc Generators (2-4).
The previous experiment on preparation of ^{99}Mo-^{99m}Tc Generators using high activity ^{99}Mo (> 10 GBq) loaded to the PZC materials indicated that the presence of a low concentration oxidizing agent ($NaOCl^-$) is necessary to maintain the high ^{99m}Tc yields and low ^{99}Mo breakthrough (5)

This paper will reports the biological evaluation (biodistribution) of some radiopharmaceutical kits labeled with ^{99m}Tc from fission and PZC ^{99}Mo-^{99m}Tc Generators.The evaluation will be done when the radiochemical purity of the kits is good and biodistribution will be done on mice and some experiment using Gamma Camera also will be done on rats.

EXPERIMENTAL.

PZC materials was synthesizes by KAKEN Co., Japan which has the adsorption capacity of Mo (Mo-99) about 280 mg/g. Molybdenum-99 was produced by irradiating 2 g natural MoO_3 targets for a week in the thermal fluxes of 1.2×10^{14} n. Cm^{-2}. S^{-1} in RSG-GAS (multi purpose reactor of BATAN). Molybdemun-99/^{99m}Tc PZC-based Generator was made by CDRR (Center for Development of Radioisotopes and Radiopharmaceuticals)-BATAN (activity ± 270 mCi) and ^{99}Mo/^{99m}Tc Fission Generator was made by P.T Batan Technology- Indonesia using alumina as adsorber with the activity is about 208 mCi.

In this experiments, normal white mouse and rats were used for biological distribution and gamma camera imaging of radiopharmaceutical kits (Pyrophosphate, HMPAO and MIBI). Biodistribution studies was carried out on healthy normal Swiss

white mice, age between 6-8 weeks and weight between 25-35 grams. The radiochemical purity of the three complexes was more than 90 % except for HMPAO more than 85 %.

The complexes of the kits were injected through the tail vein of the mice with 0.1 ml of the complex containing 3.7-7.4 MBq of activity and then the animals were killed after certain interval time. Organs were collected, weighed and counted in a well type gamma counter to calculate resident activity in different organs. The percent distribution in organs were calculated as percent injected dose per gram (% ID/g).

Imaging studies was carried out on normal white Wistar rats. The rats was injected through the tail veil of radiopharmacuticals and images were evaluated after certain interval time depend on the radiopharmaceuticals.

RESULTS AND DISCUSSION.

Radiochemical purity of ^{99m}Tc obtained from the two generator (fission and PZC generators) were ascertained by paper chromatography using methanol 85 % as eluent and Whatman I paper as a static phase. In this system more than 99 % of the two generator eluate as pertechnetate ions ($^{99m}TcO_4^-$) and the remaining as colloid of technetium (Table 1).

The data of radiochemical purity of Pyrophosphate, HMPAO and MIBI kits are summarized in Table 1 and each kits labelled with ^{99m}Tc from different generators (Fission and PZC generators). The data shown radiochemical purity for both Pyrophosphate and MIBI kits indicated more than 95 % as a complexes and about 5 % as free pertechnetate and technetium colloid. For HMPAO kit, more than 85 % as a complex and the remains as a secondary complex, free pertechnetate and technetium colloid.

The tissues and organs distribution of ^{99m}Tc Pyrophosphate summarized in Figure 1 and Figure 2 , the data obtained after 2 hours intravenous injection of the complex on mouse. The results of the biodistribution of the ^{99m}Tc-pyrophosphate from the two generators reveal that bone uptakes of the complexes at 2 hours post injection almost the same. The target/non target ratio (bone / bl;ood) were very high . It's mean that the compound will be producing a good skeletal images for visualization by gamma camera (Figures 3).

The tissues and organs distribution of ^{99m}Tc-HMPAO in selected organs summarized on Figure 4 , the data obtained after 5 minutes and 60 minutes after intravenous injection. Accumulation of the HMPAO complex on the mouse brain of the two HMPAO labelled with ^{99m}Tc from the different generator indicated not too different and ratio of the others organ also similar. The high activity in the kidney evidently due to the partial elimination of the complexes in the urine. Some radioactivity in the liver could have originated from the uptake of colloidal form ^{99m}Tc or from some biotransformation product.

Biodistribution pattern of the ^{99m}Tc-MIBI labelled with ^{99m}Tc from Fission and PZC generator is illustrated in Figure 5 and Figure 6, all of the data taken 10 minutes and 120 minutes post injection. Both ^{99m}Tc-MIBI complexes from Fission and PZC generator showed high activity in the blood at 10 minutes p.i. (2.78 % and 2.69 % ID per organ respectively) and the accumulated activity decreased 120 m p.i. (1.87 % and 1.86 % ID per organ respectively), but from these compounds indicated rapid blood clearance (0.02 % and 0.02 % ID per organ respectively).

CONCLUSIONS

In conclusions, the present paper demonstrated that ^{99m}Tc from two ^{99}Mo/^{99m}Tc Generators (Fission and PZC) indicated the same chemical characteristics. The gathered radiochemical purity and biodistribution data from the three radiopharmaceuticals kits (Pyrophosphate, HMPAO and MIBI) indicated well reflected the characteristic of the selected kits when labelled with ^{99m}Tc from the two generator.

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Table 1. Radiochemical Purity of ^{99m}Tc and Pyrophosphate, HMPAO, MIBI Kits Labelled with ^{99}Mo/^{99m}Tc Generator.

Substances	% radiochemical purity		Methods
	Fission Gen.	PZC Gen.	
^{99m}Tc	99.7	99.6	PC
Pyrophosphate	97.4	97.2	PC
MIBI	99.2	99.3	PC
HMPAO	90.22	89.93	PC

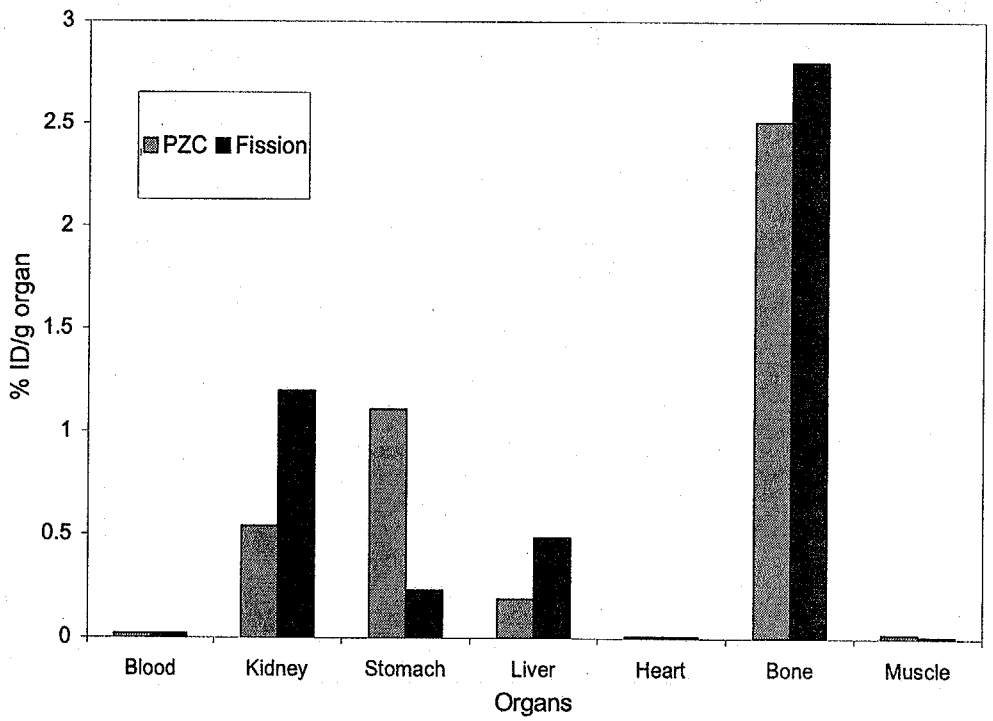


Figure 1 . Biodistribution Pattern of Pyrophosphate Labelled with ^{99m}Tc from Fission and PZC Generator

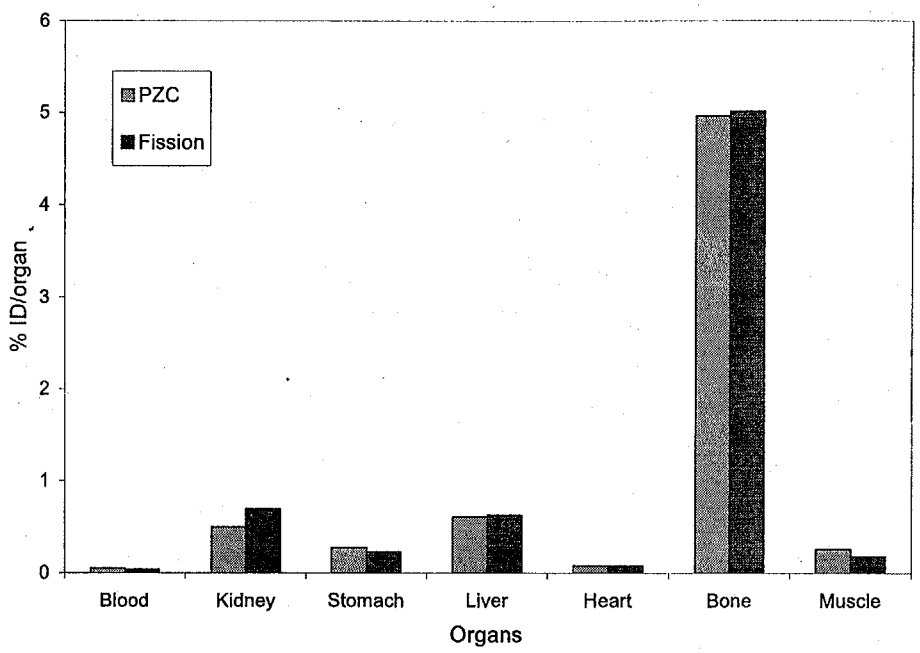
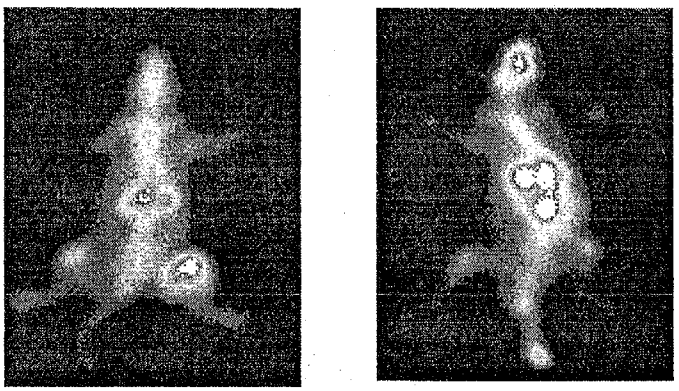


Figure 2 . Biodistribution Pattern of Pyrophosphate Kit Labelled with ^{99m}Tc From Fission and PZC Generator.



^{99m}Tc-Pyp (PZC)　　^{99m}Tc-Pyp (Fission)

Figure 3. Images of Pyrophosphate Kit Labelled with ^{99m}Tc from Fisasion and PZC Generator.

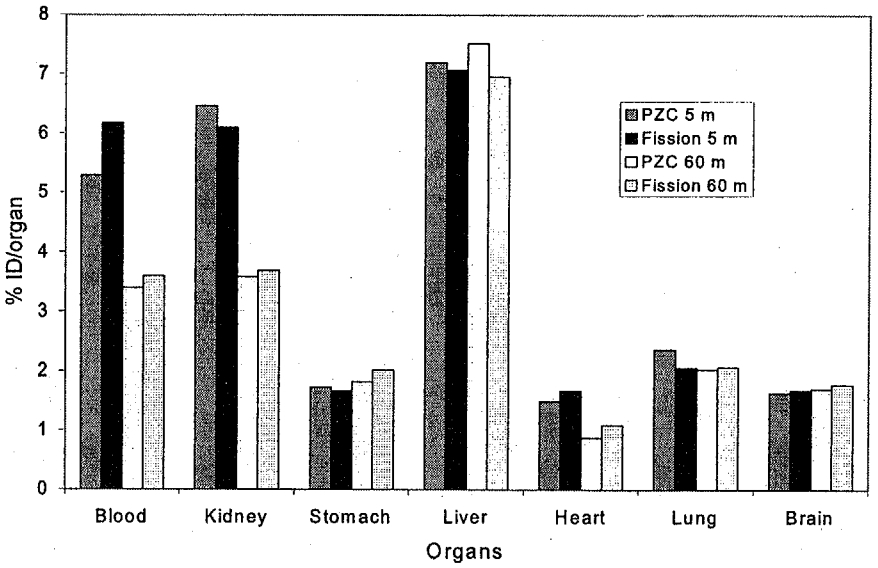


Figure 4. Biodistribution Pattern of HMPAO Kit Labelled with ^{99m}Tc from Fission and PZC Generators

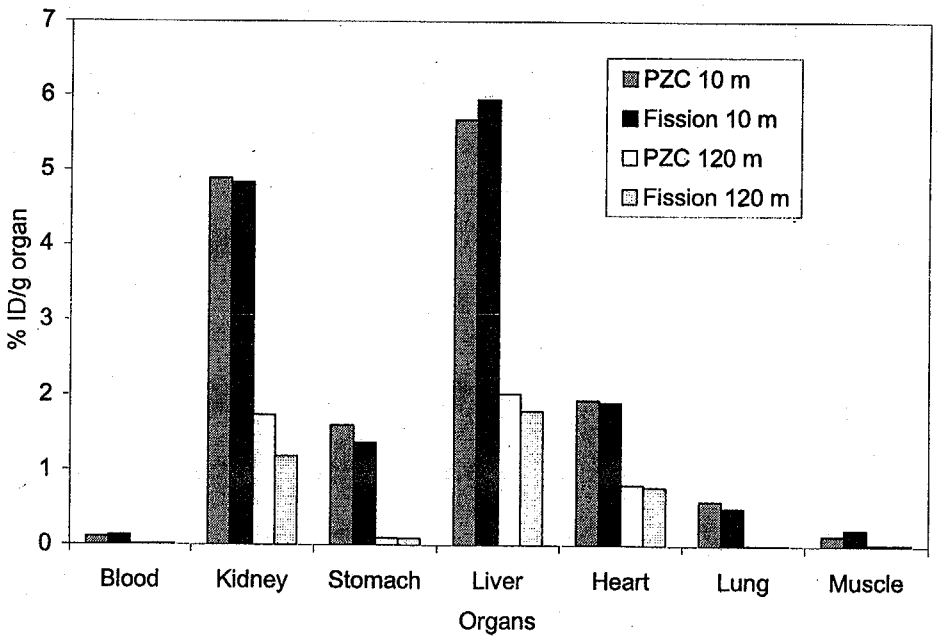


Figure 5. Biodistribution Pattern of MIBI Kit Labelled with ^{99m}Tc from Fission and PZC Generator.

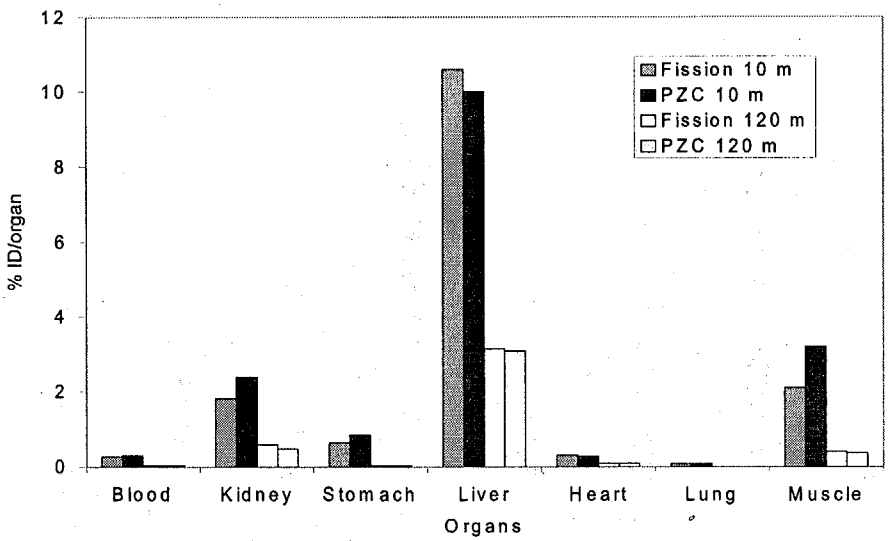


Figure 6. Biodistribution Pattern of MIBI Kit Labelled with ^{99m}Tc from Fission and PZC Generator

1.12 PERFORMANCE TESTS ON NEW CHROMATOGRAPHIC MATERIAL FOR ^{99}Mo-^{99m}Tc GENERATORS[1]

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ABSTRACT

Technetium-99m continues to be the main workhorse of nuclear medicine in the Philippines. Almost 13TBq of ^{99m}Tc was imported to the country in 2002 supplied as ^{99m}Tc-^{99}Mo generators. These generators make use of fission molybdenum adsorbed onto an alumina column. Problems associated with the alumina chromatographic generators arise due to safety and economic issues that would be remedied by gel-type generators using low specific activity reactor-produced molybdenum-99 adsorbed on a high capacity gel column material. The Philippine Nuclear Research Institute (PNRI) exerted efforts in this direction by developing a gel-type column, which showed satisfactory molybdenum adsorptive capacity. Likewise, Kaken Co. in Japan in cooperation with Japan Atomic Energy Research Institute (JAERI) developed a dried form of a gel-type polyzirconium compound (PZC). It is a ready-to-use high molybdenum capacity column material for adsorbing reactor-produced molybedum-99. The performance of this material is being tested under the framework of the FNCA project on Research Reactor Utilization.

Performance tests on four batches of PZC were performed using fission molybdenum eluted from a ^{99}Mo-^{99M}Tc generator. A total of 3.3 GBq ^{99}Mo was extracted from an alumina column of a commercial generator and mixed with carrier molybdenum solution. About 0.67 GBq was loaded into each of the 12 x 90 mm column. One batch was prepared and distributed in 1999 and tests showed very poor elution yield of 30%. Three recent batches of PZC (2002) gave elution yields of 71% (Range of 69-75). The adsorptive capacity is 99% with about 4% desorption rate. Elution volume is at 5-6 ml. Daily elution for five days gave from 1.6 to 5.5% variability. The tests were performed all at the same time and a trend of improving elution yield and consistency of daily elution yield was observed with the time of testing nearer to the sample preparation date.

[1] *Paper presented during the 2002 FNCA Workshop on the Utilization of Reasearch Reactors, Serpong, Indonesia, 13-17 January 2003*

X-ray diffraction analysis showed an amorphous structure for all three batches although PZC020522 showed the presence of a small amount of crystalline structure in the sample. X-ray fluorescence analysis gave about 40% zirconium in the sample.

These results need to be compared with the results of tests obtained at higher activity of molybdenum-99 to determine any radiation effect that may affect the elution behavior of the column material. The rate of decomposition of the gel, which is affecting its performance characteristics, needs to be defined.

I. INTRODUCTION

Technetium-99m (^{99m}Tc) remains as the main workhorse of nuclear medicine in the country. Alone or conjugated with other ligands, it is being used to show the function of major organs and other tissues such as the lung, brain, kidney, liver and bone. A three-year profile of imports of radioisotopes used as radiopharmaceutical is shown in Table 1. In addition, radioactive sources like Ir-192, Cs-137, I-125 and Sr-90 have been used in brachytherapy.

Table 1. List of Radioisotopes used in Nuclear Medicine from 2000 to 2002, in GBq[2]

Radioisotope	2000	2001	2002
Tc-99m	9.76×10^3	10.76×10^3	12.78×10^3
I-131 (solution and capsule)	3.55×10^3	5.00×10^3	6.11×10^3
I-131 (mIBG)	-	1.0×10^{-1}	3.54×10^{-1}
I-125 (RIA kits)	2.45	1.3	1.27
Tl-201	1.52×10^2	1.54×10^2	2.5×10^2
Ga-67	1.9	2.12	4.14
In-111	3.22×10^{-1}	-	-
Sm-153	-	2.5	-

About 13 TBq of ^{99m}Tc was imported in bulk as ^{99}Mo-^{99m}Tc in 2002. There are 21 hospitals with nuclear medicine facilities including a gamma scanner, most of which are located in MetroManila. Last year, St. Luke's Medical Center in Metro Manila, launched the first and only Positron Emission Tomography Scanner (P.E.T.) in Southeast Asia. Unlike conventional scans like the Magnetic Resonance Imaging (MRI) and Computerized Tomography (CT) that only provide images of organ anatomy or structures, P.E.T. can provide a direct measure of biochemistry and functional activity. At present, a cyclotron-produced Fluorine-18 FDG is used as the radioactive tracer. However, the key benefits associated with the use of technetium-99m including its lower cost in relation to Fluorine-18 FDG (PET) will continue the increasing use of ^{99m}Tc in nuclear medicine applications.

The imported commercial ^{99}Mo-^{99m}Tc generator makes use of fission product molybdenum-99 immobilized on an alumina (Al_2O_3) column. Problems associated with the alumina chromatographic generators arise due to complex and expensive technology involved in the production of both fission molybdenum as well as the generators. Added to this is the complex management of toxic fission product wastes generated in the preparation of ^{99}Mo The safety and economic issues could be remedied by gel-type generators using low specific activity reactor-produced molybdenum-99 adsorbed on a high capacity column material.

[2] Import data provided by the Licensing Section, NRLSD, PNRI.

Alternatively, ^{99}Mo can be incorporated into a gel matrix and the gel is used as the column material from which Technetium-99m is eluted. This latter approach was explored at the Philippine Nuclear Research Institute (1).

Zirconium molybdate gel was prepared by mixing molybdenum oxide with tracer amounts of ^{99}Mo. Several procedures were tried (2,3) and the products were characterised mainly for crystal structure by x-ray diffraction analysis, Zr:Mo ratio and Zr and Mo contents by x-ray fluorescence spectrometry. It was noted that the products obtained varied in the ease with which the gel can be filtered, washed and dried. It was also observed that the structure of the zirconium molybdate gel depends on one or more critical factors most important of which is the pH of the reaction mixture. An amorphous structure was found desirable for good elution yield.

Kaken Co. in Japan in cooperation with Japan Atomic Energy Research Institute (JAERI) developed a dried form of a gel-type polyzirconium compound (PZC). It is a ready-to-use high molybdenum capacity column material for adsorbing reactor-produced molybedum-99.

II. PERFORMANCE TESTS ON PZC

Three batches of PZC, 5 grams each, were sent by mail to PNRI during the year 2002 (Table 2). The performance of these materials was tested under the framework of the Forum of Nuclear Cooperation in Asia (FNCA) project on Utilization of Research Reactor. This activity was agreed upon in the previous working group meeting held in China in November 2001.

Table 2. PZC samples received and tested in November 2002

Lot no.	Quantity, grams	Date received	Sample code used
99P-1007	5	October 1999	PZC1
PZC020319	5	April 2002	PZC2
PZC020522	5	August 2002	PZC3
PZC020806	5	October 2002	PZC4

The tests should have been conducted upon receipt of the material, but due to logistical problems associated with the availability and use of high activity molybdenum-99, the tests were conducted all at one time (November, 2002) and using fission molybdenum-99 extracted from a commercial generator. A batch of PZC sent in 1999 (table 2), although way past the expiration date, was included in the test. Molybdenum-99 activity put into each column was 6.7 MBq (18 millicurie).

Stock Molybdenum and Tracer Solution
Four grams of molybdenum oxide was dissolved in 10 ml of 6M NaOH and diluted to 20 ml (130 mg/ml). Fission molybdenum was extracted from the alumina column of an imported generator (Nexta, South Africa, 150 GBq at calibration date) using 10 ml of 1:1 NH_3 solution. The extract contains about 3.3 GBq (90 mCi) of ^{99}Mo.

Adsorption of molybdenum
Two (2) ml of the stock molybdenum solution and two (2) ml of the radioactive molybdenum solution with 0.67 GBq (18 mCi) were mixed with one (1) gram of PZC sample (one trial per

batch). The pH of the mixtures was adjusted to nearly neutral pH and then equilibrated in a 90°C water bath for three (3) hours. The mixtures were lightly mixed by shaking the flask by hand intermittently during equilibration. At the end of three hours one (1) ml of the solution from the mixture was taken for molybdenum gamma measurement. The corresponding same amount of the original Mo-99 solution was counted and used as reference value for calculating the amount of molybdenum taken up by the adsorbing material.

Packing of column
The floating fine particles were removed by decantation before packing into a propylene column (12mm x 90mm). The columns were washed five times with five 10-ml saline solutions to remove free molybdenum and zirconium.

Elution yield determination
After allowing radioactive growth for 24 hours, ^{99m}Tc was eluted with 10-ml saline solution. The ^{99m}Tc activity was measured using an isotope calibrator (Victoreen CALRAD). The elution yield was calculated by comparing the activity of ^{99m}Tc with the theoretical amount of ^{99m}Tc resulting from the decay of ^{99}Mo adsorbed in the column. (Check calculations). The elution yield was measured for 5 consecutive days.

Desorption rate (molybdenum breakthrough)
The desorption rate was measured using one (1) ml of the technetium eluate. Molybdenum activity at 739 KeV was measured by gamma spectrometry using a germanium detector. The molybdenum activity corrected for PZC absorption factor was used as reference.

Elution volume
The elution profile for each column was determined by successive elution of 10 one-ml portions.

Structural analysis and zirconium content determination
Using a Xray diffractometer (Cu anode, Siemens x-ray generator, Phillips goniometer) the structure of the material was determined. The zirconium content was measured by XRF method (Kevex EDX-771)

III. RESULTS AND DISCUSSIONS

The results of the tests performed on the four batches of PZC samples are summarised in Table 3. Due to logistical limitations, only one trial for each batch had been performed. The tests were performed all at the same time and a trend of improving elution yield and consistency of daily elution yield was observed with the time of testing nearer to the sample preparation date.

The elution yield is presented graphically in Figure 1 for the four samples while Table 4 gives the daily elution yield values for the PZC samples. Figure 2 shows graphically the elution volume for each sample.

Table 3. Column properties of the PZC samples

Sample ID (Code)	% Mo Absorption	% Mo Desorption	%Elution Yield 5 days avg.	Elution Volume,ml
99P-1007 (PZC1)	97	ND	30 ± 2	6
PZC020319 (PZC2)	99	3.8	69 ± 3	5
PZC020522 (PZC3)	99	4.0	72 ± 3	5
PZC020806 (PZC4)	99	4.2	75 ± 1	5

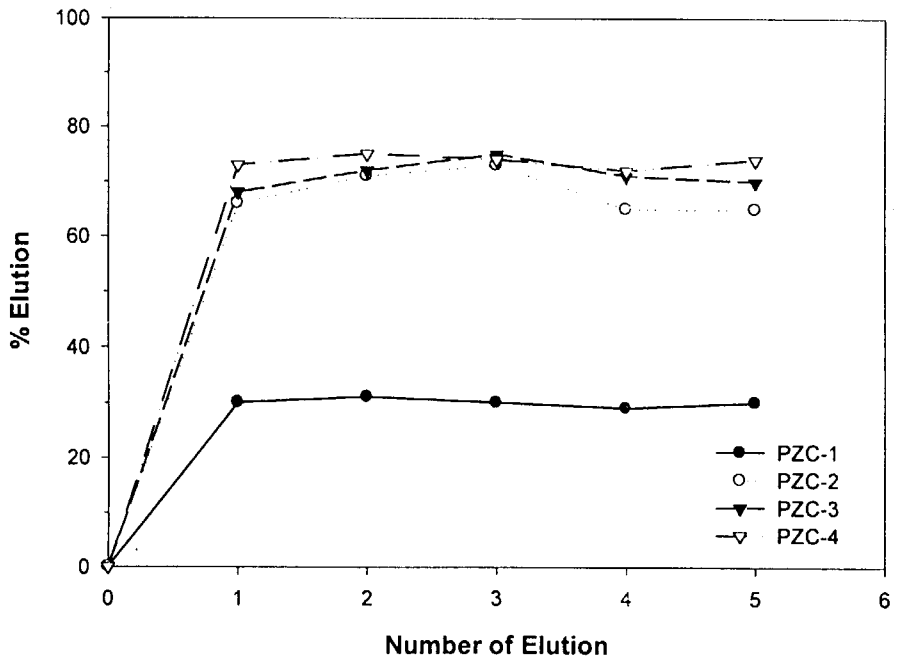

Figure 1. The elution yield of the PZC generators (2002) averages at 72%.

Table 4. Per cent elution yield and elution volume for each of the samples

Sample Code	Elution day					Avg. Elution Yield (%)	Elution Volume, ml
	1	2	3	4	5		
PZC1	30	30	30	29	30	30 ± 2	6
PZC2	66	73	73	65	65	69 ± 3	5
PZC3	68	75	75	71	70	72 ± 3	5
PZC4	73	74	74	72	74	75 ± 1	5

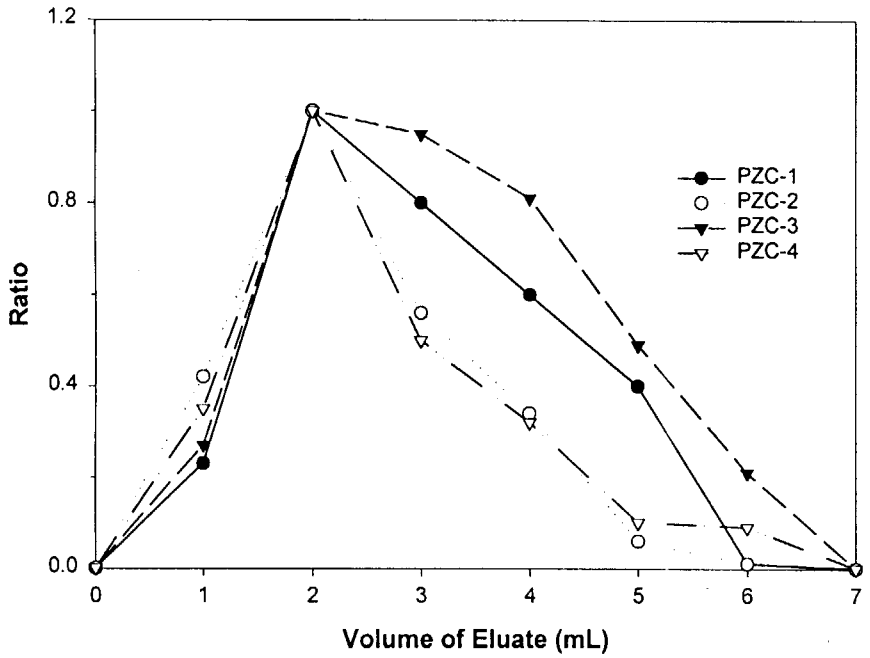

Figure 2. A five-ml saline is generally sufficient to elute the available technetium-99m from the column

The tests results of the first sample received by PNRI and which was distributed and tested in 1997 gave similar results (1). The average cumulative yield obtained (5 trials) was 73.5 ± 3.6% and the average day-to-day elution yield is 70.2 ± 1.7 %. Radiochemical assays (%pertechnetate) were performed by paper chromatography in methanol:water (75:15). The radiochemical purity of the eluate was >97%. The molybdenum-99 tracer was at nanocurie level and was also obtained from a commercial generator.

The zirconium content and structure of the samples are summarised in Table 5. Figure 3 shows the XRD patterns of the samples including that of pure ZrO. The samples were amorphous. PZC3 showed some unidentified peaks.

Table 5. Structure and zirconium content of the PZC samples

Sample ID (Code)	% Zirconium	Structure
99P-1007 (PZC1)	42	amorphous
PZC020319 (PZC2)	42	amorphous
PZC020522 (PZC3)	43	amorphous
PZC020806 (PZC4)	38	amorphous

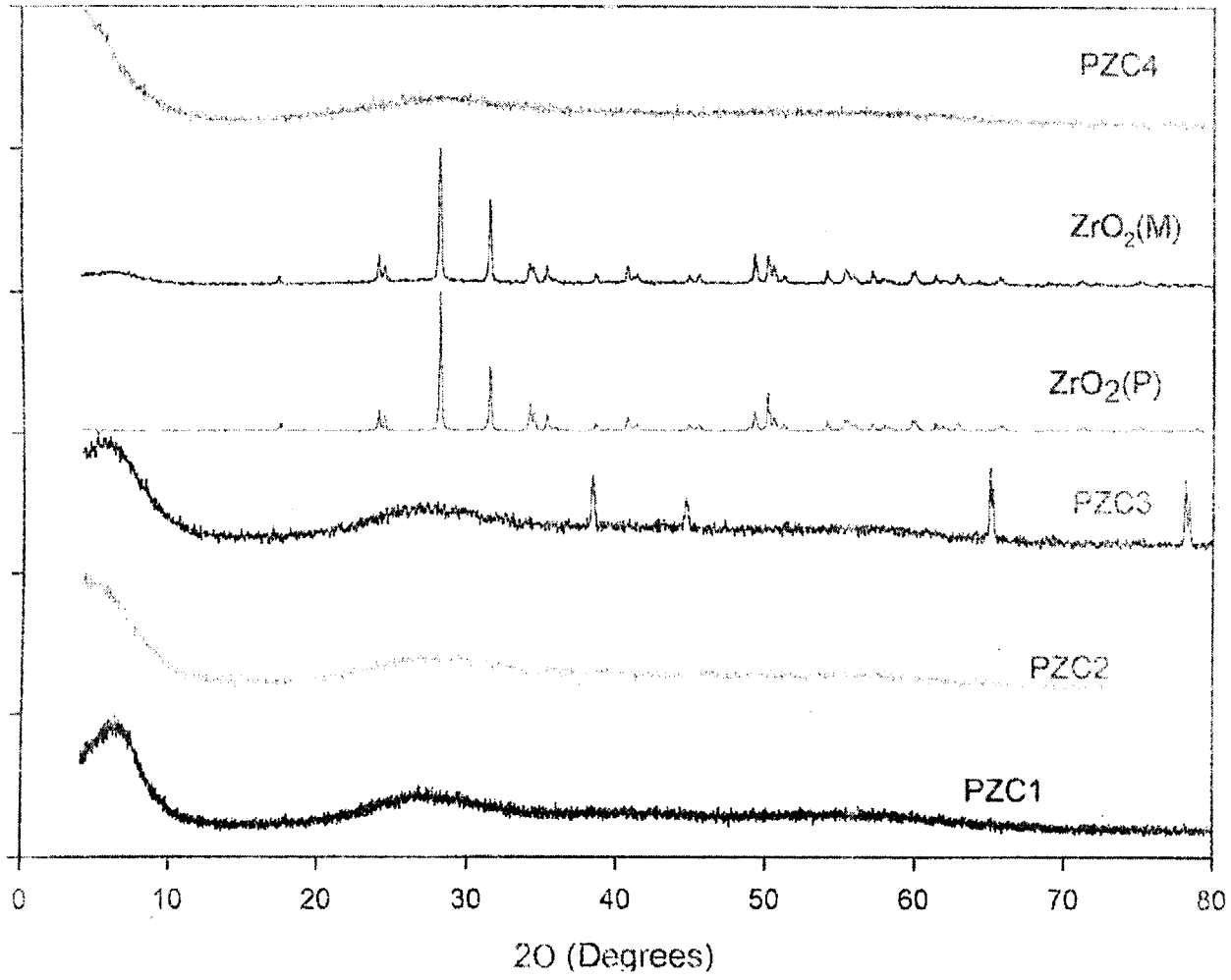

Figure 3. The PZC samples are amorphous although PZC3 exhibits some unidentified peaks.

IV. CONCLUSION

Recognizing the importance of ^{99m}Tc and ^{99m}Tc-based radiopharmaceuticals in nuclear medicine, the Philippine Nuclear Research Institute has initiated research on the development of column-type generators for ^{99m}Tc using reactor-produced ^{99}Mo. The concept is to incorporate low specific activity ^{99}Mo produced by the n,gamma reaction (with natural molybdenum as target material) into a zirconium molybdate gel matrix. This gel will serve as the column material from which the ^{99m}Tc is eluted. The advantage lies in the larger amount of molybdenum in the column material (40%maximum) compared with that of alumina currently in use (2%). This compensates for the lower specific activity attainable in the irradiation of natural molybdenum.

Although the process has been found useful for the preparation of commercial generators in some countries, the gel technology, as described, has several disadvantages: cumbersome procedures of filtering and drying of the gel, greater radiation hazard, dependence on operator skill, and long time required for the preparation. The availability of a pre-formed gel column material on which molybdenum can be adsorbed and from which technetium-99m can be eluted consistently and at high yield is a better option.

The results obtained for PZC, although limited in number of trials and the level of molybdenum activity loaded into the column, showed that the column holds promise as molybdenum column material for the generator. If the generator using this column can be available at lower cost, it is reasonable to project an increasing utilization of technetium-99 in medical applications in the country.

ACKNOWLEDGMENT
We wish to thank Syncor, Philippines for providing us with the spent commercial generators that we used for this study, the Irradiation Services Section for allowing the use of their isotope calibrator, the Applied Physics Research Section for the XRD analysis and the Analytical Measurement Research Section for the XRF analysis.

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JP0450628

JAERI－Conf 2004－010

1.13 EVALUATION OF $^{99}Mo/^{99}Tc$ GENERATOR EXPERIMENT USING PZC MATERIAL AND IRRADIATED NATURAL MOLYBDENUM

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ABSTRACT

Technetium-99m (^{99m}Tc) is the most widely used radioisotope in nuclear medicine, accounting for more than 80% of all diagnostic nuclear medicine procedure. ^{99m}Tc is almost exclusively produced from the decay of its parent molybdenum-99 (^{99}Mo). The present sources of ^{99}Mo are research reactors by using the (n,γ) nuclear reaction with natural molybdenum, resulting in inexpensive but low specific activity ^{99}Mo, or by neutron-induced fission of uranium-235, which result in expensive but high specific activity ^{99}Mo. The technology requirement for processing of ^{99}Mo from the (n, γ) "activation method" is rather simple, and is within the reach of most developing countries operating research reactors. In the "fission method" the technological and infrastructure requirements are some complex, and possibly can be sustained only by countries with advanced nuclear technology. To overcome these difficulties, Japan Atomic Energy Research Institute (JAERI) and KAKEN company have developed alternative technology for $^{99}Mo/^{99m}Tc$ generator by using a molybdenum absorbent called Poly Zirconium compound (PZC) and irradiated natural molybdenum.

The paper describes experiments for evaluation the performance of PZC as a column packing material for $^{99}Mo/^{99m}Tc$ generator from (n, γ) ^{99}Mo.

Keywords: $^{99}Mo/^{99m}Tc$ generator, polyzirconium compound, natural molybdenum

Introduction

Technetion-99m, the most widely used radioisotope in nuclear medicine, accounting for more than 80% of all diagnostic nuclear medicine procedures. Because of its excellent physical properties which make it an ideal radioisotope in organ imaging techniques. ^{99m}Tc is almost exclusively produced from the decay of its parent ^{99}Mo and most of the commercial $^{99}Mo/^{99m}Tc$ generators are still produced based on column chromatography over aluminium oxide which adsorbed ^{99}Mo obtained from fission products of ^{235}U. This type of generator has several disadvantages which limit its practical application. The major one is the limitation of the adsorption capacity for molybdate ions which requires the use of ^{99}Mo of high specific activity in the preparation of chromatographic generators of resonable size which allows rapid elution in small volume because of the limitation of the adsorption capacity of alumina for molybdate ions (20 mg. per gram of alumina). In countries which do not have a nuclear reactor with sufficient neutron flux and technology to handle fission products of ^{235}U, it is almost impossible to prepare chromatographic generators suitable for the medical use.

Recently, KAKEN Co. and JAERI (Japan Atomic Energy Research Institute) have developed a new inorganic adsorbent (Poly-Zirconium Compound : PZC) for ^{99m}Tc generator using ^{99}Mo with a low specific activity obtained by (n,γ) reaction of natural Mo. PZC synthesized from $ZrCl_4$ and isopropyl alcohol as raw materials is an organic polymer with the framework of -O-Zr-O-Zr-Cl. After the dissolution of irradiated MoO_3 target with the NaOH solution, MoO_4^{-2} is bound with Zr through the hydrolysis of a part of Zr-Cl. As a result, ^{99}Mo is chemically adsorbed (bound) strongly into the PZC together with stable Mo.

^{99}Mo on the PZC converts to ^{99m}Tc through β^- decay. The bonding -Zr-^{99m}Tc is cut easily. So ^{99m}Tc is eluted easily by the saline solution.

This paper describes experiments for evaluation the performance of PZC as a column packing materials for ^{99}Mo/^{99m}Tc generator from (n,γ) ^{99}Mo. The three batches of PZC materials are supplied by KAKEN Co. and the procedure for experiments is also specified by the company.

Experimental

Irradiate 2 g of MoO_3 with thermal neutrons. Dissolve the irradiated MoO_3 in 5 ml of 6M NaOH. Dilute the solution with water to 25 ml (53.4 mg Mo/ml); the resulting solution is called the original ^{99}Mo solution.

Pipet 5 ml of the original ^{99}Mo solution, which contains 267 mg of Mo. into a flask. Neutralize the solution to pH 7 with 1 M HCl and adjust the volume to 10-20 ml with water. 1 g of PZC is added to the solution and keep the mixture at 90°C for 3 hours in oil-bath, shake the flask slowly by hand several times.

Pipet proper amount (a few ml) of the solution from the Mo-PZC mixture into a measuring bottle for γ-ray spectrometry. Pipet the same amount of the ^{99}Mo original solution mentioned above into another measuring bottle. Measure γ-ray spectra of each of the bottles with a coaxial Ge detector. Compare the peak area at 739 keV between the two sources, and calculate the adsorption rate of ^{99}Mo.

Remove a fine powder on the surface of the solution by decantation. Pack the PZC absorbing ^{99}Mo into a glass column, with a glass filter, of 8 mm. in inner diameter and 50 mm. in length. Wash the column with 50 ml of saline solution.

After 24 hours, elute ^{99m}Tc from ^{99}Mo with 10 ml of saline solution into a measuring bottle for γ-ray spectrometry. Pipet 5 ml of the original ^{99}Mo solution into another measuring bottle. Measure γ-ray spectra of each of the bottles with a coaxial Ge detector. Compare the peak area at 739 keV between the two sources and calculate the desorption rate of ^{99}Mo.

Percent elution rate of ^{99m}Tc can be obtained from the following equation:

$$\text{Elution rate of } ^{99m}\text{Tc (\%)} = \frac{B}{A \times C \times D} \times 100$$

Where

A: the calculated activity of 5 ml of the original ^{99}Mo solution at reference time t = 0

B: the observed activity of the eluted ^{99m}Tc solution

C: the production rate of ^{99m}Tc from the ^{99}Mo in the column calculated by the following equation:

$$C = 0.86 \times \frac{\lambda_T}{\lambda_T - \lambda_M} \times [\exp(-\lambda_M t) - \exp(\lambda_T t)]$$

0.86 : Branching ratio from ^{99}Mo to ^{99m}Tc

λ_M = Decay constant of ^{99}Mo = 0.0105 hr^{-1}

λ_T = Decay constant of ^{99m}Tc = 0.1153 hr^{-1}

t = a period of time after packing Mo-PZC into a column; ≥ 24 hr

Result

Preparation of irradiated molybdenum.

Irradiation condition:

Reactor	:	Thai Research Reactor-1/Modification1 (TRR-1/M1) Operate at 1.2 MW.
Amount of target	:	2.0 g. MoO_3
Neutron flux	:	3 x 10^{13} n/cm^2 -sec
Irradiation period	:	Tuesday 6 hours and Wednesday 12 hours.
Activity of ^{99}Mo at EOB:		3400 MBq.

Table 1. Lot No. PZC020319

Exp. No.	^{99}Mo Loading (MBq)	Adsorption rate Of ^{99}Mo		1st day Elution		2nd day Elution		3rd day Elution		4th day Elution		5th day Elution	
		%	MBq^{99}Mo / PZC 1 g.	$^{99m}Tc^E$	$^{99}Mo^D$	$^{99m}Tc^E$	$^{99}Mo^D$	$^{99m}Tc^E$	$^{99}Mo^D$	$^{99m}Tc^E$	$^{99}Mo^D$	$^{99m}Tc^E$	$^{99}Mo^D$
1	512	96.49	494	60.05	0.71	61.49	0.75	66.61	0.24	67.99	0.17	69.31	0.21
2	461	90.30	416	87.02	0.39	70.16	0.36	75.02	0.21	70.49	0.30	60.17	0.20

Table 2. Lot No. PZC020522

Exp. No.	^{99}Mo Loading (MBq)	Adsorption rate Of ^{99}Mo		1st day Elution		2nd day Elution		3rd day Elution		4th day Elution		5th day Elution	
		%	MBq^{99}Mo / PZC 1 g.	$^{99m}Tc^E$	$^{99}Mo^D$	$^{99m}Tc^E$	$^{99}Mo^D$	$^{99m}Tc^E$	$^{99}Mo^D$	$^{99m}Tc^E$	$^{99}Mo^D$	$^{99m}Tc^E$	$^{99}Mo^D$
3	331	97.17	322	77.98	0.81	65.52	0.82	62.25	0.28	63.19	0.16	64.59	0.39
4	530	79.65	423	82.77	1.10	87.01	0.63	89.49	0.58	71.03	0.52	88.61	0.51

Table 3. Lot No. PZC020731

Exp. No.	^{99}Mo Loading (MBq)	Adsorption rate Of ^{99}Mo		1st day Elution		2nd day Elution		3rd day Elution		4th day Elution		5th day Elution	
		%	MBq^{99}Mo / PZC 1 g.	$^{99m}Tc^E$	$^{99}Mo^D$	$^{99m}Tc^E$	$^{99}Mo^D$	$^{99m}Tc^E$	$^{99}Mo^D$	$^{99m}Tc^E$	$^{99}Mo^D$	$^{99m}Tc^E$	$^{99}Mo^D$
5	530	87.97	467	66.92	0.85	75.85	0.60	75.95	0.53	78.09	0.40	75.58	0.38
6	530	85.89	456	71.57	1.12	74.97	1.03	76.06	0.73	75.27	0.61	78.36	0.60

^{E}Elution rate of ^{99m}Tc (%)
^{D}Desorption rate of ^{99}Mo (Bq^{99}Mo/kBq^{99m}Tc)

Mo loaded on PZC 1 gm. = 266.6 mg.
pH of eluated ^{99m}Tc = 3-4

Table 4. Using alumina as a second column (Lot No. PZC020522)

Elution time (day)	^{99m}Tc MBq	^{99}Mo breakthrough kBq	Elution rate of ^{99m}Tc (%)	pH
1	224.75	ND	74.32	6.5
2	220.38	ND	84.41	6.5
3	159.31	ND	85.13	6.5
4	123.59	ND	69.67	6.5
5	95.37	ND	80.83	6.5

ND = Not Detectable

^{99}Mo loading = 530 MBq
Adsorption rate of ^{99}Mo = 85.98 %

Table 5. Adsorption rate of ^{99}Mo

Exp No.	^{99}Mo loading (MBq)	Adsorption rate of ^{99}Mo	
		MBq ^{99}Mo/ 1g PZC	%
1	512	494	96.49
2	461	416	90.30
3	331	322	97.17
4	530	423	79.65
5	530	467	87.97
6	530	456	85.89
% Adsorption rate of ^{99}Mo			89.58 ± 6.64

Table 6. Desorption rate of ^{99}Mo

Exp. No.	Desorption rate of ^{99}Mo (Bq ^{99}Mo/kBq ^{99m}Tc)					
	1st Elution	2nd Elution	3rd Elution	4th Elution	5th Elution	Average
1	0.71	0.75	0.24	0.17	0.21	0.42±0.29
2	0.39	0.36	0.21	0.30	0.20	0.29±0.09
3	0.81	0.82	0.28	0.16	0.39	0.49±0.31
4	1.10	0.63	0.58	0.52	0.51	0.67±0.25
5	0.85	0.60	0.53	0.40	0.38	0.55±0.19
6	1.12	1.03	0.73	0.61	0.60	0.82±0.24
Desorption rate of ^{99}Mo (Bq ^{99}Mo/kBq ^{99m}Tc) : 0.29±0.09 – 0.82±0.24						

Exp. No.	% Elution rate ^{99m}Tc					
	1st Elution	2nd Elution	3rd Elution	4th Elution	5th Elution	Average
1	60.05	61.49	66.61	67.99	69.31	65.09±4.09
2	87.02	70.16	75.02	70.49	60.17	72.57±9.73
3	77.98	65.52	62.25	63.19	64.59	66.71±6.43
4	82.77	87.01	89.49	71.03	88.61	83.78±7.58
5	66.92	75.85	75.95	78.09	75.58	74.49±4.34
6	71.57	74.97	76.06	75.27	78.36	75.24±2.45
% Elution rate ^{99m}Tc : 65.09±4.09 – 83.78±7.58						

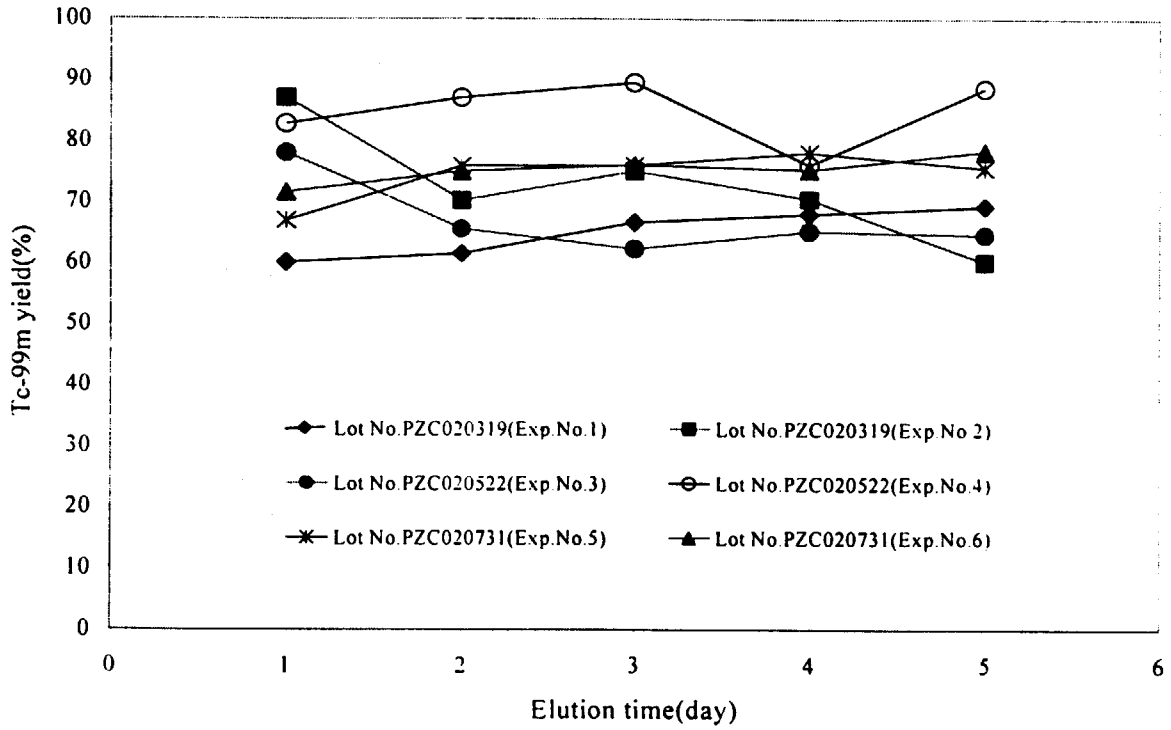

Figure 1 Elution rate of ^{99m}Tc

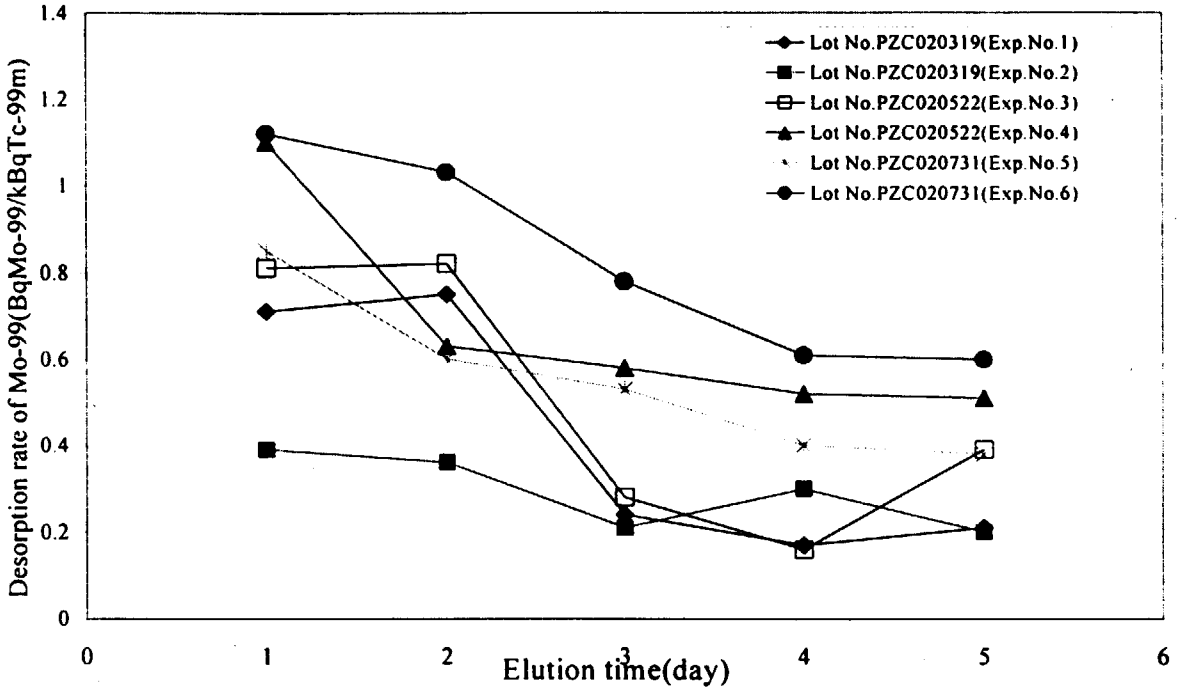

Figure 2 Desorption rate of ^{99}Mo VS Elution Time (day)

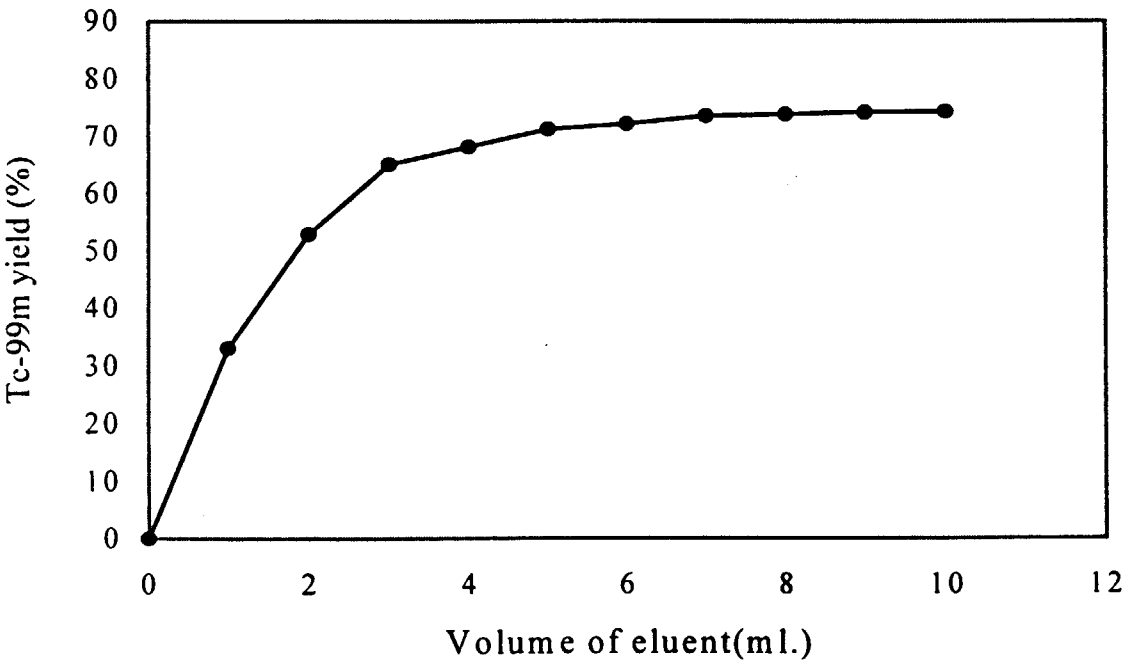

Figure 3 Elution profile of ^{99m}Tc

Conclusion

The evaluation of $^{99}Mo/^{99m}Tc$ generator experiments using PZC material and irradiated natural molybdenum were done according to the provided PZC and specified procedure from KAKEN Co. The result showed the adsorption rate of ^{99}Mo = 89.58 ± 6.64%, desorption rate of ^{99}Mo = 0.29±0.09 – 0.82±0.24 Bq ^{99}Mo/kBq ^{99m}Tc and the elution rate of ^{99m}Tc = 65.09±4.09 - 83.78±7.58 %

The PZC could adsorp ^{99}Mo with high adsorption rate and ^{99m}Tc was eluted with high efficiency but the contamination of ^{99}Mo was detected in the eluate. The problem of contamination of ^{99}Mo in the eluate was easily solved by passing the eluate through alumina column.

