ION BEAM APPLICATION

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A brief review is presented on the ion beam application in science and technology. Ion beams are used very effectively in various fields of science and technology, on the basis of advance in accelerator technology and experimental techniques for ion beam utilization.

Recent progress in this field is reviewed in terms of the direct ion beam utilization like ion beam analysis, and the utilization of neutrons as secondary particles.

KEYWORDS : Ion Beam, Utilization, Accelerators, High Quality Beam, Intense Beam, Ion Beam Analysis, Secondary Particles, Neutrons

1. INTRODUCTION

In recent years, light and heavy ion beams provided by accelerators are used in various fields and now indispensable for the modern science and technology. These beams enable various new unique techniques which were not available with existing physical or chemical techniques. Such progress is essentially owing to the development of the accelerator technology and the experimental techniques for beam utilization. Nowadays, the accelerator can provide a variety of high quality beams with sufficient reliability, and sophisticated experimental techniques are available for efficient utilization of ion beams. Therefore, utilization beam utilization will be further extending to wider fields including exotic ones.

The ion beam utilization can be categorized into two types; one is the direct use of ion beam and the other one is the utilization of secondary particles like neutrons and muons and so on. The requirement for ion beams is different according to the way of utilization.

In the direct ion beam utilization, there are various kind of application like 1) high sensitive elemental analysis with very small amount of test samples, 2) analysis of material configuration and depth profile, 3) material modification and production of new materials, 4) medical diagnosis and cancer therapy by protons and heavy ion beams, and 5) irradiation of reactor materials and microelectronics devices for development of radiation resistant material/devices. For these applications, high quality and/or well-controlled beam is more important than an intense beam: For example, a micro beam which is very effective in particular for the ion beam analysis of bio-samples requires a high quality beam with low dispersion, and the spot-scanning technique used to obtain a uniform beam in medical treatment needs a sophisticated beam control. Besides, downsizing of the accelerator system is highly desired to enable the utilization in a limited compact space with a smaller capital cost. Therefore, in the area of ion beam application, great effort has been taken to develop a compact accelerator like a "table-top accelerator" with high performance.

Secondary radiations of neutrons, radioactive beam, muons, neutrinos and so on are also used over a wide area. In particular, neutrons are of special importance both in basic science and application fields because neutrons are sensitive to light elements such as hydrogen and carbon in bio-samples which are difficult to detect with X-rays, and also useful to study of magnetic property of materials through a magnetic-moment of them. Furthermore, accelerator-based pulsed neutron sources are of special advantage over steady-state reactor-based neutron source because it enables high-precision and efficient analysis through the TOF (Time-of-Flight) method in neutron scattering studies. Neutrons also have a crucial role also in the energy area like accelerator-driven system (ADS) which transmutes minor-actinides and long-lived fission products into stable nuclides producing electricity. Neutron irradiation of fission reactor and fusion reactor materials is also important task for nuclear energy development. Radioactive beams are of increased importance for the basic study of nuclides far from the β-stable line in relation with the nucleo-synthesis and nuclear transmutation. For production of such secondary beams with required intensity, the power or the intensity of the primary beam is essential as well as the target technology to manage such high power beam.

For the reasons, intense neutron sources have been installed and utilized very efficiently in US, EU, Russia and in Japan. In recent years, demand for stronger neutron sources are quiet serious, and new neutron sources in MW class are under construction in US, Japan and Korea, and in plan in other countries. Such intense accelerators become possible owing to development of technologies on ion sources, high current low energy accelerator (i.e, radiofrequency quadrupole) and improvement of energy efficiency and beam transmission in high energy accelerators. Target technology for effective heat removal and optimum neutronics design are also essential for achievement of advanced neutron sources.

This paper presents a brief review of the recent progress in ion beam application in terms of
(1) intense neutron sources and (2) ion beam application on elemental analysis which has wide applications.

