Chapter 2

Nuclear techniques

Introduction

Nuclear techniques because of their reasonable good accuracy and precision and high specificity play an important role in environmental studies. Nuclear techniques used for environmental trace element studies in the present work were Particle Induced X-ray Emission (PIXE) and Particle Induced Gamma-ray Emission (PIGE). Both PIXE and PIGE are inherently multielemental nuclear techniques which in turn increase the speed of the analysis [1]. In this work, Nuclear techniques such as Gamma-Ray Spectrometry and Solid State Nuclear Track Detectors (SSNTD) have been used for the environmental radioactivity studies. The experimental setups used in our experiments for trace element and radioactivity studies are discussed in this chapter.

In this chapter an attempt is also made to recall the basic physical consideration of various nuclear techniques which were used in our experiments.

2.1 Trace element studies

2.1.1 PIXE

Since the demonstration by Johannson et al. in 1970 of the feasibility of Particle Induced X-ray Emission (PIXE) for elemental analysis, the PIXE technique has found
widespread success in performing routine analysis of environmental, biological and medical samples[2]. PIXE is based on the X-rays that are emitted from a sample as a high-energy beam strikes it. Normally proton beam is used for PIXE measurements. The energies of the X-rays are characteristic of the elements present while the number of X-rays of a particular energy is directly proportional to the concentration of the element in the sample. In principle, PIXE is capable of simultaneously measuring all elements from sodium to uranium in a single irradiation with relatively uniform sensitivity. In practice, up to twenty elements are commonly determined in a single irradiation at fractional mass sensitivities of 0.1 to 10 $\mu g/g$[3].

Particle Induced X-ray Emission analysis has been used more than two decades to determine trace elements in a wide variety of environmental samples including fly ash, soil, water, air, leaf, bark, tree rings etc. PIXE offers distinct advantages over more conventional analysis techniques. These include speed, sensitivity, non-destructive nature and simple sample preparation procedure.

**Interaction of charged particles with matter**

When a beam of heavy charged particle is made incident into matter it loses its energy gradually with depth, until it is finally stopped. The energy loss occurs mainly through inelastic Coulombic encounters with bound electrons[4]. Unlike an electron beam the direction of travel of an ion beam is scarcely altered during this process. The stopping power $S(E)$ of an ion with energy $E$ is defined as the energy loss per unit mass thickness traversed:

$$S(E) = \frac{-1}{\rho} \frac{dE}{dx}$$

where $\rho$ is the density of the stopping material and $x$ the distance. As defined here, $S(E)$ is expressed in units of keV g$^{-1}$ cm$^{-2}$.

Stopping powers are already experimentally measured and the data is available[5]. These form the data base to fit the parameters of semi-empirical equations, which
are now commonly used to obtain the stopping power of all elements in the periodic table[6].

For energy range of 1-4 MeV, which is most important in PIXE, the accuracy of the values calculated with the semi-empirical equations is estimated at 1-2%. The stopping power for compounds or more complex matrices is obtained from those of the constituent elements through the Brag-Kleensman additivity rule:

\[ S(E) = \sum_{i=1}^{n} W_i S_i(E) \]

where \( W_i \) and \( S_i(E) \) are the mass fraction and stopping power of constituent element \( i \).

The total path length \( R \) of an ion is obtained by integration of the stopping powers:

\[ R = \int_{E_0}^{0} \frac{dE}{S(E)} \]

where \( E_0 \) is the incident ion energy. The total path length is larger than the projected range, the difference between the two is smaller than 1% for incident protons of a few MeV.

**Bremsstrahlung**

In addition to the line spectra, a continuous spectrum of X-rays is also observed[7]. Such a spectrum is called bremsstrahlung spectrum; when energetic electrons undergo deacceleration in matter. Reducing speed of the charged particles (proton, heavy ions) in matter also generate a continuous spectrum of X-rays. In PIXE, the major sources of bremsstrahlung caused by secondary electrons scattered by the passage of the protons. The maximum energy that a heavy charged particle can give to a free electron is
where $m_e$ is the mass of an electron, $M$ is the mass of the incident particle and $E_b$ is the binding energy of the electron in the atom. Consequently most of the bremsstrahlung lies at low energies. For higher energy bombarding protons the hump extends to higher energies and therefore influence the detection of higher $Z$ element also.

The emission of characteristic X-rays

Whenever vacancies in the atomic shells are created they get refilled by electrons from outer shells. This process results in emission electromagnetic radiation with well defined energy. The energy of radiation is equal to the difference in binding energy of the electronic shells participating in the transition and is therefore characteristic of the emitting atom. The corresponding law (Moseley, 1913) relates the wave length, i.e. the energy, of the emitted radiation to the atomic number $Z$ of the respective element. In all cases the energy of the impinging ion is high enough, so that the inner shell ionization occurs and the emitted radiation belongs to the X-ray region. The emitted X-rays are characteristic X-rays.