The $^{99}Mo/^{99m}Tc$ generator using PZC material and irradiated natural molybdenum performed the satisfied result of the generator characteristics which could be developed as an alternative technology for $^{99}Mo/^{99m}Tc$ generator production.

It is strongly believed that in the near future this technology will be established and utilized under the framework of the Forum of Nuclear Cooperation in Asia (FNCA) which will benefit the countries operating small research reactors to overcome the difficulties in producing $^{99}Mo/^{99m}Tc$ generator.

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JAERI－Conf 2004－010

WORKSHOP ON THE UTILIZATION OF RESEARCH REACTOR

JANUARY 13-17 , 2003 , SERPONG , INDONESIA

1.14 INVESTIGATION ON THE PERFORMANCE OF POLYMER ZIRCONIUM COMPOUND (PZC) FOR CHROMATOGRAPHIC Tc-99m GENERATOR PREPARATION

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ABSTRACT

The performance of PZC was investigated for chromatographic Tc-99m generator preparation. Mo-adsorption of PZC in different Mo-solutions and Tc-99m elution of ^{99}Mo-PZC column were studied. Mo- adsorption capacity of higher than 250mgMo/gPZC and Tc-99m elution yield of higher than 80% were achieved with PZC adsorbent. Mo-99 breakthrough of 0.02% and Molybdenum element breakthrough of around 5μg Mo/ml were found in Tc-99m eluate. A good relationship between the Mo-content of adsorption solution and the Mo-adsorption capacity, adsorption percentage, Mo-breakthrough and Tc-99m elution yield was found. The preparation of PZC based Tc-99m chromatographic generator with 4 gram weight of PZC was successfully conducted.

INTRODUCTION

Today Technetium-99m is the main radioisotope used in Nuclear Medicine for diagnosis. More than 80% of all diagnostic nuclear medicine procedures based on using ^{99m}Tc. ^{99m}Tc (6.0 h) is almost exclusively produced from the decay of its 66 h parent ^{99}Mo.

At present nearly the entire supply of Mo-99 is based on the research reactor produced Mo-99 sources by using the (n,γ) nuclear reaction with natural Mo (^{98}Mo, ~24%), resulting in inexpensive but low specific activity ^{99}Mo or by neutron-induced fission of ^{235}U, which results in expensive but high specific activity of ^{99}Mo. In the "fission method", the technological and infrastructure requirements are more complex and possibly can be sustained only by countries with advanced nuclear technology.

The technology requirements for processing of ^{99}Mo from (n,γ) "activation method" is rather simple, and is within the reach of most developing countries operating research reactors.

Alternative technologies for ^{99}Tc^{m} generators using (n,γ) nuclear reaction with natural Mo were developed or are being under development . Although some new technology has technically been accepted in many countries, but there will have to be a substantial economic incentive for a large producers of Mo-99 or Tc-99m generators to change to a new process by reason of the existing investment in production infrastructure and in the approval of ^{99}Mo and derived products.

Among these the technologies for ^{99}Tc^{m} generators using Zirconium- or Titanium-Molybdate gel, so called "gel technology" and polymer Zirconium- or Titanium compound (PZC, PTC) based technology are considered as new ones. Under the frame of Forum for Nuclear Cooperation in Asia (FNCA) cooperation program the PZC based technology for production of Tc-99m generator developed first in Japan is a topic for further evaluation and comparison between participant countries. Any new process having a potential influence on the product quality, compared to existing process will have to be demonstrated and licensed fully. The product should be qualified by the various regulatory authorities, which is a long and expensive process adding to the market inertia referred to previously. As a contribution ,our report is aimed to investigate and evaluate the performance of PZC adsorbent for the preparation of a clinically available Tc-99m generator.

EXPERIMENTAL

Polymer Zirconium Compound (PZC) supplied by Kaken Co. (Japan) was used in all experiments. The PZC samples of four different preparation batches were investigated and compared with each other.

Totally 23 PZC columns (6 columns of 0.75 g weight for each PZC batch) were tested. Seven different conditions for adsorption and post-adsorption treatment were applied. These conditions are as follows:

a- Normal adsorption in water solution of Molybdate;

b- Adsorption in Acetate buffer solution of Molybdate (Acetate buffer solution of 0.2M acetic acid, pH=5)

c- Adsorption in H_2O_2 added Acetate solution of Molybdate (Acetate buffer solution of 0.2M acetic acid, containing 5% H_2O_2, pH=5)

d- Adsorption in CrO_4^{2-} added Acetate solution of Molybdate (Acetate buffer solution of 0.2M acetic acid, containing 2.5% K_2CrO_4, pH=7)

e- Normal adsorption in water solution of Molybdate (with different conditioning solutions) followed by sterilization in autoclave.

f- Normal adsorption in water solution of Molybdate (with different conditioning solutions) followed by drying at 100°C for one hour.

g- Normal adsorption in water solution of Molybdate followed by conditioning PZC column with different solutions.

15 ml radioactive Mo-99 solution of concentration of 13.316 mg Mo / ml, pH=7 were added to PZC samples of 0.75 g weight, then these samples were gently shaked in water bath of 50°C overnight. After shaking the samples were let to stand and a portion of clear supernatant solution was taken out to measure Mo-99 radioactivity for Mo–adsorption capacity calculation and then the remained solution was decanted to get the solid PZC part. This solid PZC sample was packed in 8ml glass column and washed with 30 ml water followed by passing column with 10 ml saline. After this step first Tc-99m elution was started after 24 hours equilibration time and daily an elution was conducted.

All experiments were carried out with PZC columns of Mo-99 radioactivity of 10 – 30 mCi.

Each column was eluted for five to seven days (one elution a day). Totally more than 120 elutions were carried out.

Elution yield, Mo-99 break-through (by Capintec Dose Calibrator), Mo element content (by photospectrometric method) were determined for each elution.

RESULTS AND DISCUSSION

A. The performance test of PZC under different column treatment

Real adsorption capacity of each PZC column and effective loading capacity (% of solid PZC loaded on the column after decanting fine PZC particles), elution profiles of each column and Mo break-through in function of standing-time were also determined. A large number of interesting results were collected and reported in the following tables and figures. (Table 1-4 and fig. 1).

Mo adsorption capacity is of about 248.1 mgMo/g PZC for the first PZC batch and 244.0 Mo mg/g PZC for the second,

254.8 mg Mo/g PZC for the third and around 265 mg Mo/g PZC for the fourth one. The capacity also varied depending on the applied adsorption conditions.

Elution yield of higher than 90% was achieved in the a, b, c, d cases and about 80% in the e, f cases.

The adsorption in Acetate buffer solution of Molybdate showed a better integrity of PZC particles (amount of fine particles is smaller) comparing with PZC particles adsorbing Mo in pure water solution of Molybdate.

Elution profile showed that the exhaustive elution would be achieved with 4 ml of saline (for a column bed of 0.75 g PZC). From these results we can estimate 25 ml saline for exhaustive elution of PZC column of 4 g.. So a real generator of 4 g PZC bed using 15 ml saline elution will be of elution yield of about 80 %.

Mo break-through remains as a problem to be discussed. This break-through is not so high (around 2-10 μg Mo/ml for a column of 0.75 g PZC and eluant volume of 8 ml). Mo-99 breakthrough is a little higher than the limit of 0.015% of Tc-99m radioactivity. An accumulation of Mo-breakthrough was observed for the elution after the equilibration time of longer than 24 hours .

If we consider a real generator of 4 g PZC column and Mo-breakthrough being dominated in the first stage of elution profile (see fig. 1), the above-mentioned value of Mo break-through is overtaking the limit required for pertechnetate solution applicable in Nuclear Medicine.

The sterilizing PZC column in the autoclave reduced to some extent the elution yield of Tc-99m, but did not affect the Mo-breakthrough of column.

It is found that the drying ^{99}Mo-PZC column brought about the shrinking of PZC. The shrinking reduced the volume of column packing .The shrinking may cause the deformation of PZC gel structure and its adsorption affinity for Molybdate ion. This effect brought about the tailing of the elution profile, the lower Tc-99m elution yield and the increase of Mo-breakthrough.

Of course, we can use Alumina clean-up column to reduce Mo-content in pertechnetate solution, but the Mo-capacity of Alumina is not so high. So the design of generator is considered as an important theme.

Table 1 : The Mo adsorption test of PZC under different column treatment

Lot No. of PZC	Sample	pH		Colour of PZC (After adsorption)	Non-adsorbed Mo-99 radioactivity (%)	Discarded fine PZC powder Mo-99 radioactivity (%)	Generator column Mo-99 radioactivity (%)	Adsorption capacity (mg Mo/g PZC)	Weight of PZC	
		Before Adsorption	After adsorption						Discarded fine powder (%)	Generator Column (%)
020522	A	7	5	White	6.3	7.9	85.8	249.7	8.5	91.5
	B*	7	5	White	7.7	6.6	85.7	246.2	7.1	92.9
	C*	7	5	White	6.7	10.5	82.8	248.6	11.3	88.7
010905	A	7	4.5	Light Brown	10.2	3.8	86.0	239.5	6.2	93.8
	B	7	4.5	Light Brown	7.2	4.1	88.7	247.4	5.9	94.1
	C	7	4.5	Light Brown	8.6	5.8	85.6	245.3	6.3	93.7
020315	A	7	4.5	White	5.3	6.6	88.1	252.3	7.7	92.3
	B	7	4.5	White	4.2	7.0	88.8	255.4	7.3	92.7
	C	7	4.5	White	3.7	7.5	88.8	256.7	7.7	92.3

Table 2: The elution test of PZC under different column treatment

Lot No. of PZC	Sample	First elution			Second to fifth elution		
		Elution Yield of Tc-99m (%)	Mo breakthrough		Elution Yield of Tc-99m (%)	Mo-breakthrough	
			μg Mo (*)	(%) (**)		μg Mo (*)	(%) (**)
020522	A	77.8	27.4	0.023	92.5 ± 0.5	27.8	0.020
	B*	69.5	132.4	0.128	84.5 ± 0.3	126.6	0.114
	C*	66.5	94.9	0.098	86.5 ± 0.5	63.4	0.050
010905	A	92.5	50.4	0.036	94.5 ± 0.5	25.1	0.017
	B	82.6	85.6	0.071	83.3 ± 0.5	85.6	0.071
	C	77.2	35.2	0.030	79.3 ± 0.6	12.6	0.010
020315	A	92.4	43.2	0.035	92.8 ± 0.5	12.2	0.010
	B	80.4	72.0	0.057	85.0 ± 0.5	52.0	0.040
	C	78.5	36.8	0.030	84.5 ± 0.5	18.1	0.014

Sample symbol:

(A) With normal condition of Mo-adsorption in water solution of Molybdate and column washing with 50 ml distilled water, eluted with saline.

(B) With normal condition of Mo-adsorption in water solution of Molybdate and column washing with 15 ml distilled water, then drying PZC powder at 100°C for one hour followed by washing with 35 ml water, eluted with saline.

(C) With normal condition of Mo-adsorption in water solution of Molybdate and column washing with 15 ml distilled water, then sterilizing PZC column at 124°C for half an hour followed by washing with 35 ml water, eluted with saline.

(B*) With normal condition of Mo-adsorption in water solution of Molybdate and column washing with 50 ml distilled water followed by conditioning column with 5 ml saline, then drying PZC powder at 100°C for one hour, eluted with saline.

(C*) With normal condition of Mo-adsorption in water solution of Molybdate and column washing with 50 ml distilled water followed by conditioning column with 5 ml saline, then sterilizing PZC column at 124°C for half an hour, eluted with saline.

(*) Total Molybdenum content in 8 ml eluate.

(**) Percentage of Mo-99 radioactivity in the eluate of Tc-99m.

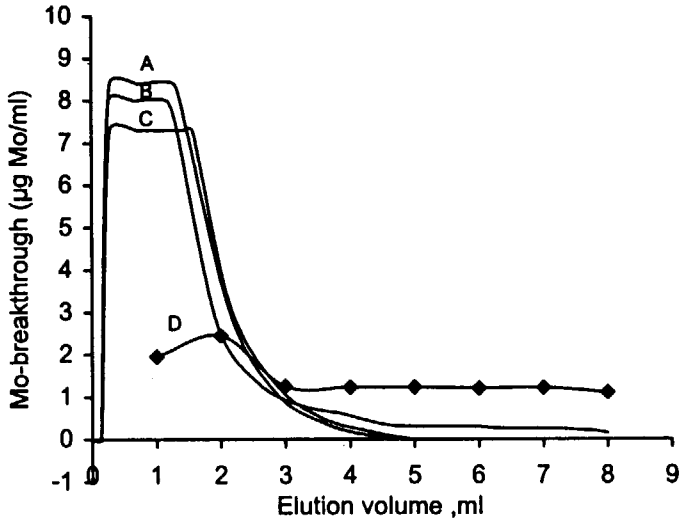

Fig. 1 : Elution profiles of ^{99}Mo-PZC column
A, B, C : The Tc-99m elution profiles of Sample A, B and C, respectively (Arbitrary Tc-99m Radioactivity Scale).
D : Mo content in the different eluate fractions of the elution profile A.

B. The performance test of PZC in different solutions

In table 3-4 the results of studies on the adsorption and Tc-99m elution performance of ^{99}Mo- PZC in different solutions were presented. It is found that the oxidizing agent added Mo-solution did not affect the Mo-adsorption and Tc-99m elution performance of ^{99}Mo- PZC columns. The conditioning ^{99}Mo-PZC columns with solution containing agent of higher Zirconium-adsorption power such as PO_4^{3-} and with a solution of high Cl^- concentration will affect the performance of ^{99}Mo- PZC column. This effect brought about higher Mo-breakthrough in Tc-99m eluate and reduced to some extent the Tc-99m elution yield.

Table 3: The Mo adsorption test of PZC in different solutions

Lot No. of PZC	Sample	pH		Colour of PZC (After adsorption)	Non-adsorbed Mo-99 radioactivity (%)	Discarded fine PZC powder Mo-99 radioactivity (%)	Generator column Mo-99 radioactivity (%)	Adsorption capacity (mg Mo/g PZC)	Weight of PZC	
		Before Adsorption	After Adsorption						Discarded fine powder (%)	Generator Column (%)
020522	I	7	5	White	6.3	7.9	85.8	249.7	8.5	91.5
	II*	7	5	White	6.3	11.7	82.0	249.7	12.5	87.5
	III	7	5	White	6.3	10.5	83.2	252.1	15.8	84.2
	IV*	7	5	White	6.3	10.6	83.1	250.2	13.8	86.2
010905	I	7	4.5	Light Brown	10.2	3.8	86.0	239.5	6.2	93.8
	II	7	5.5	Light Brown	21.8	3.5	74.7	208.4	4.5	95.5
	III	7	5.5	Light Brown	26.6	4.1	69.3	196.0	4.6	95.4
	IV	7	5.5	Light Brown	19.4	4.7	75.9	214.9	5.0	95.0
020315	I	7	4.5	White	5.3	6.6	88.1	252.3	7.7	92.3
	II	7	5.5	White	22.3	5.7	72.0	207.1	5.4	94.6
	III	7	5.5	White	21.3	4.4	74.3	209.8	5.6	94.4
	IV	7	5.5	White	24.1	5.7	70.2	202.2	5.5	94.5

Table 4: The elution test of PZC in different solutions

Lot No of PZC	Sample	First elution			Second to fifth elution		
		Elution Yield of Tc-99m (%)	Mo breakthrough		Elution Yield of Tc-99m (%)	Mo-breakthrough	
			μg Mo (*)	(%) (**)		μg Mo (*)	(%) (**)
020522	I	77.8	27.4	0.023	92.5 ± 0.5	27.8	0.020
	II*	80.7	196.6	0.168	94.5 ± 0.7	184.8	0.134
	III*	90.1	60.1	0.024	90.2 ± 0.5	34.3	0.014
	IV*	92.5	55.5	0.022	93.1 ± 0.3	31.2	0.012
010905	I	92.5	50.4	0.036	94.5 ± 0.5	25.1	0.017
	II	94.2	49.6	0.040	98.4 ± 0.6	22.5	0.017
	III	99.2	50.4	0.042	97.1 ± 0.6	21.4	0.018
	IV	99.5	52.8	0.039	98.2 ± 0.3	24.0	0.018
020315	I	92.4	43.2	0.035	92.8 ± 0.5	12.2	0.010
	II	96.4	40.0	0.033	97.5 ± 0.3	13.4	0.011
	III	98.0	35.2	0.028	98.5 ± 0.3	11.7	0.010
	IV	97.0	40.0	0.034	98.2 ± 0.3	16.2	0.013

Sample symbol:

(I) With normal condition of Mo-adsorption in water solution of Molybdate and Column washing with 50 ml distilled water, eluted with saline.

(II) With normal condition of Mo-adsorption in Acetate buffer solution of Molybdate and column washing with 50 ml distilled water, eluted with saline.

(III) With normal condition of Mo-adsorption in Acetate buffer solution of Molybdate added with H_2O_2 and column washing with 50 ml distilled water, eluted with saline.

(IV) With normal condition of Mo-adsorption in Acetate buffer solution of Molybdate added with CrO_4^{2-} and column washing with 50 ml distilled water, eluted with saline.

(II*) With normal condition of Mo-adsorption in water solution of Molybdate and Column washing with 20 ml solution of 0.2M KH_2PO_4, pH=6.2 followed by washing with 30 ml distilled water, eluted with saline

(III*) With normal condition of Mo-adsorption in water solution of Molybdate and Column washing with 20 ml solution of 9% NaCl followed by washing with 30 ml distilled water, eluted with saline

(IV*) With normal condition of Mo-adsorption in water solution of Molybdate and Column washing with 20 ml solution of Acetate buffer solution of 0.2M Acetic acid, pH=5, followed by washing with 30 ml distilled water, eluted with saline

(*) Total Molybdenum content in 8 ml eluate.
(**) Percentage of Mo-99 radioactivity in the eluate of Tc-99m.

C. Effect of the solution Mo-content on the Mo-adsorption capacity of PZC and on the Tc-99m elution yield and Mo-breakthrough of Tc-99m eluate.

The variable volumes (as specified in the Table 5 below) of the radioactive Mo-99 solution of concentration of 13.316 mg Mo / ml , pH=7 were added to PZC samples of 0.2 g weight, then these samples were gently shaked in water bath of 50 °C overnight. After shaking the samples were let to stand and a portion of clear supernatant solution were taken out to measure Mo-99 radioactivity for Mo–adsorption capacity calculation and then the remained solution was decanted to get the solid PZC part. This solid PZC was washed with 10 ml water followed by passing column with 10 ml saline. After this step first Tc-99m elution was started after 24 hours equilibration time and daily an elution was conducted.

Table 5 : Effect of the solution Mo-content on the Mo-adsorption capacity of PZC and on the Tc-99m elution yield and Mo-breakthrough of Tc-99m eluate.
(**) PZC sample, Batch Number: PZC020731
- Certified Mo- adsorption capacity: 265 mg Mo / g PZC
- Applied elution volume: 5 ml 0.9% NaCl

$$(*)\ \text{Adsorption percentage}\ (\%) = 100 \times \frac{\text{Mo adsorption capacity}}{\text{Mo content of solution}}$$

Sample **	PZC-IVA	PZC-IVB	PZC-IVC	PZC-IVD
Weight of PZC , (g)	0.20	0.20	0.20	0.20
Volume of Mo solution , (ml)	3.75	4.50	5.25	6.00
Mo-content of adsorption solution , (mgMo /g PZC)	249.67	299.61	349.54	399.48
Mo-adsorption capacity , (mg Mo / g PZC)	237.69	276.54	289.77	308.40
Adsorption percentage, (%) *	95.20	92.30	82.90	77.200
Tc-99m elution yield , (%)	74.10	92.70	84.00	81.00
Mo -Breakthrough in first elution ,(μg Mo/ml)	85.0	134.0	220.0	240.0
Mo -Breakthrough in second-to-fifth elution ,(μg Mo/ml)	2.0 ± 0.5	22.1 ± 0.8	43.5 ± 0.9	48.2 ± 0.8

The experimental results presented in Table 5 and on Figs: 2-3 revealed the fact that the Mo-adsorption capacity of PZC and Mo-breakthrough of Mo-PZC column decreased with the increasing Mo-content of adsorption solution. This is attributed to the excess of weakly bound Molybdate ion on the surface of PZC. This excess of Molybdate ion may block the pathway of Tc-99m pertechnetate ion out- diffusion and cause the lower Tc-99m elution yield observed on the right side of curve in fig. 3.

The lower Mo-content of adsorption solution has caused the unsaturated adsorption and left to some extent free active groups of high anion-affinity on PZC surface. The action of these groups may contribute a retention power to reduce Tc-99m elution yield and Mo-breakthrough in Tc-99m eluate.

In our experiment the adsorption percentage of around 90% was chosen as an optimal value for Mo-adsorption to give a ^{99}Mo- PZC column of highest performance.

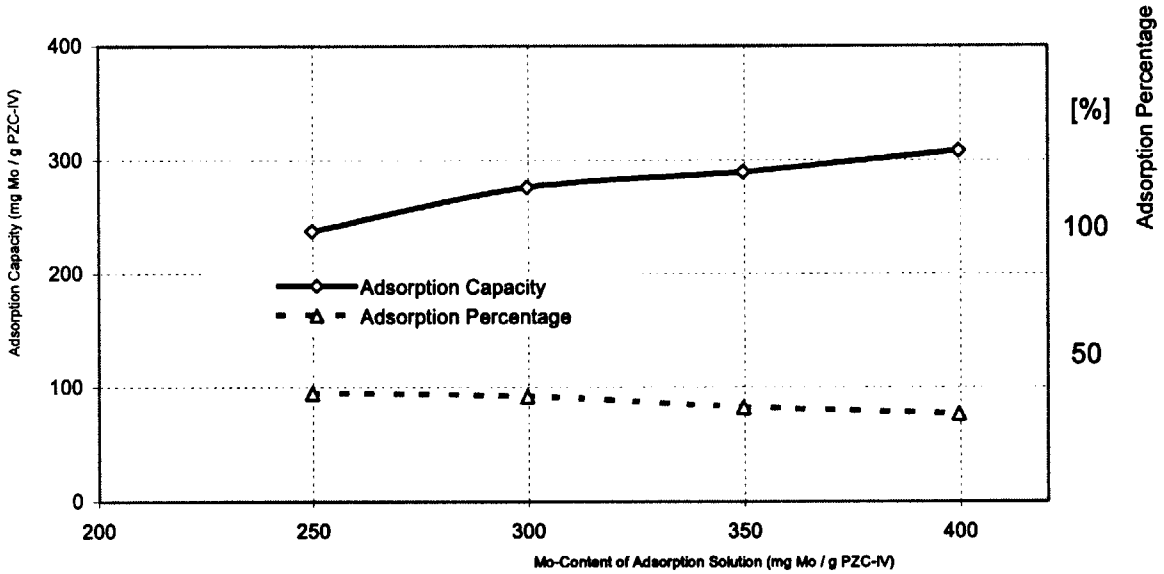

Fig.2 : Effect of Mo-content of adsorption solution on Mo-Adsorption and Adsorption percentage of PZC (Experimental Data Presented in Table:5)

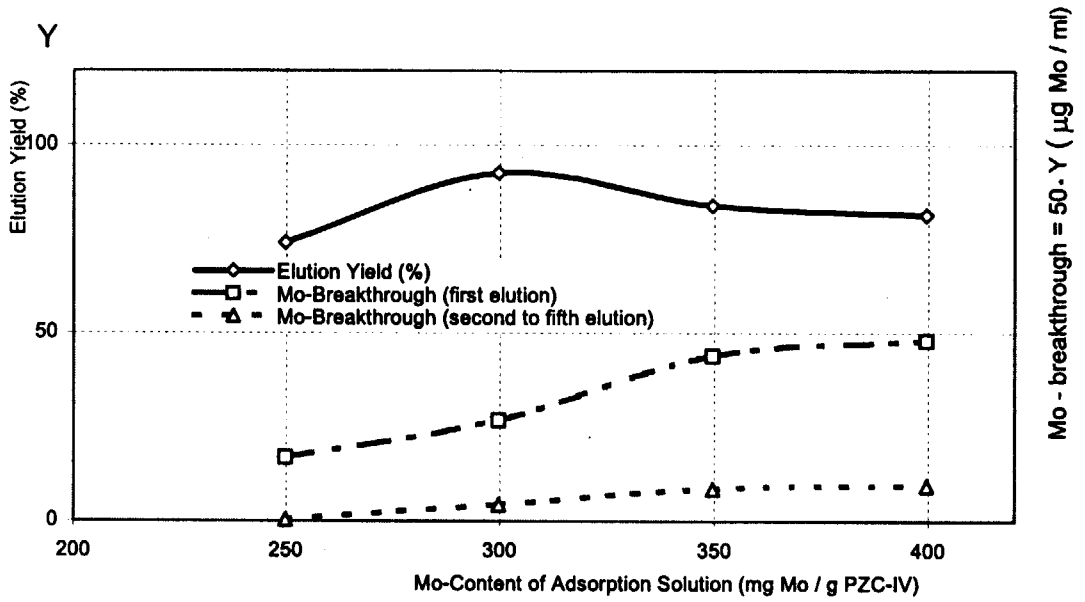

Fig. 3 : Effect of Mo-content of adsorption solution on Molybdenum element breakthrough and Tc-99m elution yield of Mo-99 PZC column (Experimental Data Presented in Table:5)

D. Practical application of PZC for Tc-99m generator preparation using Mo-99 solution of low specific radioactivity.

Based on the results obtained above and the available radioactivity of Mo-99 solution produced on the Nuclear research reactors of thermal neutron flux ranging from 10^{13} n.cm^{-2}.sec^{-1} to 10^{14} n.cm^{-2}.sec^{-1}, a column of 4 gram PZC packing was chosen to prepare the Tc-99m generators of radioactivity changing from 100 mCi to 1000 mCi Mo-99 (see Table 6 below and Fig.4).

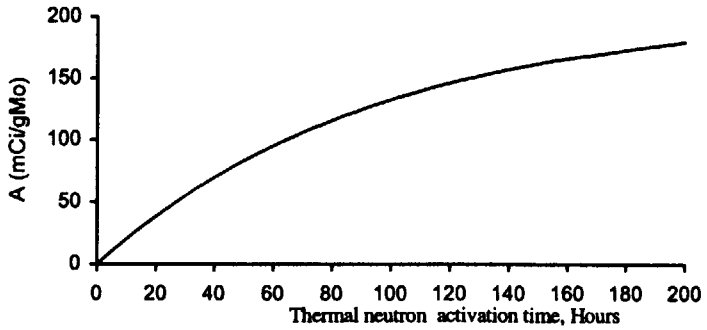

Fig.4 : Thermal neutron activation of natural Molybdenum in a nuclear reactor of thermal neutron flux of 10^{13} n.cm^{-2}.sec^{-1} .

Table 6 : Tc-99m generator production capability using a PZC column of 4 gram weight and of 250 mg Mo/g PZC Mo-adsorption capacity in the nuclear reactors of different neutron flux . Irradiation time : 186 hours continuously

Thermal neutron flux ($n.cm^{-2}.sec^{-1}$)	10^{13}	2.5×10^{13}	5×10^{13}	10^{14}
Mo-99 radioactivity at E.O.B	164.88 mCi	412.2 mCi	824.4 mCi	1648.0 mCi
Mo-99 radioactivity after one day cooling time and one day of generator preparation .	100.10 mCi	250.2 mCi	500.4 mCi	1000.8 mCi

(The values in the Table 6 were calculated for one gram weight of Molybdenum equivalent to Mo content of 4 gram PZC column.)

The following conditions for generator column preparation were established:

- PZC weight: 4g
- Mo absorption capacity of PZC: 265mgMo/g PZC
- Molybdenum adsorption percentage: 90%
- Total Mo content of PZC column: (4g PZC x 265mgMo/PZC) = 1060 mg Mo
- Total Mo content of adsorption solution: 1060: 90 % = 1178 mg Mo (Equivalent to 1768 mg MoO_3)
- Volume of 6M NaOH solution for dissolution of MoO_3 target: 4.42 ml (Concentration of Molybdenum in solution : 266.5 mg Mo/ml)
- Volume of H_2O_2 solution of 30% concentration for oxidizing Mo^{+5} to Mo^{+6} : 2,0ml
- Volume of H_2O for diluting Mo solution: 15.7ml (total volume: 22.1ml ; Mo concentration: 53.3 mg Mo/mg)
- Adequate amount of 1M HCl solution and H_2O were added to adjust pH to pH=7 and volume of final solution to V= 88.7ml, respectively (Mo- concentration: 13,325 mg Mo/ml) .
- This radioactive Mo-99 solution was added to PZC sample of 4-gram weight and gently shaked at 50^0C overnight, then let it to stand for 30 minutes. The supernatant solution was decanted and solid part of PZC was loaded in a 8 ml volume glass column of G-3 sintered glass filter, then PZC column was washed with 100ml H_2O, closed with septa and aluminum cap and autoclaved at 120°C for 30 minutes. Then PZC column was conditioned with 50ml sterile 0,9% NaCl solution. The Mo-99 radioactivity of column was measured using a Dose calibrator before being assembled in a generator body.
- The Tc-99m elution was conducted daily after equilibration of 24 hours.

As an example, a target of 1768 mg MoO_3 was irradiated in DaLat nuclear research reactor of thermal neutron flux of $2,1.10^{13}$ $ncm^{-2}.sec^{-1}$ for 100 hours continuously. After neutron activation the target was cooled for 11 hours and processed (Mo-99 dissolution and adsorption on a 4-gram PZC column) as mentioned above. This process has taken 24 hours. Following this the Tc-99m elution was conducted daily after equilibration of 24 hours. The results obtained from this generator were presented in table 7.

The good performance of this generator gave a Tc-99m pertechnetate solution reaching the requirement of European Pharmacopoeia. To fulfill the requirements of American and Japan Pharmacopoeia, this generator should couple with an Alumina clean-up column of about 2 gram weight to reduce the Mo-breakthrough lower than 0.015%.

Table 7: Testing results on the 99mTC generator preparation using PZC adsorbent (PZC , Lot No : PZC020731)

MoO_3 target weight (g)	Total Mo-99 radioactivity of Mo-solution (mCi)	Mo adsorption percentage (%)	Mo-adsorption capacity of PZC column of 4 gram weight (mg)	Mo-99 radioactivity adsorpted in solid PZC (mCi)	Mo-99 radioactivity of PZC column at delivering point of time (mCi)	Tc-99m elution yield (%)		Molybdenum element and Mo-99 breakthrough in 15 ml eluate	
						With 15 ml saline	With 25ml saline	Mo-99 radio activity percentage in Tc-99m solution (%)	(μg Mo/ml)
1.768 (1.178) gram Mo	111.20	90.00	1060.00	100.10	93.50	78.3 ± 0.6	91.2 ± 0.5	0.017 ± 0.002	4.5 ± 0.5

Production timetable:

- **21:00, Sunday:** EOB -----11 hour cooling ------**8:00, Monday:** Start MoO_3 target dissolution and PZC based generator preparation -------24 hour process -----**8:00, Tuesday:** Deliver Tc-99m generator.
- Total time elapsed after EOB until delivery: 35 hours.

CONCLUSION

The experimental results reveal the fact that PZC adsorbent has good performance for the preparation of chromatographic Tc-99m generator.
The Molybdenum adsorption capacity of PZC and its Tc-99m elution yield were not strongly affected by the column treatment conditions and composition of adsorption solution. The Molybdenum breakthrough of Tc-99m eluate eluted from PZC column was sensitive to Mo-adsorption and PZC column treatment conditions. A good relationship between the Mo-content of adsorption solution and the Mo-adsorption capacity , adsorption percentage , Mo-breakthrough and Tc-99m elution yield was found. So the optimal parameters for Mo-99 adsorption and Tc-99m generator column preparation were successfully found for the production of a Tc-99m generator giving a Tc-99m pertechnetate solution reaching the requirements of the International pharmacopoeia.
The main parameters of PZC based Tc-99m generator were stated as follows: Mo-adsorption capacity of higher than 250 mgMo/g PZC and Tc-99m elution yield of higher than 80% were found with PZC adsorbent. Mo-99 breakthrough of lower than
0.015% for a generator system of Mo-99 PZC column coupled with a clean-up Alumina column and Molybdenum element breakthrough of around 5 μg Mo/ml were found in Tc-99m eluate.

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JAERI－Conf 2004－010

WORKSHOP ON THE UTILIZATION OF RESEARCH REACTOR

JANUARY 13-17 , 2003 , SERPONG , INDONESIA

1.15 ACTIVATION ANALYSIS OF TRACE ELEMENTS IN TITANIUM-MOLYBDATE TARGET USED FOR PRE-FORMED TiMo GEL BASED ^{99m}Tc GENERATOR PRODUCTION AND RADIONUCLIDIC IMPURITY OF ^{99m}Tc PERTECHNETATE ELUATE

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FORUM FOR NUCLEAR CO-OPERATION IN ASIA
2003 - WORKSHOP ON THE UTILIZATION OF RESEARCH REACTORS

ACTIVATION ANALYSIS OF TRACE ELEMENTS IN TITANIUM-MOLYBDATE TARGET USED FOR PRE-FORMED TiMo GEL BASED ^{99m}Tc GENERATOR PRODUCTION AND RADIONUCLIDIC IMPURITY OF ^{99m}Tc PERTECHNETATE ELUATE

Tran Van HUNG, Le Van SO

ABSTRACT

A gel-type ^{99}Mo-^{99m}Tc generator using Titanium-Molybdate target (TiMo-target) as column packing was developed in Radioisotope Department, the Dalat Nuclear Research Institute [1]. The concentration of trace elements in TiMo-target and radionuclidic purity in ^{99m}Tc solution eluted from this type of generator were studied for quality control purpose . The monostandard method in neutron activation analysis has been applied to determine the concentration of trace impurities in titanium-molybdate target. For neutron activation analysis, TiMo samples were irradiated in the different channels of nuclear reactor IVV-9 at a neutron flux of maximum $2.1X10^{13}n.cm^{-2}s^{-1}$. The following chemical element impurities were determined: Na, W, Co, Fe, Zn, Ag, Sb and Cr. The gamma ray emitted nuclides found in ^{99m}Tc solution eluted from our ^{99m}Tc generator were the following: ^{65}Zn, ^{60}Co, ^{110m}Ag, ^{59}Fe, ^{51}Cr, ^{24}Na, ^{82}Br and ^{99}Mo . ^{99}Mo was found less than 2.10^{-6}mCi ^{99}Mo per mCi of ^{99m}Tc. The radioactivity of all other radionuclides was less than 5.10^{-9}mCi per mCi of ^{99m}Tc. β-emitting impurities were measured on a liquid scintillation counter and their concentration was found less than 10^{-7}%.

INTRODUCTION

Now a days, Titanium-Molybdate target is also used for the production of ^{99m}Tc radioisotope in some developing countries having low-power research reactor by following (n,γ) reaction

$$^{98}Mo(n,\gamma)^{99}Mo \longrightarrow {}^{99m}Tc$$

The supply of fission ^{99}Mo based chromatographic ^{99m}Tc generator from large nuclear center of developed countries to developing countries is restricted, because of the short half-life of ^{99}Mo parent nuclide and the large investment for using of $^{235}U(n,f)^{99}Mo$ reaction in the production of ^{99m}Tc generator. For that reason, the methods using $^{98}Mo(n,\gamma)^{99}Mo$ produced ex-natural molybdenum or ex-enriched ^{98}Mo for the production of ^{99m}Tc is suitable in developing countries having low power research reactor.

In our laboratory, chromatographic ^{99m}Tc generator using TiMo target was studied and developed [1]. This type of generator using TiMo target has a lot of advantages for developing countries with low power research reactor. The purpose of this paper is to determine the concentration of impurity elements in TiMo target by monostandard method and to study the radionuclidic impurity of ^{99m}Tc solution eluted from this type of pre-formed TiMo based generator by gamma ray and β spectrometer.

The monostandard method is based on simultaneous irradiation of samples and a monostandard for the determination of all other elements [2,3]. The concentration of the elements in a sample is determined from the following equation:

$$Ci = K\frac{Asp}{Asp^*}Ci^*$$

Where Asp = Ap/SDC

Ap - Photopeak counting rate of the radioisotope formed by (n,γ) reaction

S - Saturation factor

D - Decay factor

C - Measurement factor correcting for decay during measuring period
K - Is sum of all nuclear constants involved in activation and counting
Index (*) for monostandard element.

The thermal to epithermal neutron flux ratio can be determined by irradiating and absolute counting of a flux monitor element. The advantage of this method is the determination of a number of elements in the samples basing on only one irradiation of sample. In this work Au and Zr were used as monostandards.

For the detemination of radionuclidic impurity in ^{99m}Tc solution eluted from ^{99}Mo-^{99m}Tc generator, the gamma ray spectrometry was applied. The gamma ray spectra were measured using 4096 channel analyser (CABERA), coupled with HPGe 70 cm^3 keV to 2000 keV with an error of 2%.

EXPERIMENTAL

For neutron activation analysis of trace elements in TiMo target, the samples of weight of approximately 100 mg were packed in a polyethylene vial and in aluminum foil wrapped in quartz ampoules for short and long reactor irradiation, respectively. The samples with Au and Zr monitors were irradiated in the different channels of the nuclear reactor IVV-9, Dalat, Vietnam.

For the determination of the short-lived radionuclides, the samples were irradiated in a thermal column and 7-1 channel of reactor for 2 mins, 30 mins and 2 hrs. For the determination of the long-lived radionuclides, the samples were irradiated in neutron trap of reactor. The characteristic of neutron flux in these irradiation channels were determined and given in table 1.

Prior to radioactivity counting, the irradiated samples were cooled for about 10 mins to one day and for one month depending on irradiation time. Radioactivity of samples was measured using 4096 channel gamma ray spectrometer coupled with a HPGe detector. The irradiated samples were measured for about 5 - 10 mins and for one hour to determine the short and long-lived radionuclides, respectively.

Table 1. Characteristics of neutron flux in some irradiation channels of Dalat reactor

Chracteristics	Neutron trap	7-1 channel	Thermal neutron column
Φ_{th}	2.1 10^{13}ncm^{-2}s^{-1}	7.0 10^{12}ncm^{-2}s^{-1}	4.0 10^{10}ncm^{-2}s^{-1}
Φ_{th}/Φ_{epi}	32	14	640
α	-0.031	-0.045	0

5 gram of TiMo target for the production of a ^{99}Mo-^{99m}Tc generator were irradiated in Dalat reactor for 100 hrs at neutron flux of 2.10^{13}n.cm^{-2}s^{-1}. The neutron activated TiMo target was allowed to cool for 2 days and then packed in a generator column. This type of generator is of ^{99m}Tc activity of 300 mCi calibrated at the end of activation. This value of activity is equivalent to about 100 mCi ^{99m}Tc calibrated at first elution for clinical use. The gamma ray spectra of ^{99m}Tc eluate were measured after about one month and one hour cooling time.

RESULTS AND DISCUSSION

Table 2. shows the concentration of impurity elements in TiMo target which was synthesized for the production of ^{99}Mo-^{99m}Tc generator. Only eight impurity elements, such as Fe, Co, Cr, Sb, Zn, Na, Ag and W were found in the TiMo samples.

Table 2. The concentration of impurity elements in TiMo target

Element	Concentration g/g	Element	Concentration g/g
Fe	$(1.5 \pm 0.1)\ 10^{-5}$	Cr	$(1.2 \pm 0.2)\ 10^{-8}$
Co	$(4.9 \pm 0.5)\ 10^{-8}$	Ag	$(2.1 \pm 0.4)\ 10^{-8}$
Zn	$(1.6 \pm 0.2)\ 10^{-5}$	Na	$(8.5 \pm 0.6)\ 10^{-5}$
Sb	$(9.0 \pm 1.0)\ 10^{-8}$	W	$(4.5 \pm 0.5)\ 10^{-5}$

Two elements Na and W were determined with short irradiation time. The gamma ray spectra shown in fig. 1 were received after a 30 - minute irradiation on the thermal column of reactor and about one day cooling. In these spectra, the short half-lived radionuclides such as ^{99}Mo, ^{24}Na and ^{186}W were found. These radionuclides were formed from nuclear reaction $^{99}Mo(n,\gamma)^{99}Mo$, $^{23}Na(n,\gamma)^{24}Na$ and $^{185}W(n,\gamma)^{186}W$, respectively. Na and W were stated as impurity elements in the TiMo target. The concentration of these elements were 8.5 10^{-5} g/g TiMo for Na and 4.5 10^{-5}g/g TiMo for W.

Neutron activation analysis has been applied to the determination of impurities in the aluminum foil and in the polyethylene vial used for sample packing. The irradiation period was 2 hrs and 100 hrs for polyethylene vial and aluminum foil,

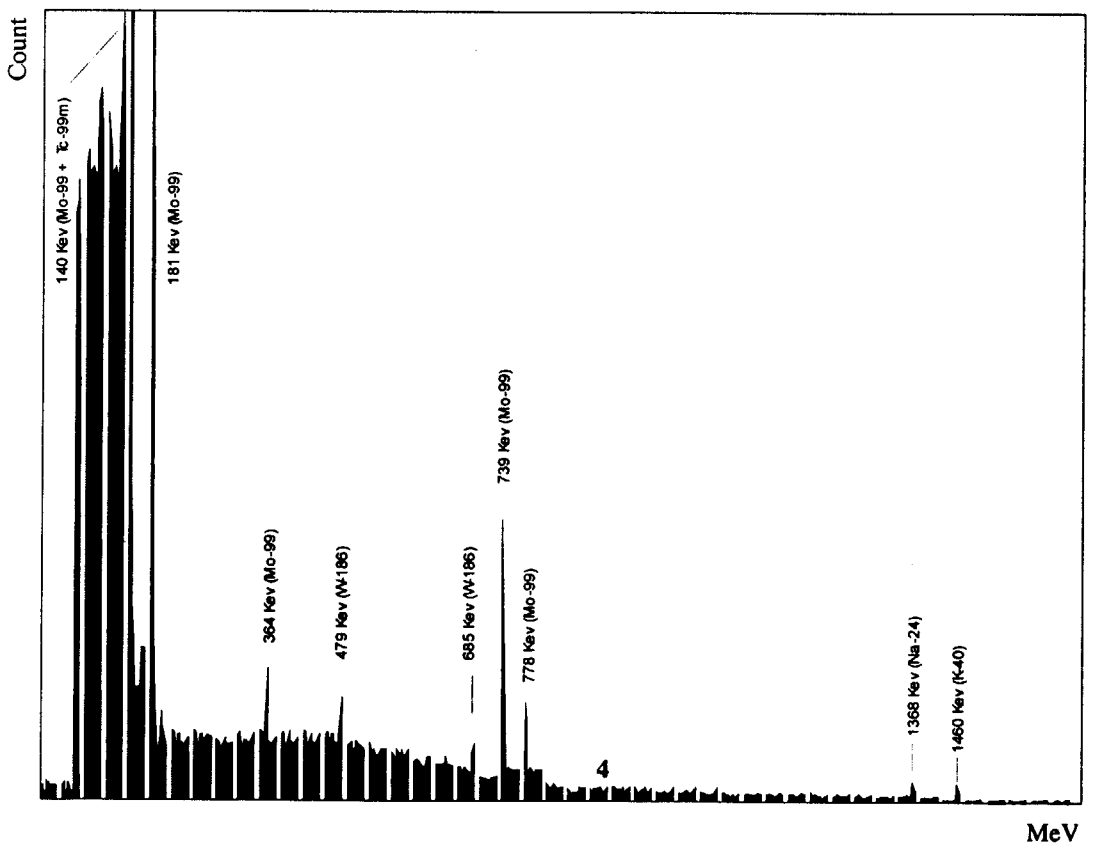

Fig. 1 Gamma spectrum of TiMo target irradiated at the thermal column of reactor

respectively. Table 3 shows the analytical result of the determination of impurities in the aluminum foil after about one month cooling and impurities in polyethylene vial after about one day cooling.

On the gamma ray spectra shown in fig. 2, the long half-lived radionuclides such as ^{59}Fe, ^{60}Co, ^{110m}Ag, ^{122}Sb, ^{65}Zn, ^{95}Nb, ^{46}Sc, ^{51}Cr, ect. were observed. Out of radionuclides ^{59}Fe, ^{60}Co, ^{110m}Ag, ^{122}Sb, ^{65}Zn, ^{51}Cr were formed from impurity elements of target as shown in table 3, while ^{95}Nb, ^{46}Sc and ^{99}Mo were produced from target element via the following reactions $^{95}Mo(n,p)^{95}Nb$ ($T_{1/2}$ = 10.2 d, E_γ = 935 keV), $^{46}Ti(n,p)^{46}Sc$ ($T_{1/2}$ = 84 d, E_γ = 889 keV, 1120 keV), $^{98}Mo(n,\gamma)^{99}Mo$ ($T_{1/2}$ = 64 h, E_γ = 140 keV, 181 keV, 739 keV, ect.). The photopeak intensities of these nuclides were very high compared to those of radionuclides formed from impurity elements.

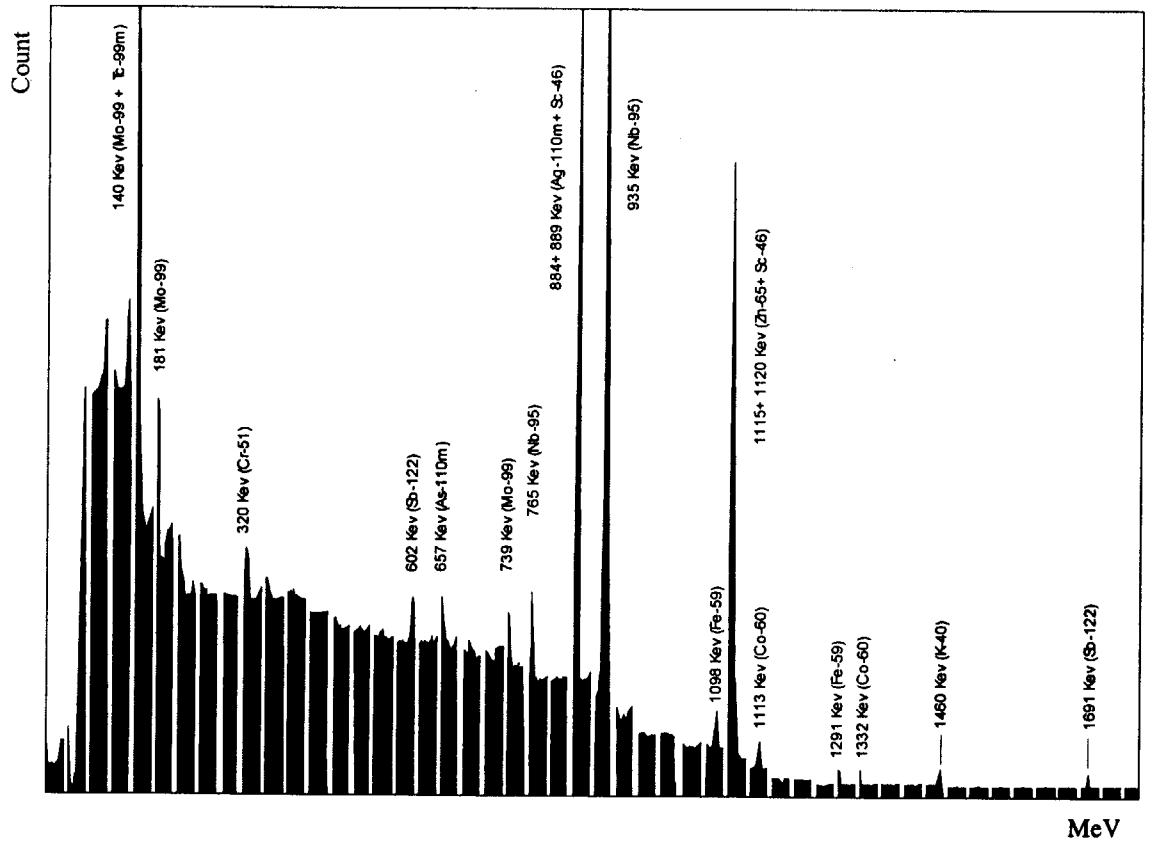

Fig. 2. Gamma spectrum of TiMo target irradiated at the neutron trap of reactor

Table 3. Concentration of trace in aluminum foil and polyethylene vial

Elements	Analytical results (ppm)	
	Aluminum foil	Polyethylene vial
Sb	0.1 ± 0.03	
Fe	8500 ± 400	
Zn	40 ± 4	
Cr	15 ± 1	
Co	4.5 ± 0.05	
Sc	0.25 ± 0.04	
Na	0.1 ± 0.03	8.2 ± 0.3

Eight trace chemical elements found in TiMo target and different radioisotopes of Ti and Mo will be the main impurities in the final ^{99m}Tc-pertechnetate solution eluted from ^{99}Mo-^{99m}Tc generator using this type of target. However, the concentration of these contaminant elements is very low and is only of trace level. The results of observation of radionuclides in activated targets were used for the identification of contaminant radionuclides in ^{99m}Tc pertechnetate solution eluted from a gel-type ^{99}Mo-^{99m}Tc generator. For determination of long half-lived nuclides, the gamma ray spectra of ^{99m}Tc eluate were measured after decay of about one month from elution. For the determination of short half-lived nuclides in ^{99m}Tc solution, 10 ml of pertechnetate eluate of radioactivity of 100 mCi were put in a standard lead pot of 3 mm thickness to reduce the activity of ^{99m}Tc and gamma spectra were recorded after about one hour of decay time.