2. INTENSE NEUTRON SOURCE

2.1 Spallation Neutron Source

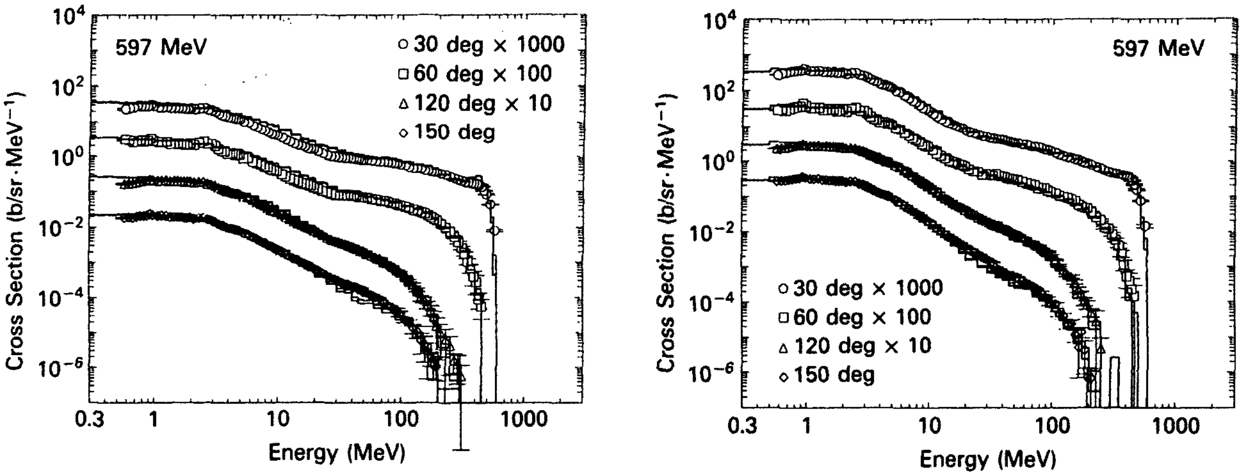
To reply the requirement for intense neutron sources, long standing endeavor has been devoted for the achievement of high beam power and efficient neutron production. For the neutron production, the energy efficiency in neutron production is very crucial to increase the neutron intensity. Table 1 compares the comparison of energy efficiency in neutron production [1,2]. The table indicates that spallation reactions have great advantage of high efficiency in neutron production which means the lowest heat deposition per neutron in the neutron production target. For the reason, the spallation reaction between a high-Z element target and an intense proton beam in the energy region higher than ten's of MeV is a most efficient neutron sources except for the case in which high energy neutrons are required.

For the reasons, a spallation neutron source has highest neutron intensity for a given beam power. In the neutron

Table 1. Neutron Yields and Deposited Heat for Some Neutron Production Reactions [1,2]

Reaction	Neutron yield n / particle	Deposited heat MeV/n
T(d,n) (Ed=0.2 MeV)	8×10^{-5} n/d	2,500
W(e,n) (Ed=35 MeV)	1.7×10^{-2} n/e	2,000
^{9}Be(d,n) (Ed=15 MeV)	1.2×10^{-2} n/d	1,200
^{235}U(n,fission)	~ 1.0 n/fission	200
(T,d) fusion	~ 1.0 n/fusion	3
Pb Spallation (Ep=1 GeV)	20 n/p	23
^{238}U Spallation (Ep=1 GeV)	40 n/p	50

* The yield per fission event is 2.4 but ~1.4 neutrons are required to maintain the reaction and compensate for parasitic losses.



(g) Fe(*p,xn*)

(h) Pb(*p,xn*)

Fig. 1. Thick Target Neutron Spectra from Spallation Reaction of 597 MeV Protons on Iron (left) and Lead (right) [3]

scattering/diffraction facilities and ADS facilities, neutrons are utilized as thermal or cold neutrons after moderation. The spallation source is advantageous also for the production of moderated neutrons because the spectrum of neutrons from heavy elements consist of, as shown in Fig.1, neutrons with an evaporation spectrum peaked around a few MeV although there are also neutrons with higher energies up to incident energies. This feature favors the design of target-moderator configuration.

As shown in Table 2, several spallation sources have been successfully operated for neutron scattering works [4-7], and now two very strong new generation sources, SNS and J-PARC, are under construction. The SNS (Spallation Neutron Source; Fig.2) [8] is under construction in ORNL and the construction of J-PARC (Japan PArticle accelerator Research Complex; Fig.3) [9] is under way in Tokai site of Japan Atomic Energy Agency (JAEA) by the collaboration of High Energy Accelerator Organization (KEK) and JAEA. The SNS will serve almost exclusively as a neutron source for neutron scattering, but J-PARC is a multipurpose accelerator complex including a nuclear/particle physics laboratory and an experimental nuclear transmutation facility (2nd phase; Fig.4). The beam powers of SNS and J-PARC reach a MW region which is not yet experienced thus far. For effective heat removal from the target, they will employ a liquid mercury target for neutron production.

The proton accelerator in the PFEP (Proton Frontier Engineering Project; Fig.5) project in Korea [10] is also under construction by KAERI for multi-purpose proton utilization and as a pilot machine for a large scale spallation source for ADS.

