Chapter 4

Development of ion beam analysis set up

The cyclotron laboratory at Panjab University, Chandigarh houses a single Dee classical cyclotron machine. The cyclotron originally belonged to Rochester University, Rochester, USA and was gifted to India and installed at Chandigarh in 1968 and was made functional in 1976 [1]. The cyclotron has arrangements for variable frequency and capable of producing protons of 2-4 MeV; deuterons of 1-8 MeV and alphas of 2-8 MeV. These charged particles in the above energy regime are ideal for ion beam analysis (IBA). The modifications and enhancements, carried out to develop a suitable IBA set-up to be used as a regional facility, are explained. The experiments which were carried out to calibrate and standardise the PIXE facility, after the successful installation and testing of the setup are discussed in detail.
4.1 Modifications in the main machine

Earlier the main magnet D.C. power was provided by a motor generator with shunt feedback amplifier. This system gave a magnetic field stability of about 1 in 10³, but this stability was not good enough for PIXE work. Therefore, a new magnet power supply from M/S Danfysik, Denmark, Model 853 was installed. The output power of this supply is 400A/125V with stability of ± 10 ppm (8hrs). This modification has improved the orbital stability and focusing which in turn has improved the beam extraction efficiency and energy stability.

The RF oscillator is driven by RCA 5771 tube, which has 25 KVA power triode with water-cooled anode. It has plate voltage 10 kV and frequency range from 10 - 20 MHz. Earlier the plate supply to the oscillator was provided by using vacuum tube rectifiers. But now the power supply has been modified replacing the vacuum tube rectifiers with high voltage silicon diodes network. This has improved the stability of the dee voltage and thereby the beam characteristics.

The generator driven analyser magnet power supply was replaced with a new power supply from M/S Danfysik, Denmark, Model 853. The output power of this supply is 20A/125V with stability of ± 10 ppm (8hrs). This modification has improved the beam current stability at the 45° beam line which was to be used for the installation of the new target chamber for carrying out PIXE experiments.

4.2 The scattering chamber

In different PIXE laboratories, the design of the PIXE chamber varies somewhat [2-7]. However, all these chambers are designed keeping in mind the reproducibility and precision of sample handling, multi target holder arrangements, irradiation procedure, accurate measurement of beam current and the mounting of different radiation detectors (for gamma rays, x-rays and charged particles). Keeping these
considerations in mind a new PIXE chamber was designed indigenously and got fabricated from M/S New Poona Industries, Pune, India. Schematic diagram of PIXE chamber is shown in Figure 4.1.

![Diagram of PIXE chamber](image)

Figure 4.1: Schematic view of PIXE chamber (not to scale)

The entire chamber is made of stainless steel shell (4 mm thickness) of 300 mm inner diameter and of 300 mm height. At 90 mm height the chamber has six ports. Its design meets all requirements of a modern PIXE facility. It has provision for: Si(Li) or Ge(Li), low energy HPGe, SSB detectors and a sample holder wheel to mount 12/24 samples at a time. This wheel is insulated from the chamber through a Teflon ring so as to enable the measurement of charge in case of thick targets. A Faraday cup is used in case of thin targets. Vacuum feedthroughs are provided to supply power to electron gun, current suppressor and SSB detectors. A view port to put close circuit TV camera; a port for attaching vacuum pump and two spare ports are also provided. A DN 100 CF flange at 90° is used to connect
the turbomolecular vacuum pump assembly to the chamber with which the final pressure of $5 \times 10^{-6}$ mbar is achieved.

The chamber has two sample holder wheels. One wheel can hold equally spaced 24 targets of 13 mm and the other can hold 12 targets of 25 mm diameter. The stepper motor with torque 12 kg-cm is made by Sanyodenki, Japan while M/S Penta Designers and Engineers Pvt. Ltd., Pune, India has designed the stepper motor drive (Model MFB-4). The stepper motor and drive units are remotely controlled from the control panel. The stepper motor is placed at 0° and rotates the sample holder wheel coaxially from 0° to 360°. The port at 135° has a guide of 35 mm ID and 100 mm length inside the chambers to bring the Low Energy HPGe detector as close as 50 mm from the target (centre of the chamber). The angle of 135° has the advantage of a two-fold reduction in bremsstrahlung as compared to 90° [8].