In case of thin targets, measured the yield of the characteristic $K_{\alpha}$ or $L_{\alpha}$ X-rays due to the passage of $N_p$ protons is

$$Y_0(Z) = \frac{N_p \, M_a(Z) \, \sigma(E_0) \, \omega_z \, b_{z,\alpha} \, \epsilon_z \, N_{av}}{A_z}$$

(2.1)

where $Z$ and $A$ are the atomic number and atomic mass of the element, $M_a(Z)$ the mass per unit area, $\sigma(E_0)$ the ionization cross section for the incident proton energy $E_0$, $\omega_z$ fluorescence yield, $b_{z,\alpha}$ the fraction of K or L X-rays that appears in the $K_{\alpha}$ or $L_{\alpha}$ line, $\epsilon_z$ the detector's intrinsic efficiency and $N_{av}$ is the Avogadro's number. The subcrips reminds us that the specimen so thin that the proton energy is constant within the target material.
It is helpful in practical terms to combine several of the quantities in equ. 2.1 into a so-called thin target sensitivity $k(Z)$

$$k(Z) = \frac{\sigma(E_0) \omega_z b_z^\alpha \epsilon_z N_{av}}{A_z}$$

The units of $k(Z)$ are X-ray counts/unit charge/µg cm$^{-2}$. Then the equ. 2.1 becomes

$$Y_0(Z) = k(Z) N_p M_a(Z)$$

In case of intermediate thick targets, say proton interaction occurring within a specimen of small but finite thickness $t$(cm). In the portion of its path from the depth $x$ to depth $x+dx$ the proton energy falls to $E$-$dE$ and the relationship between $x$ and $E$ is

$$x = \int_{E_0}^{E} \frac{dE}{\rho S(E)}$$

where $S(E)$ is the matrix stopping power. We take $\rho$ as the target density, $\rho_z$ as the trace element density in the target and hence $(\rho_z / \rho)$ is the trace element concentration $C_z$. Then the mass of trace element per unit area in the slice $(x, x-dx)$ is

$$M_a(Z) = \rho_z dx = \frac{C_z dE}{S(E)}$$

and using equ. 2.1 the corresponding characteristic X-ray yield is

$$dY(Z) = \frac{N_p C_z \sigma(E) \omega_z b_z^\alpha \epsilon_z N_{av} \exp \frac{-\mu x}{\cos \theta} dE}{S(E)A_z}$$

Integrating this and substituting the zero-thickness yield $Y_0$ which is

$$Y_0(Z) = \frac{N_p \sigma_z(E_0) \omega_z b_z^\alpha \epsilon_z C_Z \rho t}{A_z}$$
Then the correction factor is

$$F(Z) = \frac{Y_0(Z)}{Y(Z)} = \frac{\rho t \sigma_z(E_0)}{\int_{E_0}^{E_t} \left[ \frac{\sigma_z(E)}{S(E)} \right] dE}$$

This factor can be evaluated if the matrix composition and the target thickness are known.

In yield for $K_\alpha$ or $L_\alpha$ X-rays, in case of thick targets is

$$Y(Z) = \frac{N_{av} w_z b_z^\alpha N_p C_z}{A_z} \int_{E_0}^{E_t} \frac{\sigma_z(E) T_z(E)}{S(E)} dE$$

where most of the quantities are as defined above. The quantity $T_z(E)$ describes the transmission of protons from the successive depths in the target,

$$T_z(E) = \exp\left[-\left(\frac{\mu}{\rho} \cos \alpha \int_{E_0}^{E_t} \frac{dE}{S(E)} \right)\right]$$

In this expression, which describes attenuation of X-rays, $(\mu/\rho)$ is the mass attenuation coefficient per unit concentration per steradian per unit beam charge for the particular matrix, $\alpha$ is the angle of the proton beam with respect to the specimen normal and $\theta$ is the take-off angle of X-rays relative to the specimen surface.

### 2.1.2 The PIXE experimental setup

**Accelerator**

The PIXE/PIGE experimental facility at Institute of Physics, Bhubaneswar is centered around a horizontal 3 MV Tandem pelletron accelerator supplied by Electrostatics International Inc., Madison, USA [8-9]. It is equipped with two negative ion sources, RF charge exchange source and a cesium sputter ion source. While the RF source produces He beams and the Source of Negative Ion Cesium Sputtered (SNICS) produces a wide variety of negative ions from solid materials. The schematic diagram of the accelerator is given in fig. 2.1.
A stream of singly charged negative ions from the SNICS source, is accelerated to about 75 keV through a pre accelerating column before entering into the injector, which is essentially a 45° bending magnet which connects both the ion sources to the low energy end of the accelerator. It sorts the ions by mass and directs them in to the accelerating column.

An Einzel lens operating at 60 kV focuses the beam into the accelerating column. At the middle of the accelerator column is the high voltage terminal, designed for a maximum terminal voltage of 3 MV through two pelletron charging chains. This high potential pulls the negative ions towards the terminal. The beam then passes through a nitrogen gas stripper and in the process the ions get stripped of some of their electrons becoming positively charged. Since they are now at a high potential they now get pushed towards the end of the accelerating column which lies at ground potential. The stripper in general produces ions with different charge states.