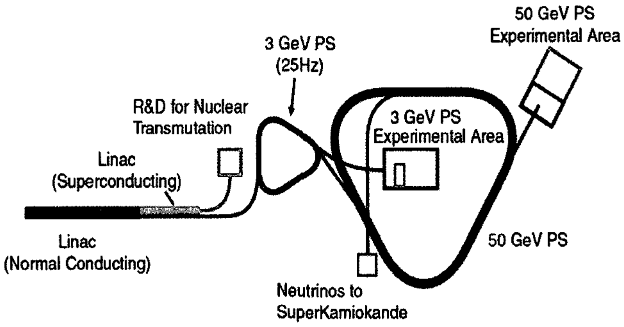


Fig. 3. Schematic View of the J-PARC Facility [9]

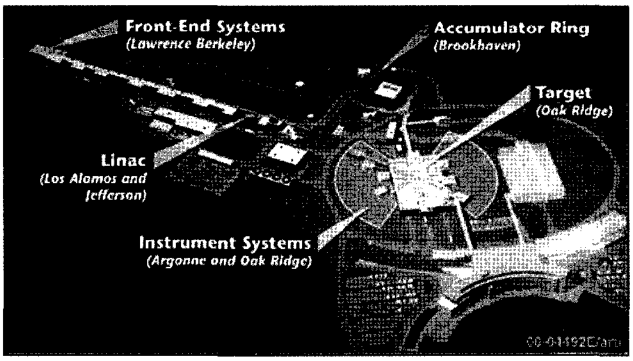


Fig. 2. Layout of SNS [8]

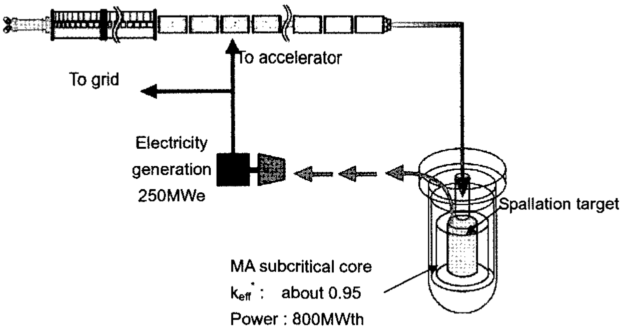


Fig. 4. Schematic of Accelerator-driven Nuclear Transmutation System [9]

Table 2. Spallation Neutron Source in the World [4-10]

Facility	Proton energy	Proton current	Beam power	Ref.
IPNS	0.45 GeV	18 μA	8.1 kW	4
LANSCE	0.8 GeV	100 μA	80	5
ISIS	0.8 GeV	200 μA	160 kW	6
KENS	0.8 GeV	200 μA	160 kW	7
SNS*	1.0 GeV	2000 μA	2 MW	8
J-PARC*	3.0 GeV	330 μA	1 MW	9
PEFP*	100 MeV	20,000 μA	2 MW	10

* under construction

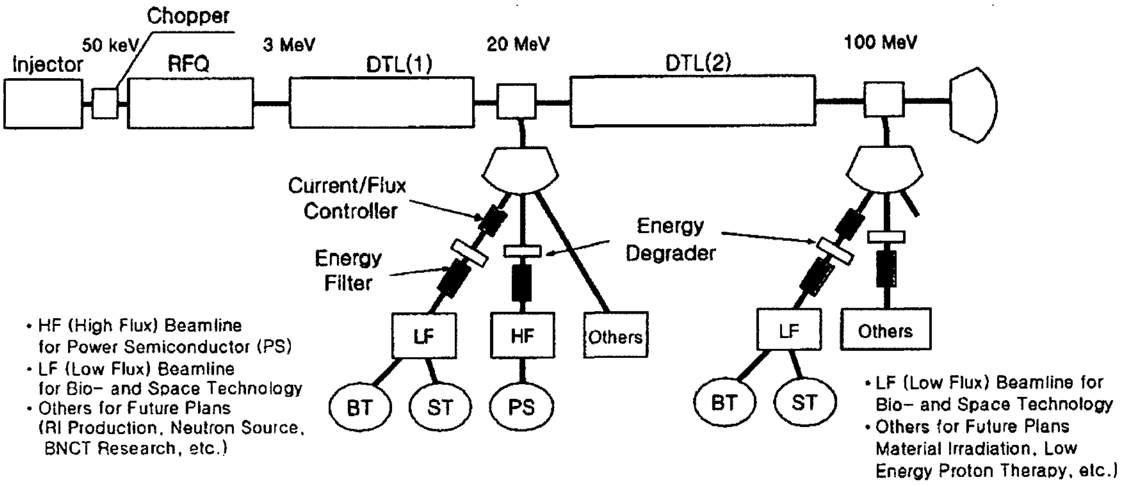


Fig. 5. Schematic View of Proton Accelerator in PEFP Project [10]

In these facilities, very high neutron flux will be achieved. To take fully the advantage of such a high neutron flux, development of experimental apparatus is required toward faster response, faster recovery and higher counting rate of radiation detectors together with multiple information an radiations. This is also very challenging task.

