

II. 18. Development of a Submilli-PIXE Camera

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Introduction

Particle-induced X-ray emission (PIXE) analysis is well known to be a novel technique for trace elemental analysis¹⁾. A beam spot size of a few mm diameters is usually used in the PIXE analysis. In this case, it is needed that the distributions of elements are uniform in the specimen. If spatial distributions of elements in a region of several cm² can be measured with a spatial resolution of submilli-meters, it will be a powerful means in the analysis of archeological samples, e.g., paintings, books and stamps. Such technique has been already developed for the PIXE analysis of the microscopic region^{1,2)}. Elemental analysis over the area of several cm² is possible by moving the specimen relative to the beam³⁾. However, it has some technical problems such as damage of sample due to local heating with intense beam and distortion of spatial distribution images of elements induced by imperfect measurement of beam current.

Here, we developed a submilli-beam line with a high-speed scanning system and a spatial distribution imaging system of n elements with a spatial resolution of submilli-meters for 3 cm×3 cm area. Furthermore, we combined this system with an in-air PIXE analysis. The high-speed beam scanning and the in-air analysis reduce the risk of damaging the specimen.

Submilli-beam line

Figure 1 shows the layout of the PIXE analysis system newly developed at a Dynamitron laboratory in Tohoku university, which consists of a submilli-PIXE camera, a vertical in-air PIXE system⁴⁾, a target chamber for nuclear spectroscopy, a low Z PIXE system using a crystal spectrometer, a polarized-PIXE system using a crystal spectrometer with a position sensitive proportional counter, a PIXE background measurement system and a vacuum PIXE system. A single-ended type 4.5 MV Dynamitron accelerator with a high-current duo-plasmatron ion source was used in this work, which accelerates hydrogen, deuteron or helium ions. The maximum beam current is larger than 100 μ A. The emittance

of the Dynamitron accelerator is $1\text{mrad}\cdot\text{cm}\cdot\text{MeV}^{1/2}$. The submilli-beam line has been settled in a -40 degrees line which had been equipped with three types of PIXE chambers in series. In order to install the submilli-beam line, two of the chambers were moved to a -15 degrees line. Ion beams are bent by -30 degrees using a switching magnet, further bent by -10 degrees and led to the submilli-beam line (see Fig. 1). Two quadrupole doublet lenses are settled at the upstream side and downstream side of the switching magnet. The beam spot size is smaller than 6 mm in diameter at the end of the beam line. Figure 2 shows the detail of the submilli-beam line. The submilli-beams were formed by using two slits with the spacing of 1.5 m. A dipole magnet to bend the beams 90 degrees was installed between the slits, which is used for a vertical in-air PIXE (ViaPIXE) system⁴⁾ (see Fig.1)⁵. The slit at upstream side is four-pole type copper jaws of 2 mm thick. The slit at downstream side consists of two wedge-shape jaws made by tantalum plates of 0.1mm thick. Both slits are remotely controlled at the control room of Dynamitron where we are monitor beam currents at each slit. The submilli-beams which are formed by passing through these two slits, are scanned horizontally and vertically on a specimen by two magnets.