A magnetic quadrupole doublet focuses the beam into the entrance slit of a high resolution double focusing bending/analyzing magnet which sorts the ions by energy and charge state. Another magnetic quadrupole doublet focuses the beam into the switching magnet which finally puts the beam into the desired beam line. There are a set of steerers, electrostatic quadrupoles and magnetic quadrupole doublets for an efficient transport of the beam into the machine from the ion source side as well as from the machine to the experimental chamber. The production, acceleration and transport of the ion beams are to be carried out in high vacuum. A vacuum of $10^{-7}$ to $10^{-8}$ Torr is maintained in all parts of the accelerator by a number of pumping stations.

**Target chamber**

The target chamber in the 45° beam line was used for both PIGE and PIGE measurements. The chamber has ports for both X-ray (45° with respect to the beam) and Gamma-ray (135°) detections. X-rays exit the chamber through a 25 μm mylar window and traverse approximately a 1cm air gap before entering the detector (fig. 45).
2.2). For charged particle detection surface barrier detector can be placed inside the chamber which can be rotated about the target. A target holder inserted from the top allows several targets to be loaded into the chamber for successive analysis.

As described earlier the ion beam emerging from the accelerator first passes through an analyzing magnet, which sorted out the ions of the correct mass and velocity, and was then normally focused by electrostatic or magnetic quadrupole lenses onto the specimen. Protons of 3 MeV, obtained from the accelerator facility at the Institute of Physics, collimated to a homogeneous beam of 3 mm diameter, were used to irradiate the targets[9]. In our setup for PIXE/PIGE analysis several samples were simultaneously loaded into the vacuum, so that a high sample throughput was facilitated.

The Si(Li) detector
A block diagram of electronics used in the PIXE experimental setup is shown in fig. 2.3. The Si(Li) detectors used for PIXE are the same as those in EDXRF. They typically have a sensitive area from 10 to 80 mm², but this area is often reduced by inserting an X-ray collimator in front of the detector. The aim of such collimator is to minimize the low-energy peak tailing that results from incomplete charge collection at the detector crystal. A Si(Li) detector(EG&G ORTEC) with full-width at half-maximum(FWHM) of 165 eV at 5.9 keV(active area of 30 mm², beryllium window thickness of 12 μm, placed at 90° with respect to the beam direction) was used in our PIXE experiment to detect characteristic X-rays emitted from the targets. A collimator with a 4 mm hole was kept before the detector.

In addition to the equipment used for quantitative PIXE analysis, target chamber also has the equipment necessary for the complementary Ion Beam Analysis(IBA) techniques such as surface barrier detector for measuring charged particles or High Purity Germanium(HPGe) detector for measuring prompt γ-rays.
The absorbers

In contrast to Energy Dispersive X-Ray Fluorescence (EDXRF) spectra, PIXE spectra have most of their X-ray intensity in the low-energy region, so that the spectral shape can advantageously be modulated by placing an X-ray absorber between sample and detector. The aim of absorbers invariably is to reduce or eliminate unwanted continuum background and/or intense X-ray peaks and their associated pileup peaks and at the same time allow bombardments at higher beam intensities, so that the elements of interest can be measured in shorter bombardment times and with fewer spectral interferences. The absorbers are usually made from organic material (e.g., Mylar or from light-element metal foils (Be and Al)). Plain absorbers are employed if complete elimination of the low-energy part of the spectrum is desired. In many cases, however, it is preferable to allow a certain fraction (e.g., a few percent) of the low-energy X-rays to pass onto the detector. This can elegantly be realized by resorting to pinhole absorbers (funny filters) [10]. Other examples in which such complex absorbers are advantageous are the analysis of metal alloys in archaeology, for example, of bronzes, and various analyses in material studies [11]. In our experiments mylar absorbers with various thickness were used for analysis of environmental samples. The choice of absorbers and their thickness are critical in PIXE as the parameters relating to the absorbers need to be properly accounted for in the data analysis.

In our PIXE analysis of environmental specimens, a mylar absorber with thickness of 75-200 μm was inserted between the specimen and the Si(Li) detector [12]. As described earlier, by doing so, the characteristic X-ray count-rate arising from the major and minor elements (and the associated sum peaks) was highly reduced and, moreover, it became possible to bombard the specimen at, much higher beam current and thus raised the X-ray count-rate for the heavier trace elements.

Charge measurement

In PIXE of thin specimens, the ions passed through and were dumped in a Faraday for charge integration. To minimize the γ-ray background originating from nuclear
reactions in the Faraday cup, long cups in which the particles were collected. Furthermore, the escape of secondary electrons from the cup as well as the entrance of secondary electrons from the specimen were prevented by placing a negatively biased ring in front of the cup. The charge integration itself was accomplished by connecting the Faraday cup to a sensitive current integrator.

In case of thick target PIXE, target holder was connected to the current integrator for charge measurement. The target holder was mounted, on an insulated stand-off, was surrounded by a cylindrical electron suppressor held at negative potential within the chamber.