The result of analysis of the impurity radionuclides in the ^{99m}Tc solution was shown in Table 4. Almost radionuclides formed in the activated TiMo target were detected in ^{99m}Tc pertechnetate eluate. However, the nuclides which were produced via (n,p) reaction such as ^{46}Sc, ^{92}Nb, ect. did not appear in the final ^{99m}Tc solution.

Table 4. The results of analysis of impurity radionuclides in ^{99m}Tc solution

Nuclide	Activity mCi per mCi of ^{99m}Tc	Nuclide	Activity mCi per mCi of ^{99m}Tc
^{59}Fe	$0.4\ 10^{-9}$	^{51}Cr	$1.0\ 10^{-9}$
^{65}Zn	$0.5\ 10^{-9}$	^{24}Na	$5.0\ 10^{-9}$
^{60}Co	$0.1\ 10^{-9}$	^{82}Br	$1.8\ 10^{-9}$
^{110m}Ag	$5.0\ 10^{-9}$	^{99}Mo	$2.5\ 10^{-6}$

From the results shown in Table 4 and the recorded gamma ray spectra of final ^{99m}Tc product, it is stated that almost the impurity nuclides in the ^{99m}Tc solution eluted from our generator are produced from trace element presented in the TiMo target material and from the Ti and Mo element via (n,p) reaction. However, in the ^{99m}Tc solution appeared a nuclide ^{82}Br, which had not been found in activation analysis. It means that concentration of this element in TiMo target has less than determination limit of analysis method.

The radioactivities of impurity nuclides in the ^{99m}Tc solution were only approximately 10^{-9} mCi for the nuclides of trace elements and 10^{-6} mCi per mCi ^{99m}Tc activity for ^{99}Mo. It means that the concentration of impurity nuclides of trace elements and ^{99}Mo in Tc-99m eluate is approximately of 10^{-7}% and of 10^{-4}%, respectively. These concentrations are much less than the limit prescribed by European Pharmacopoeia /EP/.

In our work, impurity of β emitting nuclides also was determined. The count numbers of β emitting impurity were measured on a liquid scintillation counter and its the detection efficiency determined with ^{32}P standard solution was 90%. The concentration of β emitting impurity was less than 10^{-7}% while the limit prescribed by EP is 6.10^{-4}%.

CONCLUSION

All obtained results reveal the fact that TiMo target has very small quantity of trace elements. The radioactive nuclides formed under neutron activation of TiMo target were eluted with Tc-99m pertechnetate from a TiMo based Tc-99m generator column and a trace quantity of radionuclidic impurities in Tc-99m eluate was found lower than the limit prescribed by European Pharmacopoeia.

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1.16 Performances and present situation of PZC for (n, gamma) ^{99}Mo - ^{99m}Tc Generator

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Abstract

The practicability of ^{99m}Tc generator based on (n, gamma) method using PZC (Poly Zirconium Compound), the Mo adsorbent with high adsorption efficiency, has been studied for several years by a collaboration research between National Nuclear Energy Agency of Indonesia (BATAN) and KAKEN Co., and it was also highlighted and confirmed by FNCA member countries at the last workshop held in Beijing, China. As a part of the FNCA activities, PZC has been distributed 3 times to the member countries from KAKEN within the last year (2002) to confirm the reproducibility and practicability of the ^{99m}Tc generator using PZC (PZC-^{99m}Tc generator). BATAN themselves has been continuously proceeded the advanced studies and the practical data on the ^{99m}Tc elution and ^{99}Mo breakthrough has been obtained. Even though unexpected amount of ^{99}Mo breakthrough accompanied with the ^{99m}Tc elution has been currently occurred from the PZC-^{99m}Tc generator, the problem was solved by subsequently adding an alumina column underneath the generator column contained PZC. In accordance with the realization of the practical PZC-^{99m}Tc generator, an administering tests to the mice with labeled compound of ^{99m}Tc generated by PZC method have already been started by BATAN, the plan to fabricate an automatic loading testing system to simplify the process of the ^{99}Mo adsorption to PZC and the ^{99}Mo-PZC packing to the generator column has been proceeded by BATAN and KAKEN for realizing a mass production method of ^{99m}Tc generators, and moreover the Japanese patent regarding the PZC-^{99m}Tc generator, its production process and the automatic loading system of ^{99}Mo to PZC has already been applied with joint application of BATAN and KAKEN.

The 3 main subjects, experimental results of the advanced studies on PZC-^{99m}Tc generator performed by BATAN, description of the Mo loading system and the outline of the patent applied in Japan are introduced in this paper.

1. Experimental results of advanced studies on PZC-^{99m}Tc generator

1.1 Current studies

Experiments and studies on PZC-^{99m}Tc generator have been performed for about 8 years since the Mo adsorbent with high Mo adsorption efficiency called PZC was developed by the collaboration research of JAERI (Japan Atomic Energy Research Institute) and KAKEN from 1994. Experiments using low ^{99}Mo activity such as less than 1mCi had initially been studied by the collaboration

research of JAERI and KAKEN before the collaboration of BATAN and KAKEN contracted the joint research agreement at June 2000. It is one of the advantages of the BATAN-KAKEN joint research that the experiments using a commercial level of ^{99}Mo activity such as 30-300mCi obtained by BATAN's reactor have been possibly performed by BATAN.

An outline of the current experimental results with the development steps regarding the ^{99m}Tc elution and the ^{99}Mo breakthrough from the PZC-^{99m}Tc generator is shown in Table 1.

Table 1. Current experimental results on ^{99m}Tc elution and the ^{99}Mo breakthrough of PZC-^{99m}Tc generator based on (n, gamma) ^{99}Mo method

Adsorption rate of Mo: av. 240 mg(Mo)/g(PZC)

Year	step	^{99m}Tc elution (%)	^{99}Mo breakthrough[1] (%)	^{99}Mo activity		
~99	1	av. 80	0.27	~1mCi (37MBq)	PZC only	JAERI-KAKEN
~99	2	av. 80	0.003	~1mCi (37MBq)	PZC+Alumina	↑
~00	3	av. 75	0.002	~30mCi (1GBq)	PZC+Alumina	↓
~01	4	av. 10	0.2	~300mCi (11GBq)	PZC only	BATAN-KAKEN
~02	5	av. 90	<0.00001	~300mCi (11GBq)	PZC+Alumina +0.5% NaOCl	

1) Law limit of ^{99}Mo breakthrough (in Japan) is 0.15 kBq(^{99}Mo)/MBq(^{99m}Tc); 0.015 %.

Step 1: By an experiment using PZC generator column containing a low ^{99}Mo activity less than 1mCi, the results of a high ^{99m}Tc elution of av. 80% and a high ^{99}Mo breakthrough of 0.27 were obtained. ^{99}Mo breakthrough of 0.27% was not expected because the amount is much higher than that of Radiopharmaceutical Law Limit of Japan (0.015%).

Step 2: In order to decrease the amount of unexpected ^{99}Mo breakthrough, an alumina column was subsequently set underneath the PZC column to trap the ^{99}Mo eluted together with ^{99m}Tc, and low ^{99}Mo breakthrough of 0.003% was successfully obtained.

Step 3: In BATAN, by an experiment using PZC generator column containing higher ^{99}Mo activity of ~30mCi with an alumina column, high ^{99m}Tc elution of av. 75% and low ^{99}Mo breakthrough of 0.002 were stably obtained.

Step 4: By an experiment using PZC generator column containing practical and commercial level ^{99}Mo activity of ~300mCi, a very low ^{99m}Tc elution of av. 10% and a high ^{99}Mo breakthrough of 0.2% were obtained. The low ^{99m}Tc elution obtained at a high level ^{99}Mo was assumed that

a reduction reaction of heptavalent ^{99m}Tc was occurred by a high radiation circumstance.

Step-5: From the result of Step-4, using a saline solution including NaClO as an oxidant and an alumina column for trapping ^{99}Mo breakthroughed, the results of constantly high ^{99m}Tc elution and very low ^{99}Mo breakthrough were stably obtained. By the results, the practicability and commercial activities of PZC was confirmed.

Furthermore, from the results for comparing PZC method to a gel method by experimental and desk studies, it was cleared that PZC method has practicable, environmental and economical merits, and high performances rather than gel method.

2. Description of loading system

2.1 Introduction

PZC-^{99m}Tc generator has been currently produced by the manual procedures through 5 main processes as follows;

(1) Preparation of the original ^{99}Mo solution

Mo target irradiated is dissolved in NaOH solution and the volume, pH, concentration and specific activity of ^{99}Mo are precisely adjusted.

(2) ^{99}Mo adsorption to PZC

^{99}Mo solution of about 15-20 ml is added to the PZC of 1 g in a flask, then the mixture is intermittently and mildly stirred in a heated condition at 90 degree Celsius for 3 hours.

(3) Washing treatment of PZC containing ^{99}Mo

After the process of ^{99}Mo adsorption to PZC, supernatant original ^{99}Mo solution is removed out from the flask, then the remained PZC containing ^{99}Mo in the flask is sufficiently washed by water or saline solution.

(4) Column packing of PZC containing ^{99}Mo

After the washing treatment, PZC-^{99}Mo in the flask is transferred into a glass column with a filter at near column end for packing as ^{99m}Tc generator.

(5) Condition adjustment of the column

The column containing PZC is washed again by saline solution with checking its pH (6~7), and the column is finally set in an apparatus as PZC-^{99m}Tc generator.

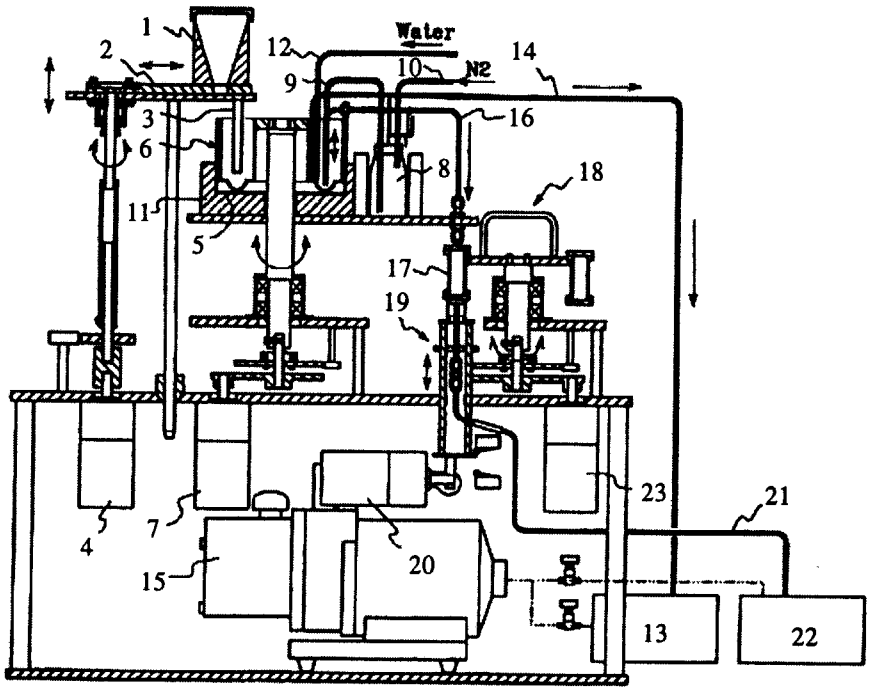
It takes about 6 to 7 hours to produce one lot generator columns by manual procedures as above, and it is not a significant problem under the experimental level to study or investigate the efficiency of PZC-^{99m}Tc generator. On the other hand, mass production of PZC-^{99m}Tc generator is expected to be realized since the technology has become practical level. Therefore, an automatic system enabling to perform the processes of ^{99}Mo adsorption and PZC packing automatically, so called a loading system, for mass production of the generator columns have been designed by KAKEN. The system

will be fabricated by BATAN-KAKEN and installed in a hot cell of BATAN in the near future.

2.2 System features

The loading system in the planning stage is mainly composed of a column packing section followed by an adsorption vessel made of glass where the ^{99}Mo adsorption to PZC is proceeded. A brief image of the loading system (side view and top view) is shown in Fig.1 and a top view and a side view of the adsorption vessel is shown in Fig.2.

Side view



Top view

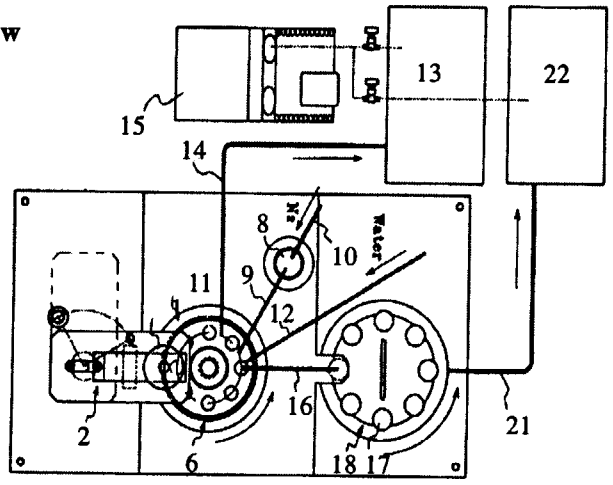


Fig.1 System of ^{99}Mo loading to PZC and column packing.

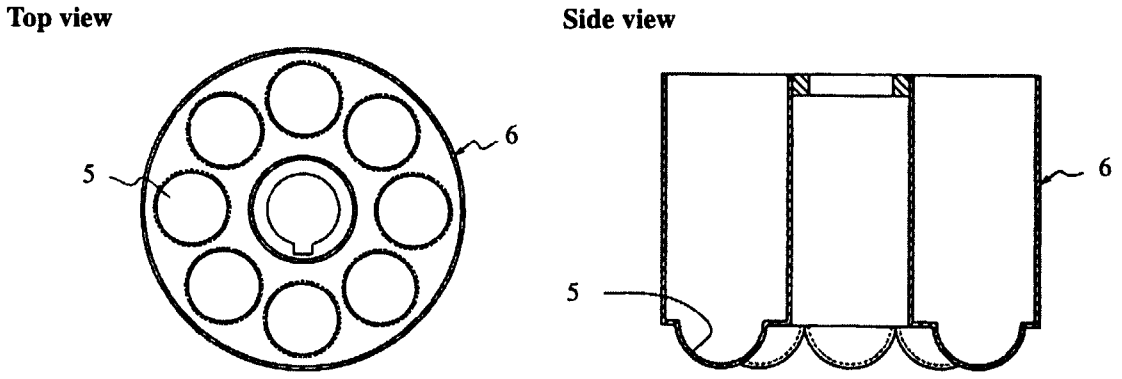


Fig.2 ^{99}Mo adsorption vessel (glassware).

The system features include mechanisms as follows;

(1) Batch off a specific amount of PZC to an adsorption vessel from PZC tank.

PZC is distributed from a container #1 to a constant volumetric hole with a slide board #2. The board #2 is horizontally slid between the bottom side opening of the container #1 and the topside opening of a guide pipe #3. As shown in Fig.1, when the bottom side opening of the container #1 and the topside opening of a hole of the slide board #2 are at the same position, PZC is fed from the container to the hole. The board #2 is then slid by a motor #4 horizontally, and when the bottom side opening of the hole placed upon the topside opening of the guide pipe #3, a specifically amounted PZC corresponding volume of the hole is then batched off and transferred into a crater (bowl shape) #5 of the adsorption vessel #6 through the guide pipe. The adsorption vessel made of glass has 8 craters at the bottom and each crater holds a specified amount of PZC. After a specifically amounted PZC (1~3 g or more) is fed into one of the craters, the vessel #6 is turned 45 degree while a back-and-forth movement of the slide board by a stepping motor #7 cooperatively. By repeated procedures, multiple sets (1-8 sets) of PZC are fed into each crater of the adsorption vessel.

(2) Add the ^{99}Mo solution into the adsorption vessel containing multiple sets of a specifically amounted PZC.

Original ^{99}Mo solution is contained in a vessel #8 and a specified amount of the solution is added to the adsorption vessel #6 through a pipe #9 by N2 gas injected to a vessel #8 through a pipe #10. Because of the characteristically designed shape of the adsorption vessel, the same ^{99}Mo solution evenly covers all and each craters containing PZC at a time.

(3) Heat the adsorption vessel and stir the mixture to promote the adsorption reaction of ^{99}Mo and PZC.

After the ^{99}Mo solution covers all crater containing PZC, the adsorption vessel is then heated

by a heater #11 and mildly stirred by a rotation with a fine wire set at the edge of a feeding pipe of ^{99}Mo or washing solution (which is not illustrated in Fig.1). Other processes after ^{99}Mo adsorption to PZC such as a washing treatment are also proceeded in the adsorption vessel. Saline solution is added to the adsorption vessel by a pipe #12 and the waste solution is removed by suction to a waste tank #13 through a pipe #14 using a vacuum pump #15.

(4) Transfer the PZC -^{99}Mo slurry to a glass column.

After the processes of ^{99}Mo adsorption to PZC and washing treatment, a pipe #16 for transfer of the PZC -^{99}Mo slurry to a glass column #17 held at a column holder #18 is lifted down and immersed to the PZC -^{99}Mo slurry placed at the crater. At the same time at the column packing section, a nipple holder #19 is lifted up by a motor #20, and a needle placed at the topside of a waste exhaust pipe # 21 is then inserted to a rubber plug placed at the bottom side of a glass column #17. By lifting up the nipple holder #19, the glass column is also lifted up from a column holder #18, and a slim pipe placed at the bottom-side of a pipe #16 is inserted to a rubber plug placed at the topside of a glass column. Followed by the glass column connection as above, a waste tank # 22 is depressurized by a vacuum pump #15 and the glass column is also depressurized, then the PZC -^{99}Mo slurry is transferred to the glass column through the pipe #16 from one of the craters. After a column packing process, the slim pipes inserted to both the topside and the bottom side of a glass column are removed out by a reverse motion of that of column connection, and the glass column loaded PZC -^{99}Mo slurry is set back to the column holder.

(5) Perform the same processes of (1)-(4) several times repeatedly.

After the glass column loaded PZC-^{99}Mo slurry is set back to the column holder, the holder is then turned 45 degree by a stepping motor #23 and the same procedures of PZC column packing is continuously proceeded for 8 times at most in the case of 8 channels. The channel number can be built up e.g. 12 channels.

2.3 Fabrication of loading system

The loading system will be fabricated by BATAN-KAKEN and installed in a hot cell located at one of the BATAN's research buildings in a near future. A production process will be simplified and significant improvement in the production capacity can be realized by employing the loading system for the production of PZC-^{99m}Tc generator. Because of the characteristically designed shape of the adsorption vessel as shown in Fig.2, 8 sets of the PZC -^{99}Mo slurries are possibly obtained in a process of ^{99}Mo adsorption, which is to say that 8 times more generator can be obtained as compared with the current manual procedures.

The loading system is designed as a prototype of actual system, in other words, the system

designed will be modified and advanced to an actual system for the commercial production of the generators in the future. For the first step of a future plan, the prototype system will be operated and examined to confirm whether the system operates the same procedures as that of manual procedures, and clinical trial will be experimentally attempted in Indonesia on the basis of about 10 generators production per week as a second step. It is expected that a mass production of the PZC-^{99m}Tc generator will be soon realized by taking the opportunity afforded by the fabrication of the loading system.

3. Patent

3.1 Introduction

A patent named "A Mo(n, gamma)^{99}Mo-^{99m}Tc generator system using Poly Zirconium Compound of selective Mo adsorbent and its Producing System" has been applied to the Japanese Patent Office dated on August 31st, 2002 by collaborated application of BATAN and KAKEN. The patent applied mainly includes the procedures of the PZC-^{99m}Tc generator production and the features a loading system.

3.2 Claims of Patent

As to the contents of the patent, there are 4 claims regarding the PZC-^{99m}Tc generator and 3 claims regarding ^{99}Mo loading and the production system of ^{99m}Tc generator column mentioned as follows.

Claim 1: PZC-^{99m}Tc generator characterized in that PZC as a ^{99}Mo adsorbent is used.

Claim 2: PZC-^{99m}Tc generator characterized in that PZC adsorbing ^{99}Mo produced by the (n, gamma) method of natural Mo is packed in a generator glass column.

Claim 3: PZC-^{99m}Tc generator characterized in that saline solution, NaOCl or other oxidants is used as an eluent to elute ^{99m}Tc from the generator.

Claim 4: PZC-^{99m}Tc generator characterized in that an alumina column to trap ^{99}Mo to be eluted from the generator simultaneously with the ^{99m}Tc elution is set underneath the PZC-^{99m}Tc generator column to obtain high purity ^{99m}Tc solution.

Claim 5-7: Loading system characterized in that the mechanisms as mentioned in 2.2 are featured.

A patent with the same contents will be also soon applied in Indonesia based on the collaboration application of BATAN and KAKEN. The patent has not been applied to obtain the exclusive right to anyone but to give an evidence for the progressive results obtained by BATAN-KAKEN joint research. Therefore the technology included in the patent is fully and freely opened to only FNCA member countries; this is KAKEN's opinion.

4. Conclusion

By the current studies and examinations, we have almost had substantial data related to the PZC method, and we are confirming the hard-and-fast conclusions for the practical use of (n, gamma)^{99}Mo-^{99m}Tc method using PZC. Hereafter, we will examine and establish the PZC method from a view point of engineering and try to spread the technology to the world.

1.17 Strategy for Commercial Application of Tc-99m Generator Produced by PZC Technology

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FNCA Coordinator of Japan

1. Introduction

Most FNCA countries import Mo-99 for the production of Tc-99m generators by paying valuable hard currency. In order to decrease the import, the establishment of technology to produce Tc-99m generator using existing research reactors, and its commercial application benefit FNCA countries. The current demand of Tc-99m and number of γ -cameras in FNCA countries are listed in Table 1.

2. Strategy

The strategic plan is proposed below. Each country should prepare its own strategy in detail to be reported to the project leader of Japan.

(1) Survey of the current and potential size of market of Tc-99m generator in cooperation with the Ministry of Health
- Number of γ -cameras
- Number of tests for diagnosis

(2) Cost of the imported generators

(3) System for the distribution of generators
- Master milker
- Individual milker at the hospitals

(4) Production plan
- Production of Mo-99 by n-γ reaction
- Design of production facilities to meet the market size to be assisted by Kaken Co.
- Manufacturing of the facilities of local technology
- Investor - national nuclear research center or private company
- Cost analysis
  - Initial investment
  - Operation cost
  - Sales income
- Market survey

In order to achieve the commercial application the good coordination and collaboration with nuclear medicine experts of hospitals and pharmaceutical company are of great importance.

3. Government Policy

The government should play an important role because the nuclear reactors are government own and governments are responsible for better utilization of reactors.

(1) Support to local products

(2) Government investment - national budget

(3) International cooperation

Table-1 Demand of Tc-99m and number of γ -Cameras in FNCA countries

	Demand of Tc-99m		Demand of Mo-99		Share	Number of Gamma-cameras
	Ci/year	Ci/week	Ci/year	Ci/week		
1) JAPAN	17,200.0	344.0	8,600.0	172.0	44.5%	1923
2) KOREA	7,000.0	140.0	3,500.0	70.0	18.1%	200
3) CHINA	13,000.0	260.0	6,500.0	130.0	33.6%	350
4) INDONESIA	220.0	4.4	110.0	2.2	0.6%	20
5) MALAYSIA	260.0	5.2	130.0	2.6	0.7%	16
6) THE PHILIPPINES	290.0	5.8	145.0	2.9	0.8%	19
7) THAILAND	430.0	8.6	215.0	4.3	1.1%	37
8) VIETNAM	250.0	5.0	125.0	2.5	0.6%	14
TOTAL	38,650.0	773.0	19,325.0	386.5	100.0%	

4. Conclusion

Through joint effort of government research institute and pharmaceutical company, commercial production of Tc-99m generator using the PZC method to be fully developed under the FNCA should be achieved in the near future.

1.18 The Trend of ^{99m}Tc Generator in FNCA Countries

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Abstract

In the 2001 Workshop on Utilization of Research Reactors held in Beijing, eight delegates from the FNCA countries, namely China, Indonesia, Japan, Korea, Malaysia, the Philippines, Thailand and Vietnam, for the ^{99m}Tc Generator Project, presented papers or orally pleaded the current status of utilization and production in terms of ^{99}Mo solution, ^{99}Mo/^{99m}Tc generator or extracted ^{99m}Tc solution in each country. This paper is a brief compilation of these topics and some additional information obtained afterward the workshop.

1. Introduction

The worldwide demand of ^{99}Mo as the parent nuclide of ^{99m}Tc exceeded 300,000 Ci. The greater part of ^{99}Mo is produced by two countries, Canada and Belgium. They are supplying more than 80% of world demand of ^{99}Mo. Their production method is based on nuclear fission of enriched uranium nevertheless crucial problems in point of view of environment, cost and safety.

A new and efficient ^{99m}Tc generator production method is under developed in the framework of FNCA project using PZC (Poly Zirconium Compound), that is

-Production of ^{99}Mo from natural molybdenum (^{98}Mo) by (n, γ) reaction,

-Adsorption of ^{99}Mo into PZC, and

-Preparation of ^{99m}Tc generator with the column of PZC/^{99}Mo.

Significant social benefits of stable supply of radioisotopes for nuclear medical diagnosis can be expected, and the local production will give economic effects to save foreign currency. In the long run, this project could formulate ^{99m}Tc supply regional network among the FNCA countries.

2. Status of ^{99}Mo production and ^{99}Mo/^{99m}Tc generators in each country

2.1 China

The China Institute of Atomic Energy (CIAE, Beijing) is routinely producing and distributing the generators with fission Mo (160 Ci/w, partially imported from overseas

because of reactor operation schedule) 4 times a month and the Nuclear Power Institute of China (NPIC, Chengdu) is producing based on zirconium molybdate gel with neutron activated (n, γ)Mo (150 Ci/w) twice a month. China is one of the two countries where the gel generator technology has been successfully established as has been done by Vietnam.

More than 800 hospitals have nuclear medicine department and more than 350 SPECT(γ -cameras) and 12 PET systems are being operated in China. Before 1996, most nuclear medicine hospitals were labeling ^{99m}Tc kits by using eluted ^{99m}Tc from fission or gel generators. After that 7 "milk stations" by which ^{99m}Tc kits were prepared and provided to nearby hospitals were established in Beijing, Shanghai, Guangzhou, Shenyang and Chengdu.

Two statistics of production and supply of ^{99m}Tc related radioisotopes in China and from CIAE are given in Table 1 and Table 2, respectively.

Table 1. Annual production of ^{99m}Tc related radioisotopes in China

Product	Actual production yield, Ci	Remarks
^{99}Mo (^{99m}Tc generaor)	6500 Ci	50% ^{99}Mo from activated ^{98}Mo 50% ^{99}Mo imported (fission)
^{99m}Tc kit	1,000,000	

Table 2. Production and supply of ^{99m}Tc related radioisotopes from CIAE (2000)

Product Production	Production/Sale	Import	Remarks
Fission ^{99}Mo	3000 Ci	4000 Ci	
^{99m}Tc generator	3500 Ci		At calibration time
^{99m}Tc cold kit	25000 vials		
Ready-to-use ^{99m}Tc kits in Beijing in Shanghai in Guangzhou	 26000 syringes 18200 18200		

2.2 Indonesia

In Indonesia the state enterprise BATAN Technologi Inc. (BATEK) is routinely producing the fission Mo generator (2 Ci/w) by using reactors of 30 MW (at moment 15 MW) RSG-GAS and TRIGA-II (2 MW). Annual shipment of generators was 265 units in 2000. In the Center for Development of Radioisotopes and Radiopharmaceuticals of BATAN, research and development of PZC generator is intensively being carried out along with the MOU with Kaken Co. Ltd. of Japan. The details of achievement will be presented and demonstrated in this workshop.

The table 3 shows the actual production rate of radioisotopes of ^{99m}Tc related by BATEK. Drastic decrease of fission ^{99}Mo production in 2000 is said to be due to

inspection by the regulatory body to secure safety of uranium target irradiation in the RSG-GAS reactor. However, the supply amount was maintained as usual by supplementary import from Australia. According to the data provided from the marketing section of BATEK, the amount of ^{99}Mo supplied in 2000 was 1103 Ci.

Table 3. Production rate by Batan Teknologi Inc.

Product	1998	1999	2000
Fission ^{99}Mo (Ci)	2002	1746	783
^{99m}Tc generator (unit)	280	260	265
^{99m}Tc kits (vial)			
^{99m}Tc-MDP	91	95	61
^{99m}Tc -HEDSPA	91	95	61
^{99m}Tc -DTPA	31	25	34

The capacity of manufacturing facility of BATEK for fission ^{99}Mo and ^{99m}Tc generators is quite impressive as shown in Table 4. The facility was designed by General Atomic Inc and production technology was licensed by US Mediphysics Inc.

Table 4. Nominal production capacity of Batan Teknologi's facility

Product	Production capacity/year
Fission ^{99}Mo	120,000 Ci
^{99m}Tc generator	12,000 units

Since 1995, Japan Atomic Energy Research Institute (JAERI) and Kaken Co. Ltd. have been collaborating to find out the ^{99}Mo adsorbent material of poly zirconium compound (PZC) fit for so called (n, γ) method. In 2000 the Kaken and the National Nuclear Energy Agency (BATAN) of Indonesia has facilitated to promote the technology of PZC based ^{99m}Tc generator under the joint research agreement.

2.3 Japan

The demand of ^{99}Mo in Japan is as large as almost one tenth of worldwide demand and whole amount is imported from Canada on weekly basis. In Japan, all of ^{99m}Tc is consumed for the preparation of radiopharmaceuticals and the amount of ^{99m}Tc based radiopharmaceuticals exceeds 80 ~ 90% of all kind of radioisotopes for nuclear medicine.

The Japan Atomic Energy Research Institute (JAERI) had been developing the technology of both fission and (n, γ) ^{99}Mo to supply to radiopahamaceutical manufacturers but because of insufficient production capacity compared with huge

demand (3000 Ci/w) of nuclear medicine in Japan, the routine distribution system failed to be established.

The followings are the selected extract of the "Statistics of Distribution of Radioisotopes, Issue 2002" compiles by the Japan Radioisotope Association.

Total amount of radiopharmaceuticals in 2001 was 59.6 billion yen (~US$ 500 million), where 46.9 billion yen (~US$ 390 million) of *in vivo* and 12.7 billion yen (US$ 106 million) of *in vitro*. The amount of *in vivo* increased slightly (0.7%) comparing with previous year while the *in vitro* decreased by 5.9%. Share of the *in vivo* and the *in vitro* was 78.7% and 21.3% of the total amount, respectively.

Table 5. Distribution of ^{99m}Tc generator and ^{99m}Tc solution in Japan

Product	1997	1998	1999	2000	2001
^{99m}Tc generator (Ci: ^{99}Mo)	4846	4876	4879	4852	4744
^{99m}Tc solution (Ci:^{99m}Tc)	6140	6691	7223	7687	8243

Total amount of sum of ^{99m}Tc and ^{99m}Tc generator exceeds 85% of all kind of radioisotopes for nuclear medicine. Amount of ^{99m}Tc solution in 2001 showed 7.3% increase comparing with the previous year 2000 but ^{99m}Tc generator decreased by 2.2%. It seems to be influenced by terrorism broken out in September 11, 2001 in US.

Table 6. Supply of cold kits for ^{99m}Tc labeling in Japan (unit: vials)

Region of diagnosis	1997	1998	1999	2000	2001
1) heart, lung	73,690	76,555	83,815	89,229	89,738
2) brain	74,763	75,997	77,560	73,338	69,027
3) liver, spleen	7,905	6,721	5,558	5,187	5,582
4) kidney	30,707	29,929	29,227	28,741	26,818
5) bone	129,618	130,997	133,581	135,856	132,539
Total	316,683	320,199	329,791	332,351	323,704

1) HAS, MAA, MIBI, Tetrofosmin

2) ECD, HM-PAO

3) Phytate, Tin Colloid

4) DMSA, DTPA, MAG_3

5) HMDP, MDP, PYP

Table 7. Supply of ready-to-use ^{99m}Tc kits (Injection) in Japan (unit: Ci)

Region of diagnosis	1997	1998	1999	2000	2001
1) heart, lung	926	1107	1228	1284	1451
2) brain	513	568	650	695	706
3) liver, gall, spleen	109	107	107	104	101

4) kidney	154	146	148	142	133
5) bone	4262	4590	4922	5302	5694
6) others	176	171	167	160	160
Total	6139	6691	7221	7687	8244

1) ^{99m}Tc HAS-DTPA, ^{99m}Tc MAA, ^{99m}Tc MIBI, ^{99m}Tc Tetrofosmin

2) ^{99m}Tc ECD

3) ^{99m}Tc GSA, ^{99m}Tc PMT, ^{99m}Tc Tin Colloid

4) ^{99m}Tc DMSA, ^{99m}Tc MAG_3

5) ^{99m}Tc HMDP, ^{99m}Tc MDP

6) $^{99m}TcNTcO_4$

2.4 Korea

The Korea Atomic Energy Research Institute (KAERI) is supplying extracted ^{99m}Tc solution by MEK method from (n, γ) ^{99}Mo produced with the HANARO (30 MW) reactor. The fission based ^{99}Mo/^{99m}Tc generator is imported from UK and Canada at moment but KAERI has launched a new project with the scope of mass production of generators with fission Mo. The tubular targets, electrodeposited with high enriched uranium oxide on inner surface, were proposed for annual production of 4,200 Ci ^{99}Mo.

The gel generator with (n, γ) ^{99}Mo has also been investigated along with the development of fission ^{99}Mo generator and now preparing the trial study of PZC type generators with ^{99}Mo from enriched target irradiated in high neutron flux.

Table 8. Status of fulfillment of ^{99m}Tc generator and eluted solution in Korea (2000)

	No. of users	Local supply	Import	Local fulfillment
^{99m}Tc generator	105	0	3210 Ci	0%
^{99m}Tc solution	105	226 Ci	0	100%

Whole demand of ^{99m}Tc generators is fulfilled by import but the demand of ^{99m}Tc solution can be met by local production. Table 9 shows an itemized breakdown of the solution distributed in 2000.

Table 9. Annual usage of ^{99m}Tc solution by KAERI, Korea (2000)

Cold kits	Supply (vials)	Cold kits	Supply (vials)
^{99m}Tc-Phytate	1,465	^{99m}Tc-PYP	610
- MDP	4,450	- HAS	150
- DISIDA	3,905	- MAA	910
- DTPA	2,905	- ASC	450
- Tin	1,600	- DMSA	1,000
		Total	17,454 vials

The total amount of distribution increased to more than 19,000 vials in 2001.

In Korea presently more than 200 γ - cameras are in use in 96 hospitals and demand for ^{99m}Tc generators is over 150 units per week. In order to overcome the situation of full rely on overseas for the ^{99m}Tc generator supply, the generator loading facility of approx. 120 m^2 in the hot cell area for assembly of the generators was constructed. Production and supply of weekly 200 units of the generators is expected by 2003.

In order to meet significantly increasing demand of ^{99}Mo, introduction of solution fuel reactor equipped with a loop system for ^{99}Mo extraction is under the feasibility study.

2.5 Malaysia

The Malaysian Institute for Nuclear Technology Research (MINT) is supplying ^{99}Mo/^{99m}Tc generators ranging 0.2 ~ 1.2 Ci per unit loaded with fission ^{99}Mo (8 Ci/w) which is imported from BATEK Indonesia to 10 hospitals every week. MINT has a facility with a capacity of 50 units assembly with additional 50 units spare. It also supplies ^{99m}Tc in form of the ready to inject radiopharmaceuticals to 2 hospitals around Kuala Lumpur. At moment import from Indonesia is halted and renovation of facility is going on. About 20 nuclear medicine and radiotherapy centers are using radiopharmaceuticals produced by MINT. The number of γ -cameras is 16 and number of in-vivo diagnosis is approximately 9,000~10,000 cases per year. The market scale of radiopharmaceuticals in Malaysia is estimated as annually US$ 1~2 million while other pharmaceuticals US$ 250 million and traditional medicines US$ 500 million. Suppliers of radiopharmaceuticals are Amarsham, Australia Radioisotope (ARI), NEN/DuPont, Mallincrodt and MINT.

Table 10. Survey on annual demand of ^{99m}Tc generators (implemented in 1995)

Institutions	Annual demand of ^{99m}Tc generators
Kuala Lumpur Hospital	52 units (600 mCi/unit)
P.J. University Hospital	52 units (800 mCi/unit)
Kubang Krian University Hospital	30 units (200 mCi/unit)
Kuching Sarawak General Hospital	24 units (200 mCi/unit)
CRC	24 units (200 mCi/unit)
Total	183 units

2.6 The Philippines

In the Philippines, the TRIGA type reactor PRR1 (3 MW) in the Philippine Nuclear Research Institute (PINRI) is under repair therefore irradiation for production of ^{99}Mo is

not available. The fission ^{99}Mo (6 Ci/w) is being imported by private companies from the Australian Radioisotope (ARI) at the moment to fulfill the minimum demand. However, research on the gel type generator has been conducted in the past motivated by the importance of wider availability at low cost and safe handling process of the (n, γ) ^{99}Mo generators.

It is very impressive to see in the Philippines the way of obtaining simulated (n, γ) ^{99}Mo without reactor operation by using fission ^{99}Mo recovered from an alumina column of a used commercial ^{99m}Tc generator and mixed with carrier molybdenum solution.

2.7 Thailand

In Thailand, the Office of Atomic Energy for Peace (OAEP) has produced ^{99m}Tc by (n, γ) reaction followed by MEK extraction process by 1~2 Ci of ^{99m}Tc per batch using TRR1/1 reactor. Because of the difficulty to meet the increased demand and limitation of reactor operation, the production of ^{99m}Tc was stopped in 1997 as can be seen in the Table 11. Study for gel generators based on zirconium molybdate and titanium molybdate were conducted in the framework of IAEA's initiative. Currently hospitals are importing generators from overseas (6~7 Ci/w).

In the ongoing project of new Ongkarak Nuclear Research Center highlighted with construction of 10 MW research reactor, associated facilities consist of clean rooms and loading/assembly plant for ^{99}Mo/^{99m}Tc generators are expected to promote enough supply to meet the domestic demand in near future.

Table 11. ^{99m}Tc consumption in Thailand

Year	Domestic (OAEP) ^{99m}Tc solution (Ci : ^{99m}Tc)	Import ^{99m}Tc generator (Ci : ^{99}Mo)
1987	96.90	94.30
1988	101.00	-
1989	118.30	87.70
1990	205.90	42.20
1991	72.20	142.00
1992	-	143.30
1993	58.90	137.10
1994	200.80	105.60
1995	166.10	130.70
1996	147.50	131.40
1997	55.00	161.10
1998	-	188.50
1999	-	212.90
2000	-	214.10

2.8 Vietnam

Dalat Nuclear Research Reactor of nominal power of 500 kW is the unique one in Vietnam. More than 90% of reactor operation time and over 80% of reactor irradiation capacity have been exploited for radiopharmaceutical production. The radioactivity of more than 150 Ci of various radioisotopes including ^{99}Mo and ^{99m}Tc was annually produced and distributed.

Number of nuclear medicine centers is 31 and they are located all over the country. Number of γ -cameras (planar and SPECT) is 14. The share of locally manufactured products of the total market is 60% and to increase market share of ^{99m}Tc generators, loading with imported ^{99}Mo solution is planned.

The Nuclear Research Institute (NRI) is supplying monthly 10 units of gel generators (sum of both zirconium and titanium molybdate gel generators). The loaded amount of ^{99}Mo for zirconium gel is possible to reach 1 Ci per unit. For the titanium gel 0.1~ 0.2 Ci of ^{99}Mo can be loaded to be irradiated after non-active molybdenum is adsorbed to make the process of preparation simple and safe. Currently hospitals are importing fission type generators from overseas (4~5 Ci/w). In near future (n, γ)^{99}Mo will be imported to produce gel type generators. The Vietnam is the unique country where the gel generator technology has been established and distributing them regularly.

Table 12. The supply / demand balance for ^{99m}Tc generator and ^{99m}Tc kits in Vietnam

Product	Supply	Demand
^{99m}Tc generator	10 units/month (500 mCi/unit)	15 units/month (500 mCi/unit)
Kits for ^{99m}Tc labeling		
- MDP	-	50 kits/month
- DTPA	20 kits/month	50 kits/month
- MA-HAS	20 kits/month	50 kits/month
- PHOSPHON	20 kits/month	50 kit/month
- PHYTATE	30 kits/month	100 kits/month
- HIDA delivatives	20 kits/month	20 kits/month
- Others (HM-PAO, MIBI, MAG_3, ECD)	-	100 kits/month

Acknowlegements

The authors are grateful for colleagues in the FNCA activity for their provision of information cited herewith. They are Luo Zhifu, Liu Yishu, Abdul Mutalib, Hark Rho Kim, Han Hyon- Soo, Mohd Shaharuddin, Wan Anuar Wan Awang, Elvira Z. Sombrito, Sakda Charoen and Le Van So

3. Conclusions

None of the participating countries can afford sufficient number of ^{99m}Tc generators to fulfill their domestic demand. In such circumstances, the present project to promote the technology of ^{99m}Tc generator using poly zirconium compound (PZC) as an adsorbent for neutron acctivated (n, γ) ^{99}Mo is well justified. Since the PZC technology still contains problematic aspects to be solved or clarified in the view point of technology, a systematic investigation with carefully planned experiment for the specific object and more importantly the intensive evaluation and review for each step of laboratory work must be implemented.

Establishment of a network among the FNCA countries should be considered to secure the supply of radioisotopes and radiopharmaceuticals including ^{99}Mo related materials.

JAERI－Conf 2004－010

Workshop

Neutron Activation Analysis

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1.19 Study on Air pollution in Beijing using aerosol samples and INAA techniques

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Abstract

In this work, a heavy traffic area were choose as a sampling place, Gent SFU sampler with nuclepore filters of 0.4 and 8 μm pore sizes were used for sampling. INAA was employed as for multielements analysis. 42 to 46 elements were determined combined with both short and long irradiations. Enrichment factor and rotated factor analysis were employed for the primary data interpretation. Seven factors have been investigated.

Sampling:

The sampler was Located at Che Gong Zhuang, Beijing. Which representative of heavy traffic places. Generally, sampling were taken twice a week by Gent SFU sampler (collecting PM10 and PM 2.5 APM on nuclepore filters of 0.4 and 8 μm pore sizes, from BDH Laboratory Supplies, CORNING Separation Division).

After sampling, the samples were put into a electronic instrument for constant humidity. Then, Mettler Toledo GmbH Microbalance (readability 1 μg) for sample weighing. One each of 0.4 and 8 μm pore-size nuclepore blank filter for every 20 samples was analyzed for corrections for contributions from multielement concentrations in blank filters.

INAA of APM Samples:

Unloaded and loaded nuclepore filters were weighed using a microbalance after equilibrated in a balance room with relative humidity of 50±5%. Samples were each packed in clean capacitor paper for short irradiation. After counting and decay for one week (or longer), they were repacked in high purity Al foil for long irradiation.

Chemical standard solutions were made of high purity metals or compounds of elements to be determined. Standards were prepared by quantitatively dispensing the solution onto ashless filter paper, drying under infrared lamp, and packing in Al foil.

Weighed Fe wire and 25 μm Zr foil were used for comparator and neutron flux ratio monitors, respectively, in k_0-NAA.

NIST SRM 1632a (Coal Powder) and GBW 07312 (Stream Sediment) were concurrently analyzed for quality control.

All the irradiations were carried out at the 15 MW HWRR in CIAE. For short

irradiation, a home-made rabbit system was used with neutron flux at irradiation position of 1×10^{13} $n.cm^{-2}.s^{-1}$. For long irradiation, arbitrary heavy water reflect channels were used with neutron fluxes of 3 to 6×10^{13} $n.cm^{-2}.s^{-1}$. A typical irradiation-counting scheme is shown in Table 1.

All countings were performed at an automated HPGe spectrometer system (Canberra 35%, 1.8 keV). Software, SPAN was used for peak analysis. A hybrid k_0-relative software, ADVNAA developed by our lab was used for elemental concentration calculations. Internal consistency and quality control were achieved by comparisons between results from k_0 and relative NAA, and analytical results and certified/information values of the elements of interest in SRMs/CRMs . It is noteworthy that S, an element commonly not considered to be determinable by NAA in APM samples, has been determined in 30 to 35% fine APM samples using our 35% efficiency HPGe detector. By using a HPGe with much higher efficiency (say, 60% or higher), S could be very well determined in most fine APM samples.

Table 1. A typical irradiation-counting scheme for APM samples

No, Counting	Flux, $n/cm^2/s$	T_i	T_d	T_c, sec	Elements determined
1	1×10^{13}	300s	300s	300	Na, Mg, Al, Cl, K, Ca, Ti, V, Mn, Cu, Br, Sr, In, I, Ba, Dy
2			1hr	500	Na, K, Sr, In, Ba, Dy, Mn
3			10hr	1000	Na, K
4	3×10^{13}	16hr	5d	1500	Na, Ca, Ga, As, Br, Mo, Cd, Sb, Ba, La, Nd, Sm, Yb, Lu, W, Au, U
5			12-17d	4000	Sc, Cr, Fe, Co, Ni, Zn,Se, Rb, Sr, Zr, Ag, Cs, Ce, Nd, Eu, Tb, Lu, Hf, Ta, Hg, Th

Note: T_i , T_d , and T_c refer to irradiation time, decay time, and counting time, respectively.

Data interpretation:

Multielemental concentrations in APM samples were statistically handled in the following approaches:

Enrichment Factors (EF) with Al as reference element, relative to earth crust average (from Mason); Time series of EF for individual elements; Factor Analysis; etc. Preliminary Findings from Statistical Treatment of Multielement Data are shown below:

a) Enrichment factors for 45 elements were calculated. The same trends as in previous report were found, i.e. Pollution indicating elements, such as As, Br, Hg, I, In, Sb, Se, etc., have prominently high EF values and EFs for fine particles are usually larger than in coarse ones. Elements mainly originated from earth dust, such as Na, Sc, REEs, have smaller EF values.

b) Factor analysis based on 90 fine APM samples (98-12 to 00-05) for 36 elements have been made. The rotated factor loading matrix is shown in Table 2. Seven factors are identified. The total communalities for 31 of the 36 elements are greater than 70%, meaning they are largely accounted

for. From the major elements in each factor, the following is implied:

In factor 1, elements with highest scores include Al, Ba, Ca, Ce, Eu, Fe, Ga, Hf, La, Mg, Sc, Sm, Th, Ti, and V, clearly indicating the soil and fly ash origin.

In factor 2, elements with highest scores include As, Cs, I, K, Rb, Sb, Se, and Zn, and at a less extent, Ca, indicating a mixture of refuse incineration and limestone, originating from large and small refuse incineration sites and numerous construction working sites in Beijing.

In factor 3, elements with highest scores include As, Br, Co, Ga, Sb, Se, U, V, and W, indicating, among other origins, the motor vehicle exhaust and coal burning origins.

In factor 4, elements with highest scores include Au, Cl, and Na, indicating a sea spray origin. The origin of Au is to be investigated.

In factor 5, elements with highest scores include Cd, I, and In, probably having a refuse incineration (e.g. used battery, etc.) and/or paint pigment origin.

In factor 6 and 7, elements with highest scores are Ag, Mn and Cu, I, respectively. They may have an origin of non-ferrous metal research institutes or factories.

During winter and spring seasons (Dec. to Apr.) factor 1 (soil/fly ash) has relatively higher scores than the rest seasons of a year. It can be very well explained by the coal burning and strong sandstorms during these seasons.

Conclusion:

INAA is one of the most important techniques for air pollution studies. Multielements fingerprint is a valuable method for pollutants origin. According to our investigation, seven factors have been found. That means the environment of Beijing city need to be improved further.

Table 2 Rotated Factor Loadings for 36 elements in Fine APM

Ele.	Factor-1	Factor-2	Factor-3	Factor-4	Factor-5	Factor-6	Factor-7	Commu-nality
Ag	-0.18	0.09	0.12	0.11	-0.03	**0.87**	0.04	0.83
Al	**0.89**	0.02	0.06	0.02	0.06	0.01	-0.15	0.83
As	0.05	**0.52**	**0.73**	0.16	0.13	-0.04	0.24	0.91
Au	0.02	-0.15	0.12	**0.60**	-0.02	0.32	0.22	0.55
Ba	**0.85**	0.09	0.24	0.19	0.08	-0.12	0.04	0.84
Br	0.24	0.17	**0.64**	0.36	0.33	0.13	-0.05	0.75
Ca	**0.78**	0.24	-0.02	0.03	-0.05	0.00	0.15	0.69
Cd	-0.03	0.02	-0.03	-0.04	**0.79**	0.08	-0.19	0.67
Ce	**0.83**	-0.04	-0.08	-0.05	0.01	0.03	0.37	0.83
Cl	0.09	0.07	0.11	**0.89**	-0.02	-0.04	-0.02	0.81
Co	0.34	0.17	**0.81**	0.29	-0.02	-0.08	0.05	0.90
Cs	0.18	**0.88**	0.26	-0.08	-0.04	0.10	0.02	0.90
Cu	0.25	0.11	0.30	0.38	-0.10	0.15	**0.64**	0.75
Eu	**0.78**	-0.01	0.13	0.22	-0.17	0.00	0.16	0.73
Fe	**0.92**	0.30	0.02	0.02	-0.02	0.07	0.07	0.95
Ga	**0.54**	0.27	**0.69**	0.21	-0.03	0.02	0.06	0.89
Hf	**0.88**	-0.01	0.14	0.10	-0.06	-0.02	-0.11	0.82
I	-0.11	**0.52**	-0.03	-0.06	**0.43**	0.09	**0.51**	0.74
In	-0.06	0.16	0.10	0.01	0.70	-0.09	0.26	0.60
K	0.29	**0.82**	-0.04	0.09	-0.02	0.01	-0.01	0.78
La	**0.82**	0.10	0.02	0.01	0.03	0.01	0.43	0.87
Mg	**0.89**	0.30	0.03	0.08	-0.07	-0.06	0.08	0.90
Mn	0.45	0.46	0.10	0.01	0.20	**0.65**	0.09	0.90
Na	0.38	0.32	0.43	**0.67**	-0.05	-0.07	0.08	0.90
Rb	0.43	**0.83**	0.17	-0.02	0.09	0.04	-0.12	0.92
S	-0.35	0.30	-0.14	-0.37	-0.34	-0.01	0.14	0.51
Sb	0.02	**0.63**	**0.61**	0.10	0.05	0.05	0.27	0.85
Sc	**0.96**	0.12	0.22	0.09	0.00	-0.01	-0.07	0.99
Se	-0.13	**0.66**	**0.63**	0.05	0.02	-0.02	0.06	0.85
Sm	**0.93**	0.03	0.25	0.11	0.06	0.00	-0.16	0.97
Th	**0.96**	0.07	0.21	0.08	0.03	0.00	-0.04	0.97
Ti	**0.94**	0.02	0.00	0.02	0.04	0.04	-0.08	0.90
U	0.30	0.00	**0.85**	0.15	0.00	0.10	-0.09	0.86
V	**0.83**	0.23	**0.46**	0.12	0.01	-0.02	-0.05	0.97
W	-0.07	0.06	**0.53**	-0.25	-0.07	0.28	0.04	0.44
Zn	0.05	0.78	0.36	0.07	0.27	0.15	0.14	0.86

Note: Data bolded and underlined indicate elements with highest scores in the factor.

1.20 Collection of Size Fractionated Particulate Matter Sample for Neutron Activation Analysis in Japan

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Abstract

According to the decision of the 2001 Workshop on Utilization of Research Reactor (Neutron Activation Analysis (NAA) Section), size fractionated particulate matter collection for NAA was started from 2002 at two sites in Japan. The two monitoring sites, "Tokyo" and "Sakata", were classified into "urban" and "rural". In each site, two size fractions, namely "$PM_{2\text{-}10}$" and "PM_2" particles (aerodynamic particle size between 2 to 10 micrometer and less than 2 micrometer, respectively) were collected every month on polycarbonate membrane filters. Average concentrations of PM_{10} (sum of $PM_{2\text{-}10}$ and PM_2 samples) during the common sampling period of August to November 2002 in each site were 0.031mg/m^3 in Tokyo, and 0.022mg/m^3 in Sakata.

Keywords: Atmospheric particulate matter, PM_{10}, Fine particles, Neutron activation analysis

Introduction

Participants of the NAA section of the 2001 Workshop on Utilization of Research Reactor, Beijing, 5-9 November 2001 (China, Indonesia, Japan, Malaysia, Philippines, Thailand, and Vietnam) agreed air pollution study as the subject to be focused on for the application of NAA in 2002. Presently, all the participating countries have capability to collect size characterized particulate matter samples, e.g. PM_2 and $PM_{2\text{-}10}$. Collection, analysis, and data interpretation of the elemental profile in the atmospheric particulate matter was adopted as a part of the activities of the project program. In order to obtain the comparable data among the participating countries, sorts of sampling device, filtering material, sampling frequency, suitable site character, and so on were discussed and specified at the Workshop.

