Available online at www.sciencedirect.com
SCIENCE DIRECT®
ELSEVIER
Nuclear Instruments and Methods in Physics Research B 210 (2003) 42–47
NIM B
Beam Interactions with Materials & Atoms
www.elsevier.com/locate/nimb

The scanning microbeam PIXE analysis facility at NIRS

Hitoshi Imaseki [a,*], Masae Yukawa [a], Frank Watt [b], Takahiro Ishikawa [a], Hiroyuki Iso [a], Tsuyoshi Hamano [a], Kenichi Matsumoto [a,c], Nakahiro Yasuda [a]

[a] National Institute of Radiological Sciences, Chiba 263-8555, Japan
[b] Research Centre for Nuclear Microscopy, Department of Physics, National University of Singapore, Singapore 119260, Singapore
[c] Faculty of Science, Toho University, Chiba 274-8510, Japan

Abstract

In March 1999, a HVEE Tandetron was installed in the Electrostatic Accelerator Building of National Institute of Radiological Sciences (NIRS) for particle induced X-ray emission (PIXE) analysis. The specifications of the Tandetron accelerator system operating at NIRS are as follows: the accelerating voltage is 0.4–1.7 MV, and the maximum beam current is 500 nA at 3.4 MeV. The accelerator facility incorporates three beam lines for conventional, in-air and microbeam PIXE analysis.

The scanning microbeam PIXE analysis line is based around an Oxford Microbeams OM2000 nuclear microscope end stage. This system provides the ability of multi-elemental mapping over sample areas up to 2×2 mm area with spatial resolutions routinely at 1×1 μm. The scheduled operation of this facility started in April 2000 and is controlled by the Division of Technical Service and Development. The result of beam resolution tests carried out in 2001 are as follows: for scanning transmission ion microscopy, the estimated beam size is 100×200 nm, measured using a 2.6 MeV proton beam scanned over a 12.7 μm repeat distance copper grid. For PIXE operation at 50 pA beam current the estimated best spot size is 0.4×0.6 μm. The microbeam facility is being used for research into the elemental distribution of small biological samples such as biological cells and tissue.

Keywords: PIXE; Particle; Microbeam; Mapping; Elemental distribution

1. Introduction

The National Institute of Radiological Science (NIRS) was established in 1957 as a special research institute, attached to the Science and Technology Agency of Japan. NIRS discarded the Van de Graaff accelerator (Model KM3000, High Voltage Engineering) in 1997 that had been operated as a device for pre-tumor therapy by neutron and particle induced X-ray emission (PIXE) analysis from 1961. In March 1999, an electrostatic accelerator, an HVEE Tandetron, was installed in the Electrostatic Accelerator Building for PIXE analysis. PIXE analysis is a trace analytical method, which has the advantage of simultaneous determination of elemental concentrations in a small specimen without any chemical separation by measuring characteristic X-rays of elements induced by accelerated charged nuclear particles such as protons.

* Corresponding author.
E-mail address: h_ima@nirs.go.jp (H. Imaseki).

0168-583X/$ - see front matter © 2003 Elsevier B.V. All rights reserved.
doi:10.1016/S0168-583X(03)01002-4

In this paper, characteristics of the new PIXE system are described [1].

2. System specifications

2.1. Electrostatic accelerator system

The installed accelerator for PIXE analysis is the Tandetron (Model 4117 MC) manufactured by High Voltage Engineering Europe Co. The accelerating voltage is 0.4–1.7 mV, and the maximum beam current for protons is 5 μA at 3.4 MeV. This system has three beam lines designed for different types of PIXE analysis: conventional, in-He and scanning microbeam [2].