Target irradiation and spectra acquisition

In PIXE two factors determine the choice of the particle bombarding energy; the X-ray production dependence and the bremsstrahlung produced by secondary electrons. Small cross section eliminate the use of low energies. On the other hand, the bremsstrahlung of secondary electrons limits the use of the higher energies. By noting these points, 3 MeV protons have been used for our both PIXE and PIGE measurements. Beam current was kept as low as possible in order to avoid target detoriation.

The target irradiation was carried out in vacuum($10^{-6}$ torr) conditions. It allows one to obtain better detection limits than when the irradiations are done in an external beam set-up(in air or helium atmosphere). The target holder can be moved in the vertical direction, which allows the analysis of different targets without changing the irradiation and measuring geometry. The targets were kept in the target chamber at $45^0$ to the beam direction. PIXE measurements were carried out with a maximum beam current of 20 nano Amps. X-rays exit the PIXE chamber through a 25 $\mu$m mylar window and traverse approximately a 1cm air gap before entering the detector. A thick mylar absorber was kept in front of the detector to attenuate the bremsstrahlung background and dominant low energy X-ray peaks. Spectra were recorded by using a Canberra series 88 multiparameter analyser.
The bombardment of an insulating thick (or semi-thick) environmental specimen in vacuum will generally give rise to charge build-up, and the specimen may reach a positive potential of up to several tens of kV before breakdown and sparking. Such a high potential will accelerate electrons up to tens of kV and as a consequence a huge bremsstrahlung background is produced in the PIXE spectrum. There are many ways to avoid this charging problem, including coating of the specimen with a thin layer of carbon (or some other conducting material), using an aluminized Mylar backing film (this approach works very well for semi-thick specimens), mixing the sample with conducting powder (this is a common method in thick target PIXE), spraying the specimen with an electron current by placing a thin foil or a grid upstream of the specimen in poor vacuum or resorting to non-vacuum PIXE[13]. Several of these approaches were compared by Goclowski el al. [14]. For Thick Target PIXE (TTPIXE) analysis, our targets have been prepared by mixing a high pure graphite with the samples as explained in detail in chapter 3.

During a PIXE bombardment the beam energy is partially or entirely deposited in the specimen. This may lead to radiation and heat-induced damage, i.e. to charring and partial volatilization of the matrix elements and even to losses of some analyte elements (e.g. As, Se, Hg). The element losses increase with increasing current density and beam fluence, but depend also on several other factors such as incident particle type and energy, specimen thickness, vacuum condition of the target chamber and chemical form (species) of the element.

2.1.3 Evaluation of PIXE spectrum

The thick/thin target PIXE analysis was performed using the GUPIX software (supplied by the University of Guelph, Canada). The GUPIX software provides non linear least-squares fitting of the spectrum, together with subsequent conversion of the X-ray peak intensities to elemental concentrations via a defined standardization technique involving fundamental parameters and user-defined instrumental constant[15]. The constants were determined using a set of Micromatter thin standards. Full account
was taken of matrix effects and secondary fluorescence contributions in both the spectrum fitting portion and the calculation of concentrations.

For a given target (specimen, standard or reference material) the measured intensity of proton-induced X-rays in the principal X-ray line of element \( z \) is

\[
I(Z) = H Q C_z \epsilon_z t_z I_1(Z, M)
\]

where \( I_1(z,m) \) is the computed theoretical intensity per unit concentration per steradian per unit beam charge for the particular matrix, \( C_z \) is the concentration of an element \( z \), \( Q \) is the measured beam charge (or equivalent), \( \epsilon_z \) is the detector's intrinsic efficiency and \( t_z \) is the X-ray transmission through any absorber present. The instrumental constant \( H \) subsumes both the Si(Li) detector solid angle and any calibration for intrinsic to whatever device is used to integrate the beam current. The \( H \) values have been calculated using Micromatter thin standards.

In case of thick targets,

\[
I_1(z) = \frac{N_{av} w_z b_z}{A_z} \int_{E_0}^{E} \frac{\sigma_z(E) T_z(E) dE}{SP(E)}
\]

where \( w_z \) is the relevant fluorescence yield, \( b_z \) is the fraction of the X-ray intensity in the line of interest, \( N_{av} \) is Avogadro's number, \( SP(E) \) is the proton stopping power and

\[
T_z(E) = \exp\left[-\frac{\mu}{\rho} \frac{\cos \alpha}{\sin \theta} \int_{E_0}^{E} \frac{dE}{SP(E)} \right]
\]

In this expression, which describes attenuation of X-rays, \( (\mu/\rho) \) is the mass attenuation coefficient per unit concentration per steradian per unit beam charge for the particular matrix, \( \alpha \) is the angle of the proton beam with respect to the specimen normal and \( \theta \) is the take-off angle of X-rays relative to the specimen surface.
In case of thin targets,

\[ I_1(z) = \frac{\sigma_z(E_0) \, N_{av} \, w_z \, b_z}{A_z} \]

where most of the terms are as defined above.

Before analysing the samples, the validity of experimental procedures adopted for sample preparations was checked by analysing the few standard reference materials. Some of our PIXE results were compared with the results obtained using an Energy Dispersive X-Ray Fluorescence (EDXRF) setup available at Punjab University, Chandigarh (India).