2.2 (d,n) Neutron Sources with Hard Spectrum

On the other hand, the rather soft neutron spectrum of spallation source is not appropriate for neutron irradiation of fusion reactor materials or production of radioactive beams by using energetic neutrons. For these purposes, (d,n) neutrons with a light element target is more favorable due to higher energies of neutrons. Figure 6 illustrates the angle-dependent neutron spectrum of the Li(d,n) reactions for 40 MeV deuterons [1,11], which are considered as the primary neutron source in IFMIF (International Fusion Reactor Material Irradiation Facility).In IFMIF, two linear accelerators of 40 MeV and 125 mA will deliver a deuteron beam on a liquid lithium target [12]. The source has peak a around 14 MeV and can be used to simulate radiation effect in the d-T fusion reactor with an intense neutron flux comparable with actual fusion reactors. This source is only one that can satisfy the required neutron flux but has deficiencies of too strong angular dependence, broad spectrum and the existence of high energy tail, which leads to requirement of further studies on the correspondence between the radiation effects in the neutron field and that in the actual fusion reactor environment.

A similar (d,n) source is under consideration for neutron production of neutron-rich radioactive beam using fast neutron induced fissions.

2.3 Neutron Sources for Medical Application

Thermal and epithermal neutrons are used successfully for therapy of cancers like brain-tumor and melanoma and so on in the reactor-based neutron fields. Recently, an accelerator-based neutron source is highly required for better access and an optimal neutron spectrum. However, such systems have not been realized yet because high neutron flux required for treatment ($\sim 10^9$ n · $cm^{-2}s^{-1}$) is very difficult to achieve with existing low energy

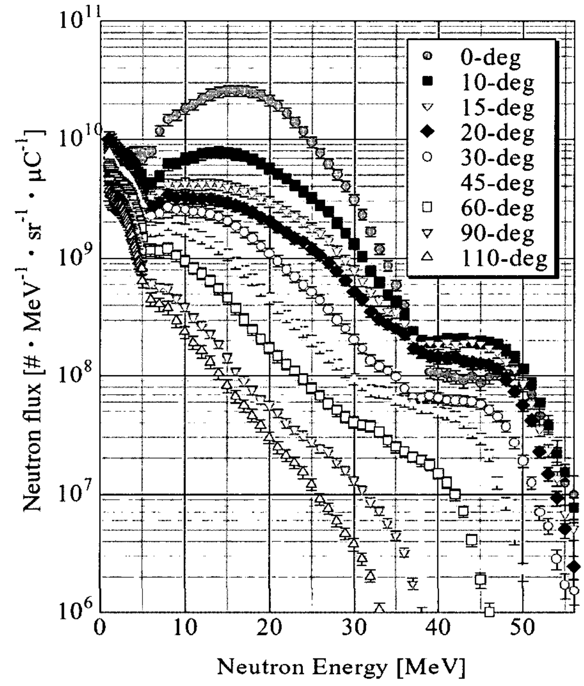


Fig. 6. Neutron Spectra from the Li(d,n) Reaction with Thick Target [1, 11]

accelerators. As a one solution to the problem, the author's group proposed an idea to employ a spallation reaction for the neutron production [13,14] and reported neutronics design of the target/moderator assembly [13] and an engineering design of the neutron production target [14] obtaining very promising result. This idea takes advantage of high energy efficiency in neutron production and a low fraction of high energy neutrons in spallation reactions.

3. ION BEAM ANALYSIS

3.1 Elemental Analysis Using RBS and PIXE Technique

Ion beams analysis is utilized over a variety of area, e.g., environmental study, bio- and life-science, material analysis, archaeology and so on. The fact that a small accelerator in a few MV range is enough for these application is advantageous for such prevalence of the technique [15].

Figures 7 shows a typical example of experimental setup for the ion beam analysis using Rutherford Back Scattering (RBS) and Elastic Recoil Detection (ERDA) [15]. Figure 8 illustrates a typical spectrum of RBS analysis. In the RBS and ERD methods, the energy spectrum of backscattered or recoiled particle are measured, and information on the element and/or its depth profile can be obtained from the energy of particles in the material.