2.2. Conventional PIXE analysis line

The beam port for normal PIXE can supply beams of 0.5–2.0 mm^2 with currents of 10–100 nA. The beam size can be regulated with a set of X–Y slits. Since two types of X-ray detectors, Si (Li) and CdZnTe detectors, are available here, elements from Na ($Z = 11$) to U ($Z = 92$) are detectable. Fifteen samples can be mounted and analysed semi-automatically using computer control. Each sample is attached to the center of a hole of 20 mm diameter on a 25 mm × 35 mm aluminum plate with an appropriate backing film. The detectors are set at 135° with respect to the incident ion beam [2].

2.3. In–He PIXE analysis line

The In–He PIXE analysis system was commissioned in March 2001, and is connected to the second beam port. The In–He beam line is used for irradiation of wet samples using a proton beam of approximately 2 mm diameter. The proton beam, which passes from the vacuum through a Kapton film, enters the analysis chamber filled with helium gas at 1 atm [2].

2.4. Scanning microbeam PIXE analysis line

The layout of microbeam scanning system (Model OM2000, Oxford Micro Beams, Ltd.) installed at NIRS is illustrated in Fig. 1. A proton beam is focused to the micron level by passing the beam through a coarse pre-slit of 0.5 mm diameter, and two more precise sets of X–Y slits that serve as object and collimator apertures [3]. The beam is focused using a set of quadrupole triplet magnets, and scanned over the sample using a scanning coil positioned before the lens system. This system enables multi-elemental mapping over a 2 mm × 2 mm area with a spatial resolution of about 1 μm. Fig. 2 shows the chamber for the scanning microbeam. Characteristics of the system are

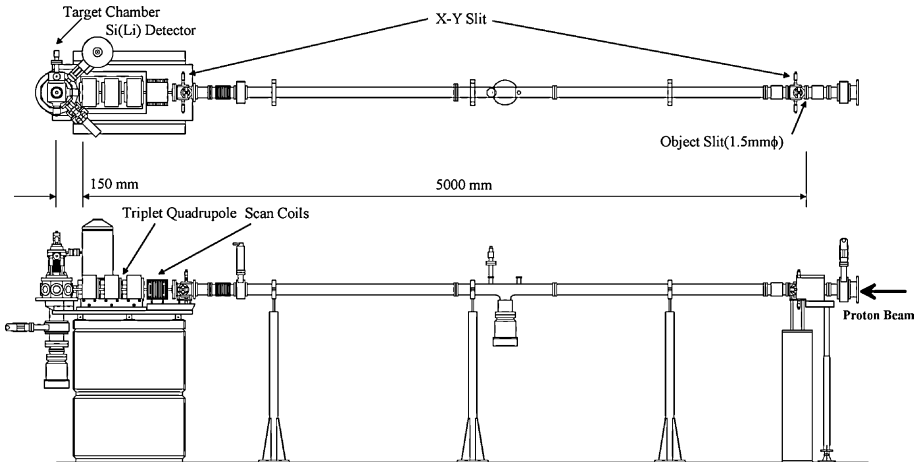

Fig. 1. Layout of scanning microbeam line.