**EDXRF**

Some of our samples have also been analysed by the EDXRF setup available at Punjab University, Chandigarh (India) for the comparison of our PIXE results[16]. It incorporates a low power (100 watt) tungsten anode, water cooled X-ray tube (KEVEX, 50 KV, 2.0 mA) as an excitation source with triaxial geometry. The X-ray tube was operated at 35 KV and 1.7 mA. The X-rays from the tube irradiated on a Mo secondary exciter and the characteristic K X-rays of Mo (17.8 keV, the weighted average energy of the Mo $K_\alpha$ and $K_\beta$ lines) were used to excite the characteristic X-rays of elements present in samples. The photopeak areas in each spectrum were evaluated using a computer programme AXIL (supplied by International Atomic Energy Agency)[17]. Matrix effects were taken into account in the spectrum deconvolution to obtain net intensities.

The elemental concentrations are determined using the relation[18]

\[ m_j = \frac{N_{ij}}{I_0 \, G \, \epsilon \, \sigma_{ij} \, \beta_i} \]  

(2.2)

where $m_j$ is the concentration (g cm$^{-2}$) of element j present in the sample, $N_{ij}$ is the
net counts per unit time for the ith group of X-rays of element j, $I_a G$ is the intensity of the exciting radiation incident on the sample visible to the detector, $\epsilon$ is the detector efficiency for the jth element, $\sigma_{ij}$ is the theoretical X-ray fluorescence cross section at 17.8 keV excitation energy and $\beta_i$ is the self absorption correction factor that accounts for absorption of incident and emitted X-rays in the sample. The X-ray fluorescence cross section values were interpolated from the tabulations of Puri et al[19].

The absorption correction factor $\beta_i$ has been evaluated experimentally by measuring the relative X-ray intensity with and without the specimen as described elsewhere[20]. The factor $I_a G$ in equation (2.2) represents the intensity of exciting radiation incident on the sample visible to the detector. In the present study the fundamental parameter approach is based on calibration with known thin standards. The value of the product $I_a G$ has been determined by measuring the K X-ray yields from spectroscopically pure thin foils of K, Ca, Sc, Ti, V, Mn, Fe, Ni, Zn, Ge, Se, Rb, Sr, Y and Mo evaporated on mylar backing and using equation (2.2). The method for evaluation in $I_a G$ is described elsewhere[18].

The validity of the EDXRF method was also checked using the coal fly ash ENO standard. It was observed that measured elemental concentration values were in good agreement with the certified values.

2.1.4 PIGE

A nuclear reaction induces the emission of gamma-rays or of various light elements or both. The possible decay or disintegration modes of the compound nucleus are called open channels[21]. Symbolically a given channel of a nuclear reactions is written:

$$a + X = b + Y + n\gamma$$

where $a$ is the projectile nucleus, $X$ is a given target isotope nucleus, $b$ is a light emitted particle, which may be absent, $Y$ is the residual nucleus and $n\gamma$ represents a
possible cascade of $\gamma$-rays (if $n = 0$, no $\gamma$-ray emitted; for $n=1$, a direct transition takes place).

As for any reaction, one can define an energy balance. For a nuclear reaction we have:

$$E_a + Q = E_b + E_Y + E_\gamma$$

where $E_a$, $E_b$ and $E_Y$ are the kinetic energies of the corresponding nuclei, $E_\gamma$ the overall energy released through $\gamma$-ray emission and $Q$ is the energy corresponding to the exothermic nature of the reaction. $Q$ is on the one hand the energy released by the reaction in the center of mass coordinates of the system; on the other hand it is equal to the change of total rest mass of all nuclei involved.

When $Q$ is positive, the nuclear reaction may take place for any $E_a$ but for small $E_a$ the reaction cross section will be negligible due to the coulomb barrier. When $Q$ is negative the corresponding channel is closed, i.e. the reaction is impossible unless enough kinetic energy $E_a$ is brought in by the projectile to open the channel. This is called the reaction threshold (slightly larger than $Q$ due to momentum conservation effects).

The nuclear reaction equation is usually compacted as $X(a, b \gamma) Y$ [22]. The nuclear reaction, such as PIGE, for analysing samples, consists of irradiating a sample with a beam having sufficient energy so that nuclear reactions are produced and detecting the prompt gamma rays. In order for a nuclear reaction to occur, the sufficient energy is required so that it can overcome the electrostatic potential energy which building up as it approached a target atom nuclei. This coulomb barrier can be taken as $(Z_1Z_2)/r$ where $r$ is the radius of the interaction and $Z_1$ & $Z_2$ are the atomic numbers of the incident and target nuclei respectively[23]. Thus the barrier is higher for incident alpha particles than it is for protons and, for a given beam, it is higher, for the higher atomic number of the target nucleus.
PIGE is based upon the detection of the prompt gamma rays that are emitted followed by a charged particle induced nuclear reaction. In environmental research the detection of the light elements in addition to PIXE analysis has special importance. PIGE method is based on the measurement of prompt gamma rays emitted from \((p, \gamma)\), \((p, p'\gamma)\) and \((p, \alpha\gamma)\) nuclear reactions\[24\].