In the PIXE chamber, there is an arrangement for a ladder to hold different absorbers in front of the detector to avoid low energy peaks if necessary and to reduce the bremsstrahlung intensity. This is sometime required to improve the count rate of elements above the background with $Z > 34$ for which the $K$ x-ray production cross section for 3 MeV protons are smaller as compared to elements between $Z = 20-30$. Mylar of thickness 100-500 m is ideal and convenient as absorber for biological specimens, since it generates no fluorescence x-ray line to interfere with PIXE spectrum. The compound filters, which acts both critical as well as funny filters, may be used in case of geological and archeological samples. The general view of PIXE setup at VEC, Chandigarh is shown in Figure 4.2.
4.3 The Detectors

Accutrol Systems Pvt. Ltd. has supplied the HPGe detector (IGLET 06135) for low energy x-ray detection. The performance region with almost 100% efficiency is from 3 keV to 60 keV x-rays, which is ideal for PIXE applications. A thin Be window of thickness 12.7 µm is provided at the entrance which is about 90% transparent to 3 keV x-rays. The detector has an active diameter of 6 mm and active depth of 6 mm with energy resolution of 135 eV at 5.9 keV. The detector placed on a retraceable bench which helps in inserting the detector arm into the 135° port of the target chamber. A hollow cylindrical Teflon adaptor was used in the 135° port which serves as a guide for the motion of detector arm. Additionally, the other end of the Teflon cylinder has a 5 mm diameter hole which serves as a
collimator for the detector crystal. This arrangement made a fixed target-detector geometry eliminating any ambiguity in this aspect for experiments carried out at different times. The distance of detector window from target was measured to be 60 mm.

A fully depleted silicon surface barrier detector of 150 µm thickness and 50 mm² area manufactured by Ortec Inc. was used to detect the scattered charged particles. The detector was placed inside a holder made up of Teflon with a heavy brass base so that it could be placed in any desired angle and distance inside the chamber.

An Ortec Inc Manufactured HPGe gamma ray detector was placed at the 45° port of the target chamber. Similar to the case of x-ray detector, this was also placed on a retractable bench. The port was fitted with an adaptor cup with 1 mm thick stainless steel base which allows the detector to move closer towards target up to a distance of 300 mm.

### 4.4 Data acquisition system

Standard NIM electronics were used to achieve the necessary shaping and amplification of the detector output signal. The amplified signal was fed to an Oxford Inc. supplied PC based multichannel analyser card named PCA2. This card is sensitive from a pulse height of 0.2 V to 8.2 V and has maximum gain of 8k. It is controlled with a versatile DOS based user interface which provides many options like changing the conversion gain, spectrum calibration, dead time monitoring and peak fitting.
4.5 Beam focusing and background minimisation

After installation of the PIXE setup the next step was to get a uniform focussed beam on the target and, thereby, avoiding any scattering of protons which may induce x-ray emission from the chamber and the target holder wheel made up of stainless steel. To achieve this, the spectra of polyatomic fluorescent material coated on aluminium target were recorded by passing the beam through the adjustable rectangular collimator made up of tantalum placed in the beam line about 30 cm before the target. A large number of counts of Cr, Ni, Mn and Fe were observed in addition to K and Au peaks of the polyatomic material as can be observed from Figure 4.3 (a). The expected source of these additional x-ray peaks was the target holder wheel made of stainless steel where the scattered protons fall. To verify this, the target was shielded with thick graphite block of thickness 10 mm. The spectrum with reduced background of Fe was observed as shown in Figure 4.3 (b). This confirms that wheel was the main source of Fe peak. To minimise further the scattered protons falling on the wheel, an additional collimator of tantalum with 2 mm diameter was placed ahead of the anti-scattering slit at a distance of 250 mm from the target. This arrangement further reduced the contribution of the elements present in the stainless steel depicted in Figure 4.3 (c). These spectra were still not free from the contribution of the elements from stainless steel. So finally we removed the tantalum collimator altogether and placed the graphite collimator inside the chamber at a distance of 35 mm from the target. This arrangement gave a clean spectrum with very little contribution of Fe x-rays as shown in Figure 4.3 (d). The negligible amount of Fe x-rays can be either from the steel chamber or from the iron impurities present in the target.

Apart from reducing the background, the collimator, also helps in defining the
Determination of beam energy

The reliability of a PIXE analysis heavily depends upon the exactness of incident proton beam energy. Because an uncertainty of 10% in the beam energy produces

beam spot size at the target. Using a 1 mm diameter collimator an optimised beam current of 22 nA was achieved whereas, with a collimator of 5 mm diameter the beam current was 100 nA.

4.6 Determination of beam energy

The reliability of a PIXE analysis heavily depends upon the exactness of incident
variable uncertainties from 6% to 26% in the observed concentration of different elements across the periodic table. In order to avoid any uncertainty about the proton beam energy, an experiment was performed to determine the proton beam energy experimentally by measuring backscattered protons from thin elemental targets.