In Japan, collection of particulate matter was carried out at two monitoring sites using same sampling system. The sampling system consists of consecutive NILU

filter holder including PM_{10} inlet, coarse filter (pore size 8 micrometer), and fine filter (pore size 0.4 micrometer) attached to the NLK-60 low volume air sampler assisted by mass flow meter. This paper presents the collection and mass concentration of particulate matter obtained in the project.

Monitoring method

- Monitoring sites

One "Urban" monitoring site is located at western side city Hachioji, of Tokyo metropolitan area. The population of Tokyo is more than twelve million, and air pollutants generated from this area is very huge. In these years, environmental conservation authority of the Tokyo metropolis focuses on nitrogen oxides and particulate matters mainly emitted from automobile exhaust, because the environmental air quality standards for these pollutants in Tokyo have not been satisfied in wide areas.

The other "Rural" monitoring site is located northeastern part of Japan, about 500km apart from Tokyo. The site is in an estuary harbor city, Sakata of Yamagata Prefecture, beside the Japan Sea/the East Sea. Population of the city is about 100 thousands.

Table1 Properties of monitoring sites

	Tokyo	Sakata
Classification of site	Urban	Rural
Postal address	Tokyo Metropolitan Univ., 1-1, Minami-Osawa, Hchioji-city, Tokyo	Tohoku Univ. of Comm.Serv. Sci. 3-5-1, Sakata-city, Yamagata
Latitude	35° 07'N	38° 55'N
Longitude	139° 23'E	139° 52'E
Altitude	126m	30m
Main Pollution Source	Highway, Building construction	Sea salts, Coal power plant

- Sampling system

Mass-flow measurement low-volume air sampler, NLK-60 (Tokyo-Dylec Corp.) combined with the NILU filter holder is used at each site. As shown in Fig.1, the filter holder consists of a PM10 aluminum impactor followed by coarse filter (pore size 8 micrometer, Nuclepore membrane filter made of poly-carbonate) and fine filter (pore size 0.4 micro meter, Nuclepore membrane filter made of poly-carbonate). Operating the system as specified flow rate of 16.7 liters per minute, the first filter collects $PM_{2\text{-}10}$ (aerodynamic particle size between 2 to 10 micrometer), and the second filter collects PM_2 (aerodynamic particle size less than 2 micrometer). This sampling system can collect particles equivalent to the Gent Stacked filter Unit PM_{10} sampler[1].

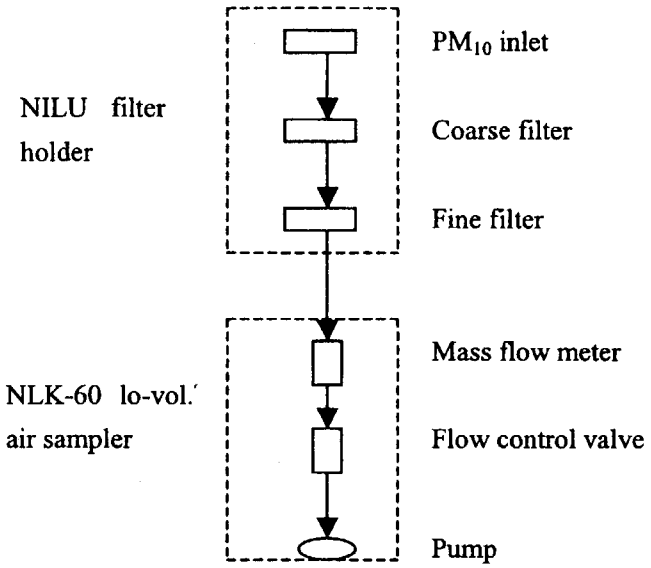

Fig.1 Sampling System

Results and discussion

Individual duration of particulate matter collection is less than a week depending on the flow rate drop caused by loaded fine filter. Particulate matter collection was carried out at each site once or twice per month. Average particulate mass concentrations during the common sampling period – from August to November 2002, were 0.031 mg/m^3 in Tokyo, and 0.022 mg/m^3 in Sakata. Higher particulate mass concentrations observed in Tokyo seem to reflect the various activities of surrounding area. Concentration ratios of the $PM_{2\text{-}10}$ particles to the PM_2 particles during the period at both sites were kept within around 0.5 to 2.0. However, during spring season in Sakata, extremely high concentration of coarse particles raised the $PM_{2\text{-}10}$ to PM_2 particle ratio up to more than 10. These phenomena were observed from March to April 2002, because of the yellow sand storms came from Asian Continent.

Particulate mass concentrations observed in this work were compared with the Japanese air quality monitoring device. In Japan, the air environmental quality standard for suspended particulate matter is specified particles which aerodynamic diameter less than 10 micrometer (equivalent to PM_7 or PM_8). As shown in Fig.2, PM_{10} sample ($PM_{2\text{-}10}$ + PM_2) of this work and parallel operated Japanese low-volume air sampler using quartz fiber filter in Sakata represents almost good relations. In average, approximately fifteen percents higher results were observed by this study. According to the environmental statistics of Japan, average concentration of Japanese suspended particulate matter was 0.045mg/m^3 (47 monitoring sites) in Tokyo[2], and 0.022mg/m^3 (12 monitoring sites) in Yamagata[3].

Table2 Mass concentration of particulate matter in Japan by this work

(unit: mg/m^3)

Site / Month	Tokyo			Sakata		
	PM_{2-10}	PM_2	PM_{10} [b)]	PM_{2-10}	PM_2	PM_{10} [b)]
Feb.				0.020	0.010	0.030
Mar.				0.037	0.007	0.043
Apr				0.151	0.013	0.164
May.				0.005	0.008	0.013
Jun.				0.007	0.007	0.014
Jul.				0.006	0.014	0.020
Aug.	0.017	0.008	0.025	0.011	0.017	0.029
Sep.	0.010	0.010	0.021	0.009	0.007	0.015
Oct.	0.018	0.020	0.038	0.010	0.012	0.022
Nov.	0.022	0.018	0.041	0.015	0.008	0.024
Average[a)]	0.017	0.014	0.031	0.011	0.011	0.022

[a)] Average concentration of August to November 2002.

[b)] Sum of PM_{2-10} and PM_2 samples.

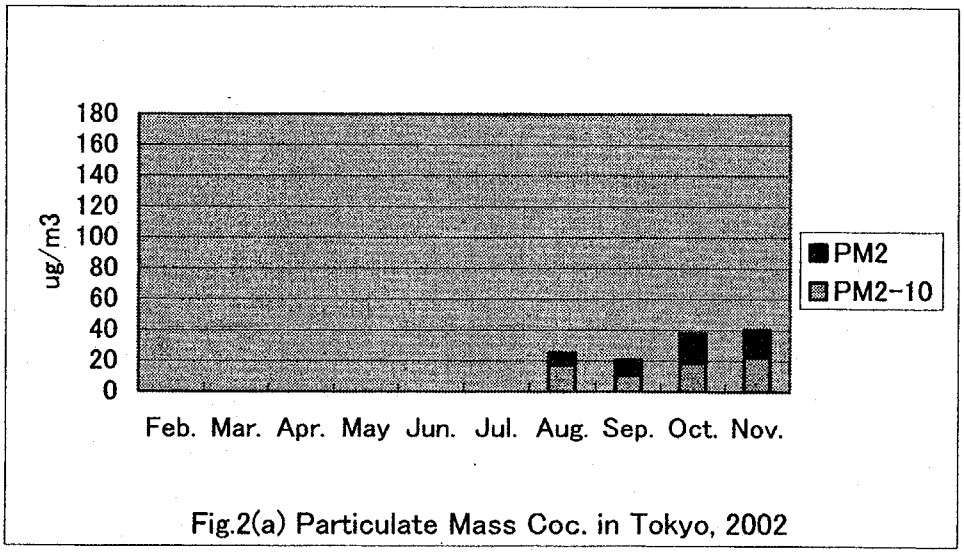

Fig.2(a) Particulate Mass Coc. in Tokyo, 2002

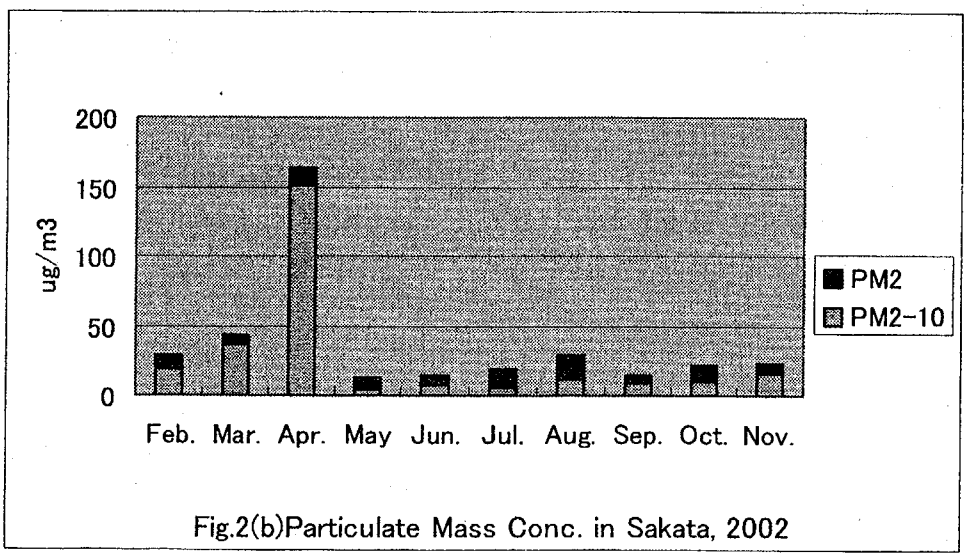

Fig.2(b)Particulate Mass Conc. in Sakata, 2002

Conclusion

Collection of size fractionated particulate matter sample was carried out in two sites Japan for NAA. Mass concentration of the particulate matter obtained by this study was compared with concentration of the Japanese suspended particulate matter that is specified as the environmental air quality standard in Japan. Collected samples seem to reflect well the situation around the monitoring site, and were provided for NAA.

References

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1.21 INAA of atmospheric particulate collected at Hachioji and Sakata, Japan

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Elemental concentration of atmospheric particulates, $PM_{2\text{-}10}$ and PM_2, collected Hachioji and Sakata, Japan in 2002 were determined by INAA using the k_0 standardization method. The concentrations of 33 elements were able to determined. In Sakata, almost elements which are index for arificial particulates were concentarted in PM_2 fraction. But this feature was not valid simply for particulates collected in Hachioji. It is suspected that more kinds of source supply these elements in Hachioiji than in Sakata.

Introduction

As part of the project on atmospheric particulate matters by NAA group of FNCA, Japanese group started to collect air particulate matter at Sakata on February and at Hachioji on August, 2002. Ohtoshi et al. reported about the sampling system and mass concentration of particulate matter in Japan in a separate article in this volume. So we will present about the method for INAA and results on elemental concentrations in this report.

Experimental

Atmospheric particulate were collected using NILU filter holder included PM_{10} inlet and two Nuclepore polycarbonate filters of different pore size at Hachioji and Sakata, Japan. Hachioji is urban area and about 40 km west of center of Tokyo. Sakata is rural area and faces to Japan Sea. The details of information of sampling in Japan were described by Ohtoshi et al.

Polycarbonate filters collected air particulate were cut in half, and the one piece was subjected to analysis by INAA. The other was kept to analysis by another technique in future. The filters to be analyzed by INAA were pressed into pellets with a diameter of 12 mm and put into double polyethylene bags. At first, the filters were irradiated for 5 min at JRR-3 PN3 or JRR-4 PN in JAERI, one by one. After irradiation, only the filter was transferred to a new polyethylene bag to avoid a contribution of an impurity in polyethelene bag, then γ-rays were measured for 5 min. After appropriate time for cooling, each filter was wrapped with double Capton® polyimide films (0.0125 mm thick) and an Al foil one by one. The stacked filters were put into a quartzes tube with 3 sets of Au and Zr monitors followed by irradiation for 6h at JRR-4 T pipe in JAERI. After

irradiation only the filter was transferred to a new polyethylene bug to avoid a contribution of an impurity in Capton film then γ-rays were measured 3 or 4 times with appropriate intervals. Elemental concentrations were determined by k_0 standardization method.

Results and discussion

Fourteen elements, Mg, Al, S, Cl, Ca, Ti, V, Mn, Cu, Br, In, I, and Ba, were determined for 5 min irradiation and 19 elements, Na, K, Sc, Cr, Fe, Co, Zn, Se, Rb, Mo, Ag, Sb, La, Ce, Sm, Hf, W, and Au, for 6 hours irradiation. Impurity in three coarse and fine filters each were analyzed. Polycarbonate filters were found to contain Na, Al, V, Mn, Cl, K, Cr, Br, and Ce and their contents were not consistent with each other. Especially the contribution of Br, Cr, and Ce in filters were severe.

Elemental concentrations of $PM_{2\text{-}10}$ and PM_2 collected at Hachioji and Sakata, Japan in 2002 were tabulated in Tables 1 and 2, repectively. For example, the elemental concentrations on October were shown in Fig.1. Elements were ordered by decreasing of concentration of PM_{10} (= $PM_{2\text{-}10}$ + PM_2) at Hachioji. The large difference between elemental concentration values between at Hachioji and Sakata was not observed totally. But, as shown in Fig.6 for example, concentrations of several elements such as Cr, Fe, Ag, Sb, Ba, Hf, and Au in Hachioji were higher than in Sakata and Na, Cl, and W in Sakata were higher. Since a part of Na and Cl are originated from sea salt particle and Sakata city faces the Sea of Japan, it is natural that concentrations of Na and Cl in Sakata were higher. The cause of high concentration of Cr to Au in Hachioji, however, is not clear at present.

Elements determined in atmospheric particulates were classified into 3 group based on enricment fraction; enrichment in $PM_{2\text{-}10}$ fraction, enrichment in PM_{10} fraction, and equal in both fractions. Classified elements were shown in Table 2 and time variations of concentration of typical elements in each class were shown in Fig.2. It is aware generally that particulates with natural origin are enriched in coarse fraction and those with artificial origin are enriched in fine fraction. In Sakata, almost observed elements distributed under such a rule. Concentrations of elements which are related with silicate rocks, for example, Al, Ca, and Sc in $PM_{2\text{-}10}$ fraction were higher than in PM_2 fraction generally at both Hachioji and Sakata. V, which is one of index elements for anthropogenic pollutant, was concentrated in PM_2 fraction in Sakata. But in Hachioji, V was not enriched in PM_2 fraction but distributed almost equally in $PM_{2\text{-}10}$ and PM_2 fractions. Like this, distributions of elements expressed by bold in Table 2 in Hachioji and Sakata were different. It is suspected that more kinds of source supply these elements in Hachioiji than in Sakata.

Table 1. Elemental concentration in ng/m^3 of atmospheric particulates collected at Hachioji in 2002.

	Aug.		Sep.		Oct.		Nov.		Dec.	
	PM2-10	PM2	PM2-10	PM2	PM2-10	PM2	PM2-10	PM2	PM2-10	PM2
Na	432	58	258	51	543	30	555	134	68	21
Mg	143	17	62	19	158	7	438	73	90	
Al	457	35	160	17	418	19	1023	145	246	29
S	1251	965		1131		286				
Cl	35	14	111	20	264	11	902	510	172	192
K	177	116	95	77	155	36	264	210	68	35
Ca	377	22	236	16	304		866	80	288	
Sc	0.10	0.0064	0.036	0.0031	0.087	0.0041	0.16	0.022	0.040	0.005
Ti	45		15		42		70		23	
V	3.2	2.6	1.0	1.3	1.8	0.5	2.4	2.4	0.79	0.47
Cr	3.1	0.29	1.9	1.55	3.6	0.47	2.1	1.2	0.83	0.50
Mn	12	3.8	6	4.4	14	1.4	19	10.0	7.2	3.9
Fe	482	73	244	64	520	33	592	140	187	43
Co	0.28	0.041	0.10	0.033	0.17	0.013	0.31	0.081	0.11	0.021
Cu	15	3.9	7	3.7		1.3		6.7	4.1	2.5
Zn	54	38	27	37	38	13	43	68	25	22
As	0.50	0.61	0.31	0.69	0.64	0.22	0.67	1.05	0.13	0.15
Se	0.66	0.70	0.25	0.52	0.47	0.20	0.44	1.33	0.11	0.20
Br	3.5	4.1	1.2	3.4	2.6	1.4	2.3	6.5	0.78	1.2
Rb			0.23	0.23	0.65	0.11	1.2	0.76	0.21	0.12
Mo	0.81	0.42	0.32	0.19	0.68	0.15	0.60	0.75	0.33	0.28
Ag	0.16	0.15	0.16	0.36	0.31	0.14	0.32	0.96	0.31	0.82
In				0.012		0.0063	0.016	0.026	0.006	0.008
Sb	3.1	2.0	1.5	1.8	2.2	0.63	1.9	2.8	0.88	0.71
I	1.1	1.0		1.7		0.69	0.89	3.4	0.52	1.0
Cs	0.051	0.043	0.024	0.024	0.059	0.011	0.096	0.081	0.018	0.011
Ba	18	5.1	8	2.4	12	1.0	14	4.5	5.9	1.5
La	0.20	0.069	0.10	0.060	0.22	0.032	0.45	0.170	0.11	0.039
Ce	0.39	0.091	0.06	0.000	0.21	0.0013	0.74	0.179	0.13	0.0057
Sm		0.0090	0.013	0.0094			0.085		0.017	
Hf	0.050		0.029	0.0043	0.045	0.0035	0.060	0.011	0.023	0.0049
Ta	0.016		0.007		0.010		0.023		0.008	
W	0.27	0.14	0.10	0.14	0.13	0.037	0.25	0.17	0.082	0.065
Au	0.0013	0.0014	0.0056	0.0009	0.0028	0.0006	0.0032	0.0021	0.0012	0.00070
Th	0.040		0.018		0.052	0.0040	0.122	0.023	0.021	0.0033

Table 2a. Elemental concentration in ng/m^3 of atmospheric particulates collected at Sakata in 2002.

	Feb.		Mar.		Apr.		May		Jun.		Jul.	
	PM2-10	PM2	PM2-10	PM2	PM2-10	PM2	PM2-10	PM2	PM2-10	PM2	PM2-10	PM2
Na	1456	361	1592	511	2402	410	100	55	570	122	624	142
Mg	344	81	404	88	821	110	39	6.0	88	20	100	23
Al	576	198	633	130	2526	266	61	15	97	31	40	23
S		878		900		1504		455				659
Cl	2337	28	2469	153	2255	119	94	6.8	717	21	771	8
K	227	133	260	134	921	223	15	20		60	81	64
Ca	320	78	337	60	1259	111			60	21	249	18
Sc	0.10	0.033	0.10	0.022	0.44	0.047	0.0050	0.0027	0.016	0.005	0.007	0.004
Ti	34	12	40	10	134	18	3.8	1.7				
V	0.94	0.74	1.1	0.66	5.8	1.9	0.22	0.89	0.29	1.60	0.14	0.83
Cr	1.0		1.1		3.5		0.43	0.055	0.60		0.83	0.31
Mn	7.4	4.4	9.1	3.8	37	6.9	1.2	0.73	1.9	1.7	0.89	1.9
Fe	335	116	320	77	1399	171	20	13	61	25	40	25
Co	0.13	0.061	0.15	0.047	0.57	0.081	0.0083	0.0079	0.027	0.016	0.014	0.020
Cu								1.2		2.11		
Zn	5.5	13	13	11	44	25	4.1	10	7.3	26	4.6	18
As	0.40	0.79	0.71	0.78	2.7	1.6	0.030	0.15	0.15	0.43	0.15	0.60
Se	0.11	0.36	0.20	0.32	0.94	0.54	0.018	0.089	0.057	0.25	0.10	0.33
Br	4.7	4.2	3.8	6.2	5.7	5.1	0.19	1.2	0.44	1.7	1.4	1.4
Rb	0.93	0.57	1.2	0.54	4.6	1.40	0.045		0.15	0.32		0.25
Mo	0.28	0.91	0.39	0.28	2.0	0.58	0.037	0.086		0.09		0.29
Ag		0.039				0.054		0.013				0.022
In		0.014		0.014		0.030						
Sb	0.12	0.38	0.27	0.156	1.5	0.86	0.064	0.33	0.16	0.52	0.16	0.38
I		1.9		1.4		2.5	0.24	0.68		0.7		0.67
Cs	0.071	0.051	0.091	0.060	0.380	0.096	0.0023		0.011	0.019	0.0094	0.033
Ba	5.0		7.2		24		0.60		2.0		1.6	1.1
La	0.28	0.10	0.30	0.079	1.1	0.15	0.013	0.023	0.041	0.02	0.02	0.02
Ce	0.50	0.063	0.61	0.048	2.5	0.0011	0.070	0.054	0.059	0.203		
Sm		0.018	0.055	0.012		0.024	0.0021	0.0017	0.0069	0.003	0.0037	0.003
Eu	0.0077	0.0024	0.0088	0.0024	0.039							
Hf		0.011	0.033	0.0074	0.13	0.015	0.0025		0.0067		0.0048	0.003
Ta			0.0081		0.035							
W	0.21	1.4	0.70	1.3	0.46	0.86	0.042	1.0		0.25		0.6
Au		0.00033	0.00047		0.0024	0.0015						0.00025
Th	0.10	0.036	0.12	0.028	0.47	0.056	0.0042		0.013	0.005	0.008	0.0063

Table 2b. Elemental concentration in ng/m^3 of atmospheric particulates collected at Sakata in 2002.

	Aug.		Sep.		Oct.		Nov.		Dec.	
	PM2-10	PM2	PM2-10	PM2	PM2-10	PM2	PM2-10	PM2	PM2-10	PM2
Na	1653	249	340	111	1572	214	1709	286	623	97
Mg	241	31	69	19	292	54	755	199	100	20
Al	157	33	148	44	394	130	1245	401	66	18
S				751		766				
Cl	1150	17	68	7	1949	27	3240	38	1263	23
K	125	104	93	78	267	213	387	240	35	21
Ca	139	33	109	27	398	62	651	149		
Sc	0.027	0.0060	0.025	0.0081	0.072	0.024	0.18	0.060	0.010	0.0027
Ti			9		24	7.3		22		
V	0.85	2.28	0.58	2.2	0.76	1.21	2.0	1.1	0.16	0.56
Cr	0.5	0.7	0.55	0.70	1.6	0.47	1.4	0.0		
Mn	3.3	2.3	3.2	2.3	8.1	5.5	17	9.8	1.5	1.2
Fe	114	39	93	40	245	101	559	198	54	13
Co	0.053	0.035	0.041	0.021	0.11	0.049	0.23	0.085	0.017	0.0093
Cu		2.3								
Zn	15	26	10	23	14	33	9.6	25	5.0	5.4
As	0.49	0.72	0.31	0.97	1.8	3.1	0.70	1.26	0.081	0.21
Se	0.33	0.70	0.13	0.32	0.27	0.80	0.16	0.54	0.023	0.08
Br	2.5	4.2	0.79	2.3	4.1	5.7	4.7	3.6	1.1	1.8
Rb	0.48	0.43	0.25	0.31	0.84	1.1	1.7	1.2		
Mo		0.36	0.20	0.44			0.48	0.72		0.17
Ag		0.071		0.047		0.081	0.082	0.145		
In		0.024				0.020		0.021		
Sb	0.33	0.74	0.41	0.88	0.38	0.90	0.18	0.54	0.094	0.14
I		1.7		1.2		2.1		0.8		0.55
Cs	0.033	0.041	0.016	0.023	0.056	0.099	0.130	0.118	0.0068	0.0049
Ba	3.1		2.4				8.7	3.3	1.2	0.61
La	0.076	0.033	0.071	0.030	0.20	0.10		0.15	0.024	0.009
Ce	0.14		0.10	0.040	0.43	0.194	1.02	0.257		
Sm	0.010		0.011	0.005	0.035	0.013	0.090	0.042	0.0040	
Eu	0.0027		0.0028		0.0060		0.0152	0.0051	0.0010	
Hf	0.012	0.006	0.0094	0.005	0.026	0.011	0.043	0.017	0.0042	
Ta							0.012			
W	0.31	3.1	0.17	0.4	0.57	0.9	0.57	7.6		0.054
Au	0.00041		0.00031	0.00039				0.00043		
Th	0.021		0.020	0.0079	0.076	0.026	0.18		0.0074	0.0032

Table 3. Elemental distribution.

	Hachioji	Sakata
Enrich in PM2-10	Al, **Au**, Ba, Ca, Ce, Cl, Co, Cr, **Cs**,Cu, Fe, Hf, La, Mg, **Mn**, **Mo**, Na, **Rb**,Sc, Th, Ti	Al, **As**, Ba, Ca, Ce, Cl, Co, Cr, Fe, Hf, La,Mg, Na, Sc, Sm, Th, Ti
Enrich in PM2	Ag, Br, I, In, S, Se	Ag, **Au**, Br, **Cs**, I, In, **Mo**, **Rb**, S, **Sb**, Se, **V**, **W**, **Zn**
equal	**As**, K, **Sb**, **V**, **W**, **Zn**	K, **Mn**

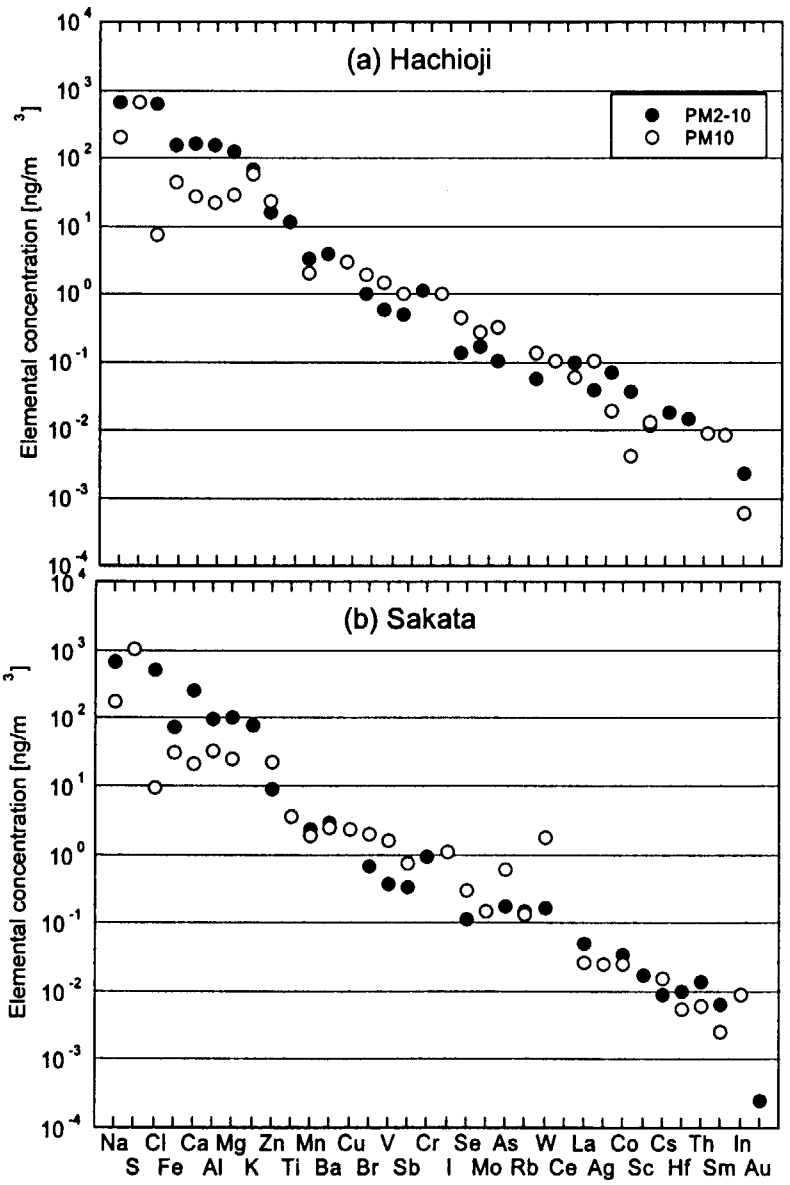

Fig.1. Elemental concentrations on October, 2002.

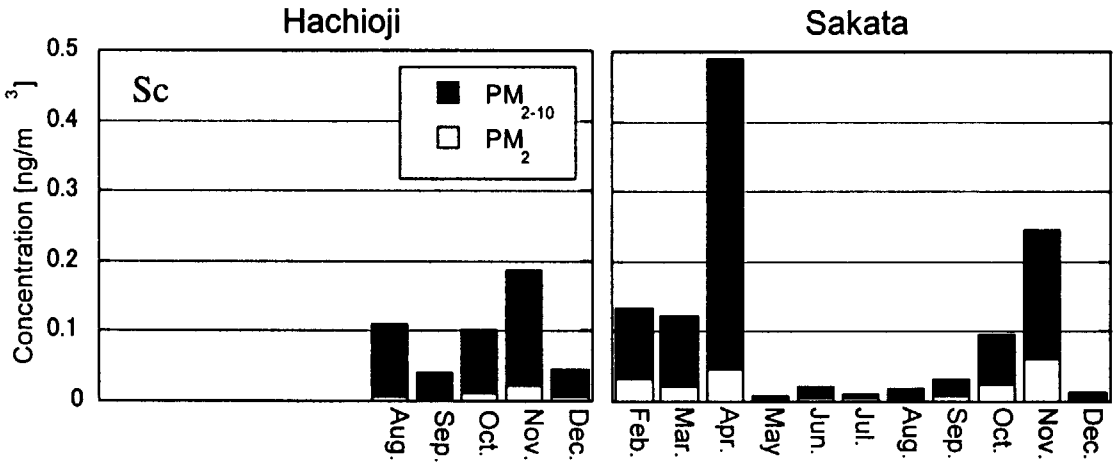

Fig.2. Scandium concentrations in 2002. Example for elements which is enriched in $PM_{2\text{-}10}$ fraction.

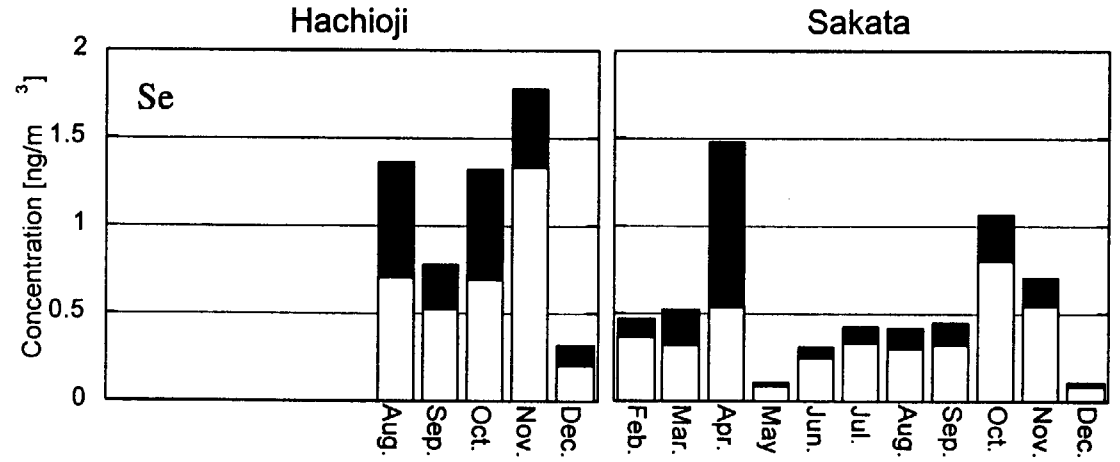

Fig3. Selenium concentrations in 2002. Example for elements which is enriched in PM_{10} fraction.

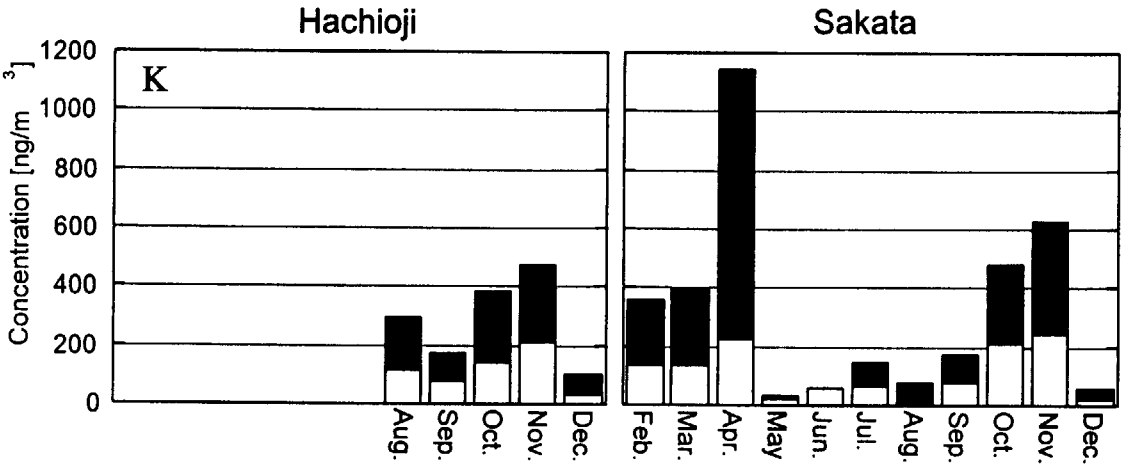

Fig.4. Potassium concentrations in 2002. Example for elements which is distributed equally in $PM_{2\text{-}10}$ and PM_{10} fractions.

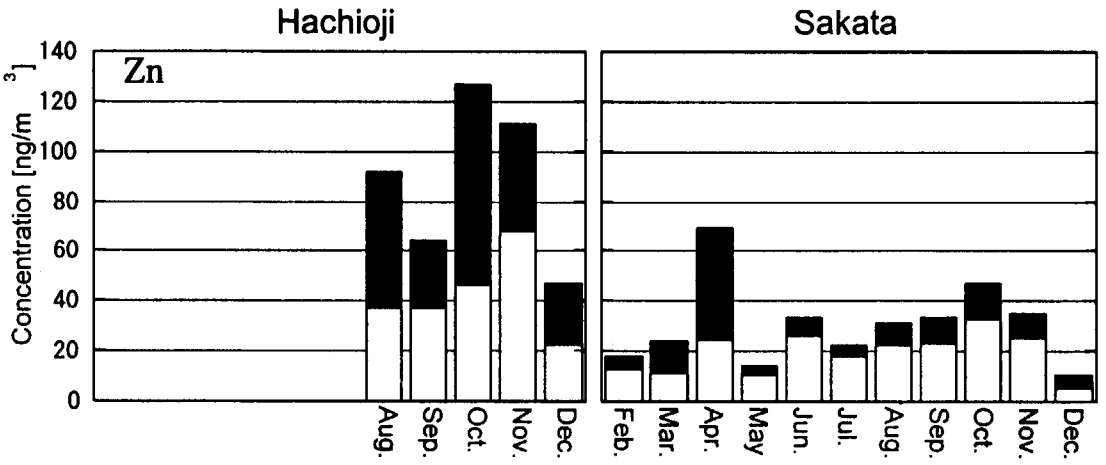

Fig.5. Zinc concentrations in 2002. Example for elements whose distribution between $PM_{2\text{-}10}$ and PM_2 in Hachioji and Sakata are different.

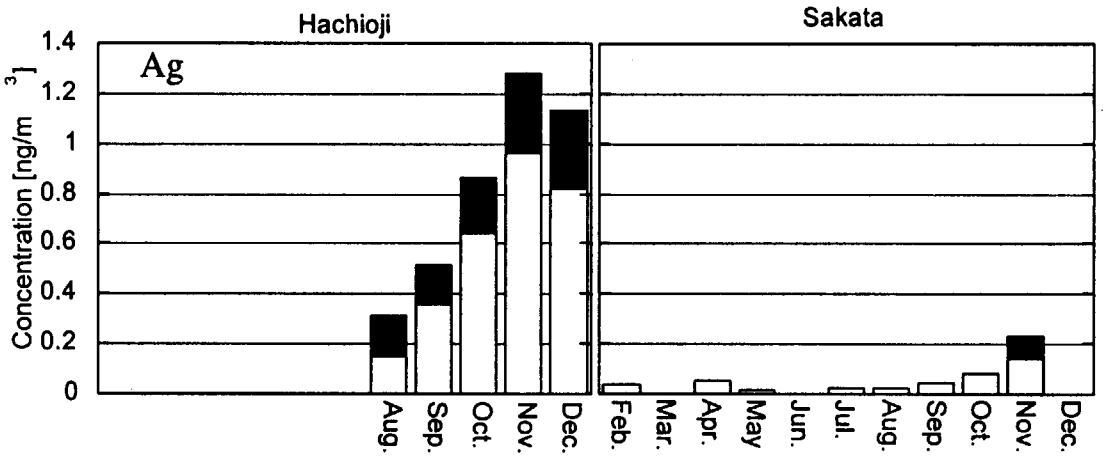

Fig.6. Silver concentrations in 2002. Example for elements whose concentrations in Hachioji and Sakata are extremely different.

JP0450637

JAERI－Conf 2004－010

1.22 ELEMENTAL QUANTIFICATION OF AIRBORNE PARTICULATE MATTER IN BANDUNG AND LEMBANG AREA*

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ABSTRACT

ELEMENTAL QUANTIFICATION OF AIRBORNE PARTICULATE MATTER IN BANDUNG AND LEMBANG REGION: The contaminated airborne particulates by toxic gases and elements have a potential affect to the human health. Some toxic elements related to air pollution have carcinogenic affect. The quantification of those elements is important to monitor a level of pollutant contained in the airborne particulate. The aim of this work is to analyze the air particulate sample using instrumental neutron activation analysis and other related technique. Two sampling points of Bandung and Lembang that represent an urban and rural area respectively have been chosen to collect the air particulate sample. The samplings were carried out using Gent Stacked Filter Unit Sampler for 24 hours, and two cellulose filters of 8 μm and 0.45 μm pore size were used. Trace elements in the sample collected were determined using NAA based on a comparative method. Elemental distribution on $PM_{2.5}$ and PM_{10} fraction of airborne particulate was analyzed, the enrichment factor was calculated using Al as reference elements, and the black carbons contents were determined using EEL Smoke Stain Reflectometer analyzed. The results are presented and discussed.

Keywords: *$PM_{2.5}$, PM_{10}, Black Carbon, Enrichment Factor, Pollutant, Airborne Particulate Matter.*

INTRODUCTION

Air pollution monitoring, especially in big cities and industrial areas, is important for several reasons. More and more aerosols and toxic gases are released to the atmosphere due to the rapid development of industrialization and urbanization,

**Presented at The 2002 FNCA Workshop on The Utilization Of Research Reactors, Jakarta, Indonesia, January 13-17, 2003.*

which lead to the increase of vehicles used for transportation. The release of aerosols may affect the human health, because it contains toxic elements and heavy metals, which contaminate the air, rainwater, and soils [1]. In order to know the source of pollutant, valid data on the elemental composition of aerosols is needed. This data is reflected by the elemental composition in airborne particulate samples taken from different area.

Instrumental neutron activation analysis (INAA) is one of the most widely used for elemental quantification in airborne particulate sample for its high sensitivity and versatility, in addition to the high accuracy and precision [2,3,4]. This analysis technique is very suitable for our airborne particulate sample, since our sample, which has been obtained by using Gent Stacked Filter unit sampler, is around 1 mg/filter for fine or coarse airborne particulate. INAA as non-destructive analysis technique can compete with, Proton Induced X-ray Emission (PIXE) and Energy Dispersive X-ray Fluorescence (EDXRF). Therefore, INAA is our choice since the analysis technique is already available.

Samples taken from Centre For Research and Development of Nuclear Technique in Bandung (urban area) and Meteorological and Geophysical Agency in Lembang (rural area). Bandung is province city of West Java categorized as industrial city with high populated of people and cars, whereas Lembang is suburb city 20 km away from sampling site in Bandung. Information on pollution levels in this area and trends over time provides a valid and scientifically based support to relevant regulatory body in making decisions to minimize the increase health risk of persons in these populated areas.

METHODOLOGIES

Sampling

Samplings of airborne particulate were carried out for 24 hours using Gent Stacked Filter Unit Sampler. The stacked Filter Unit sampler consists of fine and coarse filter that have 47 mm diameter, 0.45 μm, and 8 μm pore size. Airborne particulate matter will be deposited on 8 μm and 0.45 μm pore size filter for coarse and fine particulate respectively.

Sampling was carried out only in Centre for Research and Development of Nuclear Technique in Bandung (urban area) and Meteorological and Geophysical Agency in Lembang for rural area. In Centre For Research and Development of Nuclear Technique sampling of airborne particulate was carried out by placing the Gent sampler on the roof of the second floor of the building A about 8 m above ground level and 60 m from the nearby street of this research centre. In Meteorological and Geophysical Agency sampling of airborne particulate was carried out by placing Gent sampler on the roof of the Agency building about 6 m above ground level and about 1 km from nearby street. Exposed and unexposed filters were equilibrated in the chamber of 45-55 % of humidity and weighed in semi microbalance before using. Airborne particulate samples were placed in clean polyethylene bag.

Analysis

The elemental concentrations in airborne particulate are determined by neutron activation analysis. Samples irradiations were carried out in Serpong for short-lived radionuclides. Standards for short-lived radionuclide are prepared by pipetting suitable mixed of SPEC PURE standard solution onto Whatman filter paper no. 41 and dried using infrared. The elemental content are determined by irradiating the samples and standards for one minutes and counted directly using γ-ray spectrometer coupled with Accuspec. Gamma rays spectra from Accuspec were analyzed using Aptec OSQ/Professional.

Enrichment factor was calculated using Mason's crustal abundances [6], whereas Black Carbon (BC) was determined using EEL Smoke Stain Reflectometer as recommended by W. Maenhaut and D. Cohen [5].

RESULTS AND DISCUSSION

Mass concentration of APM

Figure 1 shows the monthly average concentration of $PM_{2.5}$, PM_{10} and BC as a function of sampling time, from January 2002 to November 2002. The distribution of $PM_{2.5}$ and PM_{10} depend on the sampling time for both sampling point. They were under the influence of the weather. Meanwhile the black carbon is independent to the

sampling time. These mean that the black carbon was released by a fixed sources on this region. The mass concentration of $PM_{2.5}$ was in the range of 9.6 $\mu g/m^3$ to 24.2 $\mu g/m^3$ and that the PM_{10} was in the range of 33 $\mu g/m^3$ to 66 $\mu g/m^3$ for the urban area (Bandung). Whereas the mass concentration of $PM_{2.5}$ was in the range of 3.5 $\mu g/m^3$ to 11.2 $\mu g/m^3$ and PM_{10} was in the range of about 4.0 $\mu g/m^3$ to 14.0 $\mu g/m^3$ for rural area (Lembang) which are lower that than of urban area (Bandung). These data much lower than that of National Ambient Air Quality Standard (NAAQ) for 24-hour average, 65 $\mu g/m^3$ and 150 $\mu g/m^3$ for PM 2.5 fraction and PM_{10} fraction respectively.

Figure 2 and 3 show the correlation of PM_{10} and $PM_{2.5}$ for both sampling area (urban and rural area). The results showed a phenomenon that have in contradiction. The obtained PM_{10} and $PM_{2.5}$ data for the period of January 2002 – November 2002 indicate a strong correlation (R^2 = 0.9157) for Lembang sampling point, whereas the result of Bandung sampling point is the opposite (R^2 = 0.1956).

Correlation analysis of black carbon with PM_{10} and $PM_{2.5}$, we presented in the Figure 4 and 5 for urban area (Bandung), and Figure 6 and 7 for rural area (Lembang). All resulted analysis show a weak correlation for APM obtained at urban area (R^2 = 0.264 and R^2 = 0.373 respectively) and strong correlation for that in the rural area (R^2 = 0.697 and R^2 = 0.6636 respectively). Correlation among $PM_{2.5}$, PM_{10} and black carbon explained that PM_{10} of Bandung city(urban area) influenced by coarse fraction, whereas PM_{10} of Lembang (rural area) influenced strongly by antrophogenic source.

Enrichment factor

As mention before, the Enrichment Factor (EF) was calculated from measured elements using Mason's crustal abundances for the sampling period using

$$EF = [(X_i/C_i)_{APM}/(X_i/C_i)_{REF}]$$

Where X_i = element of interest and C_i = reference element. The Al was used as reference element because it is supposed to be unique elements in soil. Table 1 and 2 show the elemental quantification of Al, Br, Cl, Mg, Mn, Na and V of $PM_{2.5}$ fraction taken from Bandung and Lembang sampling point. Meanwhile, Table 3 show the calculated EF for seven selected elements of Na, V, Mn, Br, Cl, and Mg contained in the sample taken from the urban area. The EF value of Br and Cl are higher compare to the other. This higher value indicated that they are not of crustal origin and they are

assumed to be anthropogenic elements. The same case is obtained for the urban area (Table 4). The EF value of Br and Cl are higher than the other. We estimate that the Br was released by fuel of the automotive, whereas the Cl probably coming from the utilization of fertilizer.

Black carbon in APM.

The monthly average concentrations of Black Carbon were determined according to suggested method [7, 8] and the obtained result show on the Table 5. The quantity of BC was calculated using formula of

$$BC\ (\mu g/cm^2) = [100/2F.e]\ Ln\ [Ro/R]$$

F indicate of correction factor of order 1, e is the mass attenuation coefficient (m^2/g), R_o and R are the unloaded and loaded filter reflectance respectively for a given wavelength.

The average concentrations of Black Carbon in urban and rural area are in the range of 3.4 $\mu g/m^3$ to 6.1 $\mu g/m^3$ and 1.3 $\mu g/m^3$ to 4.7 $\mu g/m^3$ respectively.

CONCLUSION

- The concentrations of $PM_{2.5}$ are much lower than the value of National Ambient Air Quality Standard (NAAQ) for 24-hour average.
- The quantity of Br and Cl were enriched, both for Bandung and Lembang, whereas V trend to increase.
- The concentration of Black Carbon independent to the time. It probably were released from antrophogenic sources

ACKNOWLEDGMENT

I would like to express special thank to Mr. Iman Kuntoro and Mr. Gunandjar for their support, and also I would like to thank to all members of Serpong Neutron Activation Analysis Laboratory for their cooperation.

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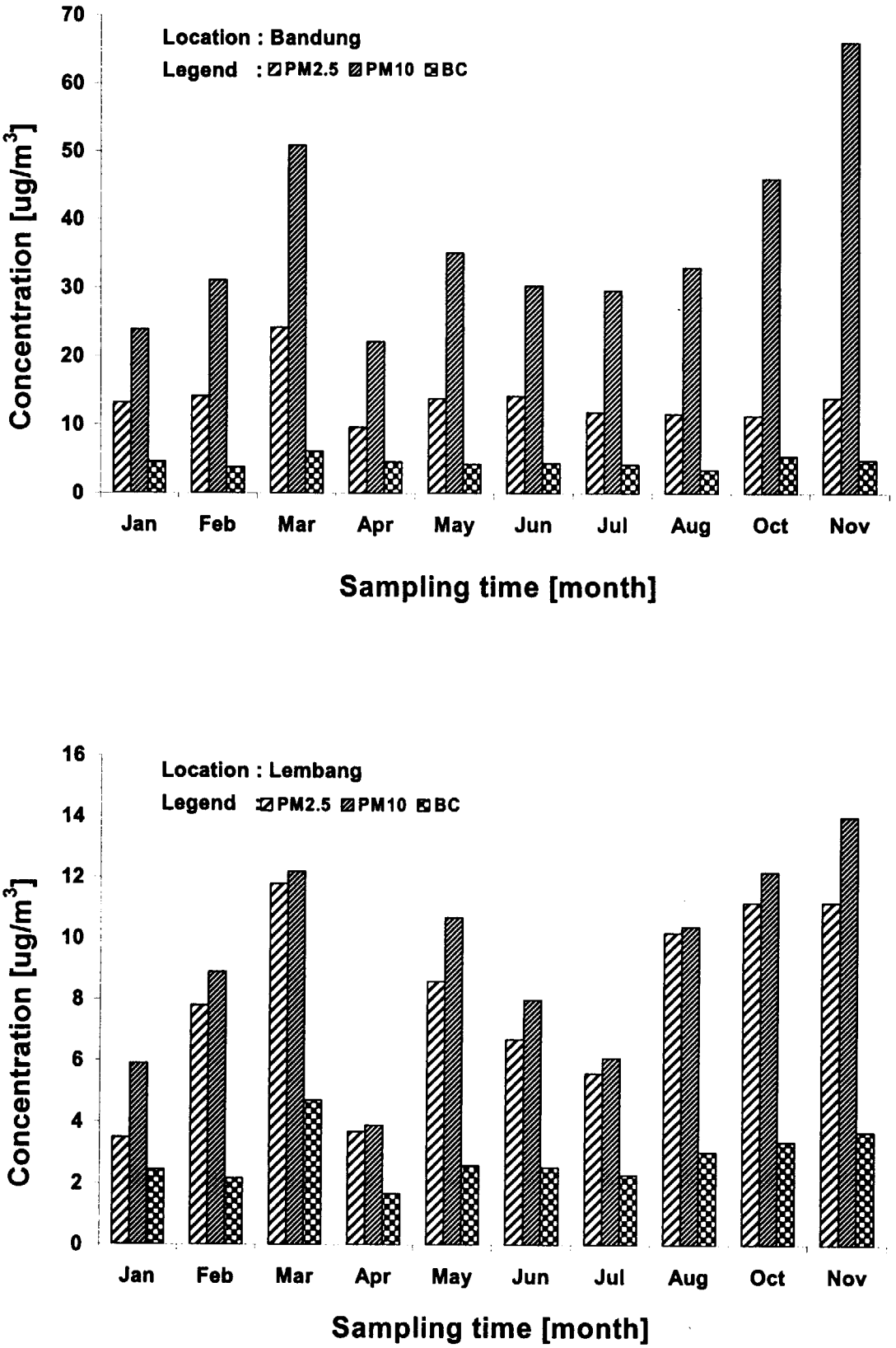

Figure 1. Monthly average of $PM_{2.5}$, PM_{10} and Black Carbon at Bandung (above) and Lembang (below) area on the period of January－November 2002.

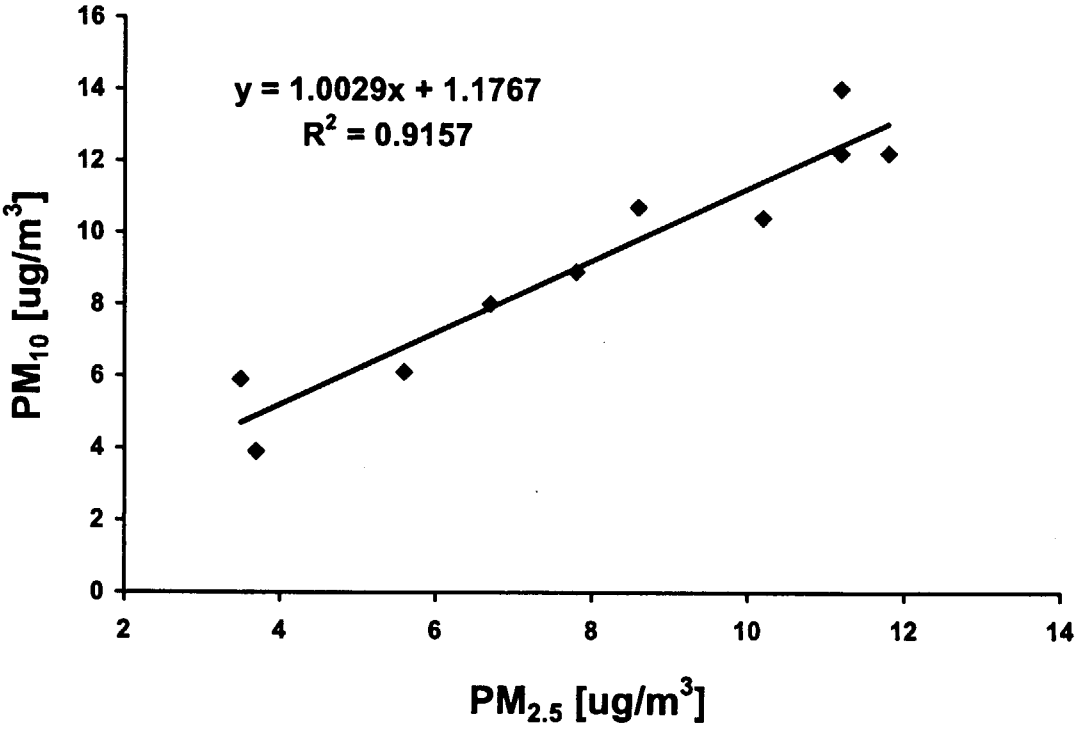

Figure 2. Relation of PM_{10} versus $PM_{2.5}$ gravimetric masses for air particulate sample taken from Lembang sampling point.