In the case of particle-Induced X-ray Emission (PIXE), elemental analysis can be done from the energy spectrum of characteristic-X-rays emitted from the sample bombarded by protons or heavier particles, as shown in Fig.9 [15]. Owing to large cross section of particle-induced X-ray emission, the measurement can be done rapidly with a small amount of sample materials in the order of nano-g [16,17]. In addition to high-sensitivity, capability of nondestructive and multi-elemental analysis is the great advantage of PIXE over chemical methods. One defect of PIXE is that it is insensitive to light elements than oxygen. It can be complemented by the combination with the RBS technique as shown below. The charged-particle activation technique is also used for analysis of light elements [15].

The important advantage of ion beams is that they can be focused very finely down to micro-m or lower (micro beam technique). Therefore, by use of micro-beam and PIXE, elemental distribution can be determined for each element in the cell scale. This technique is used very effectively to trace the elemental distribution in biomedical samples to trace the relation between element and disease. Figure 10 illustrates the system of Tohoku University group

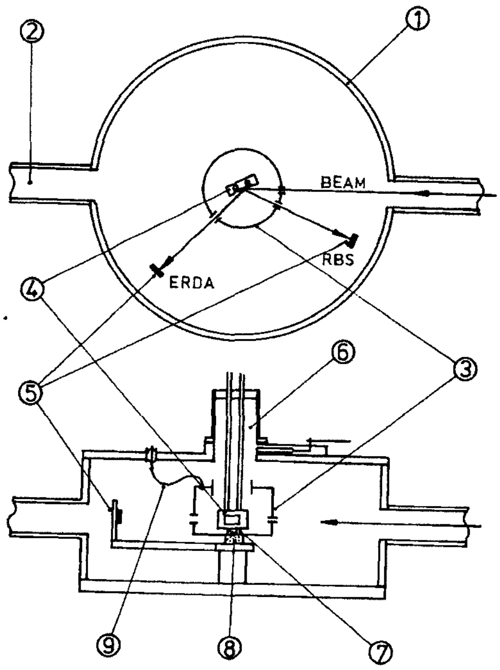


Fig. 7. Experimental Arrangement for RBS and ERD [15]

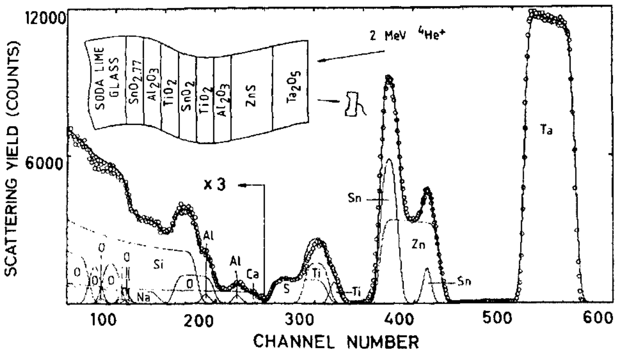


Fig. 8. Typical Example of the Particle Spectrum in RBS Experiment [15]

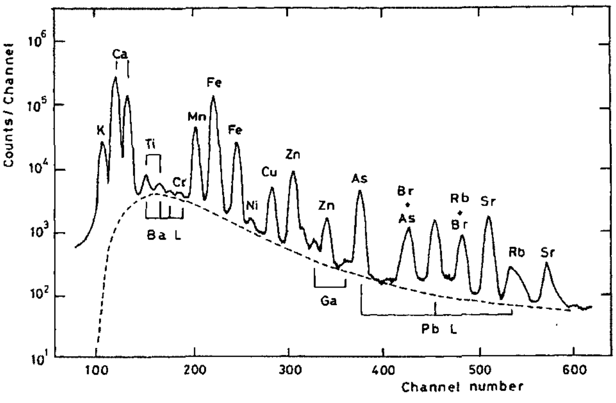


Fig. 9. Example of X-ray Spectrum in PIXE Experiment [15]