While in PIXE, electromagnetic force is involved, in PIGE short range nuclear forces are active. A basic mechanism of nuclear reactions involves the formation of compound nucleus in a highly excited state which disintegrates or decays through \(\gamma - \text{ray}\) emission within very short time. The repulsive \textit{Coulomb barrier} between the two positive charged nuclei must be overcome to achieve contact. The projectile must hence be energetic enough, the required energies for light projectiles\((p, d, ^3\text{He}, \text{etc})\) to react with medium weight nuclei\((\text{typically upto} \ \text{Fe})\) being in the MeV range. Therefore, in general for energies of 5 MeV and below nuclear reaction analysis will be suited for light elements\((Z < 15)\). For heavy nuclei the reaction cross sections tend to be negligible. Therefore PIGE is specifically sensitive to light and medium nuclei\[23\].

As in PIXE analysis, the energy of the gamma ray is indicative of the element present and the intensity of the gamma ray is a measure of the elemental concentration. PIGE technique is generally combined with PIXE to provide trace level concentration data for the light elements lithium through chlorine. Simultaneous PIXE and PIGE analyses are carried out to investigate the major, minor and trace elements composition of samples. One advantages of PIGE relative to PIXE is the high penetrability of the \(\gamma - \text{rays}\) thus diminishing matrix effects\[25\].

The principal disadvantages of nuclear reaction analyses are the high cost of the accelerator and the equipment required. The disadvantage of high cost does not always apply however because nuclear techniques lend themselves to automation. Thus if a large number of samples is being routinely analysed with a given measurement technique, and particularly if multielement analysis in a single irradiation is possible then cost per element per sample may turn out to be lower and on occasion even so much
lower then the nuclear technique may become the technique of choice because it is the least costly[23].

In general the number of $\gamma - $rays following a nuclear reaction in a homogeneous sample of thickness $t$ is given by[26].

$$N_\gamma = n_p \, n_0 \, \epsilon \int_{\Delta E} \left[ \frac{\sigma(E)}{S(E)} \right] dE$$

(2.3)

where $n_p$ is the number of incident protons, $n_0$ is the number density of target atoms, $\epsilon$ is the $\gamma - $ray detection efficiency and $\sigma(E)$ is the cross section as a function of energy $E = E_0 - S(E) \, dx$ and $E_0$ is the incident energy. The integral interval $\Delta E$ corresponds to the energy loss of the proton beam in the sample. Unfortunately the application of eq. (2.3) for the calculation of $S(E)$ and $\Delta E$ requires knowledge of the sample composition. A starting assumption should be made to get rough estimate of $S(E)$ and $\Delta E$. The analysis is then performed by iterating the calculation on the basis of the composition found at each step.

When the cross section is constant over the interval $\Delta E$, relation (2.3) takes the simple form:

$$N_\gamma = n_p \, n_0 \, \epsilon \, \sigma(E) \, t$$

where $\sigma$ is the cross section in interval $\Delta E$. In this case quantitative analysis can be carried out using reference standard samples, using the expression

$$\frac{N_\gamma}{N_{\gamma_s}} = \frac{n_0 \, t}{n_{0s} \, t_s}$$

where $s$ stands for standard.
2.1.5 The PIGE experimental setup

In our experiments PIGE was used for detection of light elements, while PIXE was used for the determination of heavier elements \((Z > 14)\). The 3 MV Tandem Pelletron Accelerator facility (as explained already) at Institute of Physics was used for PIGE analysis. A block diagram of electronics used in the PIGE experimental setup is shown in fig. 2.4. A 20% efficiency High Purity Germanium (HPGe) detector with a resolution of 1.9 keV (FWHM) at 1.33 MeV and having a volume of 109.4 cm\(^3\) was used for gamma ray measurements [27]. The detector was kept at an angle of 135° relative to the beam direction and surrounded by a lead shielding to suppress the gamma background. The beam irradiation and data collection were the same as explained in the PIXE measurement. Simultaneously both PIXE and PIGE measurements were done.

2.1.6 Analysis of PIGE spectrum

The PIGE spectra were analysed by the GANAAS program (supplied by IAEA). Peak identification and peak areas were estimated using GANAAS software [28]. The data were fitted using a least-squares procedure. As in PIXE analysis, the energy of the \(\gamma\)-ray is indicative of the element present and the intensity of the \(\gamma\)-ray is a measure of the concentration of the elements.

Mg: Several gamma ray excitation functions are measured for magnesium. Taking into account the isotopic abundances, the 1369 keV \(\gamma\)–ray has the highest yield value. Since it is also emitted in the \((p, \alpha\gamma)\) reaction on \(^{27}\)Al the interference has to be taken account [26]. Therefore 585 keV \(\gamma\)–ray has been used for quantification, in spite of its lower yield value, when the mutielemental analysis was carried out.