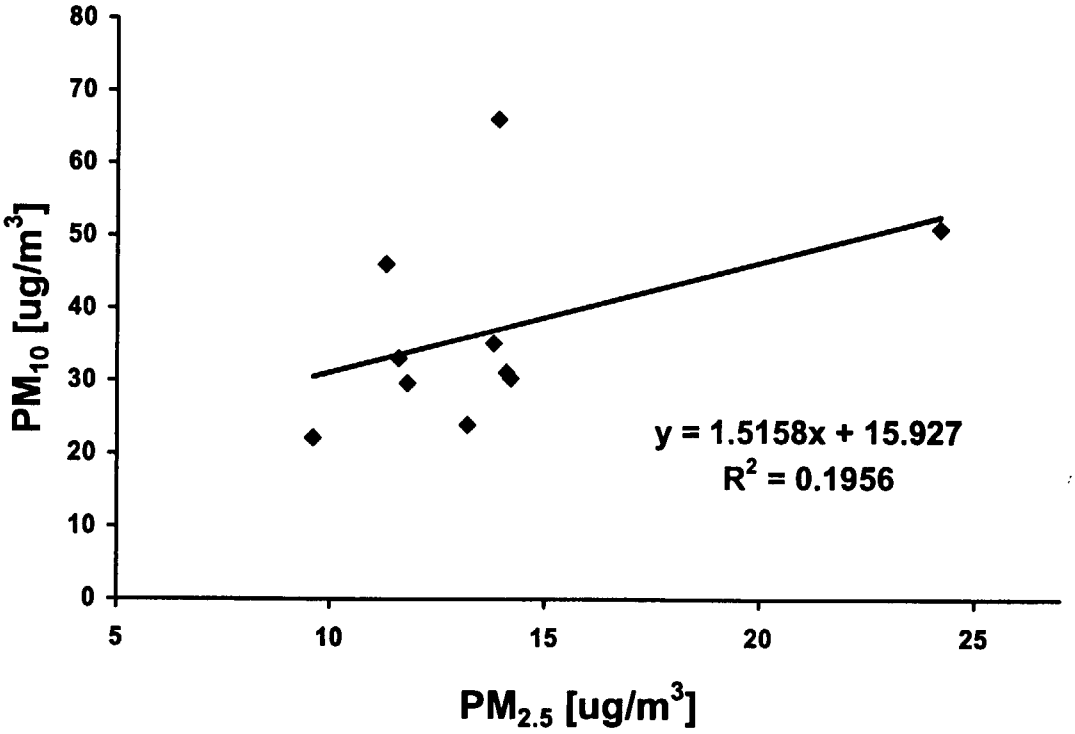

Figure 3. Relation of PM_{10} versus $PM_{2.5}$ gravimetric masses for air particulate sample taken from Bandung sampling point.

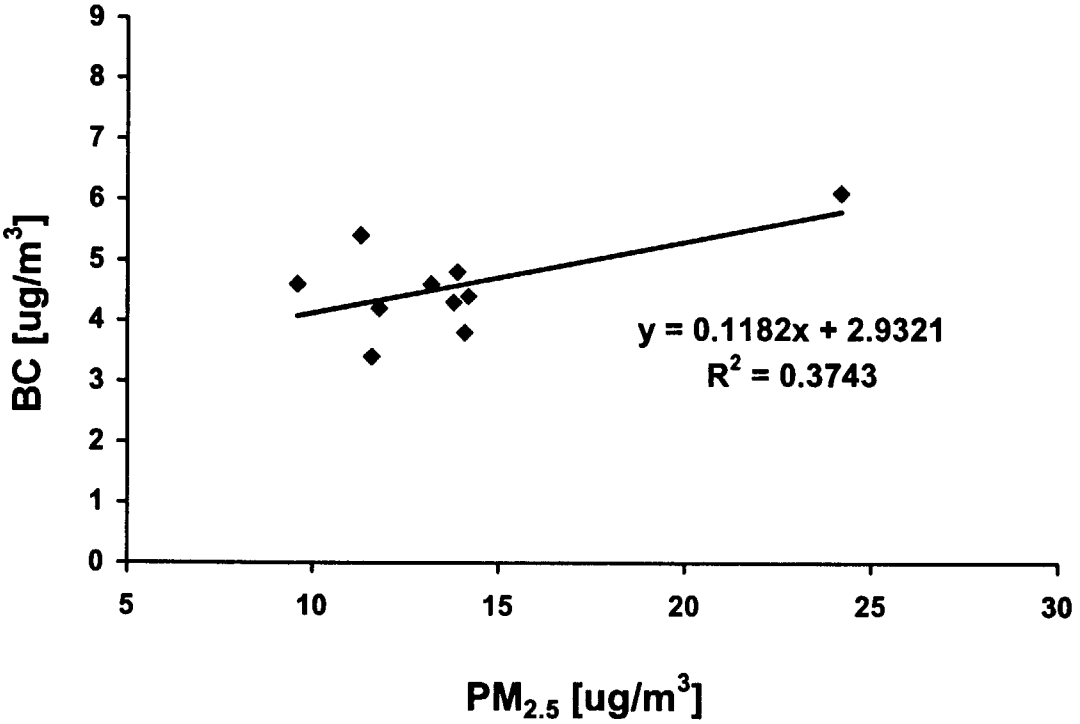

Figure 4. Correlation of Black Carbon to $PM_{2.5}$ for Bandung sampling point

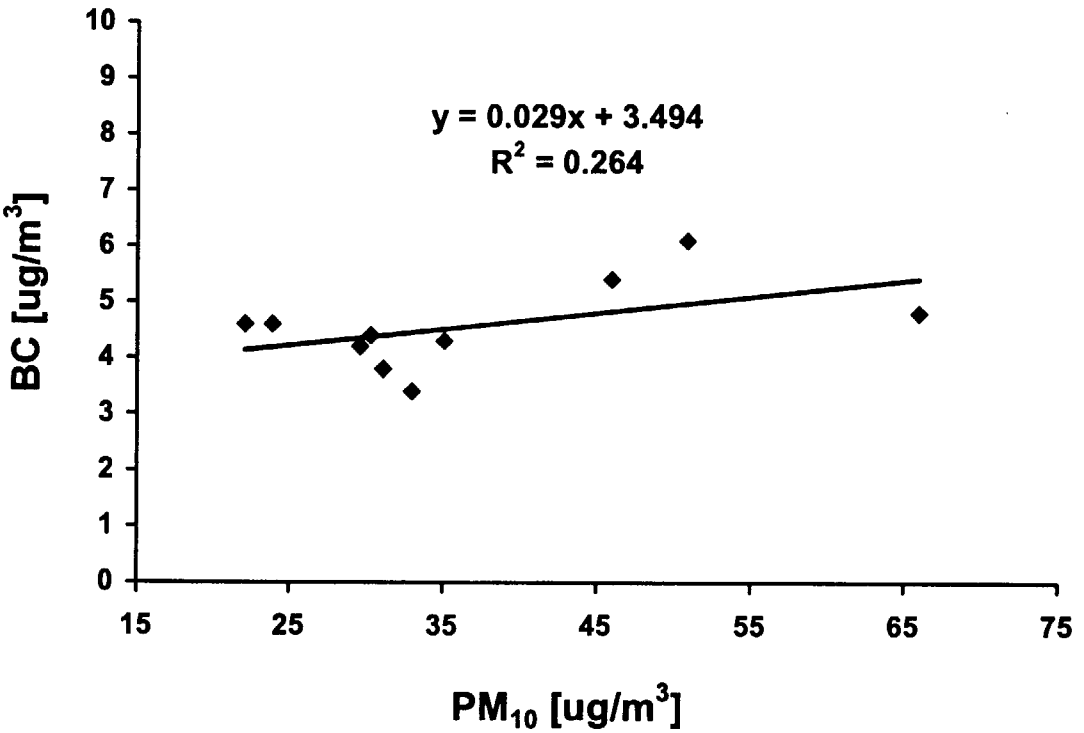

Figure 5. Correlation of Black Carbon to PM_{10} for Bandung sampling point

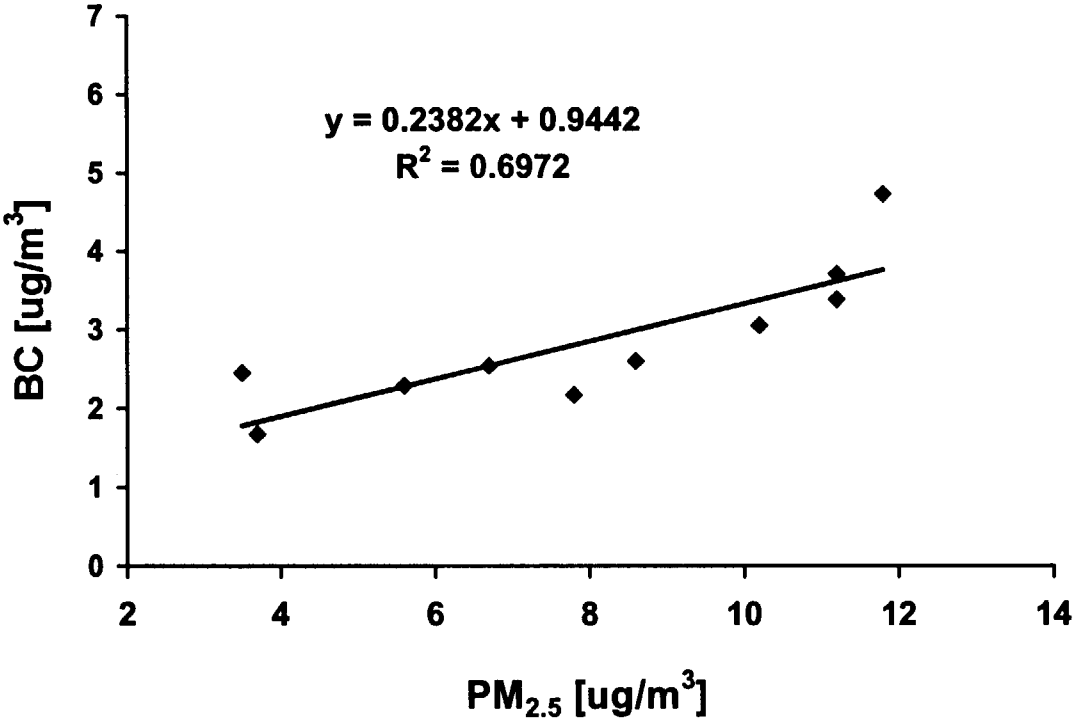

Figure 6 : Relation of Black Carbon to $PM_{2.5}$ for Lembang Area.

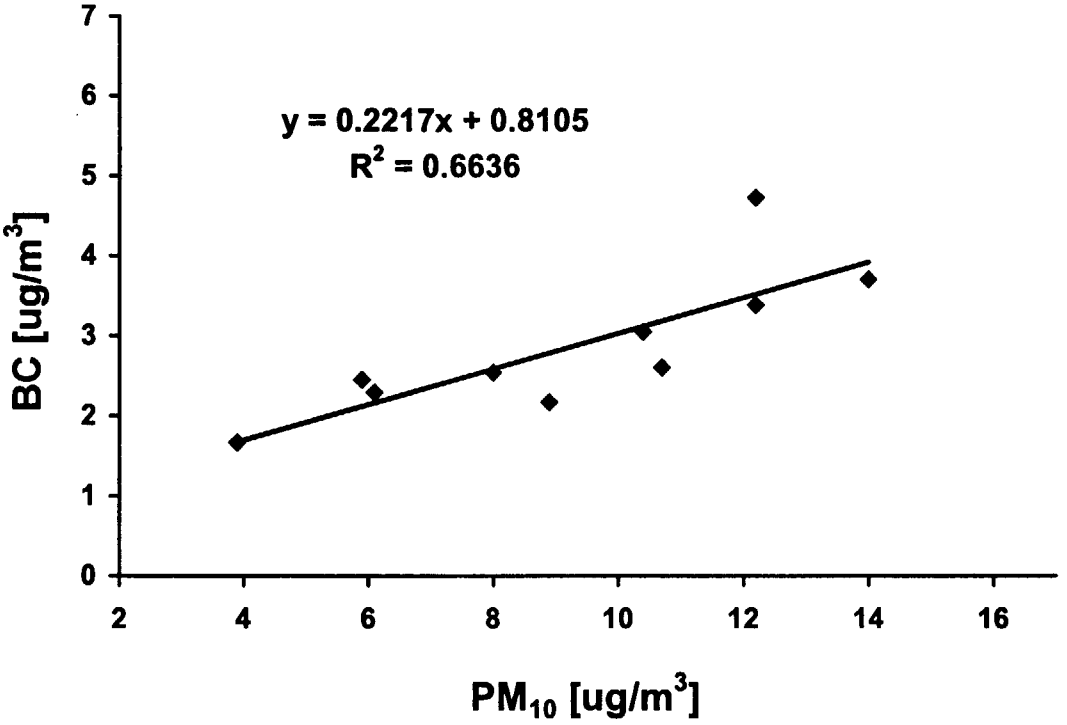

Figure 7 : Relation of Black Carbon to PM_{10} for Lembang Area.

Table 1. Average elemental concentration of $PM_{2.5}$ for Bandung sampling point.

Month	Elementary concentration, [μg/m^3]						
	Al	Br	Cl	Mg	Mn	Na	V
Jan	47	27	56	< 149	1	69	0.8
Feb	38	53	63	< 71	2	62	0.8
Mar	62	57	139	-	2	51	2.1
Apr	109	-	14	-	1	25	-
May	113	-	-	-	2	45	0.6
Jun	-	-	26	-	3	51	1.7
Jul	-	-	29	-	1	99	1.3

Table 2. Average elemental concentration of $PM_{2.5}$ for Lembang sampling point.

Month	Elementary concentration, [μg/m^3]						
	Al	Br	Cl	Mg	Mn	Na	V
Jan	13	24	56	< 53	0.5	12	0.2
Feb	13	24	65	<143	0.7	23	2.4
Mar	13	49	87	-	0.7	-	0.6
Apr	43	49	26	-	0.6	46	0.8
May	26	15	-	<77	-	-	Nd

Table 3. Enrichment factor of elements of aerosol from Bandung area (urban area) are based on Al as reference element.

Month	Na	V	Mn	Br	Cl	Mg
Jan	4	10	3	18682	745	7
Feb	5	13	3	45357	872	6
Mar	2	20	3	99134	1402	Nd
Apr	1	Nd	0.4	4177	80	Nd
May	1	3	1	No data	No data	Nd
June	1	6	1	No data	100	Nd
July	1	2	0.2	No data	76	Nd

Table 4. Enrichment factor of elements of $PM_{2.5}$ in Lembang area are based on Al reference element.

Month	Na	V	Mn	Br	Cl	Mg
Jan	7	14	3	60037	2694	9
Feb	5	14	5	60000	3125	9
Mar	2	19	5	122575	4185	Nd
Apr	3	1	1	37057	378	Nd
May	1	Nd	2	18762	289	8

Table 5. Black Carbon analysis result in $PM_{2.5}$ of Bandung and Lembang Area

Month	Bandung area		Lembang area	
	Black C [$\mu g/m^3$]	% Black C	Black C [$\mu g/m^3$]	% Black C
Jan	4.6	36	2.5	51
Feb	3.8	27	2.2	32
Mar	6.1	25	4.7	43
Apr	4.6	43	2.1	39
May	4.3	35	2.6	33
June	4.4	34	2.5	39
July	4.2	35	2.3	44
Aug	3.4	38	3.1	30
Oct	5.4	40	3.4	33
Nov	4.8	24	1.3	35

JP0450638

JAERI－Conf 2004－010

The 2002 Workshop on the Utilization of Research Reactors
13-17 Jan. 2003, Indonesia

Country Report of Korea

1.23 Recent Applications of Neutron Activation Analysis in Korea

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ABSTRACT

There are two purposes in this research; first aim is to promote the use of neutron activation analysis(NAA) as a utilization of nuclear research reactor in the field of air pollution studies through a routine and long-term monitoring. Other is to improve NAA with an experimental simplicity, high accuracy, excellent flexibility with respect to irradiation and counting conditions.

For the study on air pollution, airborne particulate matter(APM) for the fine (<2.5 μm EAD) and coarse particle (2.5-10 μm EAD) fractions were collected using the Gent stacked filter unit low volume sampler and two types of Nuclepore polycarbonate filters. Air samples were collected at two regions(suburban and industrial site of Daejeon city in the Republic of Korea) from January to December 2002. Mass concentration and elemental black carbon of APM were measured and the concentration of 25 elements were determined by Instrumental NAA. Analytical quality control is carried out using three certified reference materials(CRM). The monitoring data were treated statistically to assess air pollution source and source apportionment. The results obtained from this project can be used to investigate source identification and apportionment and its trends, and to establish a more cost-effective method for national air quality management.

Preliminary experiment for application of ko-standardization method has been carried out to determine the reactor neutron spectrum parameters, i.e. a and f-values as the main factors of irradiation quality at NAA #1 irradiation hole on HANARO research reactor, to determine peak detection efficiency for the HP Ge(EG & G ORTEC, GEM 35185) detector for the use in the ko-experiments and to compare the measured concentration results with the certified values of some CRMs applying the experimentally determined ko-parameters.

Key Words : Research Reactor, Instrumental Neutron Activation Analysis, Air Pollution Source, Source Apportionment, *ko*-NAA

1. INTRODUCTION

1.1 Air Pollution Study

Due to recognition and regulation for environmental pollution, which is caused by rapid

industrialization and urbanization, vehicle increment, etc., public concerns about clean, comfortable surroundings has been increased. For this reason, intensive efforts to control pollution sources and to examine contamination levels through the analysis of various environmental samples are being followed up. Therefore, measurement of elemental concentrations in environmental samples including air, soil and water, is very important to solve pollution problems. In particular, airborne particulate matter(APM) consists of various elements from both artificial as well as natural origin. When the level of total suspended particulate matter(TSPM) and/or the content of specific elements in PM_{10} (<10 μm EAD) are high, the short and long term effects on human health are insidious and important. Therefore, sampling and elemental analysis of PM_{10} classifying the fine particle(FPM : <2.5 μm EAD) and coarse particle (CPM : 2.5-10 μm EAD) fractions is the first step to estimate air quality and level of air pollution. Instrumental neutron activation analysis(INAA) for airborne particulate matter can analyze up to μg/g ~ ng/g level of concentrations for 30～40 trace elements simultaneously.

The results can be used to conduct epidemiological studies, to investigate source identification and apportionment, to study long-range transport and atmospheric processes, and to establish a more cost-effective air quality management. Air pollution sources can be identified by employing a statistical method of analytical data such as the Enrichment Factor, Receptor Modeling and Chemical Mass Balance Methods. If the concentrations of several elements in air samples that are specifically related to emission sources are measured, the above statistical techniques can be used to identify the fingerprinting of air pollution from the results. For these emission source studies, collection of comprehensive analytical data is required continuously. In addition, the selection of a sampling site, sampling methods and air sampler depend on the purpose of research and the elements to be analyzed.

1.2 Methodology of *ko*-Standardization

Neutron activation analysis based on *ko*-standardization method (so called *ko*-NAA) has been known as one of the most remarkable developments of the NAA with advantages of experimental simplicity, high accuracy, excellent flexibility with respect to irradiation and counting conditions, and suitability for computerization. It is not a theory describing a physical phenomenon, but a protocol for calibration procedures. It has been developed as an absolute standardization, where the unreliable nuclear data is replaced with accurate experimentally determined compound nuclear constants, so called *ko*-factor, or as a single comparator standardization which is made flexible with respect to varying characteristics of the neutron spectrum and of the detector and the source-detector configuration. *ko*-factor of analyte *a* versus monitor *m* is defined as

$$ko_{,m(a)} = \frac{M_m A_a \theta_a \sigma_{0,a}}{M_a A_m \theta_m \sigma_{0,m}} \qquad (1)$$

where, M : atomic weight, A : absolute gamma intensity, θ : isotopic abundance and σ_0 : (n,γ) reaction cross-section at 2200 m/sec.

The *ko*,Au factors experimentally determined at the INW, Gent and the KFKI, Budapest, and later with valuable contributions from other laboratories as well, can be tabulated and published in literature as a generally useful parameter. Then, the coirradiated sample with a monitor Au, the *a* analyte concentration can be obtained by following expression:

$$\rho = \frac{Asp,a}{Asp,Au} \cdot \frac{1}{ko,Au(a)} \cdot \frac{Gth,Au \cdot f + Ge,Au \cdot Q_{o,Au}(\alpha)}{Gth,a \cdot f + Ge,a \cdot Qo,_a(\alpha)} \cdot \frac{\varepsilon\ p,Au}{\varepsilon\ p,a} \qquad (2)$$

where,

Asp,a = (Np/tc)/SDCW

W : sample weight, gram

Np : measured gamma net peak area (counts)

tc : counting time

S : saturation factor, 1- exp(-λ ti)

λ : decay constant, ln2/$T_{1/2}$

$T_{1/2}$: half life

ti : irradiation time

D : decay factor, exp(-λ td)

td : dacay time

C : counting factor, [1- exp(-λ tc)]/λ tc

tc : counting time

Asp,Au = (Np/tc)/SDCw : specific count rate

w : monitor weight, gram

Gth : correction factor for thermal neutron self shielding

Ge : correction factor for epithermal neutron self shielding

f= Φ th / Φ e : thermal to epithermal flux ratio

α : parameter for the deviation of the epithermal neutron distribution form 1/E-law, approximated by $1/E^{1+\alpha}$ dependence. The α value can be positive or negative depending on the reactor condition (e.g. moderator material, geometry of irradiation site, etc.)

$$Q_0(\alpha) : I_0(\alpha)/\sigma_0 = \{(Q_0 - 0.429)/ Er'^{\alpha}\} + \{0.429/E_{cd}^{\alpha}(2\alpha + 1)\}(1eV)^{\alpha} \qquad (3)$$

where, $Q_0 = I_0/\sigma_0$, I_0 : resonance integral, Er' : effective resonance energy, Ecd : cadmium cut-off energy, 0.55 eV

ε p : full-energy(photo) peak detection efficiency

Equation (2) indicates that I_0 (or Q_0) must be converted to $I_0(\alpha)$ [or $Q_0(\alpha)$] as expressed by Eq. (3) in the *ko*-NAA method. This means that α should be known or experimentally determined. The physical and mathematical basis of the α -determination can be described as follows:

The deviations from the ideal 1/E-law can be approximated by a non-ideal, real epithermal neutron spectrum, expressed by F. de Corte et al. :

$$\Phi_{e(E)} = \frac{\Phi_e}{E^{1+\alpha}} \qquad (4)$$

where, $\Phi_{e(E)}$: epithermal flux per unit of neutron energy interval

Φ_e : energy independent proportionality constant

= integrated epithermal flux per unit of ln E-interval,

Equation (4) is rearranged as :

$$\Phi_{e(E)} E = \Phi_e E^{-\alpha} \qquad (5)$$

Thus, when plotting Φ e(E)E versus logE for some suitable monitors, a straight line will be obtained with slope -α . From Eq.(5), a formula for the α -determination based on "Cd-ratio for

multi-monitor" method in which monitors are characterized by the effective resonance energy Er', is derived as follows :

A set of N monitors are irradiated with and without Cd-covers, and the induced activities are measured on a Ge detector. If all the monitors have a $\sigma(v) \propto 1/v$ dependence up to 1.5 eV, the slope of the straight line, $-\alpha$, can be obtained by plotting :

$$\log \frac{(Er',i)^{-\alpha}}{[(Fcd,i \cdot Rcd,i - 1) \cdot Q_{0,i}(\alpha) \cdot (Ge,i/Gth,i)}\ \text{versus}\ \log Er',i \qquad (6)$$

where, i denotes isotope 1, 2, ... , n.

Two monitors with three nuclides of ^{197}Au, ^{94}Zr and ^{96}Zr, so called triple-monitor, are an optimal selection for both reliability in result and experimental simplicity. Relevant nuclear data is listed in Table 1. In practice, one can select a number of physically suitable monitors(e.g. metallic or alloyed foils and wires) with Er' ranging from low to high to check the linearity of the plotting line, thus proving that α is constant over the whole epithermal neutron energy region in the reactor irradiation position under consideration

For the determination of the parameter, f, the ratio of the thermal to epithermal neutron flux, Cd-ratio method can be used :

$$f = (RcdFcd - 1)\, Q_0(\alpha)(Ge/Gth) \qquad (7)$$

where, FCd : cadmium transmission factor for epithermal neutrons,
RCd : cadmium ratio.

The monitor used is an element with well-known Q_0 and Er' values (Au is suitable for this requirements), which is irradiated subsequently with and without Cd-cover.

2. EXPERIMENTAL

2.1 Air pollution Study

1) Sampling and sample preparation

Airborne particulates for the fine particle (<2.5 μm EAD) and coarse particle (2.5-10 μm EAD) fractions were collected using the Gent SFU sampler(low volume air sampler) with two kinds of polycarbonate filters(ϕ 47 mm, 0.4 and 8 μm pore size, Nuclepore) according to the recommended method. Flow rate of sampler is calibrated using Gillian Gilibrator2 Calibration System (Sensidyne Inc.). Samples were collected at two regions(suburban and industrial sites of Daejeon city located in the southwestern region of Korea) as shown Figure 1. Longitude, latitude and altitude of sampling site are about E127°, N36° and 50-70 m, respectively. The suburban sampling site is located several kilometers from the heavy-traffic intersection of the four-lane Honam highway and there are a small mountain in the rear of sampling site. In the industrial sampling site there are many wooden and metal fabrication factory. The sampler was placed about 6 m above the ground. Meteorological conditions such as temperature, humidity, wind direction, wind velocity and change in weather conditions were recorded regularly during the collection of samples and showed in Figure 2. The

flow rate was adjusted to 18 //min at the beginning of sampling and collected for 24 hours once a week from January to December 2002. In this period, about 50 samples for two particle fraction were collected at two sites individually. The total volume of air sampled through the filter was calculated from the reading of the volume meter and compared with the obtained value from the average of the initial and final flow rate. The collected samples were pre-stored for 24 hours in a controlled atmosphere(20℃, 50% relative humidity) and prepared in polyethylene vials after weighing in the same conditions. Electrostatic charges are controlled by ^{210}Po radioactive source. For both fine and coarse filters, each filter was cut into three parts, half for the analysis of medium and long half-life nuclides, a quarter for the short half-life nuclides and the rest for storage.

Before elemental analysis of the sample collected, the reflectance of fine particle filters was measured using Smoke Stain Reflectometer(Model 43D) according to recommended procedure for calculation of concentration of the elemental black carbon.

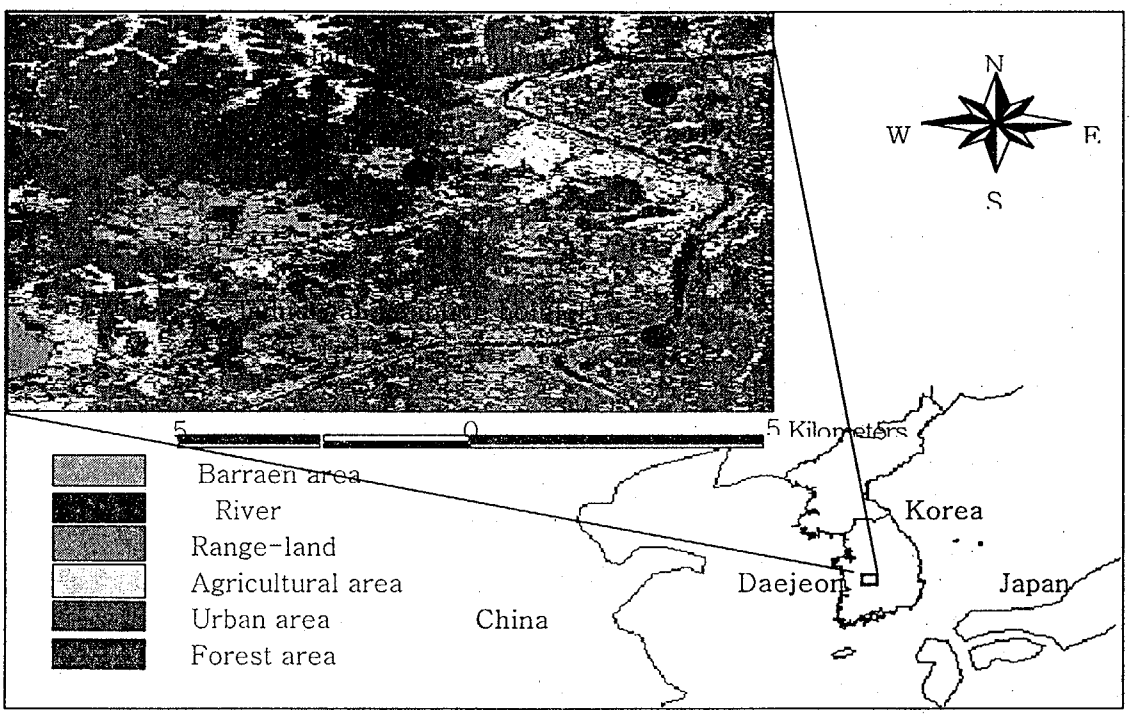
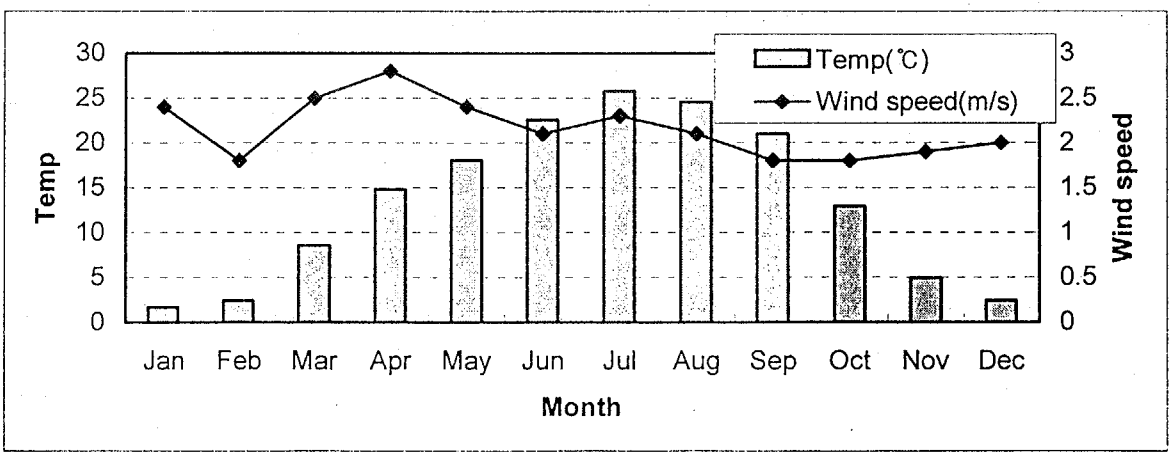


Figure 1. Sampling site of Daejeon city in Korea



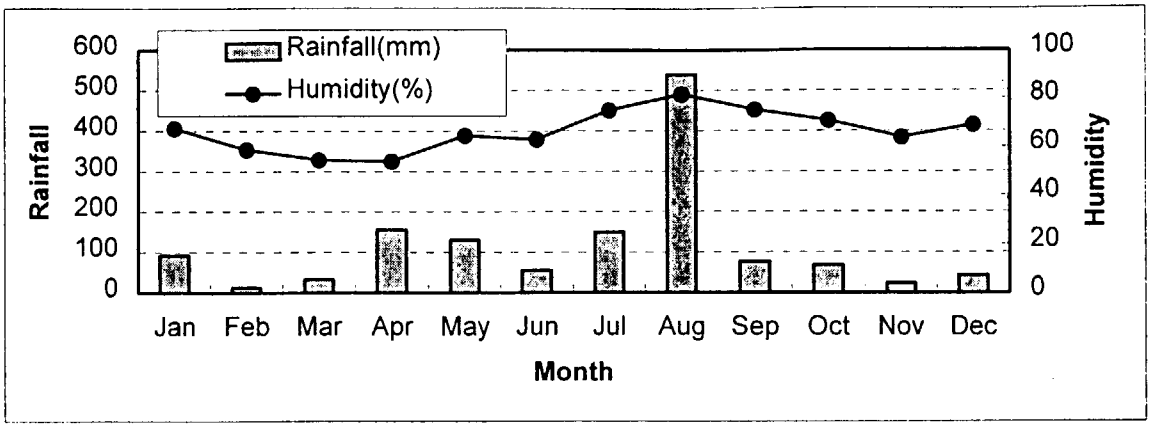


Figure 2. Meteorological condition of sampling region during sampling period.

2) INAA analysis of sample

Collected samples were irradiated with thermal neutrons using the Pneumatic Transfer System(PTS, $\Phi_t = 2.80 \times 10^{13}$ n/cm^2· s, $R_{cd} \sim 205$) at the HANARO research reactor at the Korea Atomic Energy Research Institute. Samples were irradiated at the same position to minimize geometric errors. For neutron flux monitoring, activation wires (Reactor Exp. Inc., R/X activation wire) such as Au-Al, Co and Fe were used. Analytical conditions were optimized after considering the preliminary results.

The measurements were carried out using a high purity Ge detector of 25% relative efficiency and 1.85keV resolution(FWHM) at 1332 keV of ^{60}Co and peak to Compton ratio is 45:1, coupled to a personal computer and 8k-multichannel analyzer (EG&G ORTEC, 919A MCB). Energy and efficiency calibration were done using multi-nuclide reference sources (Isotope Products Lab., ML 7500 series, 0.118" active diameter, disc type) certified by NIST. GammaVision software (EG&G ORTEC) for energy and efficiency calibrations, the acquisition of gamma spectra and peak analysis were used.

3) Data Treatment

Under the optimum analytical conditions, such as irradiation and counting time, the elemental concentrations in both samples and blank filters were measured. Concentration of the elements was calculated using the new Windows PC-code, Labview software of KAERI with the nuclear data library, which was developed at this laboratory for rapid and simple data treatment with the gamma ray spectrum obtained at preset detection conditions. The detection limits for the elements can be calculated by Currie's quantitative definition with 10% allowable uncertainty. The assessment of measurement uncertainty were evaluated as a combined uncertainty including most of the sources of standard uncertainty to be considered in INAA. The analytical data were treated statistically using Microsoft Excel.

2.2 *ko*-Standardization

1) Efficiency calibration of gamma-ray spectrometer

Gamma-ray spectrometer, HpGe detector(EG & G ORTEC, GEM35185) and an 8192 MCA, was used for this study. Eq. (2) implies that the full-energy peak detection efficiency(ε_p) of gamma-ray spectrometer used must be determined. The calibration of the gamma-ray spectrometer according to

the *ko*-NAA method has been defined as having three main problems. The first one is the determination of gamma energy peak detection efficiency using gamma-ray standard sources and the second is the correction for true-coincidence effects. The last is the correction for practical sample geometric conditions. The correction of true-coincidence effects and sample geometry was not yet done in this work due to the lack of proper *ko*-software program. Figure 3 shows the peak detection efficiency curve for three measurement positions. Distance of source-end cap of detector at position 3, 5 and 7 were 8.4 cm, 13.8 cm and 19.2 cm, respectively. In order to fit the experimental points, a fourth degree polynomial method was used.

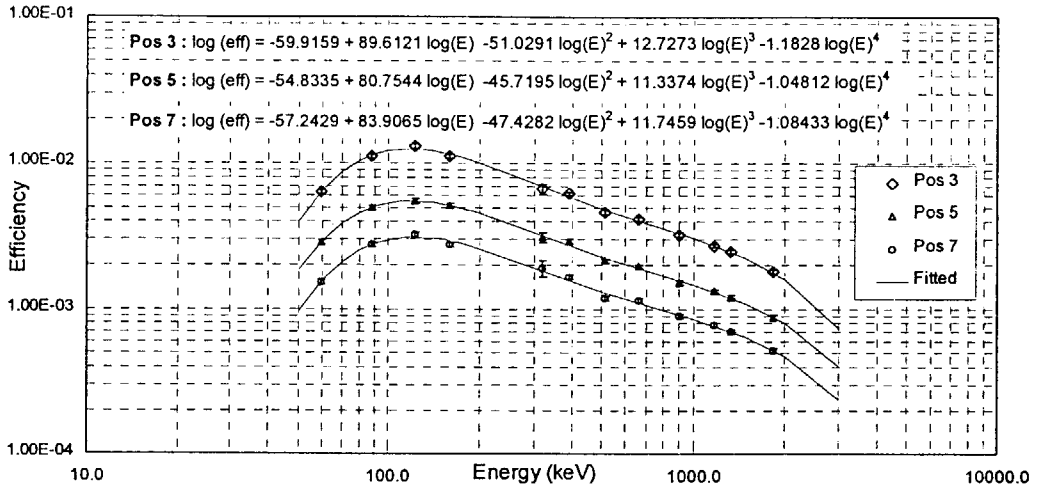


Figure 3. Efficiency curve of the HPGe detector(EG & G ORTEC, GEM35185) for three positions.

2) Determination of Cd-ratio and f-parameter

The "Cd-ratio for triple-monitor" method and Cd-ratio method were respectively applied to determine α and f parameters for NAA #1 irradiation hole of HANARO research reactor. Four sets of monitors consisting of pieces of Zr sheet (99.7329%, thickness 0.125 mm), Au-Al wire(Au 0.1124%, dia. 0.508 mm), and pure cadmium cover(thickness 1.0 mm), were applied. Each monitor set was irradiated for 10 minutes. The calibrated gamma-ray spectrometer and GammaVision 5.1 software were used for gamma-ray measurement and analysis of collected spectra.

3) Standard Reference Materials for Method Validation

In order to validate *ko*-NAA method in our system, four kinds of NIST Standard Reference Materials(SRMs), NIST SRM 1633a(Coal Fly Ash), NIST SRM 1577b(Bovine Liver), NIST SRM 2704(Buffalo River Sediment) and NIST SRM 2709(SanJoaquin Soil) were prepared and analyzed with experimentally determined *ko*-parameters. The samples with the weight of 150～200 mg for the elemental analysis using medium and long half-life nuclides were irradiated at NAA #1 hole for 30 minutes. For the elemental analysis using short half-life nuclides, NIST SRM 1633a and 1577b samples with the weight of 10～30 mg were irradiated for 1 minute. After the proper cooling time, gamma-rays from each sample were measured and the elemental concentrations were calculated by *ko*-NAA method.

3. RESULTS AND DISCUSSION

3.1 Air pollution Study

1) Analytical quality control

After the optimum analytical conditions such as irradiation time, cooling time, counting time, etc were pre-estimated according to previous procedures, the standard reference materials(National Institute of Standards and Technology, U.S.A, NIST SRM 1632c, coal fly ash; NIES CRM, No.8, urban dust) were used for analytical quality control. The analytical results are presented in Table 1. In addition, we participated in the proficiency test program of the IAEA(NAT-7, Air filter sample, 2001) and had reliable results for all measured elements as shown in Figure 4. The filter blank values of major impurities are used for the correction of individual concentrations. The concentrations of interfering elements such as Al, Cl, Mn, Br, Na were low relative to other filter types.

Table 1. Analytical results of three certified reference materials.

(a) NIES CRM No-8, Urban dust

Element	Nuclide (Energy, keV)	Experimental Value		Certi. Value	RE(%)
		Range	Mean ± SD		
Al	Al-28(1779)	3098 ~ 3365	3230 ± 119	3300 ± 200	2.1
As	As-76(559)	2.68 ~ 3.27	3.01 ± 0.30	2.6 ± 0.2	-15.7
Ba	Ba-139(165)	110 ~ 126	119 ± 6		
Br	Br-82(554)	57.7 ~ 60.5	58.7 ± 1.5	56	-4.8
Ca	Ca-49(3084)	4776 ~ 5154	4912 ± 156	5300 ± 200	7.3
Ce	Ce-141(145)	3.36 ~ 3.59	3.48 ± 0.11	3.1	-12.2
Cl	Cl-38(1642)	862 ~ 1018	908 ± 63		
Co	Co-60(1173)	3.41 ~ 3.64	3.51 ± 0.12	3.3 ± 0.3	-6.5
Cr	Cr-51(320)	26.2 ~ 30.4	27.8 ± 2.3	25.5 ± 1.5	-8.9
Eu	Eu-152(1408)	0.047 ~ 0.063	0.057 ± 0.009	0.05	-14.7
Fe	Fe-59(1099)	1099 ~ 6228	5311 ± 799		
Hf	Hf-181(482)	0.18 ~ 0.21	0.19 ± 0.02		
Hg	Hg-203(279)	0.21 ~ 0.23	0.22 ± 0.01		
K	K-42(1524)	1089 ~ 1539	1236 ± 179	1150 ± 80	-7.5
La	La-140(1596)	1.28 ~ 1.35	1.31 ± 0.04	1.2	-9.2
Mg	Mg-27(1014)	1142 ~ 1308	1220 ± 59	1010 ± 50	-20.8
Mn	Mn-56(1810)	69.72 ~ 71.55	70.3 ± 0.7		
Na	Na-24(1368)	1369 ~ 1976	1958 ± 20	1920 ± 80	-2.0
Sb	Sb-122(564)	5.77 ~ 5.88	5.81 ± 0.07	6.0 ± 0.4	3.2
Sc	Sc-46(889)	0.54 ~ 0.57	0.55 ± 0.02	0.55	-0.4
Se	Se-75(264)	1.59 ~ 1.89	1.79 ± 0.17	1.3	-37.4
Sm	Sm-153(103)	0.19 ~ 0.22	0.20 ± 0.02	0.20	0.1
Sr	Sr-87m(388)	92 ~ 105	101 ± 5	89 ± 3	-13.9
Ta	Ta-182(1221)	0.10 ~ 0.14	0.11 ± 0.02		
Th	Th-233(311)	0.36 ~ 0.42	0.39 ± 0.03	0.35	-11.4
Ti	Ti-51(320)	281 ~ 364	318 ± 36		
V	V-52(1434)	14.89 ~ 16.12	15.50 ± 0.46	17 ± 2	8.8
W	W-187(685)	5.64 ~ 6.48	6.08 ± 0.42		
Yb	Yb-169(198)	0.09 ~ 0.12	0.10 ± 0.02		
Zn	Zn-65(1115)	1116 ~ 1191	1143 ± 42	1040 ± 50	-9.9

(b) NIST SRM 1632c, Coal fly ash

Element	Nuclide (Energy, keV)	Experimental Value		Certi. Value	RE(%)
		Range	Mean ± SD		
Al	Al-28(1779)	9383 ~ 10240	9632 ± 361	9150 ± 137	-5.3
As	As-76(559)	5.71 ~ 6.39	6.02 ± 0.35	6.28 ± 0.27	4.1
Ba	Ba-139(165)	38 ~ 40	38.6 ± 0.7	41.1 ± 1.6	6.2
Br	Br-82(554)	17.4 ~ 18.2	17.8 ± 0.4	18.7 ± 0.4	4.7
Ca	Ca-49(3084)	1167 ~ 2123	1550 ± 449	1450 ± 300	-6.9
Ce	Ce-141(145)	11.6 ~ 11.7	11.7 ± 0.1	11.9 ± 0.2	1.7
Cl	Cl-38(1642)	1076 ~ 1112	1095 ± 13	1139 ± 41	3.9
Co	Co-60(1173)	3.55 ~ 3.65	3.60 ± 0.05	3.48 ± 0.2	-3.3
Cr	Cr-51(320)	14.75 ~ 15.05	14.88 ± 0.15	13.73 ± 0.20	-8.4
Cs	Cs-134(795)	0.540 ~ 0.659	0.596 ± 0.060	0.594 ± 0.01	-0.4
Dy	Dy-165(95)	0.78 ~ 0.98	0.89 ± 0.08		
Eu	Eu-152(1408)	0.2238 ~ 0.2340	0.2287 ± 0.0051	0.1238 ± 0.003	-84.7
Fe	Fe-59(1099)	7564 ~ 7730	7631 ± 88	7350 ± 110	-3.8
Hf	Hf-181(482)	0.445 ~ 0.472	0.454 ± 0.015	0.585 ± 0.01	22.3
K	K-42(1524)	1133 ~ 1183	1164 ± 18	1100 ± 33	-5.8
La	La-140(1596)	7.15 ~ 7.24	7.21 ± 0.05		
Lu	Lu-177(208)	0.16 ~ 0.17	0.16 ± 0.00		
Mn	Mn-56(1810)	12 ~ 16	14.2 ± 1.6	13.04 ± 0.53	-9.1
Na	Na-24(1368)	284.1 ~ 303.3	290.8 ± 10.9	298.8 ± 4.8	2.7
Rb	Rb-86(1076)	7.45 ~ 8.27	7.83 ± 0.41	7.52 ± 0.33	-4.1
Sb	Sb-122(564)	0.428 ~ 0.480	0.458 ± 0.027	0.461 ± 0.029	0.6
Sc	Sc-46(889)	2.719 ~ 2.767	2.746 ± 0.025	2.905 ± 0.036	5.5
Se	Se-75(264)	1.369 ~ 1.854	1.551 ± 0.264	1.326 ± 0.071	-17.0
Sm	Sm-153(103)	1.011 ~ 1.033	1.022 ± 0.011	1.078 ± 0.028	5.2
Sr	Sr-87m(388)	61 ~ 74	66.6 ± 5.8	63.8 ± 1.4	-4.4
Th	Th-233(311)	1.47 ~ 1.51	1.48 ± 0.03	1.40 ± 0.03	-5.9
Ti	Ti-51(320)	445 ~ 526	490 ± 33	517 ± 32	5.3
V	V-52(1434)	22 ~ 25	23.8 ± 1.1	23.7 ± 0.5	-0.3
Yb	Yb-175(282)	0.40 ~ 0.43	0.42 ± 0.01	0.40	-5.0
Zn	Zn-65(1115)	12.6 ~ 14.4	13.2 ± 1.3	12.1 ± 1.3	-9.2

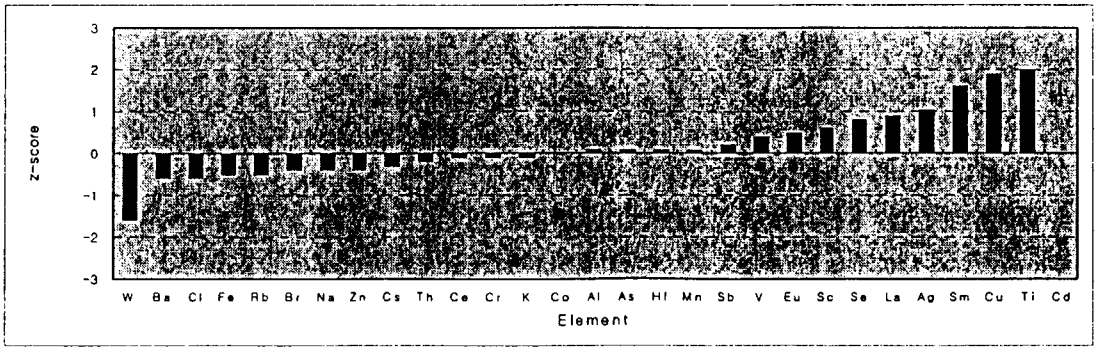


Figure 4. Results of proficiency test of (a) urban dust sample.

2) Mass concentrations of APM

The mass concentration of the coarse fraction at Daehwa industrial site(DC), the fine fraction at Daehwa industrial site(DF), the coarse fraction at KAERI site(KC) and the fine fraction at KAERI site(KF) located in the suburban area were measured from Jan. to Dec. 2002. The monitoring data during period of study are presented in Figure 5 in accordance with sampling site, date and particle size. The average concentrations of APM in industrial site were higher than those of suburban site. The mass concentration of CPM in the industrial site was higher than those of suburban site and in the case of FPM, it was reversed.

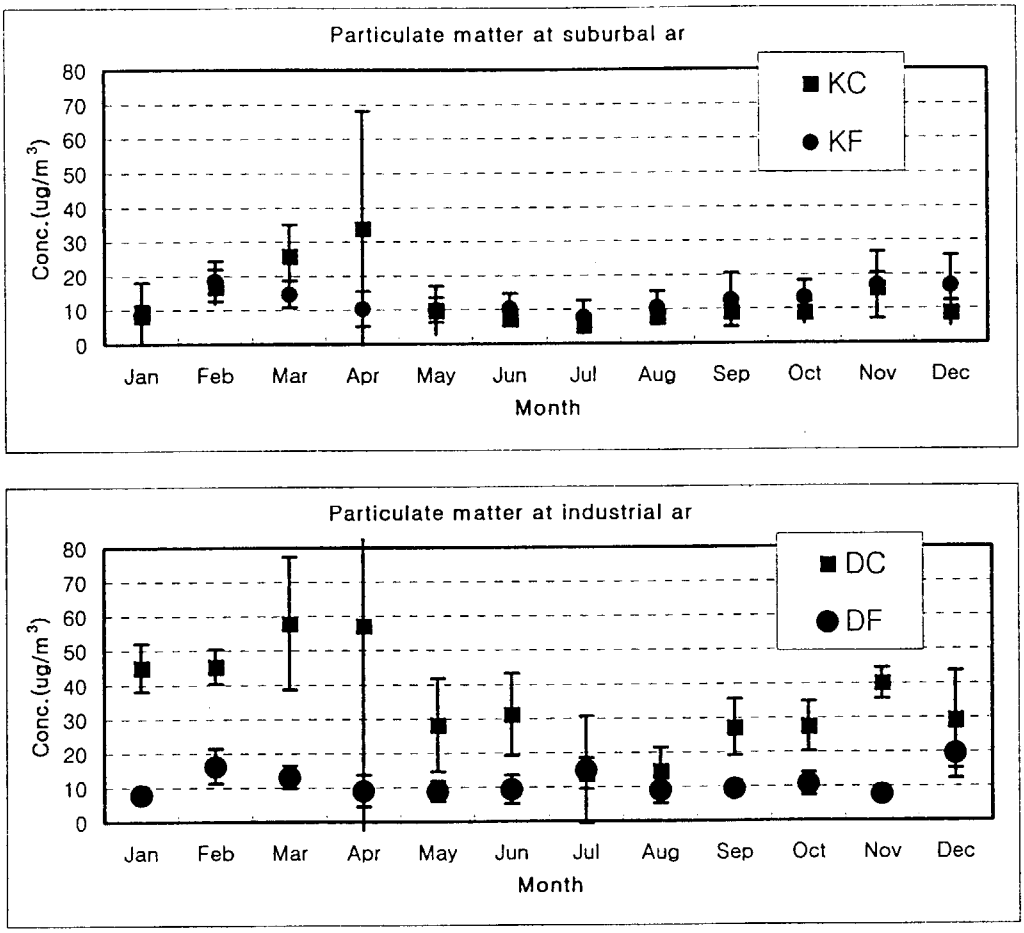


Figure 5. Variation of mass concentrations for FPM and CPM in two regions.

3) Elemental black carbon in FPM($PM_{2.5}$)

The concentrations of elemental carbon, EC, were determined for FPM($PM_{2.5}$) by W. Maenhaut and D. Cohen's suggestion using the reflection method on Nuclepore filter as follows :

$$BC(\mu g/cm^2) = [100/2F\,\varepsilon]\ \mathrm{Ln}\ [R_0/R] \quad (8)$$

where, F is a correction factor of order 1, ε is the mass attenuation coefficient in m^2/g, R_0 is the unloaded filter reflectance, 100 and R is the loaded filter reflectance for a given wavelength. The concentrations of elemental black carbon (BC) were determined for the fine fraction according to suggested methods during sampling period and are presented in Figure 6. The average concentrations of elemental black carbon at the industrial and suburban areas were 9.05 and 7.67 $\mu g/m^3$, respectively. The amounts of elemental black carbon during autumn and winter seasons were relatively higher than those of other seasons.

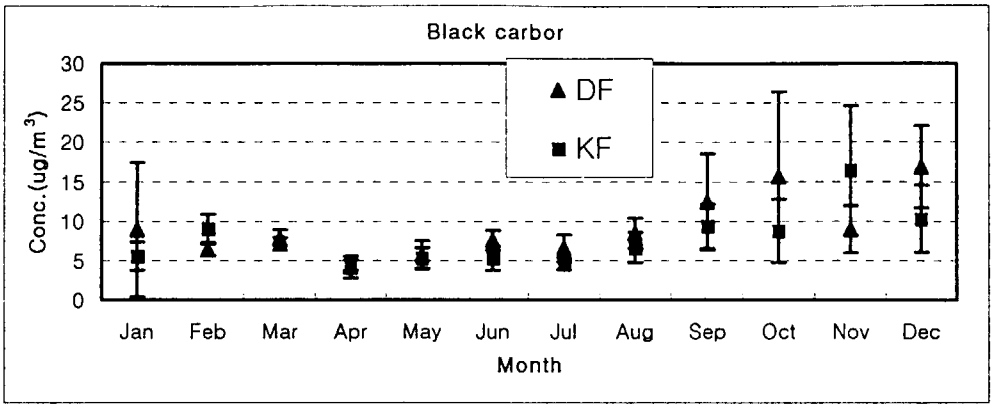
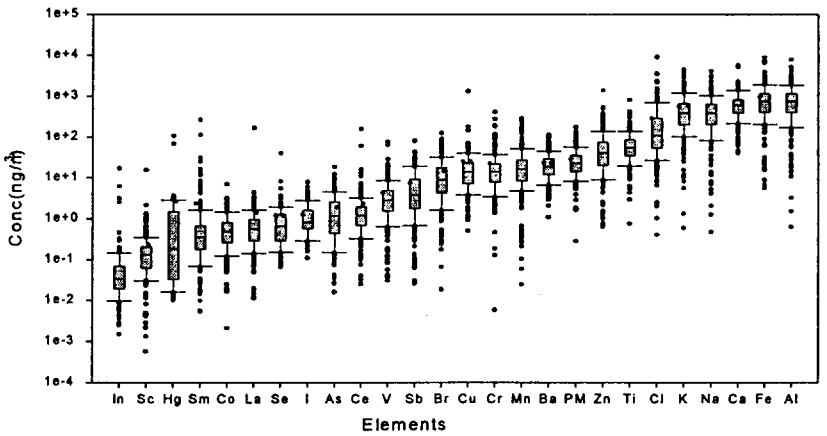


Figure 6. Variation of elemental carbon concentration for FPM in two regions.