From the scattering kinematics (Equation (3.3)) it can be observed that if the mass of the projectile and target are known then, by determining the energy of the scattered projectile at a particular scattering angle, one can obtain its incident energy. In order to achieve this, the energy of the scattered particle has to be determined quite precisely.

In the energy spectroscopy experiments, the horizontal axis in the output histogram from the ADC represents the energy of the detected particles. To determine the energy of the detected particles, the channel has to be converted into energy using proper transformation coefficients known as calibration constants. The calibration constant is determined by recording the spectrum of particles of known energy in identical circumstances as that of the experiment and fitting the channel and energy data with a straight line formula where the slope represents the increment in energy from one channel to its adjacent one and the x-intercept stands for the energy which the first channel represents. Both the spectroscopy amplifier and the ADC should provide linear response to the incoming pulse which happens to be true most of the time. But more often than not, there is a non zero x-intercept present. Generally a minimum of two known energy points are required for energy calibration of the data. In this case only one alpha particle source, $^{241}$Am, was available which emits 5.48 MeV alpha particles. So the energy represented by the first channel has to be determined indirectly.

The linearity of the data acquisition system was examined with using a ORTEC 419 precision pulse generator. First, the gain of the spectroscopy amplifier was
4.6. Determination of beam energy

adjusted so that the centroid of the 5.48 MeV alpha peak from the SSB detector came around the 4040th channel in the 4k ADC spectrum. Then the SSB detector was disconnected from rest of the electronics and the output of the pulse generator was connected to the test preamplifier. The input pulse height from the pulse generator was so adjusted that the centroid of the peak falls in the same channel as before i.e. channel number 4040. The spectrum was recorded for 5 minutes for good statistics. Then, using the pulse attenuation factor feature of the pulse generator, the amplitude of the input pulse was decreased by a factor of 2, 4, 5, 10, 20, 40, 50 and 100, and the data was recorded for the same length of time. The centroid of each peak was determined and plotted against the input pulse height. This procedure was repeated again for an initial input pulse greater than the previous one. Figure 4.4 depicts the data points and the linear fit to these data points. Linear fits to these data points show excellent linearity of the electronics and data acquisition system and the x-intercept of both the fits matches well within the fit error suggesting the stability of the ADC.

The SSB detector was kept at an angle of 120° with respect to the beam direction and at a distance of 84 mm from the target. A 2 mm thick aluminium collimator of 2 mm diameter circular aperture was used in front of the detector. Three thin foils of known thickness and composition were used as targets for the measurement. The targets were, self supporting Au and Ni foils of thickness 100 µg/cm² and 340 µg/cm² respectively and the third one was a 6 µm thick aluminised polypropylene with a 200 Å aluminium layer on the surface. The data were recorded till sufficient statistics was achieved under each peak(s) so as to minimise the fit uncertainty. Figure 4.5 depicts the backscattered proton spectra from the above three targets.

From the energy spectrum, the energy of the scattered protons from the surface of the target was determined. The scattered energy along with the kinematic factor
4.6. Determination of beam energy

was used to determine the incident energy of the protons (Table 4.1). From the table it can be observed that the derived energy of the incident protons come close to 2700 keV. The mean was found to be 2697 keV with a standard deviation of 7 keV. So for the PIXE analysis purpose the incident energy can be taken as 2.7 MeV. Energy of the protons scattered from carbon was lower than that for other three elements. It may be due to energy loss suffered by both the incident and scattered protons while passing through the top aluminium layer of the foil.

Furthermore, these data were analysed with the SIMNRA \[9\] software using the evaluated energy of 2.7 MeV for the incident protons. Excellent fit between the simulated and experimental data was achieved. As the proton energy was much higher than the Coulomb barrier energy for carbon, the elastic scattering cross sections measured by Mazzoni et. al. \[10\] was used for the simulation.
4.6. Determination of beam energy

![Graph showing counts per channel for Ni and Au elements.](image-url)
4.7. Standardisation of the PIXE setup

For the quantitative estimation of different elements by PIXE, accurate measurements of the beam charge and the detector solid angle are necessary. However, the beam charge measurement always contains certain errors and the theoretical calculation of solid angle is not reliable. Therefore, the newly developed PIXE setup was standardised for both thin and thick target analysis using different thin elemental targets of previously determined thickness namely, Al, Sc, Ti, Fe, Ni, Cu, Ge, Ag, La, Ce, Sm, Dy, Yb and Au as well as thick pellets of standard reference materials (SRM) from NIST and IAEA mixed thoroughly with graphite.