B: Both 429 and 2125 keV \(\gamma\)–ray excitation functions have high cross section values. The cross section is higher for the 429 \(\gamma\)–ray but the 2125 keV is preferred when performing a multielemental analysis of a sample where both Li and B are present.
F: Both 110 and 197 keV $\gamma$-rays have high cross section values and do not interfere with other $\gamma$-rays.

Na: The 439 keV $\gamma$-ray in really characterisation of $^{27}$Na and no interference is known for this $\gamma$-ray at proton energies lower than 3 MeV[30].

Al: The 844 and 1014 keV gamma ray cross sections have the highest value.

Si: The 1779 keV $\gamma$-ray line is preferred when performing the multielemental analysis.

**Thick targets**

The most usual use of PIGE involves a number of elements in a matrix and the effect of that matrix on the yield from a particular element must be evaluated. Because the energy loss per unit distance and reaction yield both vary with the proton energy, a technique developed by the LARN group[31, 32] of evaluating the stopping power at the energy where the yield is half that for the incident proton energy appears to give reasonably accurate results[33].

The concentration ($C$) of an element in sample of unknown composition is obtained by comparison with the homogeneous standard by the following formula[32]:

$$C = \frac{N_{\gamma} S(E)}{N_{\gamma,s} S_s(E)} C_s$$

where $s$ stands for standard.

The value $E$ is evaluated from:

$$N_{\gamma}(E) = \left(\frac{1}{2}\right) N_{\gamma}(E_0)$$

This empirical rule gives results correct to approximately 1%. To begin with, it is assumed that $S(E)$ depends only on the known constituents. If $C$ is of the order of 1% or less, the assumption is valid[32]. If $C$ is greater, an iteration calculation was done by substituting the newly calculated value of $C$ into $S(E)$ and so on.
Thin targets
In the case of thin targets, quantitative analysis was carried out using Micromatter thin standards, as

\[ \frac{N_\gamma}{N_{\gamma_s}} = \frac{n_0 \cdot t}{n_{0s} \cdot t_s} \]

where \( s \) stands for standard.

2.2 Radioactivity studies

2.2.1 Radioactivity

Thermodynamic instability of a nucleus leads to its decay through emission of different radiations to a more stable nucleus. This nuclear decay is accompanied by the emission of alpha(\(\alpha\)), beta(\(\beta\)), gamma(\(\gamma\)), neutron, proton and even heavier particles[34]. The elements that emit radiations are known as radioactive elements or radionuclides. Natural radioactivity was discovered by Henry Becqural in 1886.

The type of radioactive decay:
Following are the general type of decay process showing the changes nuclear charge and mass number:

\begin{align*}
\alpha \text{ decay} : \quad & ^AX_z \quad \rightarrow \quad ^{A-4}Y_{z-2} + \alpha \\
\beta^- \text{ decay} : \quad & ^AX_z \quad \rightarrow \quad ^AY_{z+1} + \beta^- + \overline{\nu} \\
\beta^+ \text{ decay} : \quad & ^AX_z \quad \rightarrow \quad ^AY_{z-1} + \beta^+ + \nu \\
\text{Electron capture} : \quad & ^AX_z + e^- \quad \rightarrow \quad ^AY_{z-1} + \gamma \\
\text{Isomeric transition:} \quad & ^AX_z \quad \rightarrow \quad ^AX_z + \gamma
\end{align*}

Kinetics of nuclear decay

Radioactive decay follows first order kinetics as the rate of decay depends only on the number of the atoms present.
\[
\frac{dN}{dt} = \lambda N
\]

where \(N\) is the number of atoms at any time \(t\) and \(\lambda\) is the integration constant.

The half life of the radioactive element is defined as

\[
t_{1/2} = \frac{0.693}{\lambda}
\]

**Radioactive equilibrium:** When a radioactive nuclide (parent) decays into another nuclide (daughter) which is also radioactive, then the activity initially builds up, reaches a maximum and starts decaying with parent's nuclei. Under radioactive equilibrium, the activities of parent and daughter nuclei are equal.

### 2.2.2 Gamma-ray spectrometry

It is possible that subsequent to alpha or beta decay the product nucleus may be left in either its ground state or in excited state. The excited state deexcites mainly by the emission of electromagnetic radiation known as gamma radiation. Gamma ray carries away integral units of angular radiation. The probability of gamma transition depends on the differences in the wave function of initial and final states and hence on the changes in the spin and parity following the photon emission. Gamma radiation arises from the purely electromagnetic effects in the nuclei due to: (i) changes in the charge distribution (electric moments); (ii) changes in the current distribution (magnetic moments).

\(\gamma\)-ray spectrometry is a very powerful nondestructive technique for the detection and measurement of gamma emitting radionuclides in different type of samples. Thus the technique provide a non-destructive multicomponent analysis which is extensively used for environmental samples[36]. Two types of detectors, NaI(Tl) and semiconductors are used for the purpose of \(\gamma\) — ray spectrometry. The high resolution \(\gamma\) — spectrometry is carried out using semiconductor detectors such as Ge(Li), High
Purity Germanium(HPGe) etc. The resolutions of these detectors are in the range of 1.5 to 2.2 keV at 1332 keV gamma energy depending on the quality and volume of the detector. The high resolution of Ge detectors allows one to measure absorption peak areas with little interference from neighboring peaks and several peaks from the same nuclide. Ge detector counting efficiencies are low and somewhat longer counting times are required to obtain in statistical precision comparable to that of NaI(Tl). The efficiency of the detector is quoted with respect to 3” x 3” NaI(Tl) detector. Detectors with efficiency of the order of 5 to 20 % are generally used and more recently as high as 70 – 90% is achieved.