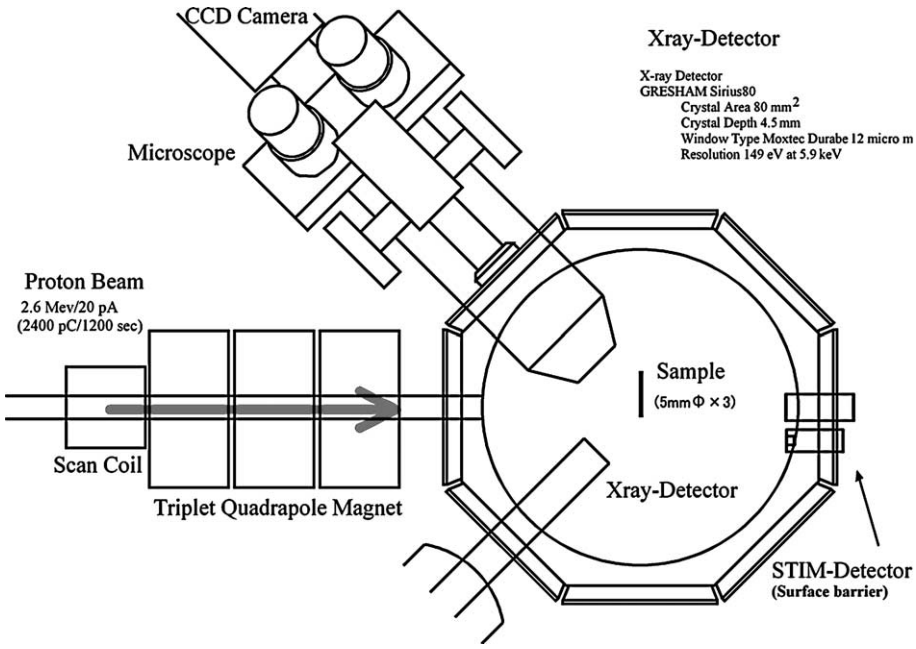

Fig. 2. Scanning microbeam PIXE chamber layout.

Table 1
Characteristics of microbeam PIXE system

Optical beam size	0.1 mm × 0.1 mm–1.0 mm × 1.0 mm	No focusing by quadrupole magnet
Beam currents	10–50 pA	
Scanning area	10 μm^2–2.0 mm^2	
Sample holder	Five samples	Sample is set at 5 mm diameter hole of 10 mm × 23 mm steel plate
Detector	Si (Li) GRESHAM Sirius80	Set at an angle of 135° from the beam line Crystal area: 80 mm^2 Crystal depth: 4.5 mm Window type: Moxtec Durabe 12 μm Energy resolution: 149 eV at 5.9 keV

summarized in Table 1. This apparatus is useful for a research into the elemental and structural analysis of small samples, for example a cell [2].

2.5. Scanning transmission ion microscopy

The STIM system was attached to the microbeam scanning line in March 2001. The function of this apparatus is illustrated in Fig. 2. An energy-loss spectrum of transmitted proton can be obtained using a surface barrier Si detector behind the sample. Since the energy-loss of transmitted protons depends on thickness and density of the sample, by scanning the sample surface with proton beams, we can get the energy-loss map to image the structure of a thin sample (up to 30 μm) with this system. Thus, we can observe simultaneously the structure of samples in the scanning area during PIXE analysis.

3. The results of beam resolution tests

3.1. First step in the year of 2000

Fig. 3 indicates the result of beam resolution tests carried out in February 2000 during the

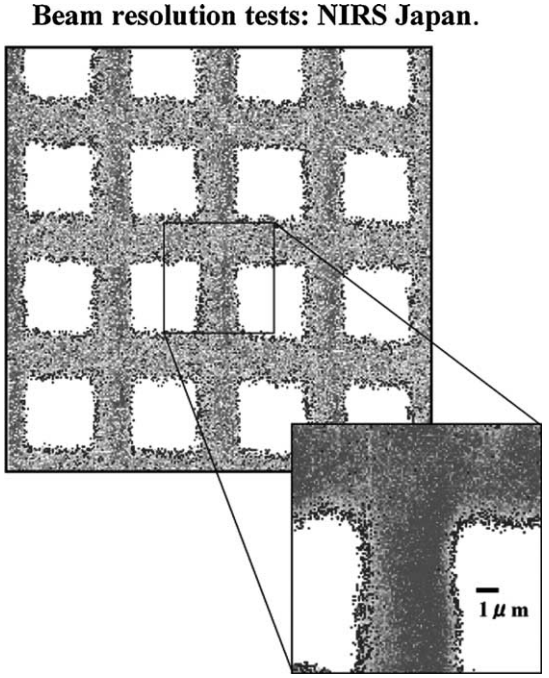

Fig. 3. PIXE scan of 2000 lines per inch Au grid. Proton energy 3.0 MeV; scan sizes 50 μm; resolution estimated beam sizes are 400 nm (horizontal) and 600 nm (vertical).

system installation. The beam size was estimated to be 0.4×0.6 μm with 12.7 μm repeat distance gold grid for 3 MeV protons. The beam current was 50 pA, the scan size was 5 μm^2 and the irradiation time was 20 min.