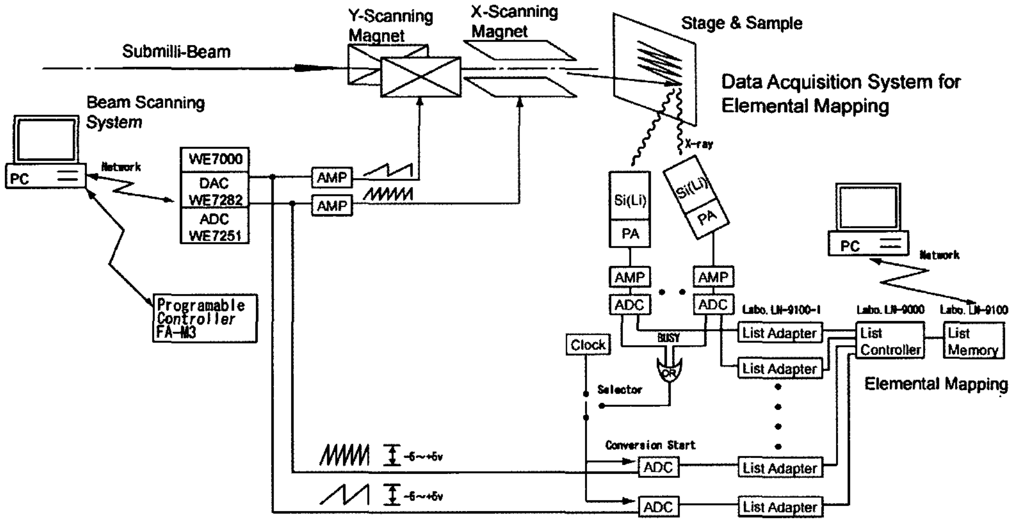


Fig. 10. Experimental Arrangement for Elemental Mapping Measurement Using Beam Scanning Technique [16-17]

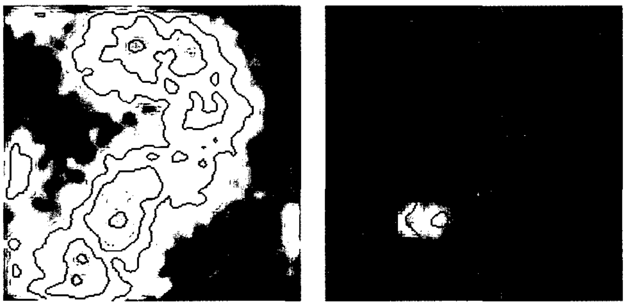


Fig. 11. Example of the Element Mapping Results for Bovine Aortic Endothelial Cells; Phosphorus (Leftt) and Iron (Right)

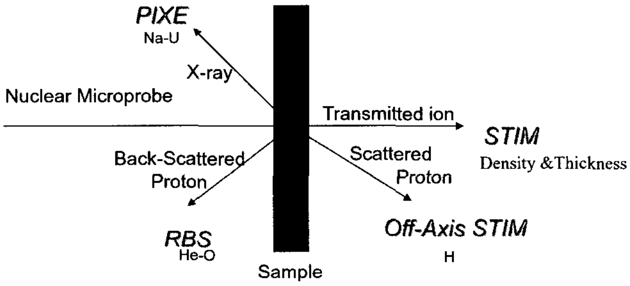


Fig. 12. Experimental Arrangement to Obtain Multiple Information from Combined Measurement of PIXE, RBS and Transmitted Ions [16-17]

for two-dimensional element trace by use of micro-beam [16,17], and Fig.11 shows the example of elemental mapping for phosphorus and iron [16,17].

Figure 12 illustrates the schematic of the experimental arrangement for the PIXE-RBS-Transmission combined technique. In this arrangement, the information on elements lighter than oxygen can be obtained with RBS, and the thickness distribution can be determined for each point of the sample by using the energy data of transmitted particles even if the sample thickness is not uniform. Therefore, this combination provides the data on mapping of almost all elements as a function of sample position as well as the thickness [16,17].

For biomedical samples, analysis in air is necessary to avoid dry up. For the reason, the analyzing system in air was developed, and the above mentioned analysis was done by using the system [16,17].

PIXE can be extended to analysis of chemical state of materials using inner-shell ionization induced by heavy ion beams [18].

3.2 Accelerator Mass Spectrometry

The accelerator-mass spectrometry (AMS) is also high-sensitive mass spectrometry which enables us to determine the isotopic abundances of long-lived radionuclide to extremely low level, 10^{-10} to 10^{-12}. In this technique, the sample atoms are ionized in the ion source and accelerated to some energy by using a tandem electrostatic accelerator and so on [19,20,21]. The accelerated particle is mass analyzed by using magnetic and/or electrostatic filed as shown in Fig.13 [19,20]. This technique was developed in 1977 for searching of heavy hydrogen isotopes and for carbon dating. Now it is employed for analysis of radionuclides with very low concentration, ^{10}Be, ^{14}C, ^{26}Al, ^{36}Cl

Table 3 Typical Radioisotope-to-stable Isotope Ratio of AMS Targets [19,20]