The x-rays coming out of the target were detected by the detector positioned...
Table 4.1: Kinematic factor ($K$) and energy of the scattered protons from the targets. The respective incident energies are calculated using the above two quantities.

<table>
<thead>
<tr>
<th>Projectile</th>
<th>Target</th>
<th>Mass (M1)</th>
<th>Mass (M2)</th>
<th>Kinematic Factor ($K$)</th>
<th>Scattered Energy (keV)</th>
<th>Incident Energy (keV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>p</td>
<td>C</td>
<td>1</td>
<td>12</td>
<td>0.778</td>
<td>2092</td>
<td>2688</td>
</tr>
<tr>
<td>p</td>
<td>Al</td>
<td>1</td>
<td>27</td>
<td>0.895</td>
<td>2420</td>
<td>2705</td>
</tr>
<tr>
<td>p</td>
<td>Ni</td>
<td>1</td>
<td>58</td>
<td>0.95</td>
<td>2558</td>
<td>2694</td>
</tr>
<tr>
<td>p</td>
<td>Au</td>
<td>1</td>
<td>197</td>
<td>0.985</td>
<td>2660</td>
<td>2701</td>
</tr>
</tbody>
</table>

at 135° with respect to the beam and the count rate was kept below 1000 counts per second to avoid any pile up. The targets were irradiated twice for two different total collected charge, i.e. for 2 and 4 $\mu$C for thin foil targets and 3 and 6 $\mu$C for the thick standards. Typical PIXE spectra for some of the thin foil targets are shown in Figure 4.6.

Analysis of the data was carried out with the GUPIX [11] software package. The fitting of the x-ray spectra by GUPIX software is done by modeling the detector response function for every x-ray line present in the spectra. The line shape ($F$) of a given x-ray peak is described analytically in terms a combination of Gaussian ($G$), shelf ($S$), truncated shelf ($TS$) and an exponential tail ($D$) functions,

$$F(i) = G(i) + D(i) + S(i) + TS(i)$$  \hspace{1cm} (4.1)  

Where, ($i$) is the spectral channel number. Except the Gaussian, all other functions contribute to the low energy tailing of the x-ray peak, due both to basic electron transport processes and also due to imperfections in the fabrication of the detector. The escape of Auger electrons and photo-electrons that are created near the front surface through that surface, are basic electron transport processes
4.7. Standardisation of the PIXE setup

![Graph a](image)

![Graph b](image)
Figure 4.6: Typical PIXE spectra from thin elemental targets (a) Sc, (b) Ge, (c) Ag and (d) Au obtained by irradiating with 2.7 MeV protons.
that cannot be removed by detector design. The intensity of the overall effect increases with decreasing x-ray energy. These effects are modelled through the shelf and the truncated shelf functions. Due to the imperfections in the detectors charge collection process the low energy tail close to the Gaussian centroid in the shape of an exponential function arises. Role of these functions is prominent in the low energy regime (< 10 keV for HPGe detector) of the spectra. A graphical representation of these functions is shown in Figure 4.7.

![Graphical representation of the components of line shape of HPGe detector](image)

**Figure 4.7:** Components of line shape of HPGe detector

The parameters were determined by modelling the detector response function for the $K$ x-rays of Sc, Ti, Fe, Cu and Ge. In the first step the spectral data were normalised with charge. Then, for background removal, the blank spectra were subtracted from each of the elemental spectra. All components, except the Gaussian FWHM, were distributed to the $K\beta$ peak by the $K\beta/K\alpha$ ratio in order to keep the number of fitting parameters small. The parameters thus obtained are added to the GUPIX detector library in order to represent the line shape of the detector used.
4.7. Standardisation of the PIXE setup

Table 4.2: Comparison of the PIXE determined thickness of thin foils with the actual one.