Submilli-PIXE camera

In the submilli-PIXE camera, specimens can be irradiated either in vacuum or in atmosphere. To scan a wide area, we use two magnets which are an air-core coil and a laminate-core magnet for vertical (Y) and horizontal (X) scanning with high-speed, respectively. These magnets are controlled by a function generator. While a raster scanning mode is used for elemental imaging, patterns of beam scanning can be freely made by computational program. The maximum area of scanning region is $3\times 3\text{ cm}^2$. A vacuum chamber (or a beam exit) assembly is placed at the end of the submilli-beam line for the PIXE analysis in-vacuum (or in-air). The vacuum chamber is a rectangular shaped aluminum chamber with a Si(Li) detector or a Si-PIN photodiode detector settled at the direction of 90 degrees with respect to the beam axis. The beam exit assembly is shown in Fig 3. A beam exit window is a Kapton foil of a thickness of $12.5\text{ }\mu\text{m}$ and its area is 16 cm^2 . The lifetime of the Kapton foil window until breakdown by beam irradiation was longer than 2000 seconds under the beam current density of $300\text{nA}/\text{mm}^2$ (100 nA for the beam spot size of 0.7 mm in diameter)⁶⁾. Since the lifetime is made to elongate by beam scanning, the endurance of the Kapton foil window is sufficient for the submilli-PIXE analysis. The specimen is set just before the exit window. X-rays from the specimen are detected with a Si(Li) or a Si-PIN photodiode detector through the exit window and a detector window of a $7.5\text{ }\mu\text{m}$ Kapton foil (see Fig. 3).

Figure 4 shows a schematic diagram of the beam scanning system and the data acquisition system for elemental imaging. In the PIXE camera, the X-ray energy and beam position are simultaneously measured in order to obtain spatial distributions of elements.

The system consists of a multi-parameter data acquisition system, ADCs for X-ray detector signals and ADCs for position signals. The position signals are derived from the control signals for the X- and Y-magnets. Analog signals from the detectors trigger the ADCs to pick up scanning voltages. After conversion, the digital data are saved in a list file. It takes 20 μ seconds for one data acquisition cycle. The list mode data acquisition is useful to observe changes in elemental imaging during irradiation. This system can sort the data for selected element/energy region and generate elemental images even while the data are accumulated. A large memory size is required in the list mode data acquisition, but it is not so serious since we can use large capacity and removable media at the present.

Performance

The proton beams were first focused at the end of the submilli-beam line by using the two quadrupole doublet lenses with opening the slit maximally, and then the four-pole slit at upstream side was closed to 0.5 mm by monitoring the beam spot size at both X and Y axes. Finally, the slit at down stream side was closed.

The beam spot size was measured in vacuum by using a sharp (#) pattern which was made of a 1.5 mm wide tungsten ribbon. The FWHM of beam spot was obtained by differentiating the spectra derived from the cross section of the spatial distribution of characteristic X-ray yields from the # pattern which was measured by the Si-PIN photodiode detector settled 90 degrees with respect to the beam axis. A measured beam spot size was $0.25 \times 0.48 \text{ mm}^2$ in FWHM for average beam currents of 1 nA. Current intensity of beam halo was three order lower than that of the beam center. Fig. 5 shows the photograph and corresponding elemental image of copper from a substrate. The printed pattern in 0.5-1.0 mm spacing is clearly seen in the measured elemental image.

Response of the magnetic field for a control voltage was studied by measuring elemental images of the copper mesh and by changing the scanning speeds. Fig. 6 and 7 show the control voltages of the magnets v.s. the positions of the copper mesh for the laminate-core magnet and the air-core coil, respectively. The position of mesh is proportional to its control voltage even at the scanning speed of 8.1 cm/sec for the laminate-core magnet. The air-core coil works at the scanning speed of 0.15 cm/sec without distortion of elemental images. It takes twenty seconds to scan the region of $3 \times 3 \text{ cm}^2$.

A shell of short neck clam and a granite were analyzed to demonstrate the application of PIXE camera to the fields of biology and geology. The PIXE analysis was performed in-air. The beam spot size was $0.54 \times 0.65 \text{ mm}^2$ and was rather large than an expected value which was estimated in consideration of diffuseness due to the scattering in the Kapton foil and air molecule. The samples were settled perpendicular to the beam axis and the X-ray detector was settled 135 degrees with respect to the beam axis. Fig. 8 shows elemental images of the Ca and Fe elements in the shell. It took 90 minutes to obtain these images

under the beam currents of 1 nA. The Ca element which is the main components of the shell is homogeneously distributed. On the other hand, the Fe element is locally distributed. Since shells concentrate the elements correlated to their circumstances, this shell had spent his life in water of Fe rich. Fig. 9 shows elemental images of the granite sample. It is seen in these images that the Fe and Ca elements are inhomogeneously distributed. Since the in-air PIXE analysis enables to measure such insulator sample without charge buildup, the submilli-PIXE camera in-air is applicable to measure elemental images contained in any sample.