3.2. Second step in the year of 2001: the installation of scanning transmission ion microscopy

Fig. 4 indicates the result in December 2001. The estimated beam size is 100×200 nm, measured using a 2.6 MeV proton beam scanned over a 12.5 μm repeat distance copper grid. The beam size measurement by using the PIXE and the STIM images is carried out in the same way with the Oxford method [3,4].

3.3. Measurement beam size: use of CR39 in August 2002

Fig. 5 shows an atomic force microscope (AFM) image on CR-39 (TNF-1, thickness: 0.9 mm), which was irradiated with the microbeam at $\sim 10^6$ ions. On an area of 40×40 μm, pin-point irradiation was

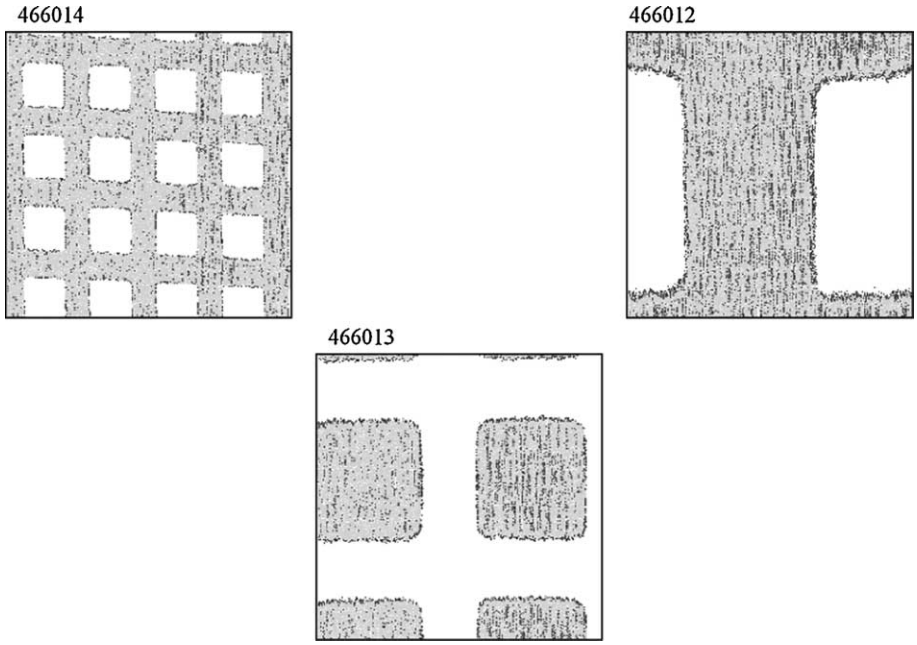

Fig. 4. STIM test by using 2000 lines per inch Cu grid. Proton energy 2.6 MeV; scan sizes 50 μm; resolution estimated visually approximately 100 nm (horizontal) and 200 nm (vertical).