<table>
<thead>
<tr>
<th>Element</th>
<th>x-ray line</th>
<th>Reported Thickness (µg/cm²)</th>
<th>Measured Thickness (µg/cm²)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sc</td>
<td>K</td>
<td>6.88</td>
<td>6.95 ± 0.1</td>
</tr>
<tr>
<td>Ti</td>
<td>K</td>
<td>9.43</td>
<td>9.35 ± 0.1</td>
</tr>
<tr>
<td>Fe</td>
<td>K</td>
<td>11.73</td>
<td>11.47 ± 0.1</td>
</tr>
<tr>
<td>Ni</td>
<td>K</td>
<td>339.00</td>
<td>340.00 ± 0.3</td>
</tr>
<tr>
<td>Cu</td>
<td>K</td>
<td>18.10</td>
<td>18.70 ± 0.2</td>
</tr>
<tr>
<td>Zn</td>
<td>K</td>
<td>176.00</td>
<td>174.80 ± 0.9</td>
</tr>
<tr>
<td>Ge</td>
<td>K</td>
<td>12.78</td>
<td>12.10 ± 0.1</td>
</tr>
<tr>
<td>Ag</td>
<td>L</td>
<td>25.90</td>
<td>25.20 ± 0.2</td>
</tr>
<tr>
<td></td>
<td>K</td>
<td></td>
<td>21.80 ± 1.2</td>
</tr>
<tr>
<td>La</td>
<td>L</td>
<td>57.30</td>
<td>54.20 ± 0.5</td>
</tr>
<tr>
<td>Au</td>
<td>M</td>
<td>59.10</td>
<td>55.60 ± 1.2</td>
</tr>
<tr>
<td></td>
<td>L</td>
<td></td>
<td>59.60 ± 0.6</td>
</tr>
</tbody>
</table>

The instrumental constant, H, which incorporates the detector solid angle and other correction factors was determined by fitting the data of both thick and thin targets. Table 4.2 represents the comparison of thickness of the targets measured by PIXE with their reported thickness.

The thick target standards of brick clay, apple leaves, coal, soil, uranium ore, rye, milk, orchard leaves, hay and blood procured from NIST and IAEA were also analysed. Typical PIXE spectra from some of the targets are given in Figure 4.8. These data were also analysed and the evaluated concentrations are found to be matching with in experimental uncertainties.
4.7. Standardisation of the PIXE setup
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Figure 4.8: Typical PIXE spectra from thick standards of brick clay, coal, orchard leaves and uranium ore.
4.8 Additional enhancements to the IBA setup

In the event of a target being electrically non-conducting it becomes difficult to monitor the beam incident on the target and determining the total number of incident projectiles. In case of some delicate targets, the generation of hot spots at the irradiated area due to a little loss of flux uniformity has to be avoided. Placing a diffuser foil just before the target serves these purposes quite efficiently. A nickel diffuser foil of 0.38 m thickness was placed in between the final collimator & target holder and was electrically isolated from the rest of the chamber. The distance of the foil from the target can be adjusted from a maximum of 80 mm to a minimum of 35 mm. The current taken directly from the diffuser foil helps in monitoring the beam current and stability whereas the backscattered particles provide the number of incident protons on the target quite precisely.

A target holder with two-dimensional precise movement for mounting targets which are to be irradiated at different places (viz. gem stones) and targets of large surface area (viz. gun short residue (GSR) samples deposited on filter paper) was also prepared. Figure 4.9 shows this target holder in position inside the scattering chamber. The movement in the x-axis is achieved by the existing stepper motor, which is attached to the target holder through a rack and pinion arrangement. The movement in y-axis is achieved with the help of a A.C. motor mounted on the target holder assembly.

The linearity of the current integration from the diffuser foil was tested with the help of the SSB detector. The collimator - target and diffuser foil - target distance are 110 mm and 60 mm respectively. The foil was such positioned that it is well beyond the field of view of the x-ray detector. The detector was positioned at an angle of 120° with respect to the diffuser foil with a 2 mm diameter collimator. The distance between diffuser and detector was of 84 mm.

Two sets of data for backscattered protons from the Ni foil was recorded.
4.8. Additional enhancements to the IBA setup

Figure 4.9: Arrangements for the target holder with two dimensional motion. The inset picture shows a closeup view around the centre of the scattering chamber. In this figure; (a) the target holder assembly, (b) SSB detector, (c) graphite collimator, (d) diffuser foil, (e) an Indian bank note positioned on the target holder.

The first was with the current integrator connected to the Faraday cup where the proton beam is being dumped after passing through the Ni foil and the second was with the current integrator connected to the diffuser foil which is being charged up due to the passage of proton beam through it. Total area under the back scattered proton peak was extracted and plotted against the total collected charge as shown in Figure 4.10. The linear fits to the data shows good linearity of the total integrated charge and the ratio between the slopes of both the curves represents the normalisation constant. This suggests that the reliability of the quantitative estimation will be retained while using the diffuser foil for beam current normalisation.

The only challenge in this technique is to maintain the diffuser foil in good
Additional enhancements to the IBA setup

Figure 4.10: Comparison of the two sets of data taken with different current integrator position.

condition throughout the experiment because of the thickening of the foil due to the carbon deposits from the oil vapour. Therefore the foil has to be changed periodically and for every foil one has to determine the normalisation constant before using it in the experiment.
4.8. Additional enhancements to the IBA setup

References


3. N. Hertel, Nucl. Inst. and Meth., B14, (1986) 58