Nuclides	Ratio	Ratio values
^{10}Be	^{10}Be/ ^{9}Be	~1 x 10^{-11} (rock) ~10^{-9} (sea water) ~10^{-6} (extra-terrestrial)
^{14}C	^{14}C/ ^{12}C	~1 x 10^{-12} (modern carbon)
^{26}Al	^{26}Al/ ^{27}Al	~10^{-14} - 10^{-13} (rock) ~10^{-9} (extra-terrestrial)
^{36}Cl	^{36}Cl/ Cl	~1 x 10^{-15} (sea water) ~10^{-12} (rock) ~10^{-9} (extra-terrestrial)
^{129}I	^{129}I/ ^{127}I	~10^{-12} (atmosphere; natural) >10^{-13} (anthropogenic; seawater) >10^{-9} (modern soil)

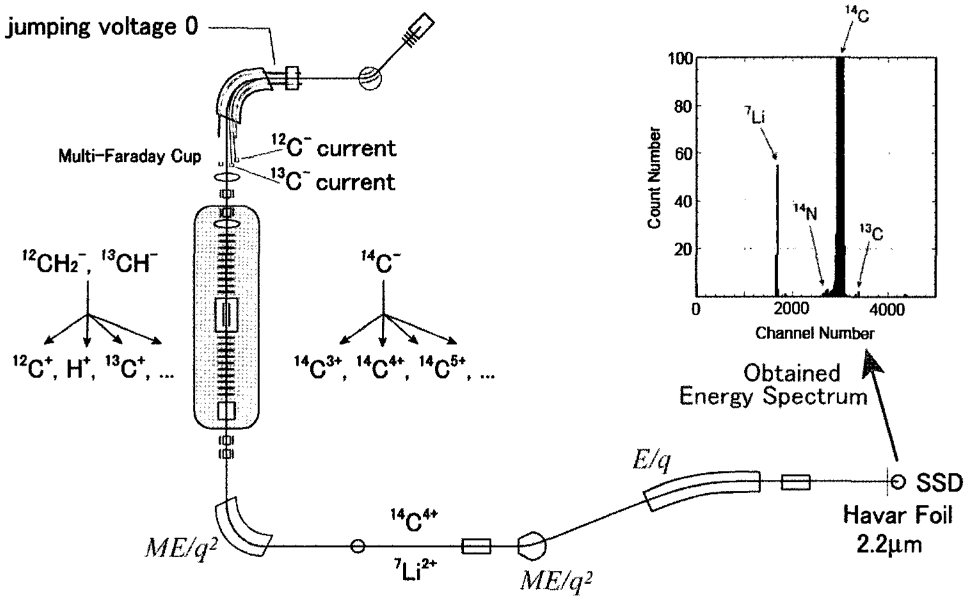


Fig. 13. Experimental Arrangement for AMS [19-20]

and ^{129}I. These nuclides are formed by spallation reactions of cosmic-rays with oxygen and silicon. Therefore, the isotopic- ratio of these isotopes includes various information in the geology scale.

In the case of ^{14}C, the AMS technique enables dating with much less sample amount (~1mg) and within shorter period compared with the traditional β-counting technique. The sensitivity of ^{14}C detection in AMS is higher than the β-counting technique by about 10^{6} [19]. The dating using ^{10}Be is applicable to older ages in the order of ten's of thousand years because the half life of ^{10}Be is much longer that ^{14}C and its production rate is rather high.

Originally, AMS employed a cyclotron and a tandem accelerator for nuclear physics experiment to achieve mass resolution required. However, it proved that lower beam energy was enough for AMS except for some cases like ^{36}Cl and so on. For the reason, at present, about one half of AMS accelerators are the electrostatic ones of 3 MV in terminal voltage, and that of 1 MV terminal voltage is successfully employed for AMS of ^{14}C, ^{10}Be, ^{26}Al, ^{129}I and ^{244}Pu [19]. Further, downsizing of the accelerator is now on going, and "table-top accelerator" with 0.5 to 0.25 MV terminal voltage was developed (Fig.14). Downsizing of accelerators will lead to further extension of the AMS utilization.

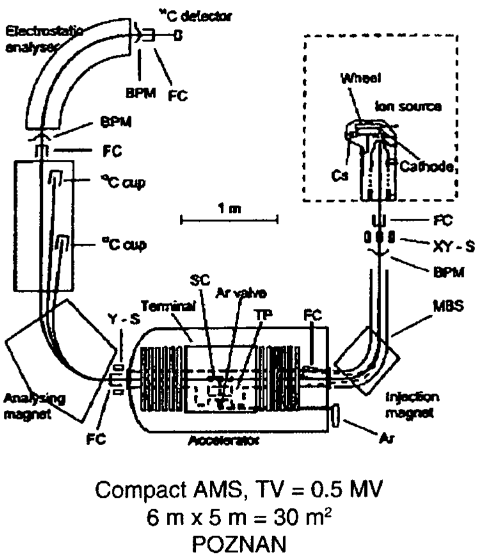


Fig. 14. Down-sized AMS System of 0.5 MV [19]

4. SUMMARY AND OUTLOOK

As described above, ion beam utilization is indispensable as one of the basic infrastructure in modern science and technology. It will find further filed of application and extend over various fields along with the technical development in accelerator technologies and beam utilization.