Summary

The submilli-PIXE camera has been developed for measuring spatial distribution of elements in a specimen, which consists of the high-speed scanning system of the submilli-beam and the data acquisition system. The beam spot size of 0.25 mm×0.48 mm is obtained with a beam current of 1 nA. The maximum beam scanning area is 3 cm×3cm. The maximum scanning speed is 0.15 cm/sec and 8.1 cm/sec for vertical and horizontal scanning, respectively. In order to improve the spatial resolution, we are planning to install quadrupole doublet lenses at the downstream side of the slit of the submilli-beam line.

As the application of the submilli-PIXE camera to the fields of biological and geological sciences, a shell of short neck clam and granite were analyzed in-air. We got clear elemental images for both samples. This system will also be a powerful means for archeology and other fields.

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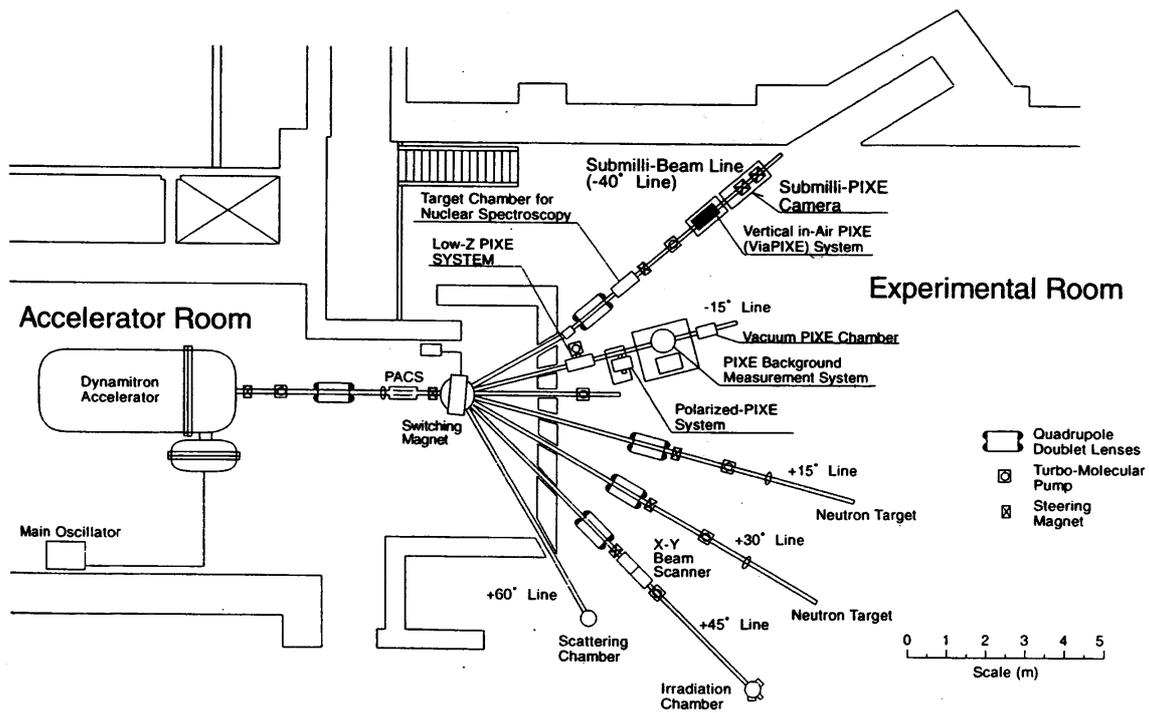


Fig. 1. Layout of the PIXE analysis system newly developed at the Dynamitron laboratory in Tohoku University.