The successful measurement of low activities required an optimum combination of high detection efficiency and low (and equally important) stable detector backgrounds. The sensitivity of measurements varies(approximately) directly with detector efficiency and inversely with square root of the background. Increased sensitivity can also be obtained by increasing the sample size to the optimum level[36].

2.2.3 The Gamma-ray spectrometer: Experimental setup

In this work, gamma-ray spectrometer was used for the environmental radioactivity studies of coal, fly ash, geological samples(soil, sediment, minerals, rock etc) and water.

Gamma-ray spectrometer used for gamma-emitting radionuclide measurements consists of a High Purity Germanium(HPGe) detector coupled with a PC based Multi Channel Analyser(MCA). A 20 % efficiency HPGe detector with a resolution of 1.9 keV at 1.33 MeV and having a volume of 109.4 cm$^3$ was used for gamma ray measurements. The detector was housed in a 3 inch lead shielding coated with 1mm Al inside to reduce the system background. Signals from the detector was connected to a PC-based MCA. The energy and efficiency calibrations of the system was done using standard radioactive sources[37].

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Data analysis

The analysis of gamma spectra is fairly simple if the peaks are well separated. The data were analysed using the GANAAS software. The peak area is related to the disintegration of the radionuclides as\[38\]

\[ I = A \ (B.R.) \ \epsilon \]

where \( I \) is the peak area per sec(cps), \( A \) is the disintegration per sec(dps), \( B.R. \) is branching intensity or gamma ray abundance of the corresponding gamma photon and \( \epsilon \) is the efficiency of the detector.

2.2.4 Solid State Nuclear Track Detectors (SSNTD)

Ionising radiations, having high linear energy transfer (LET), while passing through a dielectric material create a submicroscopic tracks along their path. These submicroscopic latent tracks consist of radiation darraged material[39-40]. They can be suitably developed by chemical etching to sizes convenient for viewing under the optical microscope or for counting using spark counter.

Some important features of SSNTD are:

i) The damage tracks produced by charged particles are permanent record, which can be viewed at a later stage; ii) The dimension of latent tracks can be enlarged by preferential chemical etching to enable them to be viewed under optical microscope; iii) These detectors are less prone to environmental changes; iv) Fading of latent tracks occur at high temperatures; v) They can be used in any size; vi) They are good sensitive to alpha but not to beta and gamma radiations; vi) They are passive detectors and do not require energy to operate.

Several techniques are in use to measure the Radon and/or Thoron levels in air. This includes their collection on a filter paper and subsequent alpha counting. But SSNTD technique has been increasingly used for the measurement of radon or thoron in

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soil gas, uranium exploration, earthquake prediction, and geological and geophysics studies. This technique is the most reliable method for the integrated and long-term measurement of indoor radon activity[39]. The commonly used track detectors for radon and thoron measurements are thin plastic materials such as Cellulose Nitrate(CN) and polycarbonate like CR-39. In this present work Solid State Nuclear Track Detectors such as CR-39 were used to study activity levels of Radon(Rn) and its progeny in indoor-air.

2.2.5 SSNTD: Experimental setup

Solid State Nuclear Track Detectors(SSNTD) technique was used for measurement of aerosol activity(indoor). The measurements were carried out using the aluminium cups of 60 mm in diameter and 90 mm in height fabricated in collaboration with Health Physics Unit, Jaduguda and Radiation Safety Systems Division, Bhabha Atomic Research Centre, Bombay. Each cups was separated into two chambers by a thin latex membrane. The membrane helps one to discriminate radon against thoron[41-42]. The bottom end of the cup was covered using a Whatman filter paper and a perforated aluminium lid. At the other end, a aluminium cap with a loop on the outer surface, acts as a cover. Solid state nuclear track detector films(CR-39) were loaded inside the each chamber of the cup. The Solid State Nuclear Track Detector inside a cup with a latex membrane determines the radon concentration alone and the SSNTD inside the cup with a Whatman paper measures the sum of radon & thoron concentration in air. The exposed films were chemically etched in 6N KOH solution for 6 hours at 60 °C constant temperature[42]. Etched tracks on the films were counted using an optical microscope and indoor radon concentration was estimated using the appropriate calibration factors as explained in chapter 6. This type of passive dosimeters were used due to their low cost and good alpha sensitivity.
Bibliography


Fig. 2.1 The schematic diagram of accelerator
Fig. 2.2 Target chamber
Fig. 2.3 A block diagram of electronics used in the PIXE experimental setup
Fig. 2.4 A block diagram of electronics used in the PIGE experimental setup