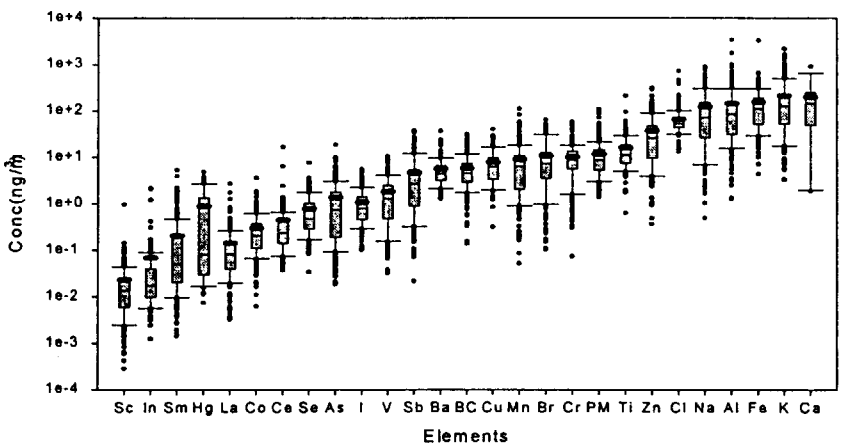
4) Elemental concentrations in APM

The range and annual average of elemental concentration were obtained from the measurement values of individual sample and the results together with number of samples measured for each element are summarized. The concentrations of 25 elements, Al, As, Ba, Br, Ca, Ce, Cl, Co, Cr, Cu, Fe, Hg, I, In, K, La, Mn, Na, Sb, Sc, Se, Sm, Ti, V and Zn were determined. Using Box-Whisker plot, distribution of elemental concentration with sampling site and sample size during sampling period are presented in Figure 7.

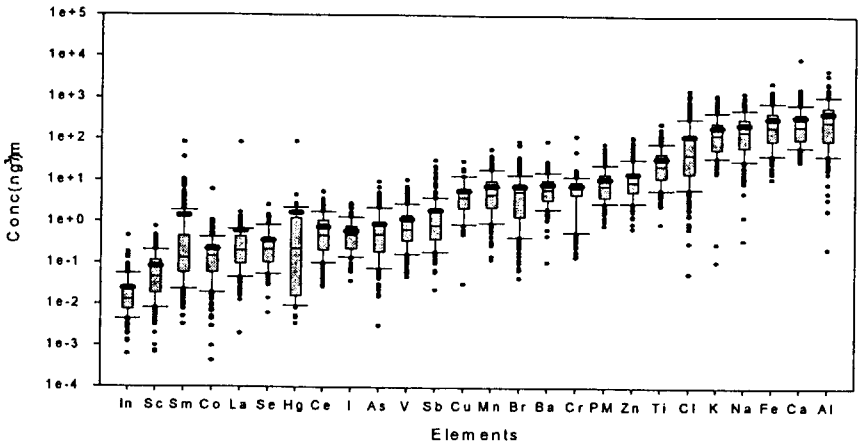
(a) Coarse fraction at Industrial area



(b) Fine fraction at Industrial area



(c) Coarse fraction in suburban area



(d) Fine fraction in suburban area

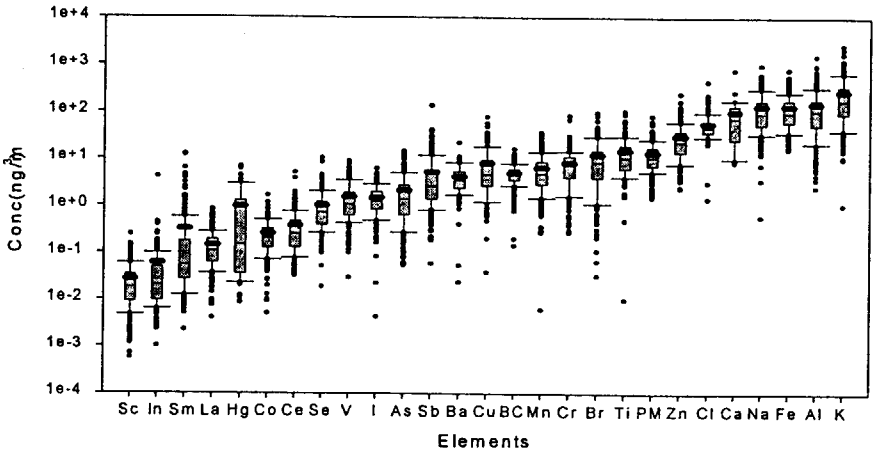
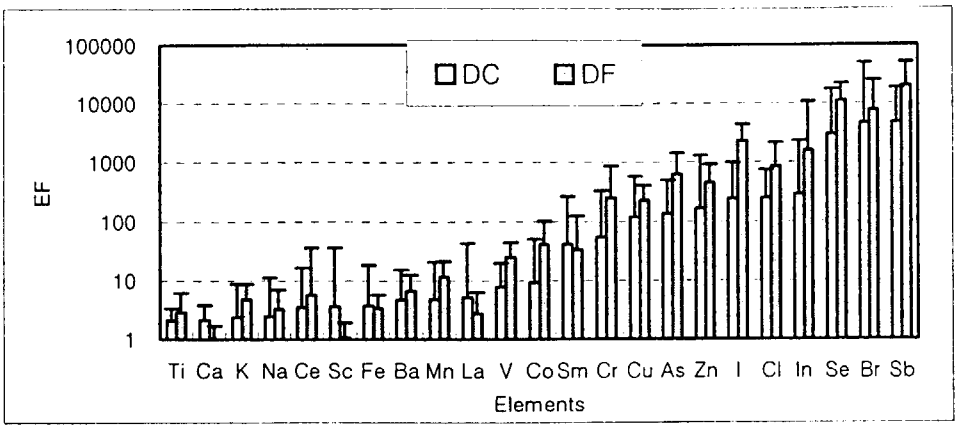


Figure 7. Box-Whisker plot of elemental concentration with sampling site during sampling period.

5) Evaluation of Enrichment factor

In order to find out the major emission sources, enrichment factors(EF) were calculated from measured elemental concentrations using Mason's crustal abundances for the sampling period. An enrichment factor is given by the double ratio of the element of interest in the sample to a reference element in the sample divided by the ratio of the same element found in a reference material(e.g. rock, soil, etc) ; $EF = (X_i/C_i)_{APM} / (X_i/C_i)_{REF}$. Sc is taken as the reference element because it is supposed to be a unique element in soil. In general, most of the crustal elements (Al, Si, Ca, K, Fe, Mn, Ti, etc.) have EF values close to unity, higher EF values of elements indicates that they are not of crustal origin. Therefore, it can be classified to two groups from the EF value of measured element ; the anthropogenic origin and the crustal origin. The average EF values calculated for particle size and sampling site are presented in Figure 8. These values were in the range 10^0 - 10^4 and the industrial area showed generally higher values than those of the suburban area. High EF values of V to Sb are observed at both areas, and they are assumed to be anthropogenic elements. The EF values of the FPM are higher than those of the CPM in anthropogenic elements with respect to the elements of crustal origin generally. These trends suggest that this result is available to investigate air pollution sources and levels for the evaluation of environmental conditions.

(a) Industrial area



(b) Suburban area

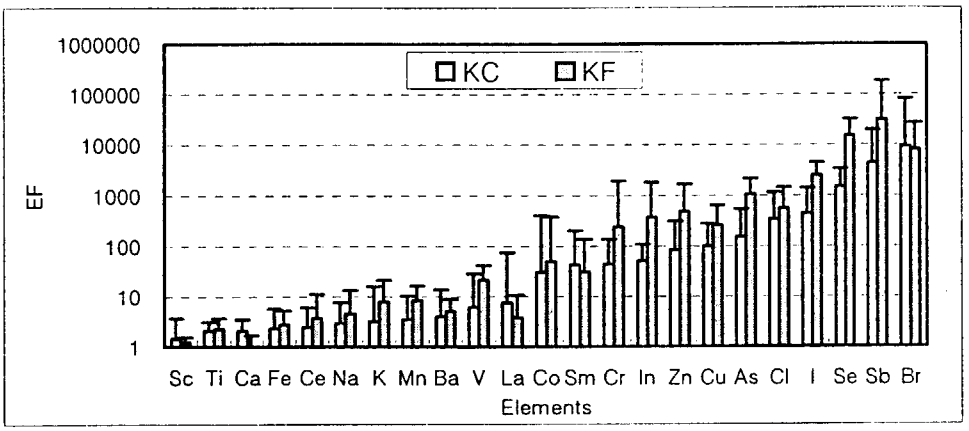
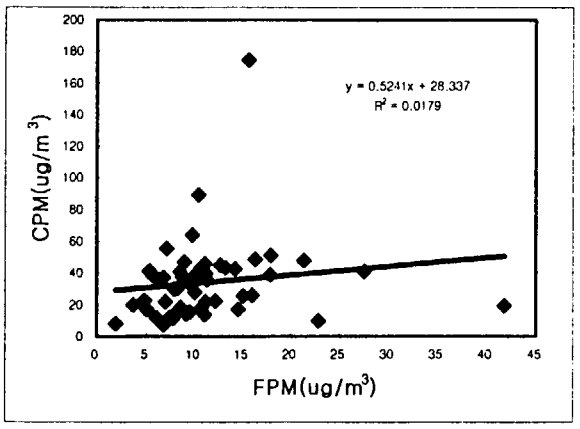


Figure 8. Enrichment factors of CPM and FPM at two sampling site.

6) Correlation analysis and Source identification

Regression analysis is carried out to identify relationship between elements, BC and APM, and to elucidate pollution sources. The correlation of CPM and FPM as well as those of elemental black carbon and fine APM with sampling site are presented in Figure 9 and 10. Factor analysis and receptor modeling for the chosen data set was performed using general statistical program(SPSS).

(a) Industrial area



(b) Suburban area

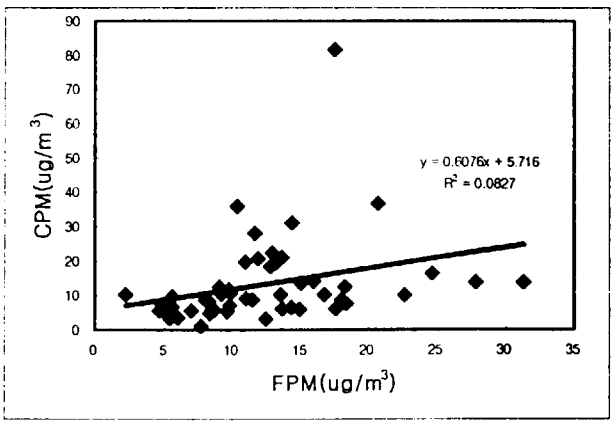
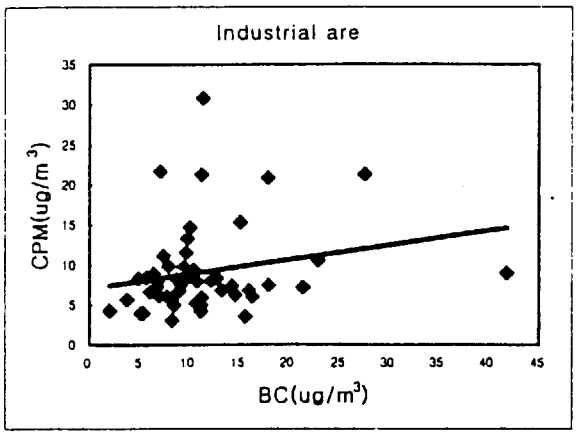


Figure 9. Correlation between CPM and FPM at sampling sites.

(a) Industrial



(b) Suburban

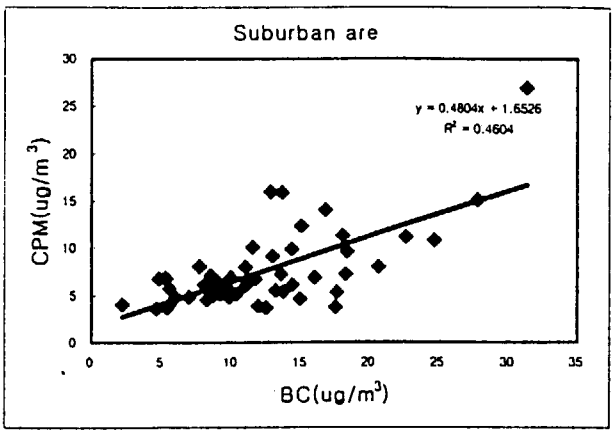


Figure 10. Correlation between elemental carbon and FPM at sampling sites.

3.2 *ko*-Standardization

1) The values of α and f parameters

Calibration of the reactor neutron spectrum parameters related to *ko*-NAA at the NAA #1 irradiation hole in HANARO research reactor was carried out by the "Cd-ratio for triple-monitor" method. Because neutrons of NAA #1 irradiation hole are well thermalized, there are only small fraction of epithermal neutrons in comparison with thermal neutrons (RCd,Au≒82). In this case, the activities induced by epithermal neutrons in (n,γ) reactions are very low and then overlapped by the statistical fluctuations of the ones induced by thermal neutrons. Therefore, it is advised to use of the Cd-ratio methods for both determination of α and f parameters, which gives the most reliable results if a proper choice of monitors is made. Table 2 shows the Cd-ratio determined by three nuclides of ^{198}Au, ^{97}Zr/^{97m}Nb & ^{95}Zr for four sets of monitor and the values of α and f parameters calculated from Eq. (6) and Eq. (7). Furthermore, Figure 11 shows the value of α parameter determined considering the experimental variation and fitting line with a good correlation coefficient of 0.98.

Table 2. Experimental values of Cd-ratio, α and f parameters for irradiation hole(NAA#1) applying triple-monitor method

Monitor	Cd-ratio				Mean ± SD*
	Set #1	Set #2	Set #3	Set #4	
^{197}Au(n,γ)^{198}Au	83.03	88.38	76.31	79.96	81.92 ± 5.11
^{96}Zr(n,γ)^{97}Zr/^{97m}Nb	11.04	11.03	10.99	10.60	10.94 ± 0.23
^{94}Zr(n,γ)^{95}Zr	533	588	496	550	542 ± 38
α	0.122	0.127	0.126	0.133	0.127 ± 0.005
f	1032	1105	938	984	1014 ± 71

* indicates standard deviation

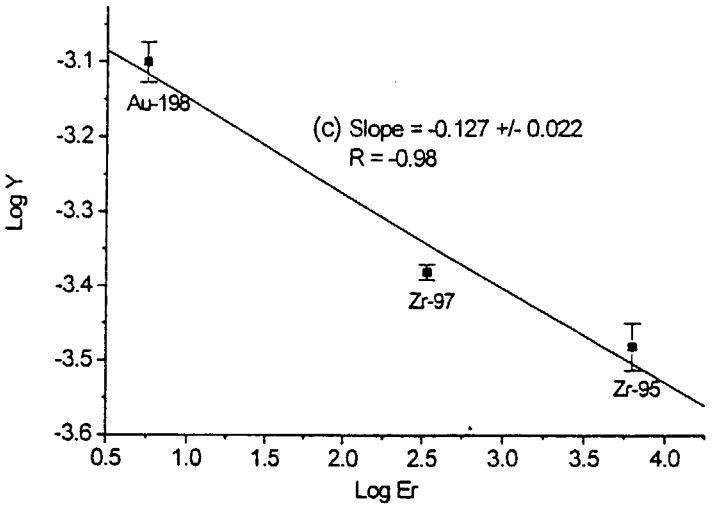


Figure 11. The result of α -determination by "Cd-ratio triple-monitor" method.
($Y = (Er,i)\text{-}\alpha$ / [(Fcd,i · Rcd,i - 1)· Qo,$i(\alpha)$· (Ge,i/Gth,i))

2) The Analytical results of SRMs

Analysis of the SRMs applying a *ko*-NAA procedure with the determined neutron spectrum parameters (i.e. α and f parameters) along with Asp,Au (specific count rate for ^{198}Au) and the detector efficiency values were carried out. Comparison between experimental results and certified values was done in terms of the ratio of the experiment to certified values. For the accuracy evaluation, Z-score values is used frequently. Z-score is defined by :

$$\text{Z-score} = Vc - Ve. / [(Uc)^2 + (Ue)^2]^{1/2} \quad (9)$$

where, Vc : certified value, Ve : experimental value,
Uc : uncertainty in the certified value
Ue : uncertainty in the experimental value

The analytical results of 4 SRMs in various elemental composition are presented in Figure 11 with Z-score plotted as Y-error bar. The analytical results can be briefly summarized as follows :

Figure 12(a) shows the analytical results of NIST SRM 1633a(Coal Fly Ash). 20 elements were determined. The ratio of certified to experiment value was in the range of 0.81(Sr)～1.20(Cs) and the highest Z-score was 1.89(Fe). Figure 12(b) shows the analytical results of NIST SRM 1577b(Bovine Liver). 9 elements were determined. The ratio of certified to experiment value was in the range of 0.83(Co)～1.14(Zn) and the highest Z-score was 1.04(Na). Figure 12(c) shows the analytical results of NIST SRM 2704(Buffalo River Sediment). 15 elements were determined excluding short-lived nuclides. The ratio of certified to experiment value was in the range of 0.90(Yb)～1.12(Cs) and the highest Z-score was 1.30(Na). Finally, Figure 12(d) shows the analytical results of NIST SRM 2709(SanJoaquin Soil). 15 elements were determined excluding short-lived nuclides. The ratio of certified to experiment value was in the range of 0.83(Zn)～1.12(Co) and the highest Z-score was 1.71(Zn).

From the certified/experiment ratio and Z-score, accuracy of the elements, Co, Zn and Fe, was lower than the other elements for the application of *ko*-NAA.

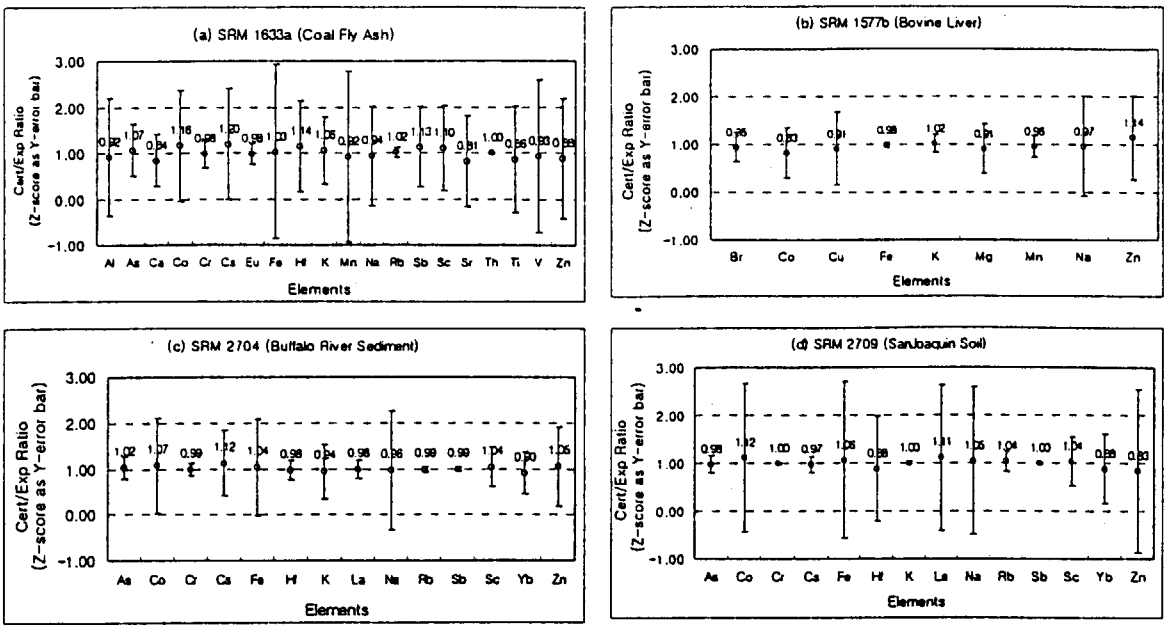


Figure 12. Comparison of experimental results with certified values for NIST SRMs.

4. CONCLUSIONS

4.1 Air pollution Study

To enhance the use of neutron activation analysis(NAA) and to assess the feasibility of the use of NAA as a routine monitoring tool for air pollution study, the concentrations of trace elements in airborne particulate matter were measured with two particle sizes at two regions.

Air pollution and relevant environmental information can be obtained from the statistical results of analytical data and neutron activation analysis using a research reactor proved to be a useful tool for environmental pollution monitoring and management as a cost-effective and benefit method.

4.2 Application of *ko*-Standardization

The assessment of nuclear characteristics to the NAA #1 irradiation hole of the HANARO research reactor for application of *ko*-method was carried out through the determination of neutron spectrum parameters, i.e. α and *f*-factor by the "Cd-ratio for triple-monitor" method using Au & Zr monitors, the calibration of a gamma-ray spectrometer using the HPGe detector(EG & G ORTEC, GEM35185), and the analysis of various SRMs applying the experimentally determined *ko*-parameters is carried out to compare the measured concentration results with the certified values. The experimental results revealed that the *ko*-NAA method is applicable on NAA #1 of HANARO research reactor.

ACKNOWLEDGEMENTS

This research has been carried out under the Nuclear R&D Program by Ministry of Science and Technology, Republic of Korea.

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1.24 Characterization of the finest and coarse airborne particulate matter in Kuala Lumpur's ambient air

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Abstract

We will report the mass concentration of the finest (PM2.5) and coarse (PM2.5-10) airborne particles in ambient air of Kuala Lumpur area collected using GENT Stack filter unit fitted with appropriate polycarbonate filters. The sampling site (Lat: 03° 10' 30" Long: 101° 43' 24.2") is about 1 km from Kuala Lumpur City Center. Elemental concentration in the samples collected once every month throughout the year 2002 has been analyzed using the currently available NAA facilities at MINT. Elements determined in the study include Al, As, Br, Co, Cr, K, Lu, Mn, Na, Sb, Sc, Ti, V, and Zn. Quality control material used to control the quality of the analytical procedures was NBS 1633a.

1.0 INTRODUCTION

Regular monitoring of the finest (PM2.5) and coarse (PM2.5-10) airborne particulate matter in Kuala Lumpur ambient air have been carried out since 1997 implementing the IAEA/RCA regional air pollution study. In this study the elemental concentration of the collected APM has been determined by various available analytical technique including INAA, reflection method (black carbon), ICP-MS and ion chromatography (anion and cation). As of our discussion in Beijing, 2001, it was decided to revisit the study with the primary aim of optimizing the applicability of currently available NAA facilities for analyzing trace amount of the collected APM on the appropriate polycarbonate air filters. Options for applying new technique and using new detector with relatively good efficiency may be identified and be used in future for such study. The K_o technique for quantification is one of the options under consideration.

In this presentation, we report the mass concentration of finest and coarse particles throughout 2002 in Kuala Lumpur collected once every month. Elemental concentration determined in both group of particle sizes, which have been determined by the currently available NAA facilities at MINT will also be presented.

2.0 EXPERIMENTAL

2.1 Sampling site

The sampling station is located in University Teknologi Malaysia compound (Lat: 03° 10' 30" Long: 101° 43' 24.2"), about 1 km northeast of the city center of Kuala Lumpur (Fig. 1). The site is a mixture of residential and commercial activities which is surrounded by various industrial estates within the Klang Valley Region (KVR). The valley comprises total area of 3,000 km^2 with total population of 3.0 million people, is the most densely populated and

industrialized area in the country. Its climate is typical of the humid tropics with mean daily temperature of between 26° and 30° C, the average precipitation is about 2,500 mm. The winds are generally light throughout the year between 0.5 m s^{-1} and 1.0 m s^{-1}.

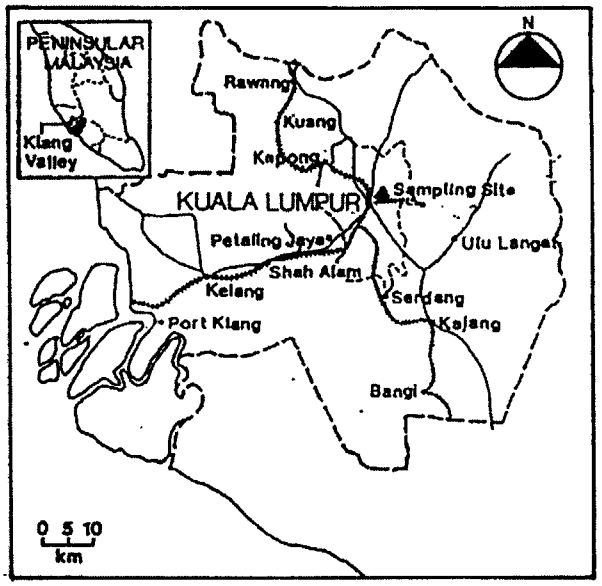

Fig. 1: The sampling site at Kuala Lumpur.

2.2 Sampling time and analytical methods

The sampling has been done once every month (24 hr. average, morning-to-morning) throughout the year 2002. A total of 12 pairs of filters were used where each pair consists of finest and coarse airborne particulate matter which was collected for the study. However due to technical problem, only the first six pairs of the filters were analyzed for the elemental content.

The mass of the air particulate matter was gravimetrically determined using microbalance. All the filters were conditioned for about 24 hours before and after sampling prior to weighing to ensure no effect of moisture in the quantification of the mass of the airborne particulate matter. The total mass was divided by the volume of air passing through the filter to provide micrograms of the atmospheric air particulate per cubic meter of air sampled.

The elemental analysis (except black carbon) has been done using INAA technique. The elements determined include Al, As, Br, Co, Cr, K, Lu, Mn, Na, Sb, Sc, Ti, V, and Zn. The NBS 1633a have been used as the quality assurance material for the analytical procedure.

3.0 DISCUSSIONS

The fine and coarse particles concentrations throughout the year 2002 are presented in Table 1. The data on PM2.5 is comparable to data collected from previous study, i.e. 33.6 ugm^{-3}, 29.2 ugm^{-3} throughout the year 2000 and 2001, respectively. The same refer to concentration of the coarse particulate matter, i.e. 21.4 ugm^{-3} and 16.6 ugm^{-3}, throughout the year 2000 and 2001, respectively. A slight haze episode was sighted during routine sampling conducted in March, April and October, however there were no significant increment of the particulate

matter concentration detected except on October's sample where the PM2.5 concentration significantly increase to 46 ugm^{-3} whilst the coarse particulate matter concentration remain comparable to other data.

The average elemental concentration in the collected air particulate matter is presented in Table 2. In general the results of analysis are also comparable to the previous analytical results from the previous study. It is always a trend that the expected elements originated from soil such as Al and Eu are always at higher concentration in coarse particulate matter fractions.

The results on the analysis of quality assurance material NBS 1633a are presented in Table 3. The material of different amount (mostly determined via short-lived radioisotope) was analyzed to test the applicability of the material when used at smaller amount than recommended by the manufacturer, i.e. 100mg. It was found that the concentration of each element was lower when the amount of the material tested is in the range of 20－25 mg. The difference might be influenced by the inhomogeneity of the material when tested in smaller amount.

4.0 CONCLUSION

Fourteen elements were quantitatively determined using the NAA technique in the segregated fine and coarse particle fractions using the NAA technique. It is envisaged that by applying the K_0 method combined with more efficient detector one may improve number of elements analyzed by this technique.

Table 1: Mass Concentration of APM at sampling site in Kuala Lumpur for 2002

Month	Vol. of air (m^3)	Flow rate (l/min)	Particulate	Weight (ug)	Conc. (ug/m^3)
January	7	15	Fine	177.00 ± 2.10	25.29 ± 0.30
			Coarse	154.67 ± 2.42	22.10 ± 0.35
February	7	15	Fine	206.33 ± 3.44	29.48 ± 0.49
			Coarse	169.00 ± 1.10	24.14 ± 0.16
March	7	15	Fine	277.67 ± 2.66	39.67 ± 0.38
			Coarse	122.00 ± 3.10	17.43 ± 0.44
April	7	15	Fine	216.67 ± 2.73	30.95 ± 0.39
			Coarse	120.33 ± 2.94	17.19 ± 0.42
May	8	15	Fine	149.00 ± 2.76	18.63 ± 0.35
			Coarse	95.67 ± 8.16	11.96 ± 0.10
June	7	15	Fine	255.33 ± 2.07	36.48 ± 0.29
			Coarse	138.00 ± 4.00	19.71 ± 0.57
July	7	15	Fine	159.33 ± 3.50	22.76 ± 0.50
			Coarse	137.00 ± 2.10	19.57 ± 0.30
August	7	15	Fine	226.00 ± 1.51	32.29 ± 0.21
			Coarse	150.33 ± 1.97	21.48 ± 0.28
September	7	15	Fine	286.67 ± 2.73	40.95 ± 0.39
			Coarse	173.00 ± 2.10	24.71 ± 0.30
October	7	15	Fine	322.67 ± 3.01	46.10 ± 0.43
			Coarse	136.67 ± 3.50	19.52 ± 0.50
November	7	15	Fine	169.33 ± 8.62	24.19 ± 0.12
			Coarse	94.67 ± 3.95	13.52 ± 0.56
December	9	15	Fine	250.33 ± 3.88	27.81 ± 0.43
			Coarse	113.67 ± 2.34	12.63 ± 2.60
Average			Fine	224.69 ± 55.26	31.22 ± 8.23
			Coarse	133.75 ± 25.64	18.66 ± 4.27

Table 2: Elemental concentration in first six pairs of filters analyzed by NAA.

Element	Fine (ng/m^3)		Coarse (ng/m^3)	
	Range	Average ± SD	Range	Average ± SD
Ti	0 - 704	117 ± 287	477 - 877	611 ± 169
Al*	0.56 - 1.79	1.23 ± 0.14	3.29 - 13.70	6.83 ± 3.48
V	25.50 - 61.57	40.3 ± 13.09	10.29 - 18.29	17.72 ± 3.25
Mn	60.29 - 673	244 ± 253	71.38 - 176	121 ± 42.44
Na	93.29 - 749	277 ± 259	107 - 1037	385 ± 361
K	0 - 1000	432 ± 401	0 - 720	196 ± 289
Br	7.88 - 20.14	12.29 ± 4.42	2.00 - 4.75	3.62 ± 2.54
Lu	0 - 0.29	0.14 ± 0.12	0 - 0.29	0.10 ± 0.11
As	0 - 4.38	1.90 ± 2.10	0 - 7.00	2.11 ± 3.39
Sb	0.57 - 7.71	1.85 ± 4.06	0.71 - 2.43	1.23 ± 1.02
Cr	3.14 - 11.86	2.58 ± 7.37	0 - 1.25	0.21
Cs	2.14 - 3.14	2.72 ± 0.40	0.29 - 1.75	1.17 ± 0.60
Zn	53.71 - 76.43	36.82 ± 47.30	0 - 98.86	38.59 ± 44.49
Co	2 - 5.43	3.12 ± 1.22	0.75 - 4.43	2.29 ± 1.34
Eu	0.43 – 1.29	0.72 ± 0.29	1.29 – 1.86	1.42 ± 0.23
Black Carbon*	3.55 – 8.31	6.87 ± 1.20	1.50 – 2.03	1.67 ± 0.14

* expressed in ug/m^3

Table 3: Analytical results of analysis of quality assurance material NMS 1633a

Elements	Analytical Results		Certified Values (ug/g)
	60 – 100 mg	20 -25 mg	
Dy	14.6 ± 5	9.7 ± 4.3	15.6 ± 1.2
Ti	6850 ± 1000	6687 ± 1177	8.230 ± 3.90
V	289 ± 20	232 ± 21	297 ± 16
Al	13.5 (%) ± 1.4	12.5 (%) ± 0.6	14.3 ± 1.0 (%)
Mn	181 ± 15	148 ± 14	179 ± 8
Cl	< 100	< 100	< 69
Na	1760 ± 200	1663 ± 229	1700 ± 100
K	1.85 (%) ± 0.3	1.68(%) ± 0.3	1.88 ± 0.06 (%)
Sc	14.8 ± 2		17.0 ± 1.5
U	10.9 ± 2		10.2 ± 0.1
Sb	6.4 ± 0.6		6.8 ± 0.4
La	74 ± 3		84 ± 8
Cr	159 ± 8		196 ± 6
Zn	227 ± 77		220 ± 10
Co	40 ± 3		46
Eu	3.2 ± 0.1		4.0

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2002 Workshop for Utilization of Research Reactors BATAN, Jakarta, Indonesia, January 13-17, 2003

1.25 Airborne Particulate Matter Collection and Analysis by XRF

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ABSTRACT

The Philippine Nuclear Research Institute (PNRI) continues to pursue its air pollution research in support of the implementation of the 1999 Clean Air Act. The primary tool for analysis is X-Ray Fluorescence spectrometry (XRF) since the PRR-I is still on extended shut down. Following the workplan approved during the 1991 Workshop on Utilization of Research Reactors, the PNRI collected airborne particulate matter using the Gent sampler. The sampling site selected for the program was Poveda Learning Center, located beside a major highway, the Epifanio delos Santos Avenue (EDSA) where the principal source of pollution is vehicular emissions. Samples collected up to August were analyzed by XRF using three sets of analytical parameters to allow optimized analysis of a wider range of elements including Na and Pb.

Although the PNRI has no operating reactor, it has personnel who have trained in NAA but are unable to apply the technique. As mentioned in the 2001 Workshop, the PNRI is considering several options to resume reactor-related activities. Thus, it is necessary to ensure continuing availability of expertise in NAA in the PNRI. It looks forward to collaborating with other Institutes through the FNCA program for the analysis of samples by NAA and using reactor parameters from collaborating Institute, to obtain experience in the use of Ko. This would also allow validation of XRF data obtained for these samples. In return it can analyze samples for collaborating institutions to generate data on Pb and S, which are important for pollutant source apportionment.

INTRODUCTION

In Metro Manila the importance and urgency of addressing the air pollution problem is recognized. A new Clean Air Act was passed in 1999 and enacted at the end of 2000 with support from the Asian Development Bank. Implementation of the Clean Air Act is being made in partnership with different government institutions and the private sector. Following its mandate to promote nuclear technology application to meet local needs, the Philippine Nuclear Research Institute implements the project "Metro Manila Air Pollutant Characterization and Source Identification". The project has made tangible contributions to the national programme on air quality in terms of data generation, analytical services and capability building among collaborating end-user institutions.

PROJECT OBJECTIVES

The project is being undertaken in support of the national programme on air quality. It aims to apply nuclear and related analytical techniques to generate data useful for air quality management These include:

- $PM_{2.5}$ and PM_{10} ambient levelsParticulate elemental compositionPollutant source contributions

SAMPLING LOCATION

Since 1996, the project has operated air monitoring stations at 6 sites in Metro Manila. Table 1 gives the location and inclusive dates of sample collection for the different stations. With the exception of La Mesa Dam, all the sampling sites are close to major thoroughfares. However, care was taken to located the sampler as far away from the road as possible. Figure 1 shows the location of the PNRI stations as well as the regular monitoring stations of the Environmental Management Bureau, where TSP is measured.

Table 1. Philippine Nuclear Research Institute sampling stations equipped with a Gent sampler

Sampling site	Description	Inclusive dates of sampling
Ateneo de Manila University	Institutional/ residential area	June 1996 – October 1997, October 1998 to date
Poveda Learning Center, EDSA	Secondary school at a commercial district, beside a major thoroughfare	October 2001 to date
University of Sto. Tomas	Commercial/ residential area	March to September 1999, June 2000 to September 2002
Manila Waters Services treatment facility	Forested watershed	November 1996 to February 1998
Vista Verde Homes	Residential area close to industrial zone	February – July 1999

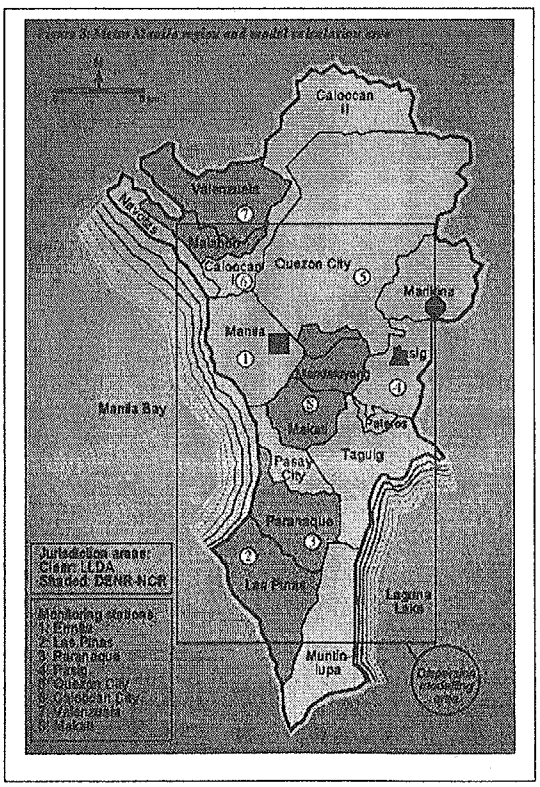


Figure 1. Location of PNRI air monitoring stations.

METHODOLOGY

Air sample collection

Air sample collection is done using the Gent dichotomous sampler. Samples are collected in two fractions: fine, with mean aerodynamic diameter less than 2.5 microns and coarse, with mean aerodynamic diameter in the range 2.5 – 10 microns. Due to filter clogging, the sampler is set to go on for 2 hours and then off for 4 hours giving a total of 8 hours sampling over a 24 hour period.

Particulate mass determination

Particulate mass is determined from the difference of the equilibrated weight of the filter before and after loading. Equilibration is done in a desiccator kept at a relative humidity of 35%. Weighing is done with a Mettler MT5 microbalance. Equilibration and weighing are repeated until a constant weight is obtained. Equilibration time in-between weighings is 24 hrs.

X-ray fluorescence spectrometry

Air filter analysis is done using a KEVEX 771 Secondary target X-ray fluorescence spectrometer, with a Rh X-ray source. Analysis is done in secondary target mode. The secondary target materials (Gd, Ag, Zr, Ge, Ti and Al) are mounted on a turret and can be selected automatically. The air filter sample is placed in an EPA-type sample holder, a 4 x 4 cm^2 plastic cassette and mounted on a sample position on the 16-position sample wheel.

The IAEA software, AXIL is used for spectrum processing and subsequent quantitation by the method of elemental sensitivities.

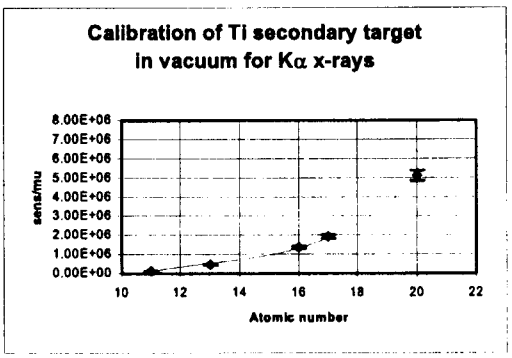
Instrument calibration is done using the following standards:

Ti secondary target: NaCl, Al, CaF2, ScF3
Ge secondary target: NaCl, CaF2, ScF3, V, Fe, Ni, CuSx, ZnTe, GaAs, Se, RbI, MoO3, CdSe, Sn, BaF2, PrF3, SmF3, GdF3, TbF3, LuF3, Pt
Ag secondary target: CaF2, ScF3, V, Fe, Ni, CuSx, ZnTe, GaAs, Se, RbI, MoO3, CdSe, Sn BaF2, PrF3, SmF3, GdF3, TbF3, SmF3, LuF3, Pt, Pb, ThF4

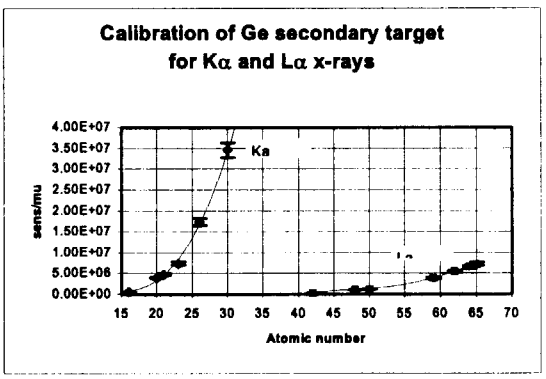
Current analytical parameters presently used for aerosol analysis at the PNRI are given in Table 2. Counting time with Ti secondary target has been increased to improve detection of light elements. Figure 2 shows typical calibration curves for Ti (vacuum), Ge and Zr secondary targets. The advantage of using multiple secondary targets for enhanced detection of a wider range of elements in air particulate matter is seen.

Table 2 Analytical paramenters for analysis of air filters using the KEVEX 771

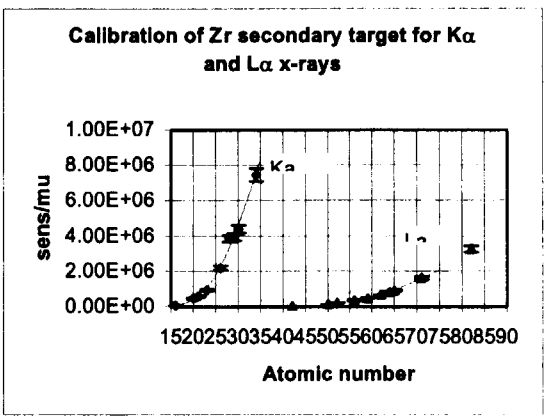
Secondary target	Atmosphere	kV setting	mA setting	Counting time (sec)
Ag	Air	45	3	1000
Zr	Air	30	3	1000
Ge	Air	25	3	1000
Ti	Vacuum	10	3	2000



(a)



(b)



(c)

Figure 2. Calibration curves for Ti in vacuum (a), Ge (b) and Zr(c) secondary targets using thin film standards

RESULTS AND DISCUSSIONS

Particulate mass concentration

Figure 3 summarizes the particulate mass concentration obtained for 2002 at the Poveda Learning Center The station is at an area exposed to vehicular emission, although the sampler is at about 100 m form the road side. It is good to note that neither the Philippine short term standard for PM10 (150ug/cu m) nor the USEPA PM2.5 standard of 60 ug/cu m are exceeded The mean for PM2.5 and PM2.5-PM10 are 16.5 and 21.0 respectively. The mean for PM2.5 is slightly higher than the annual long term standard of the USEPA (15 ug/cu m.).

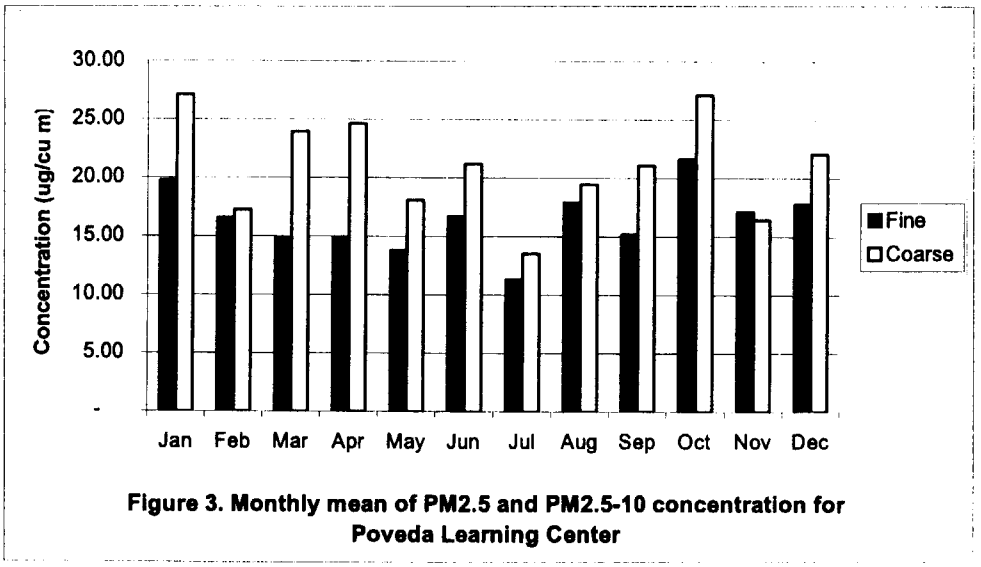


Figure 3. Monthly mean of PM2.5 and PM2.5-10 concentration for Poveda Learning Center

XRF spectrometry

The calibration curves obtained for the KEVEX 771 show that the use of several secondary targets in XRF spectrometry allows optimized detection of a wide range of elements in airborne particulate matter. Typically 13-20 elements can be measured in air particulate matter including light elements Na and Mg. Tables 3 and 4 give concentration of different elements measured in samples collected from the Poveda monitoring station.

Table 3. Elements measured in PM2.5

Element	Mean (ng/cu m)	Range (ng/cu m)
Al	646	348-895
S	700	270-1465
Cl	30.4	15.4-44.7
K	109	59-247
Ca	105	51.7-155
V	17.2	4-28
Mn	2.79	1.4-5.6
Fe	103	1.4-300
Ni	3.77	1.39-8.37
Cu	4.19	1.4-6.98
Zn	77	21-296
Br	5.92	1.4-12.6
Pb	36	15.3-76.8

Table 4. Elements measured in PM2.5-10 fraction

Element	Mean (ng/cu m)	Range (ng/cu m)
Na	479	239-817
Mg	702	160-3974
Si	856	176-1204
P	76.8	48.9-132
S	454	251-853
Cl	618	138-1273
K	154	94-251
Ca	792	478-1314
Ti	54.2	14.0-251
V	11.7	3.59-23.7
Mn	13.3	5.58-22.3
Fe	479	318-749
Ni	9.5	6.98-12.6
Cu	7.22	3.59-12.6
Zn	65	23-127
Se	5.7	4.2-9.0
Br	23.7	20.9-25.1
Pb	42.5	33.5-81.0

CONCLUSIONS

XRF spectrometry with variable secondary target excitation is a useful tool for particulate characterization leading to pollutant source characterization. The PNRI has made tangible contribution to the national effort to improve air quality by generating data and providing analytical services to local groups. It is participating in the FNCA program, in order to maintain continuing expertise in NAA even without a reactor, using the opportunities for exchange made possible by the FNCA and other regional programs such as the RCA/IAEA.

Although the PNRI has no operating reactor, it has personnel who have trained in NAA but are unable to apply the technique. Maintaining this expertise and continuing reactor-related activities would improve the chances of getting funding for reactor repair or the construction of a new one. It looks forward to collaborating with other Institutes through the FNCA program for the analysis of samples by NAA and using reactor parameters from collaborating Institute, to obtain experience in the use of Ko. This would also allow validation of XRF data obtained for these samples.

The 2002 Workshop on the Utilization of Research Reactors
Serpong, Indonesia
January 13-17, 2003

1.26 Study of Urban Air Pollution in Thailand

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Abstract

The Office of Atoms for Peace has conducted a monitoring study of urban air pollution in Thailand for years. The primary objective of the project was to support the use of nuclear-related techniques for research and monitoring studies on air pollution. The databases obtained have been analyzed and interpreted by statistical methods including source identification using receptor model. This paper reports the work of 2002 at a heavy traffic area in Bangkok. A Gent sampler was set at the curbside of a major road in Bangkok to collect fine and coarse particles routinely on a weekday for 24 hours, once a week. The filter samples were analyzed for elemental concentrations by use of instrumental neutron activation analysis. Black carbon was separately determined by means of the reflectance measurement of the filter sample. In the report, the methodologies and the results of analyses of fine and coarse particles on filters collected in 2002 are presented. The study of the applicability of certified reference material was done by analyses of two standard reference materials provided by JAERI, i.e., NIST 1632c and NIES No.8. The comparisons of the measured and certified values are also given in the paper.

Key words : Air pollution in Thailand; INAA; Particulate matter

1. Introduction

It is generally known that air pollution problem commonly occur in the big cities by one way or another. In Thailand, the situation is most severe in Bangkok metropolis where there are huge population, very dense traffic and continuity of the city development. Concerning to the pollutants in the atmosphere, Thailand regulate and monitor for the major pollutants, i.e., SO_2, NO_2 and CO, O_3, Pb and suspended particulate matter. It has been reported that the lead content in the atmosphere decreases satisfactory due to the regulation of using unleaded gasoline since 1991. The levels of those pollutant gases, on the other hand, still need to be monitored carefully. O_3, in particular, has the increasing tendency. At present, however, the most important pollutant that causes the most serious problem in Thailand, especially in Bangkok, is the atmospheric particulate matter. In recent years, greater attention has been to characterize the nature of particulate matter (PM) since it has the adverse impact on human health and also on the environment. Particles with aerodynamic diameter less than 10 μm, termed PM_{10}, can be transported over large distances and can enter human respiratory system. Particles with aerodynamic diameter less than 2.5 μm, termed as $PM_{2.5}$, are most effective in scattering light and are the major cause of visibility impairment. Regarding the fact that PM_{10} and $PM_{2.5}$ or coarse- and fine-mode PM, in addition to falling into different size ranges, differ in formation mechanisms, chemical compositions, sources, and exposure relationships, the studies on particular modes will have result on the effectiveness of pollution control and air quality management.

Beginning in 1994, the Environmental Research Group, Office of Atoms for Peace (OAP) has conducted the study on air pollution. The Gent stacked filter unit (Gent SFU), provided by International Atomic Energy Agency (IAEA), has been used for the project. Gent SFU is the standard air sampler[1] that collects simultaneously the coarse- and fine-mode PM, i.e., $PM_{2.2\text{-}10}$ and $PM_{2.2}$. The study areas were mainly at urban sites and sometimes at the suburb of Bangkok. The samples were then measured for mass and were analyzed for elemental composition. A receptor model has been applied to some set of data to investigate for airborne particulate sources. In this paper, the work of 2002 at a business area in Bangkok is reported.

2. Methodologies

2.1. Aerosol sampling

Started from January through December 2002, the sampling site was at a business area in Bangkok where is located at latitude 13° 45′ N and longitude 100° 29′ E. A Gent sampler was set at the curbside of a major road in the city center. Figure 1 shows location of the selected area for the study. The map in Figure 2 gives a closer view of the sampling site. Coarse and fine particles, i.e., $PM_{2.2\text{-}10}$ and $PM_{2.2}$, were collected on two sequential 47 mm diameter Nuclepore polycarbonate filters (8 μm and 0.4 μm pore size). The sampling was operated at flow rate about 16 lpm for 24 hours basis on a weekday, normally once a week.

2.2. Analytical technique

The air filter samples were first measured for mass concentrations using a Microbalance. The Smoke Stain Reflectometer (Model 43D of EEL) was used for the determination of elemental carbon in the samples. The filter samples were then analyzed for elemental concentrations by Instrumental Neutron Activation Analysis (INAA).

For INAA, the air filter samples including standards and filter blanks were packed in polyethylene vials and irradiated in 1.2 MW TRIGA MARK III Research Reactor at the thermal neutron flux in the order of 10^{12} n/cm^2.sec. All irradiated samples were then transferred to new vials and counted for gamma-ray activities. Two different irradiations and four gamma ray counts after appropriate decay times were conducted in order to determine short-, medium-, and long-lived radionuclides. Up to 27 elemental concentrations, their uncertainties and detection limits were obtained.

2.3. The study of the applicability of certified reference material

Following the plan set in WURR 2001, the analyses of certified reference materials (CRMs) provided by JAERI, i.e., NIES No. 8 and NIST 1632c were implemented. Sets of different amounts of such CRMs, each of which three samples were weighted and analyzed for elemental compositions by INAA. Four sets consisted of 1, 5, 10, and 15 mg were used for determination of short-lived radionuclides while three sets of 5, 20, and 50 mg were used for determination of those medium- and long-lived radionuclides. For this work, comparative method using three standard reference materials, i.e., ECH (coal fly ash), PACS (marine sediment), and SD-M-2/TM (marine sediment) were performed.

3. Results and discussion

3.1. Particulate composition data

Totally 50 pairs of fine and coarse particulate matter were collected and analyzed. The results of mass concentrations for both fine and coarse particles are shown in Figure 3. The

annual average and the 24-hour maximum values of those mass concentrations are compared with the ambient air quality standard of Thailand[2] as shown in Table 1. The black carbon content of the fine and coarse fraction filters is given in Figure 4. Figures 5 and 6 display the box and whisker plots to indicate the median, the 10^{th}, 25^{th}, 75^{th}, and 90^{th} percentile of the elemental concentrations for the fine and coarse fractions, respectively.

The results indicate the rather high level of particulate mass at the sampling site in the business area of Bangkok City. Particularly, both the annual average and the maximum value of PM_{10} derived, as summarized in Table 1, are higher than the ambient air quality standard of Thailand limited by Pollution Control Department. Nevertheless, it can be observed from Figures 3 and 4 that the fine particulate mass and black carbon have the declining trends in rainy season. A few high peaks of both fine particulate mass and black carbon content occur correspondingly. This information can indicate the vehicle combustion source. Elemental data as shown in Figures 5 and 6 are also important for an investigation of pollution sources of fine and coarse particulate matter in atmosphere. In our preliminary study on elemental concentrations of airborne particulate matter, it indicates that the main sources are most likely city dust, emissions from vehicle combustion and refuse incineration.