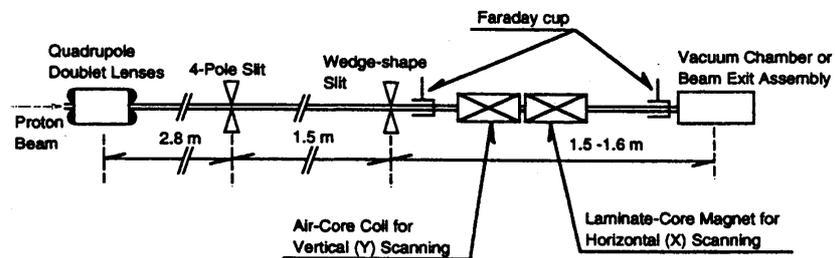


Fig. 2. Detail of the submilli-beam line.

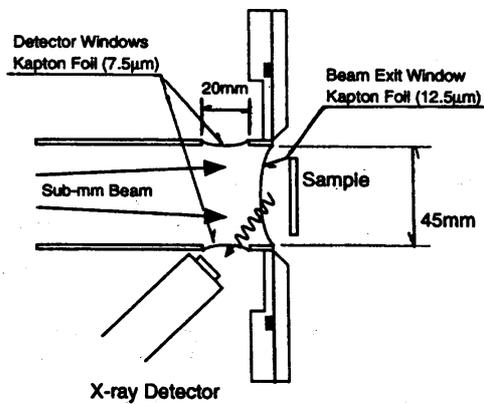


Fig. 3. Cross section view of the beam exit assembly.

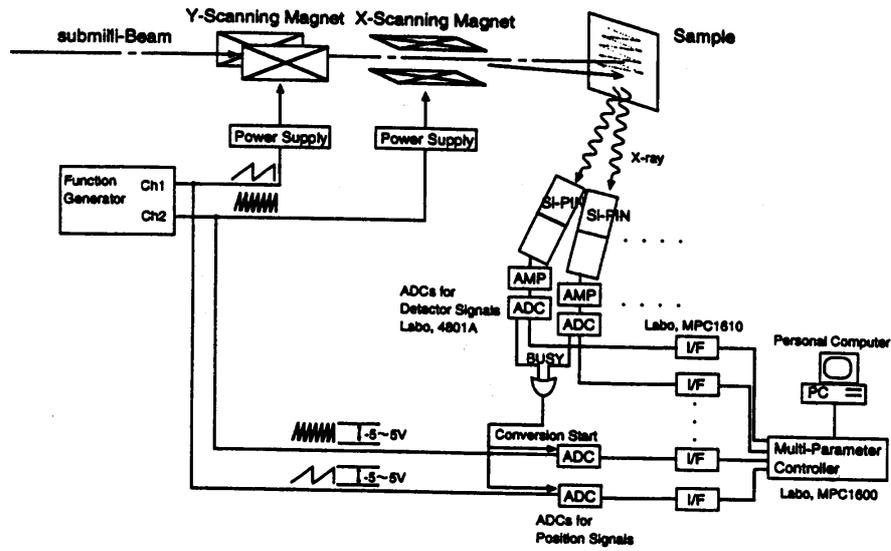


Fig. 4. Schematic diagram of the beam scanning system and the data acquisition system for elemental imaging

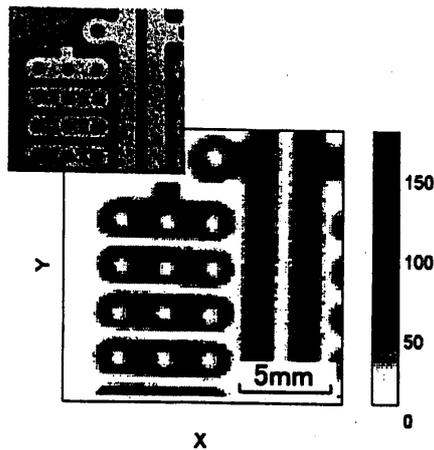


Fig. 5. Photograph of the substrate and the corresponding elemental image of Cu.