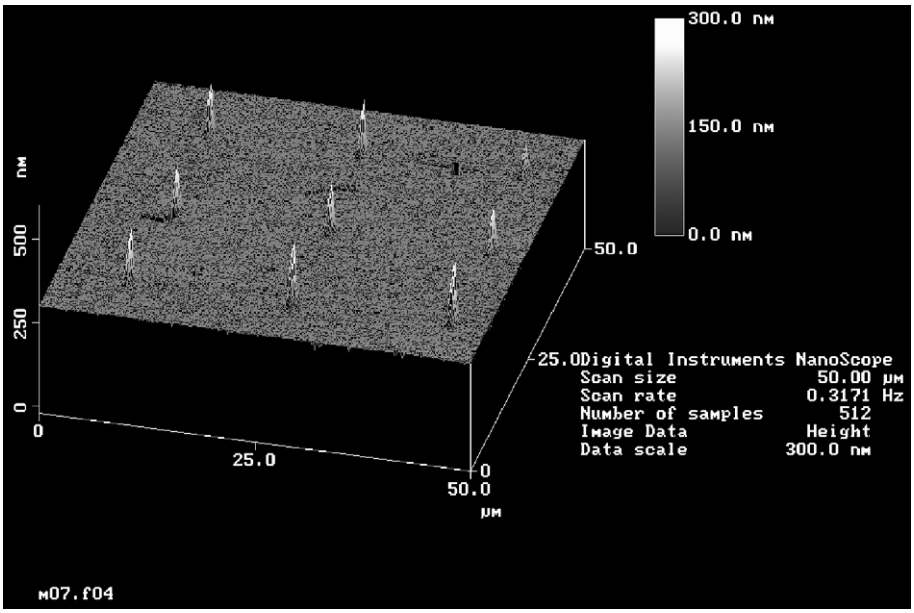

Fig. 5. AFM image of etched-pits on a CR-39 film. The CR-39 film is irradiated with the microbeams at $\sim 10^6$ ions. These pits are formed by etching for 1 min in 7 N sodium hydrate solution. So the etching time is very short, it is considerable that the pits sizes are almost equivalent to the real beam size. On the area of 40×40 μm, pinpoint irradiation is made by shifting the CR-39 film at the intervals of 20 μm. As the each grid distance of the etched-pits is measured 20 μm by using the scale on the AFM, we can determine that the aiming accuracy of the scanning microbeam PIXE analysis line is good.

made by shifting the CR-39 at intervals of 20 μm. In these images, the beam-halo is not shown. The spot size measured in this way was 0.8×1.5 μm.

4. Future research plan: microbeam irradiation of cells

The use of high energy ion microbeams is a new avenue of radiation research especially in radiation biology and radiation protection. For this purpose, we have started the construction of a new microbeam irradiation facility (named as SPICE) which will be connected to our Tandem accelerator (3.4 MeV $^2H^+$ and 5.1 MeV $^4He^{2+}$). For our primary goal, ''irradiation of cell organelles by a single high energy ion within a position resolution of 2 μm in a reasonable irradiation time'', special features must be considered. Using a 90° magnet, a vertical beam line will be branched from the present microbeam PIXE beam line. The focusing system will be installed in a cradle which is hung on a rigid frame structure. At the end of the vertical beam line will be mounted a focusing triplet quadrupole-magnet system (Oxford Microbeams, Ltd.), an automated X–Y stage for cell dishes and a video microscope will be mounted.

5. Summary

As shown above, the scanning microbeam PIXE analysis line, which is based on Oxford Microbeams OM2000 nuclear microscope, provides better performance (it has good spatial resolution and reliable beam aiming accuracy). Though we have such instruments, we measured only the biggish samples as against the spatial resolution. We are planning to use this microbeam system to the measurement of a cell nucleus. In order to irradiate the minute sample on an extremely larger holder than that, absolute positioning system, which works with cell viewing

system, is needed. At first, this absolute positioning system will be mounted on the SPICE system.

Acknowledgements

Authors would like to express sincere thanks to Dr. Geoff Grime, Oxford University, for their excellent help in the installation and the system training of our PIXE system of NIRS.

References

[1] M. Yukawa, H. Imaseki, Int. J. PIXE 10 (1–2) (2000) P71.
[2] H. Imaseki, M. Yukawa, Int. J. PIXE 10 (3–4) (2000) P77.
[3] G.W. Grime, M. Dawson, M. Marsh, I.C. McArther, F. Watt, Nucl. Instr. and Meth. B 54 (1991) 52.
[4] F. Watt, G.W. Grime, Principles and Applications of High Energy Ion Microbeam, Adam Hilger, Bristol, 1987.