3.2. Data of select elements in CRMs

The results of the elemental analyses of NIES No. 8 and NIST 1632c are summarized in Tables 2 and 3. Over 20 elements in both materials which have certified or reference values are listed for comparison. In an overview for all elements analyzed, there is not much significant difference in data found for different sets of weight of the samples. However, the set of 1-mg samples analyzed for those short-lived nuclides result in rather high values with higher standard deviations than the other sets. This may imply that too small amount of the material should not be used nor give the reliable result. On the other hand, it may depend, more or less, on the facility and method used and need individual lab's consideration on how to use those CRMs effectively.

4. Conclusion

During 2002, sampling and analysis of both fine and coarse particles in Bangkok metropolitan area at the selected site were conducted and yield a database of chemical information. It shows the trend of atmospheric particulate as a major problem of urban air pollution. After complete the data analysis and validation, this database can be used to provide an estimate of major source types contributing to airborne particulate matter. Therefore, the next step to accomplish this project is to utilize the database with an advance receptor model for source identification and apportionment which is necessary for the development of effective and efficient urban air quality management.

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2. Ambient air standards of Thailand (1995). Pollution Control Department, Ministry of Natural Resource and Environment.

Table 1 Mass concentrations of fine and coarse particles and air quality standard of Thailand[2] for PM_{10}

	Mass concentration in $\mu g/m^3$			
	Fine : $PM_{2.2}$	Coarse : $PM_{2.2-10}$	PM_{10} (this work)	PM_{10} (standard)
Annual average	20.8	35.8	56.6	50
24 hour (max)	60.7	96.2	156.9	120

Table 2 Comparative data of measured values and certified/reference values for selected elements in NIST 1632c

Element	Certified/Reference Values (μg/g)	Measured values (μg/g) of samples			
		1 mg	5 mg	10mg	15 mg
Al	9150 ± 137	10433 ± 1071	9139 ± 205	9196 ± 96	8901 ± 235
Ca	1450 ± 300	1657 ± 650	1541 ± 135	1547 ± 296	1571 ± 221
Cl	1139 ± 41	1148 ± 31	1053 ± 41	1143 ± 167	925 ± 30
Mg	384 ± 32	490 ± 128	380 ± 42	376 ± 85	334 ± 25
Mn	13.0 ± 0.5	17.1 ± 3.1	13.7 ± 0.5	12.2 ± 0.1	12.7 ± 0.8
Na	299 ± 5	269 ± 23	283 ± 9	278 ± 14	279 ± 12
Ti	517 ± 32	636 ± 102	630 ± 68	540 ± 47	490 ± 9
V	23.7 ± 0.5	22.1 ± 3.8	22.3 ± 1.0	21.8 ± 1.1	20.9 ± 1.1
			5 mg	20 mg	50 mg
As	6.18 ± 0.27		6.18 ± 0.22	6.68 ± 0.33	7.17 ± 0.07
Br	18.7 ± 0.4		17.6 ± 1.41	18.9 ± 0.46	19.4 ± 0.25
Ce	11.9 ± 0.2		11.5 ± 0.61	11.8 ± 0.64	12.3 ± 0.18
Co	3.48 ± 0.20		3.35 ± 0.10	3.41 ± 0.07	3.50 ± 0.09
Cr	13.7 ± 0.2		13.4 ± 1.14	13.7 ± 0.72	13.3 ± 0.46
Cs	0.59 ± 0.01		0.57 ± 0.06	0.70 ± 0.06	0.67 ± 0.06
Eu	0.124 ± 0.003		0.122 ± 0.027	0.084 ± 0.007	0.083 ± 0.003
Fe	7350 ± 110		7327 ± 78	7310 ± 125	7536 ± 134
Hf	0.58 ± 0.01		0.57 ± 0.19	0.65 ± 0.12	0.60 ± 0.05
K	1100 ± 33		1050 ± 33.7	1154 ± 12.6	1159 ± 12.4
Na	299 ± 5		263 ± 12	295 ± 3	316 ± 2
Rb	7.52 ± 0.33		7.68 ± 1.18	6.98 ± 1.67	7.56 ± 0.70
Sb	0.46 ± 0.03		0.43 ± 0.03	0.47 ± 0.01	0.46 ± 0.01
Sc	2.90 ± 0.04		2.63 ± 0.11	2.74 ± 0.01	2.82 ± 0.05
Sm	1.08 ± 0.03		0.94 ± 0.01	1.04 ± 0.01	1.12 ± 0.01
Th	1.40 ± 0.03		1.38 ± 0.09	1.34 ± 0.04	1.41 ± 0.04
Zn	12.1 ± 1.3		11.8 ± 0.9	13.8 ± 4.96	12.7 ± 2.33

Table 3 Comparative data of measured values and certified/reference values for selected elements in NIES No.8

Element	Certified/Reference Values (μg/g)	Measured values (μg/g) of samples			
		1 mg	5 mg	10mg	15 mg
Al	3300 ± 200	3759 ± 325	3721 ± 28	3798 ± 84	3822 ± 101
Ca	5300 ± 200	5880 ± 711	5522 ± 209	5554 ± 335	5800 ± 212
Mg	1010 ± 50	970 ± 210	878 ± 51	946 ± 137	937 ± 31
Na	1920 ± 80	2020 ± 417	1925 ± 7	2076 ± 305	1992 ± 94
V	17 ± 2	15.9 ± 1.6	15.6 ± 1.1	16.3 ± 1.9	15.9 ± 0.8
			5 mg	20 mg	50 mg
As	2.6 ± 0.2		2.76 ± 0.28	2.86 ± 0.11	2.81 ± 0.10
Br	56		56.1 ± 2.1	60.9 ± 3.0	61.7 ± 0.8
Ce	3.1		3.38 ± 0.17	3.62 ± 0.35	3.75 ± 0.21
Co	3.3 ± 0.3		3.62 ± 0.60	3.31 ± 0.38	3.28 ± 0.09
Cr	25.5 ± 1.5		30.5 ± 6.1	27.6 ± 1.7	26.5 ± 0.7
Cs	0.24		0.49 ± 0.05	0.32 ± 0.15	0.31 ± 0.07
Eu	0.05		0.11 ± 0.01	0.08 ± 0.01	0.06 ± 0.003
K	1150 ± 80		1351 ± 171	1187 ± 57	1245 ± 88
La	1.2		1.16 ± 0.09	1.28 ± 0.01	1.29 ± 0.11
Mo	6.4		7.55 ± 1.04	7.29 ± 0.39	6.57 ± 0.67
Na	1920 ± 80		2018 ± 45	2032 ± 153	2100 ± 89
Rb	4.6		4.66 ± 0.38	5.06 ± 0.49	4.96 ± 0.08
Sb	6.0 ± 0.4		6.38 ± 0.22	6.51 ± 0.30	6.58 ± 0.09
Sc	0.55		0.58 ± 0.03	0.58 ± 0.03	0.58 ± 0.01
Sm	0.2		0.21 ± 0.03	0.22 ± 0.004	0.21 ± 0.01
Th	0.35		0.42 ± 0.06	0.40 ± 0.02	0.36 ± 0.01
Zn	1040 ± 50		881 ± 163	890 ± 183	936 ± 79

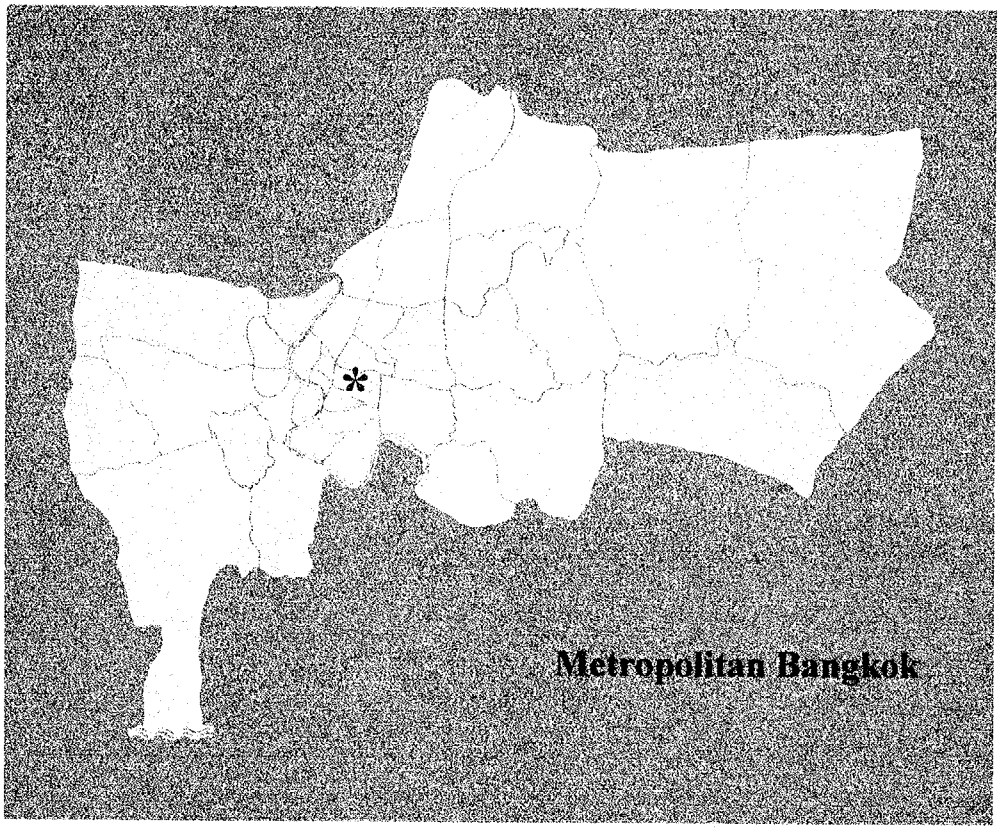

Figure 1 Location of sampling site at Pathumwan district, Bangkok, Thailand.

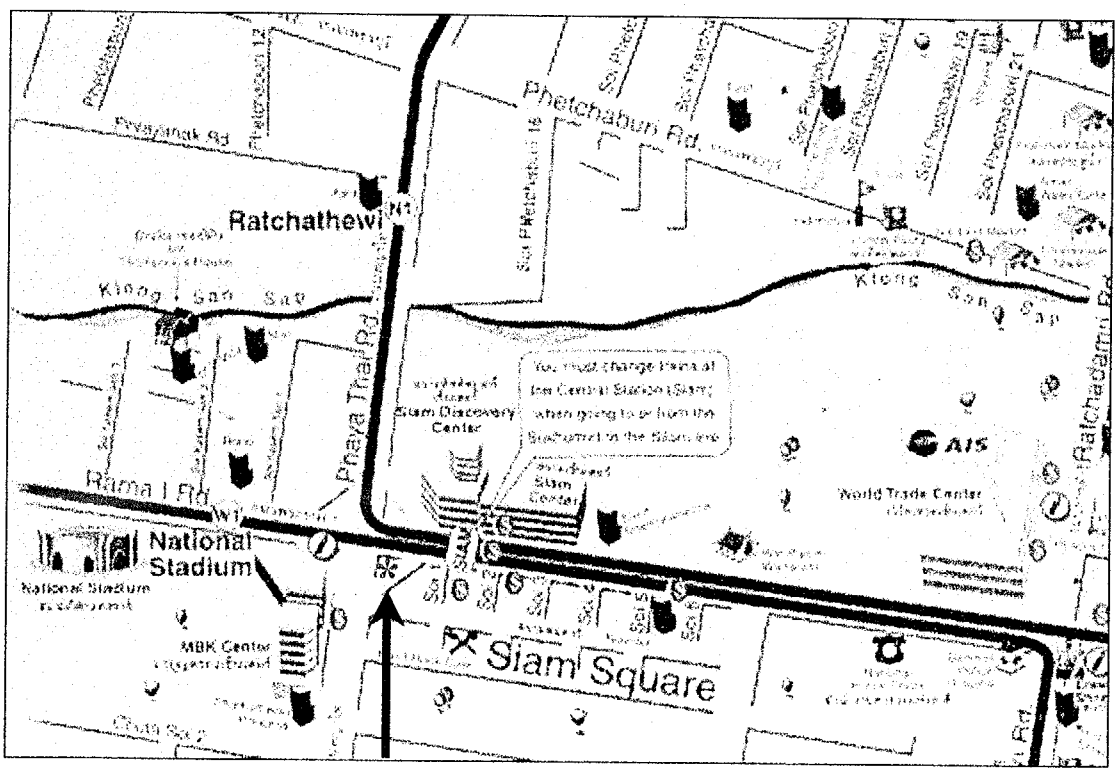

Figure 2 Map of the sampling site at Pathumwan, Bangkok. (* marks for the sampling site)

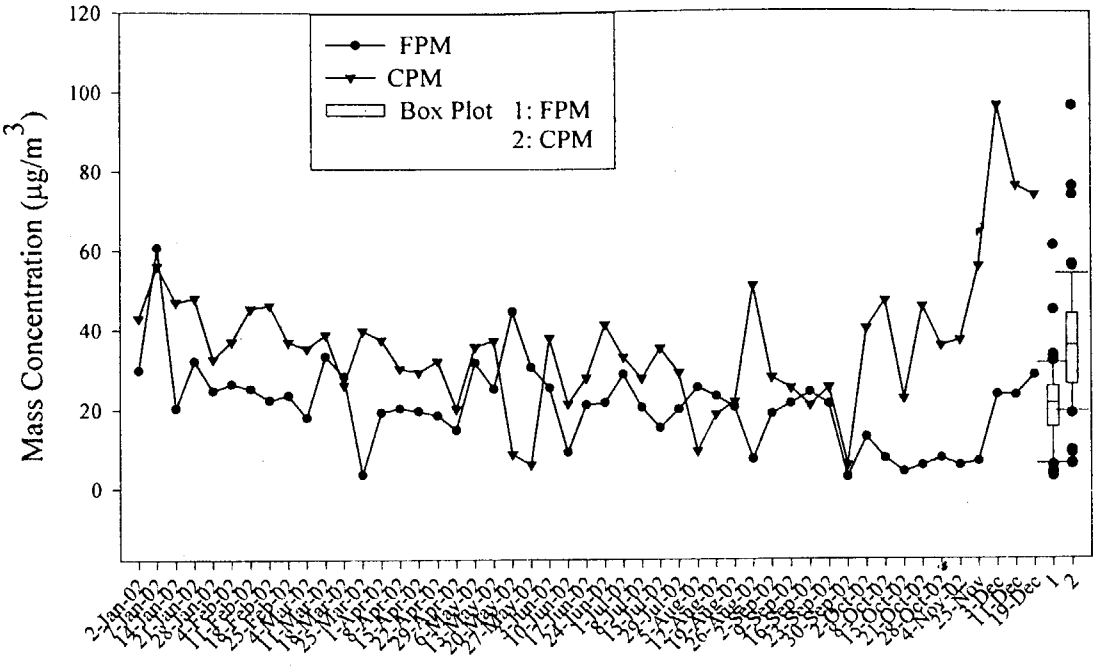

Figure 3 Time series and box-whisker plots of the fine and coarse particle fraction mass concentrations.

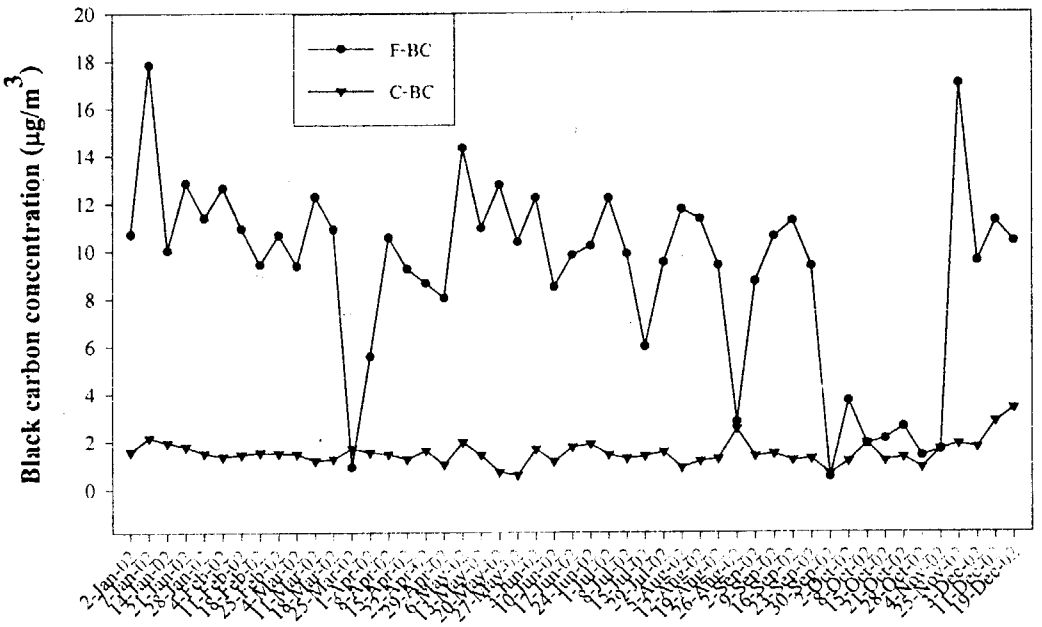

Figure 4 Time series plots of the black carbon concentrations in the fine and coarse particle samples.

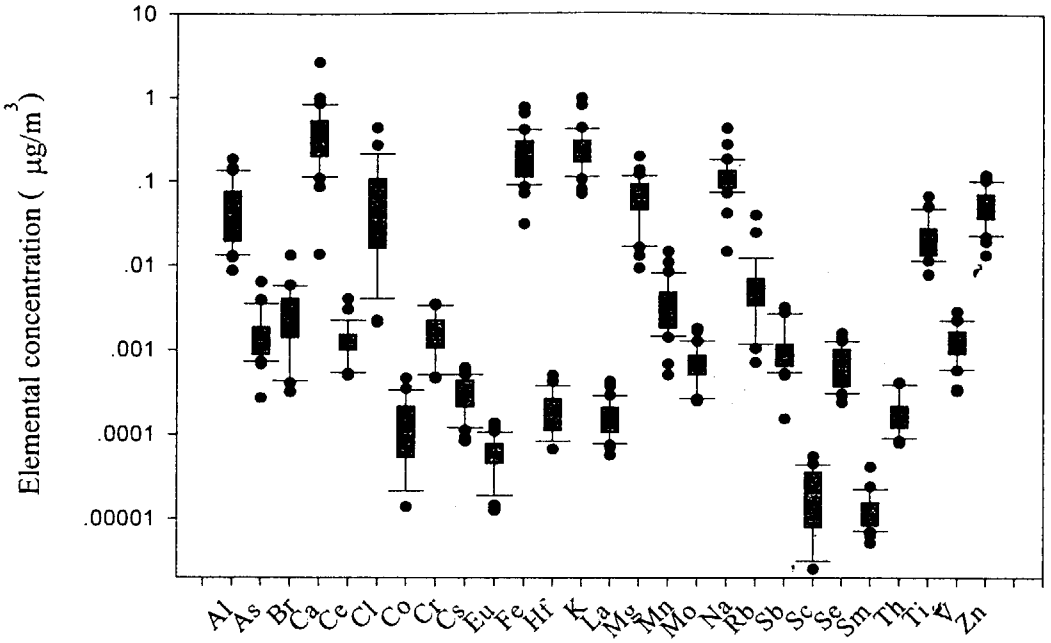

Figure 5 Box-whisker plots of the elemental concentrations in fine particle samples.

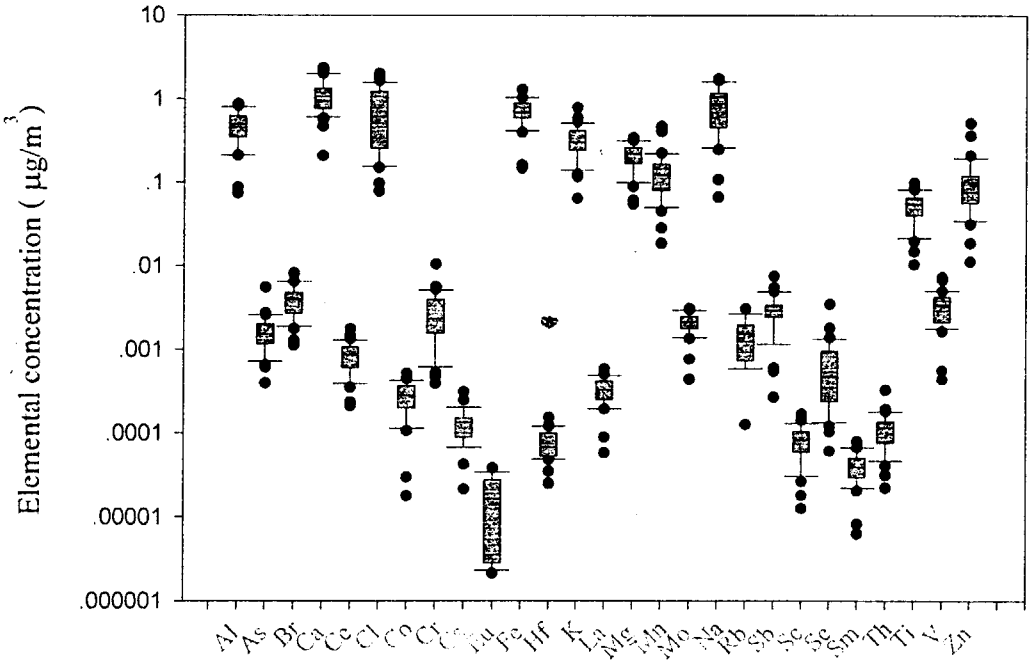

Figure 6 Box-whisker plots of the elemental concentrations in the coarse particle samples.

1.27 APPLICATION FOR AIRBORNE PARTICULATE MATTER AS A DEMONSTRATION USING k_0-NAA METHOD IN DALAT NUCLEAR RESEARCH INSTITUTE OF VIETNAM

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ABSTRACT

The airborne particulate samples have been collected using two types of polycarbonate membrane filter $PM_{2.5}$ and $PM_{2.5-10}$ in two typical sites of industrial (Ho Chi Minh City) and rural (Dateh) regions in south of Vietnam. The concentration of trace elements in the samples has been determined by the k_0-NAA procedure developed in Dalat NRI. In order to check the developed k_0-NAA procedure for the airborne particulate matter, two standard reference materials (SRMs) Urban Particulate NIST-1648 and Vehicle Exhaust Particulates NIES-8 were analyzed and the obtained results have been compared and interpreted in term of deviation between experimental results and the certified values.

INTRODUCTION

In the research programme on monitoring of air pollution in Vietnam, some sites have been selected to collect the airborne particulate samples using two types of polycarbonate membrane filter $PM_{2.5}$ and $PM_{2.5-10}$. The airborne particulate samples collected from two sites of industrial (Ho Chi Minh City) and rural (Dateh) regions in south of Vietnam have been analyzed. The number of interested elements in the airborne particulate matter are large of about 30, so when using the conventional relative NAA method the number of used SRMs must be more than one (about three). This leds to the systematical error of the analytical results depend on the different SRMs affecting to the result interpretation.

The k_0-NAA is a technique capable of multielemental analysis, relatively simple to carry out and in particular for that systematical error is stable. The k_0-NAA method allows the concentration determination of all elements of which activated radionuclides can be quantified by γ-ray spectrometry, even if this element determination was not foreseen (for instance, in the case of a sample contamination).

This report briefly describes the application of the k_0-NAA method for airborne particulate matter in order to show an overtop advantage of the k_0-NAA method for this object.

EXPERIMENTAL

Sampling and sample preparation

Sampling sites: Industrial (Ho Chi Minh City) and rural (Dateh) regions

Sampling period: January – October 2002

Sampling frequency: Each week

Sampling time: 24 h (flow rate: 18 lpm)

Coarse fraction: $PM_{2.5-10}$ and Fine fraction: $<PM_{2.5}$

Filter: Polycarbonate membrane filter 47 mm diameter, 8 μm and 4 μm pore size

Weighing: Maintain the same condition before and after sampling.

Sampling address: (1) Dateh District, Lamdong Province (Vietnam), latitude: 11°31' N, longitude: 107°28' E, altitude: 800 m; (2) Truong Dinh Street, 1st District, Ho Chi Minh City (Vietnam), latitude: 10°46' N, longitude: 106°41' E, altitude: 9 m.

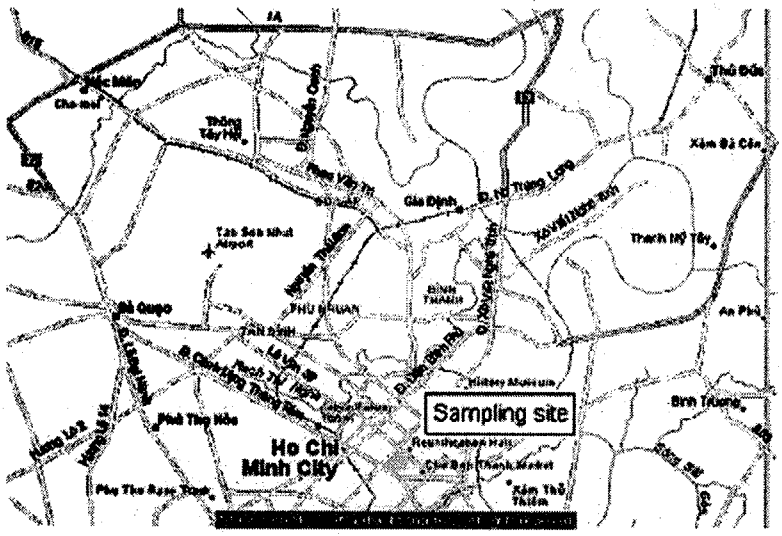

Fig 1. The rough map of Ho Chi Minh City site

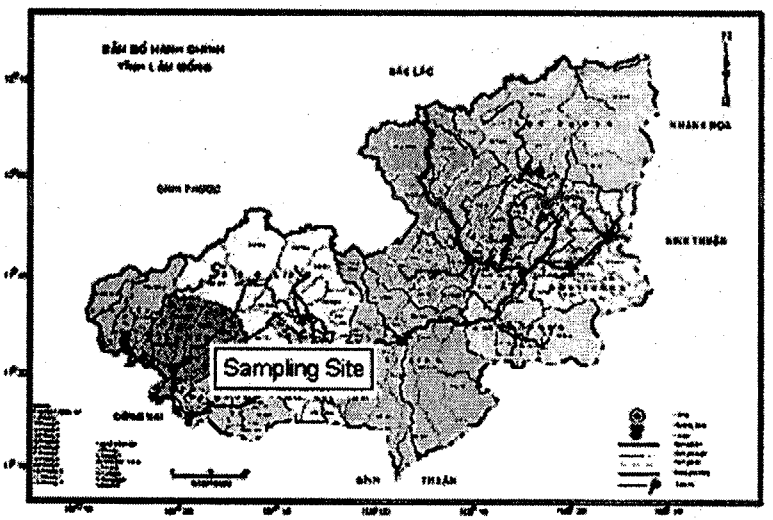

Fig 2. The rough map of Dateh site

The samples (filters containing airborne particulate matter) were folded so that they can put into 1cm × 1cm polyethylene vials and then sealed by heating. Before measurement, the samples have been transferred to new vials. Themselves of the short irradiated samples were used repetitively for the long irradiation.

The monitors used were sheet of Zr (99.8%, thickness 0.125 mm, ADVENT Research Materials Ltd.), wires of Al-0.11%Au (dia. 0.5 mm, IRMM) and Ni (99.98%, dia. 0.076 mm for determination of fast neutron).

The k_0-NAA procedure

Neutron source: Dalat research reactor of 500 kW power.

Irradiation facilities: Dry channel 7-1 using pneumatic transfer system for short irradiation down to 45 seconds and 40 wet irradiation holes in rotary rack using manual for long irradiation up to 20 hours.

Thermal neutron flux: 4.6×10^{12} n/cm^2.sec and 3.5×10^{12} n/cm^2.sec for channel 7-1 and rotary rack, respectively.

Counting system: Ortec GMX-30190 detector using automatic sample changer model ASC2 possibly for 40 samples in a measurement batch, and a Canberra GX1520 detector using manual. Acquisition and analysis software using Ortec GammaVision 5.32 for both gamma-ray spectrometers connected to an Ortec 919E MCB.

Data processing: The "Ko-DALAT" software developed in NRI.

Quality control: Urban Particulate NIST-1648 and Vehicle Exhaust Particulates NIES-8. Data intercomparison work has been participated by IAEA.

The irradiation, decay and counting time parameters for the airborne particulate sam ples are showed in Table 1.

Table 1. Irradiation, decay and counting time for the airborne particulate samples

	Irradiation time	**Decay time**	**Counting time**	**Nuclides**
Short	5 m	5 m	300 s	Al, Ca, Cu, Mg, Ti, V
(Channel 7-1)		30 m	900 s	Ba, Cl, I, In, K, Mn, Na
Long	10 h	2-3 d	1800 s	Au, Br, La, ^{122}Sb, Sm
(Rotary rack)		3 w	7200 s	As, Au, Ce, Co, Cr, Cs, Fe, Hf, Lu, Rb, ^{124}Sb, Sc, Se, Th, Yb, Zn

The measurements are performed on the calibrated γ-ray spectrometers at the positions where the dead time of system is controlled as less than 5%.

Calculation of concentrations and detection limits

The concentration ρ of the element x in the sample is calculated from the following equation[1]:

$$\rho = \frac{\left(\frac{N_p/t_c}{SDCW}\right)_a}{A_{sp,m}} \cdot \frac{1}{k_{0,m}(a)} \cdot \frac{G_{th,m} \cdot f + G_{e,m} \cdot Q_{0,m}(\alpha)}{G_{th,a} \cdot f + G_{e,a} \cdot Q_{0,a}(\alpha)} \cdot \frac{\varepsilon_{p,m}}{\varepsilon_{p,a}}$$

where,

m - coirradiated neutron fluence rate monitor;

W - sample mass (in grams);

$k_{0,m}(a)$ - k_0-factor, $= \frac{M_m \gamma_a \theta_a \sigma_{0,a}}{M_a \gamma_m \theta_m \sigma_{0,m}}$ of analyte a vs. monitor m;

f $= \Phi_{th}/\Phi_e$ ratio;

$Q_0(\alpha)$ $= \frac{Q_0 - 0.429}{\overline{E}_r^{\alpha}} + \frac{0.429}{E_{Cd}^{\alpha}(2\alpha+1)}(1eV)^{\alpha}$, where $Q_0 = \frac{I_0}{\sigma_0}$, with

I_0 is the resonance integral, E_{Cd} is the cadmium cut-off energy (E_{Cd} = 0.55eV);

$\overline{E}_r$ - effective resonance energy in eV;

α - expression for the deviation of the epithermal neutron distribution from $1/E$ shape, approximated by a $1/E^{1+\alpha}$ dependence.

The calculation of detection limits is done in the same way. For a peak not found, the detection limit for the corresponding element can be calculated by replacing the term N_p in the expression for concentration calculation with the detection limit in counts, according to Currie: $DL\ (counts) = 2.706 + 4.653\sqrt{B}$. Where, B is the background counts at the expected peak energy. Peak width is taken equal to 3 times the detector resolution at this energy.

All these nuclear parameters such as k_0, Q_0, $\overline{E}_r$, and decay data have been introduced in the PC nuclear data library[2] integrated in the "Ko-DALAT" software in order to perform a fully automatic k_0-NAA treatment.

Sources of error

- Sample preparation: We carried out the tests with the aim of evaluating the mass imprecision and the possible losses during the sample preparation. The error calculated from the results is under 1.5%.

- Measurement conditions: The sample shape is well defined and the sample position with regard to the detector is defined better than 0.5 mm. The errors due to the sample position during the measurement are generally about 1 to 2%.
- Efficiency curve: The efficiency curve was established for the counting positions, using the γ-ray point sources (^{241}Am, ^{133}Ba, ^{109}Cd, ^{60}Co, ^{137}Cs and ^{152}Eu) and the multinuclide standard source including ^{241}Am, ^{133}Ba, ^{109}Cd, ^{139}Ce, ^{57}Co, ^{60}Co, ^{137}Cs, ^{203}Hg, ^{113}Sn, ^{85}Sr and ^{88}Y from Isotope Products Laboratories (USA). These errors are of the order of 1 to 2% for energies over 200 keV and of 3 to 5% for energies around 100 keV region.
- Nuclear data: The errors is evaluated for k_0 and Q_0 factors of about 1% [1].
- Flux monitor: The error has been estimated at 1.5% for α and 1% for f [1].
- Time: The experimental conditions of this work, the error of time is negligible for long irradiation, but for short irradiation is estimated of about 1 to 2%.

The most important error occurs in the γ-ray spectrum processing of the net peak area determination, which depends on the counting statistic and the spectral matrix. As a consequence, the irradiation, decay and counting times should be optimized for each element group as showed in Table 1.

RESULTS AND DISCUSSION

About 27 elements in the airborne particulate samples were determined. Tables 2 & 3 show mean concentration of trace elements in airborne particulate samples collected fro m two sites of Dateh and Ho Chi Minh City, respectively.

Table 2. Mean concentration of trace elements in Dateh region for fine and course fractions (unit ng/m^3).

Elements	Samples	Coarse		Fine	
		Mean	STD	Mean	STD
Al	35	1003	350	203	120
As	35	3.7	1.1	2.1	1.4
Br	40	13.4	1.9	5.5	0.9
Ca	40	752	140	523	120
Ce	35	4.2	1.3	3.4	1.2
Cl	35	1238	305	985	230
Co	35	0.65	0.31	0.58	0.23
Cr	40	4.76	1.52	4.23	1.35
Cs	40	0.35	0.14	0.30	0.12
Cu	35	156	22	59	18
Fe	40	1120	410	740	240
Hf	40	0.14	0.06	0.11	0.05
K	40	920	310	771	320
La	40	1.40	0.55	1.30	0.45
Mg	40	352	85	235	75
Mn	40	24.1	4.3	15.2	3.6
Mo	35	1.32	0.31	1.01	0.27
Na	40	430	75	230	67
Rb	35	3.1	0.9	2.3	0.7
Sb	40	11.9	1.7	5.6	1.4
Sc	40	0.3	0.1	0.2	0.1
Se	35	2.1	0.4	1.1	0.4
Sm	40	0.2	0.1	0.2	0.1
Th	40	0.35	0.14	0.31	0.11
Ti	40	87.2	16.5	65.7	12.6
V	40	3.6	0.8	2.1	0.7
Zn	40	102	22	75	11

Table 3. Mean concentration of trace elements in Ho Chi Minh region for fine and course fractions (unit ng/m^3).

Elements	Samples	Coarse		Fine	
		Mean	STD	Mean	STD
Al	35	1957	460	1756	360
As	30	5.7	1.9	4.1	1.7
Ba	30	56.2	2.7	32.1	2.2
Br	35	30.2	6.1	25.8	3.9
Ca	35	956	180	755	215
Ce	30	5.4	1.5	4.6	1.4
Cl	30	1751	324	1520	305
Co	30	0.72	0.35	0.66	0.21
Cr	35	6.02	1.89	5.22	1.41
Cs	35	0.45	0.16	0.40	0.13
Cu	30	278	24	215	21
Fe	35	1644	330	1350	116
Hf	35	0.3	0.1	0.2	0.1
K	35	1211	135	1144	120
La	35	2.1	0.7	1.5	0.4
Mg	35	542	57	329	67
Mn	35	41.7	4.2	37.1	3.6
Mo	30	1.42	0.10	1.21	0.09
Na	35	944	85	785	63
Rb	30	6.1	0.9	5.3	0.7
Sb	35	19.4	1.8	15.2	1.5
Sc	35	0.5	0.2	0.4	0.1
Se	30	3.2	0.5	2.7	0.3
Sm	35	0.4	0.1	0.3	0.1
Th	35	0.51	0.16	0.45	0.13
Ti	35	126.7	14.3	108.4	11.2
V	35	6.8	0.9	5.2	0.8
Zn	35	121	20	104	13

The SRMs Urban Particulate NIST-1648 and Vehicle Exhaust Particulates NIES-8 have been used to check for the developed k_0-NAA procedure.

Figures 3 and 4 show a comparison of experimental results and the certified values. The Z-scores are showed as error bars for each point on the chart.

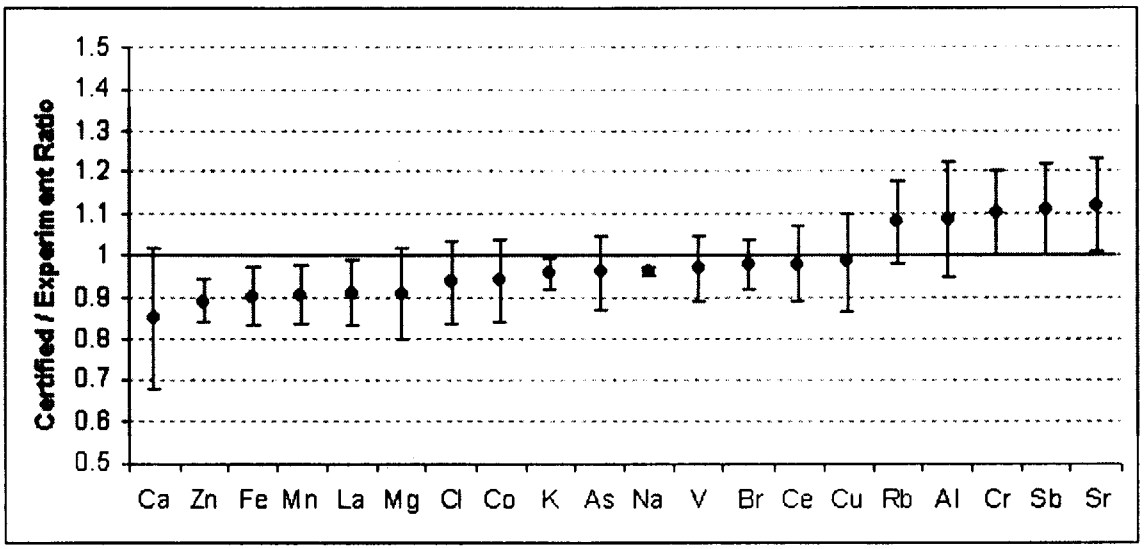

Fig 3. Comparison of experimental results and the certified values (NIST-1648)

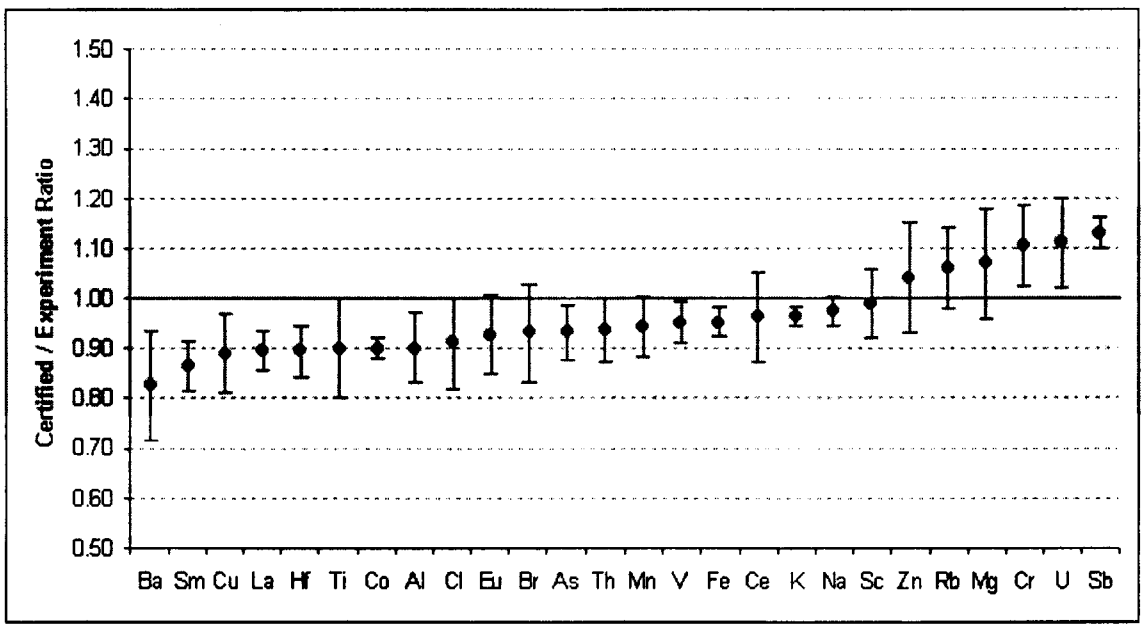

Fig 4. Comparison of experimental results and the certified values (NIES-8)

CONCLUSIONS

A multi-element analysis procedure based on the k_0-NAA method allowing to simultaneously determine concentrations for about 27 elements (Al, As, Ba, Br, Ca, Ce, C l, Co, Cr, Cu, Fe, Hf, K, La, Mg, Mn, Na, Rb, Sb, Sc, Sm, Sr, Th, Ti, U, V, and Zn) in airb orne particulate matter collected using the polycarbonate membrane filters, was developed at the Dalat research reactor.

The obtained results have revealed that the k_0-NAA procedure enables an absolute direct analysis, avoiding the use of a lot of standards and thus simplifies the preparation for analysis.

The application of k_0-NAA procedure can be generally applied for to the characterization of airborne particulate matter, which are difficult to find the proper reference materials and therefore regarded as a successful application of the method.

ACKNOWLEDGEMENTS

The FNCA organizers of the 2002 Workshop on Utilization of Research Reactors are gratefully acknowledged.

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1.28 Comparison of Chinese and European k_0 Software

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Abstract

The element determination by neutron activation analysis is commonly done by the relative method using comparison standard materials. However, when a simultaneous multi-element analysis of an unknown sample is carried out, this method requires an advance preparation of reference material for each content element, their simultaneous irradiation with the sample, and measurement in a same condition. It is indeed a demanding technique with the laborious work such as arranging reference materials. On the other hand, the k_0 method does not usually require reference materials, and allows easier and more accurate simultaneous multi-element analysis, therefore it is widely practiced in many countries including European nations. This report describes two kinds of k_0 software (KAYZERO/SOLCOI, ADVNAA) on their characteristics and the results of environmental standard sample (NIST 1632c, NIES No.8, JB-3) analyses using those software. As a result, both of those software accomplished an accuracy within about 10% in analysis of all but a few elements. They have both drawbacks and advantages in their characteristics and features, although it might not be reasonable to compare two products with different development purposes, commercial, or personal use.

keywords : Neutron Activation Analysis, k_0 method, INAA, KAYZERO, SOLCOI, ADVNAA

1. k_0 software

The two software products being analyzed here are KAYZERO/SOLCOI and ADVNAA. KAYZERO/SOLCOI was produced by DSM Research as a commercial product with the cooperation by Prof. De Corte (INW RUG, Gent, Belgium) and Dr. A. Simonits (KFKI, Budapest, Hungary) based on the k_0 method developed by them. It consists of KAYZERO for reactor-neutron activation analysis using k_0-standardization method and SOLCOI for the calibration of Ge detectors and the coincidence correction factor calculation. ADVNAA was developed by Dr. Ni Banfa and Dr. Tian weizhi of CIAE in China for personal use.

2. Comparison of software functions

(1) System requirements

An IBM-compatible Personal Computer with DOS 3.21 or higher, mathematical coprocessor, and 2 Mega Byte of free extended memory is recommended for KAYZERO/SOLCOI. Therefore in many cases it does not run on the latest computers with Windows. Also, it is normally necessary to have a gamma ray analysis software SAMPO-90 which is supported by this k_0 software, since KAYZERO/SOLCOI requires

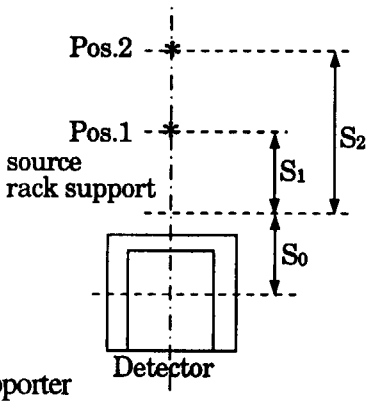
Peak Table File (PTF) and spectrum file (SPE) for the analyses. SAMPO-90 runs on DOS, and it requires a computer with a similar operating environment as with KAYZERO/SOLCOI. ADVNAA runs on Windows, and thus it runs on the newer computers. The ADVNAA software that was used this time supports two types of analysis software, SAMPO-90 and RES-PK (from Kanazawa University) as an exception.

(2) Calibration of Ge detectors

SOLCOI is used for efficiency computation and coincidence correction factor calculation. Calculated effective solid angles are used to convert well-measured point source reference efficiency to the efficiency for cylindrical vials containing all kinds of matrices at different positions relative to the detector. SOLCOI is a software product for conducting Ge detector calibration (calculation of the solid angle) with the capability of calculating the counting efficiency of a desired measurement point by inputting the counting efficiency measured at a reference position more than 20cm from a Ge detector and the peak to total ratio at the desired measurement point. It can also calculate counting efficiency in accordance with the sample shape by inputting data such as the sample dimensions and density, when the sample has a large size or a large thickness. In addition, coincidence correction factors of gamma ray can be calculated, although it requires a detailed data input such as Ge detector shape, material, and density for the calculation.

ADVNAA calculates counting efficiency of an arbitrary measurement point with a technique called EID (Effective interaction Depth) law that uses counting efficiencies calculated in advance for 2 points to calculate and input EID into the program. In this version, a user cannot change the counting efficiency since this software was developed for personal use. EID law can be briefly illustrated in the following equation.

$$\frac{\varepsilon_1(E)}{\varepsilon_2(E)} = \frac{((S_2 + S_0(E)))^2}{((S_1 + S_0(E)))^2}$$



Where,

$\varepsilon_1(E)$, $\varepsilon_2(E)$: Full energy efficiency of E at counting position 1 and 2, respectively

S_1, S_2 : Distances from source rack supporter to source position 1 and 2, respectively

$S_0(E)$: Effective interaction depth for E, measured from source rack supporter

(3) Calculation of flux parameter etc.

KAYZERO calculates flux parameters α and f by the calculation based on the "Bare Triple Method" and and "Bare bi-isotopic monitor" method using Au and Zr respectively. Moreover, comparator factor (Fc) for the comparator element used can be calculated.

ADVNAA can calculate f by the same method using Au and Zr. However, flux parameter α can not be calculated nor the correction by that calculation, therefore the user has to know the spectrum of the irradiation position.

Flux parameter (α) is calculated by the next expression.

$$(a-b)Q_{0,1}(\alpha)G_{e,1}/G_{th,1} - aQ_{0,2}(\alpha)G_{e,2}/G_{th,2} + bQ_{0,3}(\alpha)G_{e,3}/G_{th,3} = 0$$

$$a=\left[1-\frac{A_{sp,2}}{A_{sp,1}}\cdot\frac{k_{0,Au}(1)}{k_{0,Au}(2)}\cdot\frac{\varepsilon_{p,1}}{\varepsilon_{p,2}}\right]^{-1} \qquad b=\left[1-\frac{A_{sp,3}}{A_{sp,1}}\cdot\frac{k_{0,Au}(1)}{k_{0,Au}(3)}\cdot\frac{\varepsilon_{p,1}}{\varepsilon_{p,3}}\right]^{-1}$$

Where,

A_{sp} : the specific count rate

$Q_0(\alpha)$: resonance integral ($1/E^{1+\alpha}$) to 2200m/s cross-section ratio (= $I_0(\alpha)/\sigma_0$)

$$Q_0(\alpha)=\left\{(Q_0-0.429)\cdot\bar{E}_r^{-\alpha}+0.429/\left[(0.55)^{\alpha}(2\alpha+1)\right]\right\}$$

$I_0(\alpha)$: resonance integral for a $1/E^{1+\alpha}$ epithermal spectrum

G_e : correction factor for epithermal neutron self-shielding

G_{th} :correction factor for thermal neutron self-shielding

k_0 : k_0 factor of analyte

ε_p : full energy peak detection efficiency

Moreover, flux parameter (f) is calculated by the next expression.

$$f=\frac{G_{e,1}\cdot\frac{k_{0,Au}(1)}{k_{0,Au}(2)}\cdot\frac{\varepsilon_{p,1}}{\varepsilon_{p,2}}\cdot Q_{0,1}(\alpha)-G_{e,2}\cdot\frac{A_{sp,1}}{A_{sp,2}}\cdot Q_{0,2}(\alpha)}{G_{th,2}\cdot\frac{A_{sp,1}}{A_{sp,2}}-G_{th,1}\cdot\frac{k_{0,Au}(1)}{k_{0,Au}(2)}\cdot\frac{\varepsilon_{p,1}}{\varepsilon_{p,2}}}$$

Where, 1 : $^{97}Zr/^{97m}Nb$(743.3keV) and 2 : ^{95}Zr. (724.2+756.7keV)

(4) Calculation of element concentration based on k_0 method

KAYZERO calculates the element concentration by using the efficiency corresponding to the sample shape and the coincidence correction factor calculated by SOLCOI. And it uses the correction of gamma interference, self shielding, self absorption for Φ_{th}/Φ_{epi}, for accurate calculation.

ADVNAA can carry out correction of the neutron self shielding, non 1/v reaction, ^{235}U fission interferences and gamma interferences, although it does not do the correction of the sample shape, coincidence, or neutron spectrum using the flux parametor(α).

The concentration of an element in a sample based on the k_0 method is calculated by the following equation.

$$\rho_a(\mu g/g)=\frac{\left(\frac{N_p/t_c}{SDCW}\right)_z}{A_{sp,m}}\cdot\frac{1}{k_{0,m}((a))}\cdot\frac{G_{th,m}\cdot f+G_{e,m}\cdot Q_{0,m}(\alpha)}{G_{th,a}\cdot f+G_{e,a}\cdot Q_{0,a}(\alpha)}\cdot\frac{\varepsilon_{p,m}}{\varepsilon_{p,a}}\cdot 10^6$$

ρ_a : concentration of analyte a (in $\mu g/g$)

N_p : measured net peak area

t_c : counting time

S : saturation factor (= $1-\exp(-\lambda t_{irr})$)

D : decay factor (= $-\exp(-\lambda t_d)$)

C : counting factor (= $(1-\exp(-\lambda t_c))/\lambda t_c$)

W : sample mass (in grams)

f : the thermal to epithermal neutron fluence rate ratio (=ϕ_{th}/ϕ_e)

(5) Report output

KAYZERO can output concentration, SD, DL using some options (Per gamma of one or several measurements, Mean per radioisotope of several measurements, all gammas / nuclides / measurements). Moreover, the concentration of the element into the element, the nuclide, and energy can be saved in the file by CSV (Comma Separated Value) format.

ADVNAA can save the concentration and the error of each element to sample ID by CSV format.

(6) Other functions

In addition, KAYZERO/SOLCOI has the following functions.

- Displaying and printing of PTF file
- Energy calibration
- Gamma rays spectrum display and output with SPE file
- Monitoring of the flux parameter and detector calibration for QC
- Calculation of self-shielding factor with thermal neutron
- Fitting of calculated efficiency and P/T ratio
- Fine tuning for adjustment of Ge detector
- etc.

ADVNAA has the following functions.

- Three kinds of analysis methods (the k_0 method, the relative method, and the both method) can be selected for analysis.
- Many samples can be analyzed consecutively by creating the Batch file.
- Deduction of background
- etc

(7) Operativeness

Although KAYZERO is a DOS application, its interactive input style makes it very user-friendly. Since inputting of samples, irradiation, measurement information necessary for analysis are required for each gamma ray spectrum file, it is designed to save the previous data to save time of inputting when input of the same information is needed multiple times. In addition, its features such as the display of the manual from the "help" file and selections from the list show the developer's consideration for the user.

Since ADVNAA was originally developed for the purpose of personal use, it is unavoidable that there are some restrictions in operativeness. However, this software can automatically carry out analysis of many samples with fewer steps than KAYZERO, with minimum necessary inputs and batch file selection. On the other hand, it needs caution upon input in the case of separate irradiation such as this experiment, since it is designed for the condition of simultaneous irradiation of monitor, comparator, and the sample.

3. Comparison of analysis results

Analyses were carried out in the PN-3 irradiation facility of JRR-3 (ϕ_{th}= about 1.6×10^{13} n/cm^2.s, ϕ_f= about 4×10^9 n/cm^2.s, R_{cd}= about 200) to determine the accuracy of KAYZERO/SOLCOI and ADVNAA that are based on the k_0 method that does not require standard material as with the comparison method. 50mg(N=3) each of NIST 1632c, NIES No.8, and JB-3's environmental samples were analyzed after irradiation for 10 seconds for short life nuclides and 20 minutes for medium/long life nuclides. In this experiment, the differences in element concentrations appear to depend on each software product's calculation procedure, library, and correction, since we used the same irradiation/measurement condition and the same γ-ray peak analysis results. For KAYZERO, the flux parameters of the JRR-3 PN-3 were α=0.092 obtained based on

the Cd covered Triple monitor method and f=3880 calculated with the Bare bi-isotopic monitor method. For ADVNAA, the value f=6540 which was calculated with ADVNAA was used because the correction of α cannot be calculated. The effect of this f value difference was within 0.2% from a comparison with calculation for each element of interest, which was small enough to disregard. Element concentrations and deviations of standard environmental samples (NIST 1632c, NIES No.8, JB-3) by KAYZERO and ADVNAA are shown in Table- 1-3. In this experiment, KAYZERO and ADVNAA analyzed 27 elements and 29 elements respectively with accuracy within 10% excluding a few elements.