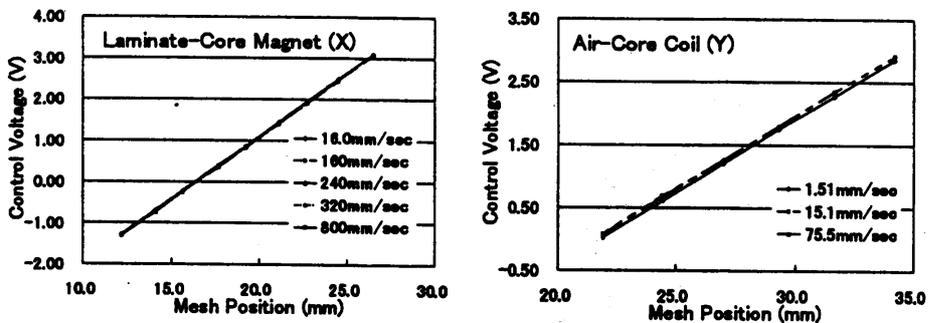


Fig. 6. Control voltages of the magnets v.s. the positions of the copper mesh for the laminate-core magnet.

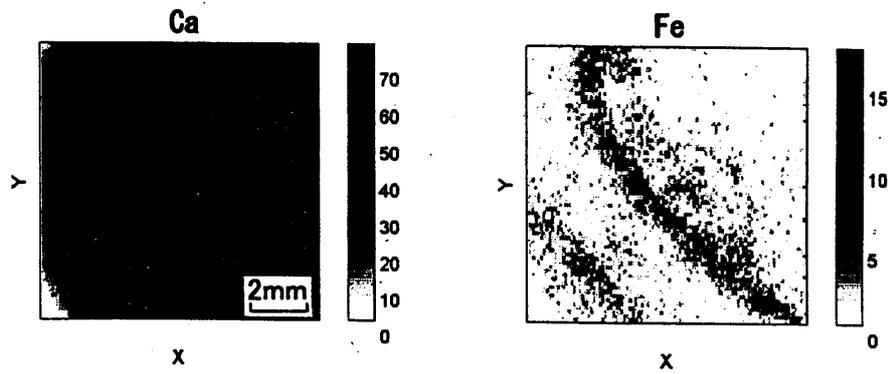


Fig. 7. Elemental images of Ca and Fe in the shell.

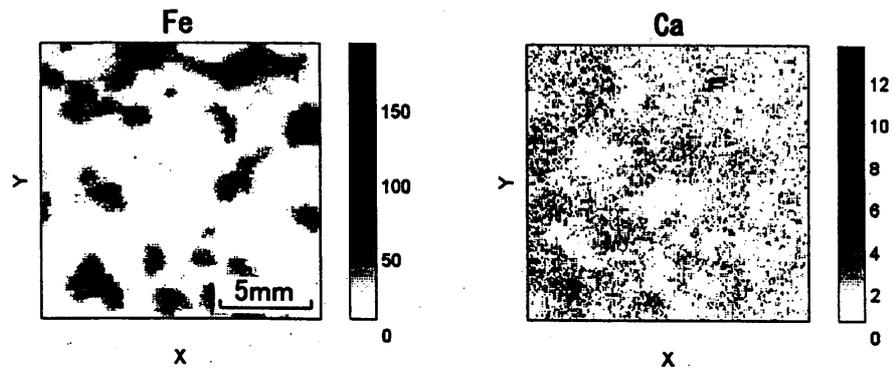


Fig. 8. Elemental images of Fe and Ca in the granite sample.