In the nuclear engineering field, development of instrumentation and radiation measurement method should be promoted to support and promote the works and also to open new possibility of application.

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REFERENCES

[1] S.Cierjacks (Ed) "Neutron Sources For Basic Physics and Applications", (An OECD/NEA Report), Pergamon Press (1983)

[2] IAEA-TEDOC-**410** "Properties of Neutron Sources" (Proceedings of an Advisory Group Meeting on Properties of Neutron Sources organized by IAEA 1986)

[3] W.B.Amian, R.C.Byrd, D.A.Clark, C.A.Goulding, M. M.Meir, G.L.Morgan, and C.E.Moss, "Differential Neutron Production Cross Sections for 597 -MeV Protons", *Nucl. Sci. Eng.*, **104**, 339 (1990)

[4] http://www.pns.anl.gov/

[5] http://lansce.lanl.gov/

[6] http://www.isis.rl.ac.uk/

[7] http://neutron-www.kek.jp/index_e.html

[8] http://www.sns.gov/, SNS, Oak Ridge National Laboratory

[9] http://j-parc.jp/, J-PARC, Japan Atomic Energy Agency/ High Energy Accelerator Research Organization (KEK)

[10] http://www.kaeri.re.kr/npet/ccc_eng/index.htm

[11] M.Hagiwara, T.Itoga, N.Kawata, N.Hirabayashi, T.Oishi, T.Yamauchi, and M.Baba, "Measurement of Neutron Emission Spectra in Li(d,xn) Reaction with thick and Thin Targets for 40 MeV- Deuterons", *Fusion Sci. & Technol.*, **48** (1320-1328) 2005

[12] http://www.frascati.enea.it/ifmif/

[13] S.Yonai, T.Aoki, T.Nakamura, H.Yashima, M.Baba, H.Yokobori, Y.Tahara, "Feasibility Study on epithermal neutron field for cyclotron-based boron neutron capture therapy", Med. Phys. **30**(8), 2021-2030 (2003)

[14] Y. Tahara, Y. Oda, T. Shiraki, T. Tsutsui, H. Yokobori, S. Yonai, M. Baba, T. Nakamura, "Engineering Design of a Spallation Reaction-Base Neurton Generator for Boron Neutron Capture Therapy", *J. Nucl. Sci. Technol.*, **43** (1) 9-19 (2006)

[15] Zeev. B.Alfassi and Max Peisach (Ed) "Elemental Analysis by Particle Accelerators", CRC Press (1992)

[16] S.Matsuyama, K.Ishii, H.ohtsu, H.Yamazaki, Y.Kikuchi, Ts.Amartaivan, K.Inomata, Y.Watanabe, A.Ishizaki, Y.Barbotteau, A.Suzuki, T.Yamaguguchi and G.Momose, "Microbeam analsyisis at Tohoku University for biological studies", *Int. J. PIXE* **15** No.1-2, 41-45 (2005)

[17] S.Matsuyama, K.Ishii, H.Yamazaki, Y.Kikuchi, Ts. Amartaivan, S.Abe, K.Inomata, Y.Watanabe, A. Ishizaki, R. Oyama, Y. Kawamura, A. Suzuki and G. Momose, T.Yamaguchi and H.Imazeki, "Microbeam analsyisis of aerosol particles at Tohoku University", *Int. J. PIXE* **15** Nos.3&4, 257-262 (2005)

[18] H.Yamazaki, K.Ishii, Y.Takahashi, S.Matsuyama, Y.Kikuchi, Ts. Amartaivan, T.Yamaguchi, G.Momose, S.Abe, K.Inomata, Y.Watanabe, A.Ishizaki, "Identification of Qxidation States of Trace-Level Arsenic in Environmental Water Samples Using PIXE", *Int. J. PIXE* **15**, Nos.3&4, 241-248 (2005)

[19] W. Kutschera "Progress in isotope analysis at ultra-trace level by AMS" Int. J. of Mass Spectrometry 242 (2005) 145-160

[20] http://www.malt.rcnst.u-tokyo.ac.jp/indexj.html (MALT facility, Univ. of Tokyo)

[21] H.Matsuzaki, Private communication