4.Evaluation

The comparison between the two software products was conducted with manuals and documents on them, and with actually operating them and making analyses. As has been already mentioned, it might not be appropriate to compare their specs, operationality, etc. due to their differing development purposes, but it is summarized below nonetheless.

- The commercial product KAYZERO shows user-friendliness in its operationality. Its downside is that it would not run on recent hardware. It needs update for Windows.
- ADVNAA, with its original "Home made, Home use" development purposes, needs modification in its user interface for the general users. However, the procedure for analysis is simple and it enable the user to choose the relative method.

They have both demonstrated similar accuracy for the number and concentration of elements. The differences in element concentration were within the range of about 10% for all but a few elements with both of those software products. It is suspected that these discrepancies were caused by the factors such as the efficiency calculation method, the coincidence correction, and the flux parameters, since the basic analysis procedure is identical. In addition, as the large deviation between the standard samples and a few elements is due mainly to the large counting errors, it suggests that analysis with longer irradiation time is necessary.

5.Conclution

Results of the comparison between these two types of k_0 software show that they have both drawbacks and advantages in their performances and characteristics. The analyses of element concentration gave similar results within about 10% accuracy with both of those software products. However, a method such as conducting an analysis with checking each element concentration using standard samples needs to be established for users to carry out an activation analysis based on the k_0 method, as a quality control of analysis values. And, it is obvious but a user needs to know the software fully upon operation. I would like to hope that users will be able to carry out analysis more easily and accurately with these types of k_0 software becoming more sophisticated and widespread in the near future.

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Table. 1 Comparison of analytical results by KAYZERO and ADVNAA in NIST 1632c

Elements	Certified Value		KAYZERO		ADVNAA		Deviation (%)		
	Conc. (ppm)	±Error (ppm)	Conc. (ppm)	SD (%)	Conc. (ppm)	Err (%)	(K-C)/C	(A-C)/C	(K-A)/A
Na	298.8	4.8	287	0.031	294	0.68	-4.0	-1.6	-2.4
○ Mg	384	32	LD		318	28		-17	
○ Al	9150	137	8880	0.65	9550	0.76	-3.0	4.4	-7.2
Cl	1139	41	1060	0.74	1190	1.2	-6.9	4.5	-11
K	1100	33	1030	0.24	1060	2.4	-6.4	-3.6	-2.8
○ Ca	1450	300	LD		1400	10		-3.5	
○ Sc	2.905	0.036	2.56	0.088	2.63	1.7	-12	-9.5	-2.7
○ Ti	517	32	531	1.4	510	4.9	2.7	-1.4	4.1
○ V	23.72	0.51	23.0	0.52	24.0	0.92	-3.0	1.2	-4.2
○ Cr	13.73	0.2	13.0	1.1	13.2	4.0	-5.3	-3.9	-1.5
Mn	13.04	0.53	12.7	0.32	12.1	2.8	-2.6	-7.2	5.0
○ Fe	7350	110	6680	0.48	7140	2.1	-9.1	-2.9	-6.4
Co	3.48	0.2	3.09	2.1	3.24	3.8	-11	-6.9	-4.6
Zn	12.1	1.3	LD		15.5	11		28	
△ Ga	3		3.35	0.69	3.16	0.55	12	5.3	6.0
○ As	6.18	0.27	5.18	0.72	4.94	1.3	-16	-20	4.9
○ Br	18.7	0.4	16.3	0.24	15.8	1.4	-13	-16	3.2
Rb	7.52	0.33	LD		8.25	16		9.7	
Sr	63.8	1.4	LD		65.4	18		2.5	
Sb	0.461	0.029	0.512	2.73	0.542	7.4	11	18	-5.5
○ Cs	0.594	0.01	LD		0.623	15		4.9	
○ Ce	11.9	0.2	11.1	0.77	11.3	5.3	-6.7	-5.0	-1.8
○ Sm	1.078	0.028	0.806	1.5			-25		
○ Eu	0.1238	0.0033	0.201(*)	0.31			62		
○ Hf	0.585	0.01	0.447	2.2	0.482	4.4	-24	-18	-7.3
Th	1.4	0.03	1.27	3.7	1.28	4.8	-9.3	-8.6	-0.78

(*) : Not recommend value in KAYZERO

○ Reference value

△ Information value

Table. 2 Comparison of analytical results by KAYZERO and ADVNAA in NIES No.8

Elements	Certified Value		KAYZERO		ADVNAA		Deviation (%)		
	Conc. (ppm)	±Error (ppm)	Conc. (ppm)	SD (%)	Conc. (ppm)	Err. (%)	(K-C)/C	(A-C)/C	(K-A)/A
Na	1920	80	1990	0.13	2000	1.7	3.7	4.2	-0.50
Mg	1010	50	1020	5.2	1010	8.3	0.99	0	0.99
Al	3300	200	3390	0.11	3690	0.15	2.7	12	-8.1
K	1150	80	1160	0.53	1230	3.0	0.87	7.0	-5.7
Ca	5300	200	5190	4.4	5990	5.9	-2.1	13	-13
○ Sc	0.55	*	0.54	1.0	0.558	3.0	-1.8	1.5	-3.2
V	17	2	16.0	0.12	16.9	1.5	-5.9	-0.59	-5.3
Cr	25.5	*	33.6	4.4	32.3	9.9	32	27	4.0
Co	3.3	0.3	3.42	4.0	3.39	5.3	3.6	2.7	0.88
Cu	67	3	LD		71.8	3.9		7.2	
Zn	1040	50	1040	3.4	1060	8.2	0	1.9	-1.9
As	2.6	0.2	2.70	1.3	2.75	3.2	3.9	5.8	-1.8
○ Se	1.3	*	LD		2.02	34		55	
○ Br	56	*	57.0	0.11	55.3	1.2	1.8	-1.3	3.07
Sr	89	*	LD		107	9.9		20	
○ Mo	6.4	*	8.14	21			27		
Sb	6	0.4	5.30	0.16	5.46	1.0	-12	-9.0	-2.9
○ La	1.2	*	1.09	0.75	1.19	5.6	-9.2	-0.83	-8.4
○ Ce	3.1	*	3.40	4.4			9.7		
○ Sm	0.2	*	0.168	0.77			-16		
○ Eu	0.05	*	0.0421(*)	22			-16		
○ Th	0.35	*	0.449	23	0.452	2.0	28	29	-0.66

(*) : Not recommend value in KAYZERO

○ Reference value

Table. 3 Comparison of analytical results by KAYZERO and ADVNAA in JB-3

Elements	Certified Value	KAYZERO		ADVNAA		Deviation (%)		
	Conc. (ppm)	Conc. (ppm)	SD (%)	Conc. (ppm)	Err. (%)	(K-C)/C	(A-C)/C	(K-A)/A
Na	20300	20100	0.018	20600	0.99	-0.99	1.5	-2.4
Mg	31300	31800	0.44	31700	1.8	1.6	1.3	0.32
Al	91000	91900	0.26	99900	0.45	0.99	9.8	-8.0
○ Cl	259	LD		321	15		24	
K	6500	6260	2.9	6460	3.7	-3.7	-0.62	-3.1
Ca	70000	62400	0.46	71200	1.3	-11	1.7	-12
Sc	33.8	31.9	0.39	33.2	1.3	-5.6	-1.8	-3.9
Ti	8600	8750	0.52	8720	1.3	1.7	1.4	0.34
V	372	411	0.26	427	0.70	10	15	-3.8
Cr	58.1	58.4	1.8	59.8	1.9	0.52	2.9	-2.3
Mn	1370	1370	0.24	1360	0.35	0	-0.73	0.74
Fe	82700	76000	0.12	82000	0.80	-8.1	-0.85	-7.3
Co	34.3	33.5	1.4	34.3	3.4	-2.3	0	-2.3
Zn	100	133	7.2	124	12	33	24	7.3
La	8.81	7.48	0.20	8.50	2.8	-15	-3.5	-12
Ce	21.5	20.2	4.8			-6.1		
Sm	4.27	3.58	0.11			-16		
Eu	1.32	1.07	0.21(*)	1.33	2.7	-19	0.76	-20
Dy	4.54	4.74	1.4			4.4		
Yb	2.55	2.47	0.27	2.25	1.5	-3.1	-12	9.8
Hf	2.67	2.64	13	2.99	12	-1.1	12	-12
Th	1.27	LD		1.65	8.8		30	

(*) : Not recommend value in KAYZERO

○ Preferable value

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2. Summary Report

Workshop

Tc Generator

Neutron Activation Analysis

Work Plans and Recommendations by the 2002 FNCA Workshop on Utilization of Research Reactors

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SUMMARY REPORT OF THE 2002 FNCA WORKSHOP ON THE UTILIZATION OF RESEARCH REACTORS

January 13 –17, 2003

Jakarta/Serpong, INDONESIA

This document presents a summary on the implementation of the 2002 FNCA Workshop on the Utilization of Research Reactors held in Jakarta/Serpong from January 13 to 17, 2003. The workshop discussed two different fields of activities, namely, Technetium Generator technology (TCG) and Neutron Activation Analysis (NAA). It was attended by scientists and technical specialists from China, Indonesia, Japan, Korea, Malaysia, the Philippines, Thailand, and Vietnam. A summary of the discussions, progress reports and future programs of each group are presented below.

1. Workshop Report of Tc-99m Generator Group

In the 2002 Workshop on Utilization of Research Reactors held in Serpong Indonesia, one delegate from each of China, Korea, Malaysia, the Philippines, Thailand and Vietnam, five delegates from Japan were hosted by Indonesia to participate and promote the technology of Tc-99m generator using poly zirconium compound (PZC) as an adsorbent for neutron activated (n, γ) Mo-99.

1.1 Evaluation of the experiments conducted by each country

Following to the agreement of the workshop held in Beijing China in November 2001, PZC materials have been distributed to participating countries in April, August and October 2002 from Kaken Co. In the present workshop, the results of the experiments in terms of Mo-99 adsorption, Tc-99m elution and Mo-99 breakthrough were reported and discussed to evaluate the performance of PZC based Tc-99m generator. All data of the Mo-99 adsorption was fairly good and elution of Tc-99m was also reasonable. The Mo-99 breakthrough were also reasonable but suggested the second column of alumina adsorbent is necessary. In case the higher radioactive Mo-99 is loaded, use of saline containing oxidizing agent gives good elution rate and very low breakthrough of Mo-99.

The comments from each country for its result are as follows.

China

The experiments show that the PZC has a good adsorbing performance at 90℃ for 3 hours (enough adsorption capability to Mo, high adsorbing rate and very low desorption rate of Mo-99). However, the cause of lower elution rate is unknown at moment.

Indonesia

The performance of high-activity Mo-99 dry-bed generators loaded with a reaction product obtained by reacting PZC materials and neutron irradiated natural Mo in the molybdate form shows that the

Tc-99m yields, Mo-99 breakthrough and radioactivity concentration meet the requirements for use in the labeling of radiopharmaceutical kits. The presence of a low-concentration oxidizing agent NaOCl during the elution step is necessary in order to maintain the high Tc-99m yields and low Mo-99 breakthrough. The future program is concerned in establishing quality control methods for total contents of Mo, Zr, and oxidizing agent in the eluate.

Korea

In order to use PZC as the adsorbent for the commercial supply of Mo-99/Tc-99m generator, the following problems should be taken into account:

(a) Attempts should be made to make granular insoluble product, which resists swelling and does not peptize in the presence of liquid. This will not only improve the flow rate but also reduce the chance of using peristaltic pump.

(b) Suitable steps should be taken to improve the stability of the polymer after the absorption of Mo-99 with respect to its particle sizes and flow characteristics.

Malaysia

Elution profile for fourteen generators prepared show acceptable elution efficiency when compared with commercially available fission product Tc generators. Mo content is higher than specified by British Pharmacopea, which is should be less than 0.1%. In this case, second alumina column is required to reduce the Mo-99 breakthrough.

Philippines

The results obtained for PZC, although limited in number of trials and the level of Mo activity loaded into the column, showed that the column holds promise as Mo column material for the generator. If the generator using this column can be available at lower than fission cost, it is reasonable to project an increasing utilization of Tc-99m in medical applications in the country.

Thailand

The PZC could adsorb Mo-99 with high adsorption rate and Tc-99m was eluted with high efficiency but the contamination of Mo-99 was detected in the eluate. The problem of contamination of Mo-99 in the eluate was easily solved by passing the eluate through alumina column.

It is strongly believed that in the near future this technology will be established and utilized under the framework of the Forum of Nuclear Cooperation in Asia (FNCA) which will benefit the countries operating small research reactors to overcome the difficulties in producing Mo-99/Tc-99m generator.

Vietnam

A good relationship between the Mo-content of adsorption solution and the Mo-adsorption capacity, adsorption percentage , Mo-breakthrough and Tc-99m elution yield was found. The preparation of PZC based Tc-99m chromatographic generator with 4 gram weight of PZC was successfully conducted. And a large number of interesting results were collected.

Table 1. Results of experiments conducted by each participating country

	^{99}Mo loading	^{99}Mo adsorption	^{99m}Tc elution	^{99}Mo breakthrouh
China	68.5 mCi	98.3%	7~17%	0.04~0.06%
Indonesia	273 mCi	94~95%	>90%	<0.03%
Korea	10 ~500 μCi	67~88%	67~70%	0.03~0.5%
Malaysia	2 mCi	82~92%	74~94%	0.2~0.4%
Philippines	18 mCi	99%	69~75%	0.09~0.20%
Thailand	80 mCi	89%	65~84%	0.3~0.8%
Vietnam	100 mCi	90%	78~91%	0.017~0.02%

1.2 Demonstration of PZC based Tc-99m generator

Demonstrative experiment was carried out during the present workshop as agreed in the 2001 Workshop in Beijing. The results of the experiments are summarized in Table 2 whereas the results of each countries' are summarized in Table 1.

Two PZC column assemblies were prepared for the experiment. The results showed slightly lower Mo adsorption ratio which might be due to instability during storage of PZC material. The Tc-99m elution rate, however, was around 80% and breakthrough of Mo-99 in eluate was low enough to clear the criteria of medical application.

Table 2. Results of demonstration experiment*

Assembly	^{99}Mo-loading	^{99}Mo adsorption	^{99m}Tc elution*	^{99}Mo breakthrough**
1	311.8 mCi	79.2%	79.6%	0.00029%
2	311.8 mCi	82.0%	83.8%	0.00032%

* Oxidizing agent (NaOCl) is used in the elution step.

** Secondary column (alumina) is applied.

1.3 Work plan for 2003

1.3.1 Standardization of production procedure

Standardization of production procedure for PZC based Tc-99m generator should be studied and established for the activities in 2003. Draft of the procedure shall be made by Kaken and Project Leaders of Indonesia, Japan and Vietnam to be circulated to all participating countries for comments. In this connection, importance of the automatic Mo loading and column packing system which is planned to install in the cell of BATAN facility cooperated with Kaken was stressed.

1.3.2 Distribution of PZC material

Distribution of the PZC material will be continued for three times in the year 2003 for the experiment of Mo adsorbent PZC for Tc-99m generator with strictly standardized experimental procedure instruction as mentioned in 1.3.1. The amount of PZC material to be sent by Kaken Co. is 15 grams each time. For these experiments, use of secondary column of alumina and saline containing NaOCl for Tc-99m elution process will be compulsory.

1.3.3 Supply of (n, γ) Mo-99

China (NPIC), Indonesia (BATAN) and Korea (KAERI) can provide Mo-99 produced by irradiation of natural Mo-98 with free of charge except expense of container and transportation fee.

1.3.4 Comparison with the other generator

Comparison of technological aspects for production of PZC type Tc-99m generator is expected to be compiled by Indonesia for fission type generator whereas China and Vietnam for gel type generator.

1.3.5 Survey of the market of Tc-99m generator

The potential size of market of Tc-99m generator in terms of number of gamma cameras, number of tests for diagnosis, cost of the imported generators and system for the distribution of generators should be surveyed by project leaders of FNCA countries and compiled by Japanese Project Leader as early as possible.

1.3.6 Setting of Mo-99 loading/PZC column packing machine

An automated Mo-99 loading/PZC column packing machine is to be set in the hot cell facility of BATAN in cooperation between Kaken and BATAN.

1.4 Work plan for 2004

1.4.1 Labeling test

Confirmation of quality of Tc-99m solution obtained from PZC generator through labeling test with various ligands

The work plans for 2003 and 2004 are summarized in Table 3.

Table 3. Work Plan

Year	Work plan	Remarks
2003	1) Standardization of production procedure	Japan, Indonesia, Vietnam
	2) Distribution of PZC material	15 grams x 3 times, Japan
	3) Supply of (n, γ) Mo-99 in case of necessary	China, Indonesia, Korea
	4) Comparison with the other type generators	China, Indonesia
	5) Survey of the market of Tc-99m generator	All participating countries
	6) Setting of Mo-99 loading system	Japan, Indonesia
2004	1) Labeling test	For Quality Assurance

1.4 Comments and suggestions

1) In order to secure the supply of radioisotopes and radiopharmaceuticals in each country, a network among the FNCA countries is desirable to facilitate mutual supply of target materials and chemical reagents and also mutual use of reactors as well.

2) Publication of the outcome resulted from the activities of Tc Generator Group should be done at least at the final stage of the workshop.

3) It is hoped that FNCA sends a letter for each government to appeal the importance of setup of the

automatic loading system in order to spread high quality PZC based Tc-99m generators.

1.6 Others

1) The Kaken Co, the manufacturer and patent holder of PZC material, presented the detailed plan of automatic Mo loading / PZC column packing machine as well as the future plan for realizing the Tc-99m generator production based on PZC technology.

It was also reported by Kaken Co that a joint-patent shared by Kaken Co and BATAN concerning the Mo-99/Tc-99m Generator System based on PZC materials with neutron irradiated natural molybdenum and the automatic Mo loading / PZC column packing machine was registered both in Japan in August 2002 and in Indonesia in November 2002.

2) As the invited talks, the current status of production of fission Mo-99 and Tc-99m generator in Indonesia and a comparative study on the labeling of Tc-99m kits using fission type generator and PZC type generator were presented by BATAN Teknologi and BATAN, respectively. And also the experimental results of application of PZC to W-188/Re-188 generator was presented by JAERI.

3) The FNCA coordinator of Japan made a lead-off speech on strategy for commercial application of Tc-99m generator produced by PZC Technology. In succession, the paper on the trend of Tc-99m generator in FNCA countries was presented by Japanese Project Leader and the plan for realizing the PZC generator was proposed by the President of Kaken Co.

2. WORKSHOP REPORT OF NAA GROUP

The 2002 FNCA workshop on utilization of research reactors for NAA group was held at Serpong, Indonesia on 13-17, January 2003, with participants from China, Indonesia, Korea, Japan, Malaysia, Philippines, Thailand and Vietnam. On the first day of the workshop, Prof. Tsunehiko Otoshi, Tohoku University of Community Service and Science presented a general lecture on neutron activation analysis entitled "NAA as Analytical Tool of Environmental Issue".

This reports consists of three parts, (i) summaries of invited and country reports, (ii) summaries of demonstration of ko-method experiment and reports related with ko-softwares and (iii) future program.

2.1 SUMMARIES OF INVITED AND COUNTRY REPORTS

(Invited speeches)

1) "Improving Air Quality in Indonesia Study of Fine and Coarse Particulates" by Dr. Puji Lestari, Bandung Institute of Technology, Indonesia. Fine and coarse portions of atmospheric particulates were collected in 5 locations in Bandung and their constituents were determined by NAA and other methods. Results indicate that the concentration of coarse particles increases in the daytime while there is no significant difference for fine particles between daytime and nighttime.
2) "Current Status of Air Pollution Monitoring in Indonesia" by Ms.Novy Farhani of Environmental Management Center of Indonesia. The ambient air pollution monitoring network has been installed since 1999 at about 10 districts in Indonesia. Automatic equipments monitor 5 parameters (SO_2, PM_{10}, CO, NO_2 and O_3) continuously, and the status of air pollution is evaluated by the Pollutant Standard Index (PSI).

(Country reports)

1) "Air Pollution Study in Beijing 2002 by using NAA and PM_{10} Particulates" by Prof. Bangfa Ni of China Institute of Atomic Energy. Particulate matter samples collected at Che Gong Zhuang, Beijing are analyzed by k0-NAA method. Around 45 elements per sample are determined by this method. Multi elements data are evaluated by enrichment factors, factor analysis and so on.
2 "Collection of Size Fractionated Particulate Matter Sample for NAA in Japan" by Tsunehiko Otoshi, Tohoku University of Community Service and Science, Japan. At two monitoring sites in Japan, particulate matter samples are collected for NAA. While higher concentration of coarse particulates is observed during spring season, average concentration of PM_{10} during the common sampling period is 0.031mg/m^3 in urban site, and 0.022mg/m^3 in rural site.
3 "INAA of Atmospheric Particulate Collected in Hachioji and Sakata", by Dr.Yasuji Oura of Tokyo Metropolitan University, Japan. Particulate matter samples collected at two sites in Japan are analyzed by NAA. For the determination of Cr, Cl, and Br, impurity of blank filter should be considered, and blank values should be subtracted in calculation. Among elements determined, concentrations of Cl, Ag, Zn, Cu in individual size fractions are different between two monitoring sites. It depends on the difference in emission sources at each site.

4) "Elemental Quantification of Airborne Particulate Matter in Bandung and Lembang Regions", by Mr.Sutisna, Research and Development Center for Materials Science and Technology, National Nuclear Energy Agency of the Republic of Indonesia. NAA is applied to particulate matter samples collected at Bandung (urban) and Lembang (rural). In rural site, fine portion of particulates is dominant. However, in urban site, higher concentrations of coarse particles are observed. Enrichment factors of elements such as V, Br, and Cl differ depending on the season.
5) "Recent Application of NAA in Korea" by Dr. Yong Sam Chung of Korea Atomic Energy Research Institute, Daejeon, Korea. Regarding activities of the project, air pollution study and preliminary experiments with an application of ko-method are carried out. Air particulate samples collected at two regions (suburban and industrial area of Daejeon city) are analyzed by NAA and concentrations of 25 elements in 50 filter samples are determined. Two certified reference materials (CRMs) distributed from Japan are used for analytical quality control. Higher mass concentrations of airborne particulate matters are observed during spring season due to Asian dust. Concentrations of black carbon are measured using reflectometer. For the evaluation of emission source for multi-element data, enrichment factors and correlation coefficients are calculated for fine and coarse particulates.
6) "Characterization of the Finest and Coarse Airborne Particulate Matter in Kuala Lumpur's ambient air" by Dr.Khalik Haji Wood of Malaysian Institute for Nuclear Technology Research (MINT), Malaysia. Particulate samples collected at the site about 1km from Kuala Lumpur City Center are analyzed by NAA method. Average mass concentrations of fine particulates of 0.031mg/m^3 are higher than those of coarse particles of 0.019mg/m^3. High particulate concentrations which may be caused by haze episodes are not observed during the sampling period. Quality control of NAA is carried out using NIST SRM 1633a. Some elements such as Mn and Cr in fine portion of particulates indicate the presence of anthropogenic pollution sources around the monitoring site.
7) "Airborne Particulate Matter Collection and Analysis by XRF" by Ms.Flora Lopez Santos of Philippine Nuclear Research Institute (PNRI), Philippines. Particulate samples collected at three monitoring sites in Manila Metropolis are analyzed by XRF method. The sampling stations designated for the FNCA program are at a commercial area close to a major highway. PNRI is considering several options to resume reactor-related activities, and it is necessary to ensure continuing availability of expertise in NAA. Obtained data indicate the decrease of Pb concentration in fine particles because of the use of unleaded gasoline in Philippines.
8) "Study of Urban Air Pollution in Thailand" by Dr.Wanna Chueinta of Office of Atoms for Peace, Thailand. Particulate matters collected at downtown Bangkok are analyzed by NAA and 20 elements can be determined. Black carbon is one of major constituents of fine particulates. Quality control study of NAA using SRMs indicate that too small amounts of SRM size, say less than 1mg, may yield unreliable or highly uncertain values, especially for such elements determined by using short-lived nuclides.
9) "Airborne Particulate Matter as a Demonstration using ko-INAA at Nuclear Research Institute of Vietnam" by Mr.Ho Manh Dung of Nuclear Research Institute, Dalat, Vietnam. Particulate matter samples collected at two monitoring sites (industrial site in Ho Chi Minh City, and rural site in Dateh) are analyzed by ko-method.

About 28 elements for both sites are determined by ko-method. The ko-method has an advantage to simplify the routine calculation process in NAA. It is insisted that ko-NAA method works better than the conventional NAA using SRMs when evaluating emission sources, considering a higher reproducibility in the relationship between elements.

(General Discussion)

After the presentation of the above country reports, general discussion was held as follows;

(i) Considering the importance of QA and QC in NAA, it was confirmed that some efforts related with QA/QC (inter-laboratory comparison, laboratory round robin test and/or validation of method) would be continuously made using the two CRMs provided and their results would be included in the country report at future workshops.

(ii) Several opinions were exchanged for sampling methodologies and sampling sites of PMs. (It was decide to adopt "PM2" or "fine particles" instead of "PM2.5", considering a poor accuracy of the cut off of a Gent SFU sampler.)

(iii) Future visions of researches using NAA and its needs related to marketing were discussed.

(iv) It was emphasized that the utilization of ko-method would greatly improve the performance of NAA.

2.2 SUMMARIES OF KO EXPERIMENTS AND REPORTS RELATED WITH KO SOFTWARES

(ko-method experiments)

A demonstration experiment of NAA using ko-method was performed. Although this was the first performance of the application of ko-method to real samples at BATAN, reasonable data were finally obtained for geological standard rock powder samples after tremendous efforts of Indonesian scientists. It may be mentioned here that the success of this demonstration experiment was achieved by an excellent collaboration of Indonesian and Japanese peoples. Especially, the contribution of Mr. F. Sasajima of JAERI was highly appreciated.

(Reports related to ko-software)

1) "Demonstration of NAA ko-software, ADVNAA" by NI Bangfa, China Institute of Atomic Energy, China. A ko-software developed by the speaker at CIAE, which is named ADVNAA is demonstrated. Several features which are not admitted for the European software (KAYZERO/SOLCOI) are explained.

2) "Comparison of Chinese and European ko-softwares" by Fumio Sasajima, Japan Atomic Energy Research Institute, Japan. Two kinds of ko-softwares (KAYZERO/SOLCOI and ADVNAA) are compared by applying these softwares to environmental reference standard samples (NIST 1632c, NIES No. 8 and GSJ JB-1). Basically, there is no essential difference between analytical results obtained by them.

3) "Evaluation of CIAE-ko software" by Y. Oura, Tokyo Metropolitan University, Japan. A ko-software developed at CIAE, China, which is mentioned as ADVNAA above, is critically evaluated in

comparison with a DSM ko-software (KAIZERO/SOLCOI). Being similar to the conclusion by Sasajima, no essential difference is found between two softwares.

4) "Development of ko-standardization method of NAA (ko-NAA) regarding software and experiment in Dalat Research Reactor of Vietnam" by Ho Manh Dung, Nuclear Research Institute, Vietnam. A ko-software developed by the speaker at NRI (Dalat) is demonstrated. By applying the ko-software to several standard reference materials including NIST-1632c and NIES-8, it is shown that the ko-method using the software and experimental procedure developed by the speaker is reliable and practical for air particulate matters.

2.3 FUTURE PROGRAMS

(1) Air particulate study

During 2002, each country achieved a satisfactory progress, as reported in country reports of air particulate study. Up to now, no apparent problems were admitted in conducting the program of air particulate study. Following such a promising situation, we have set the following plan for 2003.

1). We continuously collect and analyze air particulate matters according to the agreement at the 2001 FNCA workshop.

2). To summarize experimental outcomes of 2002, we agreed to prepare a joint report in which all data for air particulate matters collected in 2002 are to be included. We set several stages with their deadline as follows;

(i) (Stage 1; June 30, 2003) *(Submission of data and experimental conditions)* All data are to be reported to a responsible person in Japan, whose name will be announced later. At most, two sampling sites per each country. Each country is required to report all necessary conditions regarding sampling in addition to all analyses data (with errors). A type of analytical uncertainties needs to be specified. A format for describing sampling conditions will be distributed to each country. Thailand is responsible to prepare and distribute the format. Earlier submission is highly advised.

(ii) (Stage 2; August 15, 2003) *(Submission of comment/discussion)* Once all data are accommodated in one table (probably in EXCEL file), it will be sent to all persons who have sent their analytical data. Then, they are required to send scientific comments and discussions for compiled data to a NAA group leader (M. Ebihara).

(iii) (Stage 3; September 30, 2003) *(Finalization of draft)* A NAA group leader is responsible to appoint an appropriate person to organize all comments and discussions sent from contributors and finalize a report (reports). Modification of the draft can be done by communicating with each contributor by email.

(2) QA/QC

We have agreed to conduct some experiments related with QA/QC issue at the 2001 workshop. Unfortunately, only three countries (Korea, Malaysia, and Thailand) reported this subject at this workshop. We discussed again this subject and set a new plan for the year of 2003 as follows;

In order to evaluate uncertainties involved in analyzing a small size of sample like air particulate

matters, a small-sized sample is to be analyzed repeatedly. For this purpose, 1 mg of two SRM sample (NIST 1632C and NIES No.8) is to be analyzed more than ten times. Either ko method or comparison method (or both) can be applied by using either chemical standards or other standard reference materials for obtaining absolute contents of elements.

Apart from this, a concern of impurities in filter papers was stated. This issue is also important in evaluating our QA/QC. Therefore, it is desired to analyze a blank filter occasionally

(3) Installation and development of ko-software

Following the discussion at the last workshop in Beijing, we continuously discuss a possibility of the introduction of ko-method to our project of air particulate matters. After extensive discussion, we came to the following conclusions;

1) Introduction of ko method in analyzing air particulate matters is highly desired in all participating countries. Note that the introduction of ko method includes installing of ko software and performing of experiment with use of the software.
2) Considering advantages and disadvantages for using a common software and individual softwares among our community, it was admitted that we would choose any appropriate software from three existing ko softwares; DSM version, CIAE version and Dalat version. The last two versions are so-called home-made sofrwares and their developers announced that these softwares could be distributed for inspection without any charge among participating countries of the workshop. Each country who wants to install these home-made softwares may contact directly to their developers for their installation and use.
3) At the next meeting, each country is supposed to present INAA data for air particulate matters calculated by using ko method.
4) In future, it is desired that a common upgraded software of ko method, which will be shared among our community, will be developed based on CIAE and Dalat versions. Their developers agreed to make effort to complete such a software.
5) It is requested that a training course for ko method is held, hopefully before the next workshop. It may be considered that such a course is attached to the next workshop although earlier opening is more practical and, hence, desired.

2.5 Others

1) (Future activities) For future activities of the NAA group, the following themes were raised by participants;
   (i) INAA of marine samples for heavy metals.
   (ii) INAA of solid waste samples.
   (iii) Application of NAA for mining.
   (iv) Development of prompt gamma-ray analysis (PGA).

   As the study of air particulate matters is expected to be performed without any problem among all the participating countries in coming years, we may extend our activities to some of the above listed themes in (near) future. Among them, the marine sample was suggested and agreed to be the most feasible subject for co-operative analysis of the NAA group. Detailed procedures may be discussed at

the next workshop. In the meanwhile, some common samples are to be analyzed and their data are desired to be presented at the next workshop. Malaysia is responsible to prepare such samples and distribute them to all participating countries.

2) (The miscellaneous)

(i) It is aimed that our analytical data are reflected in the environmental management policy. To make this successful and effective, we must make our best efforts to find any way to approach to appropriate officials, agencies, institute, etc, which are related environmental pollution problems.

(ii) It is encouraged that participating countries actively apply for any exchange program like the former STA fellowship program to expand our activities within the frame of FNCA. The Japanese (sub)group-leader is requested to make an appeal to MEXT that such an exchange program is essential to promote the FNCA activity.

3. WORK PLANS AND RECOMMENDATIONS BY THE 2002 FNCA WORKSHOP ON UTILIZATION OF RESEARCH REACTORS

3.1 In accordance with the FNCA "Proposal on the Next three year Activity in the Field of Utilization of Research Reactors" by the FNCA coordinator meeting on March 2001, the workshop agrees to make the following work plans and recommendations for the next workshop in the area of Utilization of Research Reactor to meet needs of the FNCA countries and to achieve results with socio-economic impact to end users.

(1) Tc-99m Generator

- Standardization of production procedure for PZC based Tc-99m generator should be studied and established. Draft of the procedure shall be made by Kaken and Project Leaders of Indonesia, Japan and Vietnam to be circulated to all participating countries.
- Distribution of the PZC material will be continued for three times in the year 2003 for the experiment of Mo adsorbent PZC for Tc-99m generator with strictly standardized experimental procedure instruction. The amount of PZC material to be sent by Kaken Co. is 15 grams each time. For these experiments, use of secondary column of alumina and saline containing NaOCl for Tc-99m elution process will be compulsory.
- China (NPIC), Indonesia (BATAN) and Korea (KAERI) can provide Mo-99 produced by irradiation of natural Mo-98 with free of charge except expense of container and transportation fee.
- Comparison of technological aspects for production of PZC type Tc-99m generator is expected to be compiled by Indonesia for fission type generator whereas China and Vietnam for gel type generator.
- Survey of the potential size of market of Tc-99m generator in terms of number of gamma cameras, number of tests for diagnosis, cost of the imported generators and system for the distribution of generators.
- An automated Mo-99 loading/PZC column packing machine is to be set in the hot cell facility of BATAN in cooperation between Kaken and BATAN.

(2) Neutron Activation Analysis

- Air particulate matters are to be continuously collected and analyzed in accordance with the agreement at the 2001 FNCA workshop. To summarize experimental outcomes of 2002, a joint report in which all data of air particulate matters collected in 2002 are included is to be prepared in 2003.
- At the next workshop, each country is supposed to present INAA data for air particulate matters calculated by using ko-method. In order to put this goal into practice, it is desired for a training course for ko-method to be held, preferably prior to the 2003 workshop or in connection with the workshop.
- Each country may choose any of DSM version, CIAE version and Dalat version as a ko-software when introducing the ko-method. However, it is highly desired that a common software be used among our community for assessing INAA data effectively and efficiently. Such a software should be developed in collaboration with each other of the NAA workshop members.
- Besides air particulate matters, marine sediment samples will be included within the scope of our activities. The term of 2003 will be used for working out our new strategy.

3.2 Additional remark

The workshop agrees that the following countries be proposed in the FNCA Coordinators Meeting as possible venues for the next year's workshop.

(1) Vietnam as the hosting country for the workshop. Themes: 1) Application of NAA technology to the environmental analysis. 2) Information exchange on research reactors and their applications, and discussion on related topics.

(2) Indonesia as the hosting country for the sub-workshop. Themes: 1) Establishment of the Tc Generator technology. 2) Application of Neutron Scattering technology for the development of TPE.

Appendix

Schedule of
The 2002 FNCA Workshop on the Utilization of Research Reactors
January 13 – 17, 2003, Jakarta & Serpong, Indonesia

Plenary Session, Sahid Hotel , Jakarta

Monday, 13 January 2003		
09.00 – 09.30	**Registration**	
09.30 – 10.00	**Welcoming/Opening Remarks:**	
	- Chairman of Organizing Committee	
	- MEXT Representative	
	- Japanese Project Leader	
	- Chairman of BATAN	
10.00 – 10.30	**Coffee Break and Group Photo**	
General Lectures:		**Chairperson**
10.30 – 11.00	Dr. S. Machi, JAIF: Nuclear Technology of Sustainable Development and FNCA Activities.	**Dr. Hudi Hastowo**
11.00 – 11.45	Dr. Soebowo Soemewo (Indonesia): Medical Uses of ^{99m}Technetium Radioisotope in Indonesia	
11.45 – 12.30	Prof. Dr. Tsunehiko Otoshi (Japan): Neutron Activation Analysis as Analytical Tool of Environmental Issue.	
12.30 – 13.40	**Lunch**	
Country Report on Research Reactor Operation		**Chairperson**
13-40 – 14.00	Mr. Yuan Luzheng (China): The Current Status of Utilization of Research Reactors in China.	**Mr. Isao Takeshita**
14.00 – 14.20	Mr. Iman Kuntoro (Indonesia) : Operation and Utilization of Indonesian Research Reactors.	
14.20 – 14.40	Mr. Isao Takeshita (Japan) : Status of Research Reactors in Japan.	**Mr. Sakda Charoen**
14.40 – 15.00	Dr. Yong Sam Chung (Korea) : Current Status of HANARO Reactor and its Application.	
15.00 – 15.30	**Coffee Break**	
Country Report on Research Reactor Operation		**Chairperson**
15.30 – 15.50	Dr. Abdul Khalik Haji Wood (Malaysia) : Utilization of MINT's Research Reactor.	**Mr. Iman Kuntoro**
15.50 – 16.10	Ms. Elvira Z. Sombrito (The Philippines): Status of Research Reactor Utilization and their Related Activities.	
16.10 –16.30	Mr. Sakda Chaeron (Thailand) : Research Reactor and its Application in Thailand.	**Mr. Yuan Luzheng**
16.30 –16.50	Dr. Le Van So (Vietnam) : Current Status of Opreation and Utilization of the Dalat Research Reactor.	
19.00 –21.00	**Reception hosted by MEXT and BATAN**	

Appendix

Paralell Session, Serpong

RADIOISOTOPE GROUP

Tuesday, January 14, 2003		
Country Report on PZC-Based ^{99}Mo/^{99m}Tc Generator		**Chairperson**
09.00 – 09.30	Dr. Liu Yishu (China) : The Performance of Gel ^{99m}Tc-Generator	**Ms. Elvira Z. Sombrito**
09.30 – 10.00	Mr. Hyon Soo Han (Korea) : Status on Development and Production of Tc-99m Radioisotope and Its Generator in Korea	
10.00 – 10.30	Dr. Zulkifli Mohamed Hashim (Malaysia) : The Utilization and Performance of Poly-Zirconium Compound (PZC)as Chromatographic Column for ^{99m}Tc- Generator	
10.30 – 11.00	**Coffee Break**	
Country Report On PZC-Based ^{99}Mo/^{99m}Tc Generator		**Chairperson**
11.00 – 11.30	**Invited Speaker:** Mr. Adang H. Gunawan : A Comparative Study on the Labelling of Radiopharmaceutical Kits with ^{99m}Tc-Obtained from F.P. ^{99}Mo/^{99m}Tc Generator and PZC-Based ^{99}Mo/^{99m}Tc Generator	**Dr. Zulkifli Mohamed Hashim**
11.30 – 12.00	Ms. Elvira Z. Sombrito (The Philippines) : Performance Test on new Chromatographic materials for ^{99}Mo/^{99m}Tc Generator	
12.00 – 13.00	**Lunch**	
	Country Report On PZC-Based ^{99}Mo/^{99m}Tc Generator	**Chairperson**
13.00 – 13.30	Mr. Shakda Charoen (Thailand): Evaluation of ^{99}Mo/^{99m}Tc Generator Experiment Using PZC Material and Irradiated Natural Molybdenum	**Dr. A. Mutalib**
13.30 – 14.00	Dr. Le Van So (Vietnam): Investigation on the Performance of Polymer Zirconium Compound for Chromatographic ^{99m}Tc Generator Preparation	
14.00 – 14.30	**Coffee Break**	
Invited Speakers		**Chairperson**
14.30 – 15.00	Mr. H. Matsuoka: Application of PZC to ^{188}W/^{188}Re generator	**Dr K. Tatenuma**
15.00- 15.30	Dr. Mulyanto Suryosubroto: Current Status on Production of Fission Product ^{99}Mo and ^{99}Mo/^{99m}Tc Generator in Indonesia	
	Country Report On PZC-Based ^{99}Mo/^{99m}Tc Generator	**Chairperson**
15.30 – 16.00	Dr K. Tatenuma (Japan) : Performances of PZC for (n, γ) ^{99}Mo-^{99m}Tc generator	**Dr. Le Van Soo**
16.00 – 16.30	Dr. A. Mutalib (Indonesia): The Performance of ^{99}Mo/^{99m}Tc Generator Based on PZC Materials and Neutron Irradiated Natural Molybdenum	

Appendix

Parallel Session, Serpong

Neutron Activation Analysis Group

Tuesday , January 14, 2003

Invited Speakers		Chairperson
09.00 – 09.45	Dr. Puji Lestari (Indonesia) : Improving Air Quality in Indonesia: Study of Fine and Coarse Particulate	Dr. Khalik Wood
09.45 – 10.30	Ms. Novy Farhani (Indonesia) : Current Status of Air Pollution Monitoring in Indonesia	
10.30 – 11.00	**Coffee break**	
Country Report of Air Particulate Study		**Chairperson**
11.00 – 11.20	Prof. Bangfa Ni (China) : Summary of Progress Report	Dr.Yong Sam Chung
11.20 – 11.40	Prof. Tsunehiko Otoshi (Japan): Collection of Size Fractionated Particulate Matter Sample for Neutron Activation Analysis in Japan	
11.40 - 12.00	Dr.Yauji Oura (Japan): INAA of Atmospheric Particulate Collected at Hachioji and Sakata	
12.00 – 13.00	**Lunch**	
Country Report of Air Particulate Study		**Chairperson**
13.00 – 13.20	Mr. Sutisna (Indonesia): Elemental Quantification of Airborne Particulate Matter in Bandung and Lembang Regions.	Ms. Flora Lopez Santos
13.20 – 13.40	Dr.Yong Sam Chung (Korea): Recent Application of Neutron Activation Analysis in Korea	
13.40 – 14.00	Dr. Khalik Haji Wood (Malaysia): Characterization of the Finest and Coarse Airborne Particulate Matter in Kualalumpur's Ambient Air	
14.00 – 14.30	**Coffee break**	
Country Report of Air Particulate Study		**Chairperson**
14.30 -14.50	Ms. Flora Lopez Santos (Philippine): Airborne Particulate Matter collection and Analysis by XRF	Dr.Yauji Oura
14.50 - 15.10	Ms. Wanna Chueinta (Thailand): Study of Urban Air Pollution in Thailand	
15.10 – 15.30	Mr. Ho Manh Dung (Vietnam): The Application for Airborne Particulate Matter as a Demonstration using k_o-INAA at Nuclear Research Institute of Vietnam.	
15.30 – 16.30	General Discussion	Prof. Tsunehiko Otoshi

Appendix

Parallel Session

RADIOISOTOPE GROUP

Wednesday, January 15, 2003

09.00 – 09.20	Discussion on preparation of demonstration of ^{99m}Tc Generator Production	**Chairperson: Dr K. Tatenuma, Dr. A. Mutalib**
09.20 – 10.30	Demonstration on the production of ^{99}Mo/^{99m}Tc Generator based on PZC materials and neutron irradiated natural molybdenum	
10.30 – 11.00	**Coffee Break**	
11.00 – 12.00	Demonstration on the production of ^{99}Mo/^{99m}Tc Generator based on PZC materials and neutron irradiated natural molybdenum	
12.00 – 13.00	**Lunch**	
13.00 – 14.00	Demonstration on the production of ^{99}Mo/^{99m}Tc Generator based on PZC materials and neutron irradiated natural molybdenum	
14.00 – 14.30	**Coffee Break**	
Discussion of Activity Report and Future Program		**Chairperson**
14.30 – 15.00	Dr. Sueo Machi (Japan) : Strategy for Commercial Application of ^{99m}Tc-Generator Produced by PZC Technology	**Mr. Hyon Soo Han, Dr. Liu Yishu**
15.00 – 15.30	Dr. Tsuguo Genka (Japan) : The Trend of ^{99m}Tc-Generators in FNCA Countries	
15.30 – 16.00	Dr. Katsuyoshi Tatenuma (Japan) : Proposal and Future Plan for Realizing ^{99m}Tc-Generator based on the (n, γ) method using PZC	

Parallel Session, Serpong

Neutron Activation Analysis Group

Wednesday , January 15, 2003

	k_o-Software and Its Application	**Chairperson**
09.00 – 10.30	Demonstration of k_o Method	Ms. Sri Wardhani
10.30 – 11.00	**Coffee break**	
11.00 – 12.00	Demonstration of k_o Method	Ms. Sri Wardhani
12.00 – 13.00	**Lunch**	
Invited Speaker		**Chairperson**
13.00 – 13.30	Prof. Bangfa Ni (China) : Demonstration of NAA k_o method software, ADVNAA	Mr. Sutisna
13.30 – 14.00	Mr. Fumio Sasajima (Japan): Comparison of Chinese and European k_o Software	
14.00 – 14.30	**Coffee break**	

Appendix

Invited Speaker		Chairperson
14.30 -15.00	Dr. Yasuji Oura (Japan): Evaluation of CIAE-k_0 Software	Prof. Mitsuru Ebihara
15.00 – 15.30	Mr. Ho Manh Dung (Vietnam): Development of k_0-standardization of INAA (k_0-INAA) Regarding Software and Experiment in Dalat Research Reactor in Vietnam.	
15.30 – 16.00	General Discussion	

Paralel Session

Radioisotopes Group

Thursday, January 16, 2003		
Discussion		**Chairperson**
09.00 – 10.30	Discussion of Activity Report and Future Program	**Dr. T. Genka**
10.30 – 11.00	**Coffee Break**	
11.00 – 12.00	Demonstration on the production of $^{99}Mo/^{99m}Tc$ Generator based on PZC materials and neutron irradiated natural molybdenum	
12.00 – 13.00	**Lunch**	
13.00 – 14.00	Demonstration on the production of $^{99}Mo/^{99m}Tc$ Generator based on PZC materials and neutron irradiated natural molybdenum	
14.00 – 14.30	**Coffee Break**	
Report Preparation		**Chairperson**
14.30 – 15.10	Workshop Report Preparation	**Dr. T. Genka, Dr. A. Mutalib**
15.10 – 16.30	Workshop Report Preparation	**Dr. T. Genka, Dr. A. Mutalib**

Paralel Session

Neutron Activation Analysis Group

Thursday , January 16, 2003

	Round Table Discussion	**Chairperson**
09.00 – 10.00	Round Table Discussion on Activity Report and Future Program	Prof. Mitsuru Ebihara
10.00 – 10.30	**Coffee break**	
10.30 – 12.00	Round Table Discussion on Activity Report and Future Program	Prof. Mitsuru Ebihara
12.00 – 13.00	**Lunch**	
13.00 – 14.30	Workshop Report	Prof. Mitsuru Ebihara
14.30 – 15.00	**Coffee break**	
15.00 – 16.30	Workshop Report	Prof. Mitsuru Ebihara

Appendix

Plenary Session, Serpong

Friday, January 17, 2003		
Plenary Session		**Chairperson**
09.00 – 10.30	Round Table Discussion Workshop Summary Recommendation to the Coordinators Meeting	**Dr. Hudi Hastowo**
10.30 – 11.00	**Coffee Break**	
11.00 – 11.30	Closing Remarks: - Japanese Project Leader - Japanese FNCA Coordinator - Indonesian FNCA Coordinator	
11.30 --	**LUNCH**	

Appendix

List of Participants
2002 Workshop on the Utilization of Research Reactors
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国際単位系 (SI) と換算表

表 1　SI 基本単位および補助単位

量	名称	記号
長さ	メートル	m
質量	キログラム	kg
時間	秒	s
電流	アンペア	A
熱力学温度	ケルビン	K
物質量	モル	mol
光度	カンデラ	cd
平面角	ラジアン	rad
立体角	ステラジアン	sr

表 2　SI と併用される単位

名称	記号
分，時，日	min，h，d
度，分，秒	°，′，″
リットル	l，L
トン	t
電子ボルト	eV
原子質量単位	u

1 eV = 1.60218 × 10^{-19} J

1 u = 1.66054 × 10^{-27} kg

表 5　SI 接頭語

倍数	接頭語	記号
10^{18}	エクサ	E
10^{15}	ペタ	P
10^{12}	テラ	T
10^{9}	ギガ	G
10^{6}	メガ	M
10^{3}	キロ	k
10^{2}	ヘクト	h
10^{1}	デカ	da
10^{-1}	デシ	d
10^{-2}	センチ	c
10^{-3}	ミリ	m
10^{-6}	マイクロ	μ
10^{-9}	ナノ	n
10^{-12}	ピコ	p
10^{-15}	フェムト	f
10^{-18}	アト	a

表 3　固有の名称をもつ SI 組立単位

量	名称	記号	他の SI 単位による表現
周波数	ヘルツ	Hz	s^{-1}
力	ニュートン	N	m·kg/s^2
圧力，応力	パスカル	Pa	N/m^2
エネルギー，仕事，熱量	ジュール	J	N·m
工率，放射束	ワット	W	J/s
電気量，電荷	クーロン	C	A·s
電位，電圧，起電力	ボルト	V	W/A
静電容量	ファラド	F	C/V
電気抵抗	オーム	Ω	V/A
コンダクタンス	ジーメンス	S	A/V
磁束	ウェーバ	Wb	V·s
磁束密度	テスラ	T	Wb/m^2
インダクタンス	ヘンリー	H	Wb/A
セルシウス温度	セルシウス度	℃	
光束	ルーメン	lm	cd·sr
照度	ルクス	lx	lm/m^2
放射能	ベクレル	Bq	s^{-1}
吸収線量	グレイ	Gy	J/kg
線量当量	シーベルト	Sv	J/kg

表 4　SI と共に暫定的に維持される単位

名称	記号
オングストローム	Å
バーン	b
バール	bar
ガル	Gal
キュリー	Ci
レントゲン	R
ラド	rad
レム	rem

1 Å = 0.1 nm = 10^{-10} m

1 b = 100 fm^2 = 10^{-28} m^2

1 bar = 0.1 MPa = 10^5 Pa

1 Gal = 1 cm/s^2 = 10^{-2} m/s^2

1 Ci = 3.7 × 10^{10} Bq

1 R = 2.58 × 10^{-4} C/kg

1 rad = 1 cGy = 10^{-2} Gy

1 rem = 1 cSv = 10^{-2} Sv

（注）

1. 表 1 − 5 は「国際単位系」第 5 版，国際度量衡局 1985 年刊行による。ただし，1 eV および 1 u の値は CODATA の 1986 年推奨値によった。
2. 表 4 には海里，ノット，アール，ヘクタールも含まれているが日常の単位なのでここでは省略した。
3. bar は，JIS では流体の圧力を表わす場合に限り表 2 のカテゴリーに分類されている。
4. EC 閣僚理事会指令では bar，barn および「血圧の単位」mmHg を表 2 のカテゴリーに入れている。

換算表

力	N(=10^5 dyn)	kgf	lbf
	1	0.101972	0.224809
	9.80665	1	2.20462
	4.44822	0.453592	1

粘度　1 Pa·s(N·s/m^2) = 10 P(ポアズ)(g/(cm·s))

動粘度　1 m^2/s = 10^4 St(ストークス)(cm^2/s)

圧力	MPa(=10 bar)	kgf/cm^2	atm	mmHg(Torr)	lbf/in^2(psi)
	1	10.1972	9.86923	7.50062 × 10^3	145.038
	0.0980665	1	0.967841	735.559	14.2233
	0.101325	1.03323	1	760	14.6959
	1.33322 × 10^{-4}	1.35951 × 10^{-3}	1.31579 × 10^{-3}	1	1.93368 × 10^{-2}
	6.89476 × 10^{-3}	7.03070 × 10^{-2}	6.80460 × 10^{-2}	51.7149	1

エネルギー・仕事・熱量	J(=10^7 erg)	kgf·m	kW·h	cal(計量法)	Btu	ft·lbf	eV
	1	0.101972	2.77778 × 10^{-7}	0.238889	9.47813 × 10^{-4}	0.737562	6.24150 × 10^{18}
	9.80665	1	2.72407 × 10^{-6}	2.34270	9.29487 × 10^{-3}	7.23301	6.12082 × 10^{19}
	3.6 × 10^6	3.67098 × 10^5	1	8.59999 × 10^5	3412.13	2.65522 × 10^6	2.24694 × 10^{25}
	4.18605	0.426858	1.16279 × 10^{-6}	1	3.96759 × 10^{-3}	3.08747	2.61272 × 10^{19}
	1055.06	107.586	2.93072 × 10^{-4}	252.042	1	778.172	6.58515 × 10^{21}
	1.35582	0.138255	3.76616 × 10^{-7}	0.323890	1.28506 × 10^{-3}	1	8.46233 × 10^{18}
	1.60218 × 10^{-19}	1.63377 × 10^{-20}	4.45050 × 10^{-26}	3.82743 × 10^{-20}	1.51857 × 10^{-22}	1.18171 × 10^{-19}	1

1 cal = 4.18605 J (計量法)

= 4.184 J (熱化学)

= 4.1855 J (15 °C)

= 4.1868 J (国際蒸気表)

仕事率　1 PS (仏馬力)

= 75 kgf·m/s

= 735.499 W

放射能	Bq	Ci
	1	2.70270 × 10^{-11}
	3.7 × 10^{10}	1

吸収線量	Gy	rad
	1	100
	0.01	1

照射線量	C/kg	R
	1	3876
	2.58 × 10^{-4}	1

線量当量	Sv	rem
	1	100
	0.01	1

（86 年 12 月 26 日現在）

Proceedings of the FNCA 2002 Workshop on the Utilization of Research Reactors January 13-17, 2003 Jakarta & Serpong, Indonesia (Contract Research)