

Ion Beam Analytical Technique PIXE for Pollution Study at Dhaka Van de Graaff Accelerator Laboratory

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Abstract: The pollution is increasingly becoming a serious problem in Bangladesh for unplanned urbanization and industrialization, have severely affected the environmental components like soil, water and air. Recently, the experimental set-up of 3 MV VDG accelerator laboratory has been renovated to achieve better control and high quality research and to perform different ion beam techniques simultaneously to extract maximum information from the target in a single experiment. Atomic fluorescence based ion beam analytical technique 'Proton Induced X-ray Emission' (PIXE) has been used for environment pollution study. The calibration and standardization experiments have been also done to be ensuring that, the experimental setup is capable for high quality research works and of international standard level. The atomic fluorescence based PIXE technique has been applied to analyze the soil samples around the Jamuna Fertilizer area of Sarishabari. The detected elements and their average concentrations are Al(5081.76 ppm), Ca(4169.98 ppm), Cu(16.66 ppm), Fe(11264.78 ppm), S(40.6 ppm), K(9176.8 ppm), Si(1798.58 ppm), P(146.8 ppm), Ti(2975.92 ppm), Cr(82.74 ppm), etc. Comparing with the elemental concentration of IAEA soil-7 standard, the experimental value of maximum elements is higher that may be harmful for the environment as well as human being.

Keywords: Analytical, GUPIX, MCA, MAESTRO-32, PIXE, [Si(Li)] detector, X-rays.

I. Introduction

The adequate modernized facilities available in the Accelerator Laboratory of Atomic Energy Centre, Dhaka have been used for the research works, from sample preparation to data analysis. Through the protection of contamination probabilities, the samples have been collected from the surrounding area of Jamuna Fertilizer, Sarishabari of Jamalpur. Microprocessor control Freeze Drying system (Flexi-Dry), Oven (0 – 220 °C), Mortar grinding machine, Microbalance and Hydraulic Pellet Maker have been used for sample preparation. The experimental set-up with the Si(Li) detector and the other associated circuitries have been used for experiments. The proton beam of energy 2.3 MeV has been used for sample irradiation, MAESTRO-32 software for data acquisition and well established GUPIX software has been used for data analysis. The standardization of PIXE setup has been done using a set of thin films standards. A number of incidents may occur, if the high-energy charged particles hit a target, e.g. emission of electrons, X-rays and gamma-rays emission, alteration of the nuclear structure of the sample, the ion may scatter in the backward or the forward direction, produce luminescence in the sample or simply pass through the sample by losing part of its energy [1]. In the present day of global economic activity, and the global concerns of environmental degradation with ever increasing industrialization and introduction of newer practices in agricultural developments, the need for ensuring material qualities in relation to economic developments and human health, has become an important issue. For such an issue to be addressed in right perspective, what is essential is to perform chemical compositional analysis of any material does fulfill the primary requirement to study the properties of a material, whether it is a product from an industrial manufacturing process or environmental and biological specimens. Elemental analysis of any material, be it of physical, chemical, biological or environmental origin, provides a holistic view about the intrinsic quality of the material to be used or consumed in different sectors of national economy, sustainable for desired quality of life [1, 2].

II. Detector [Si(Li)] For PIXE

Semiconductor detectors consisting of large crystal of very pure germanium or silicon or compound crystals are now widely used in many laboratories all over the world. For particular research, an appropriate detector selection is important and it depends upon the efficiency, resolution, and count rate performance, the stability of the detector and the range of photon energy of interest. The Lithium Drifted Silicon Si(Li) detector model SL30165 has been used for our experiments and data acquisition. The [Si(Li)] detector is a P-I-N diode whose intrinsic (I) region is sensitive to ionizing radiation, particularly X-rays. As Lithium is an electron donor, its presence makes the Si crystal a better semiconductor and minimizes effects of impurities. Active diameter of the detector is 6.2 mm, area 30 mm², thickness 3 mm, and distance from the Be window is 5 mm. The Beryllium

Cryostat window thickness is 0.025 mm. The depletion voltage for the detector is -100V dc and recommended bias voltage is -700 V dc.

III. Pre-Amplifier Unit

For our PIXE experiments, pre-amplifier Model 2008B (CANBERRA) has been used. The pre-amplifier unit is located as close as possible to the detector and the input circuit is designed by matching to the detector characteristics, avoiding the degradation of signal fidelity due to the length of the interconnecting coaxial cable. The pre-amplifier unit is used to extract the signal from the detector without significantly degrading the intrinsic signal-to-noise ratio. Shaping pulses and matching impedance are also done by pre-amplifier. Bias supply to the detector is also provided through this unit.

IV. Spectroscopy Amplifier

In data acquisition system, the spectroscopy amplifier is one of the most important electronic instruments that perform several key tasks in pulse processing system. We used spectroscopy amplifier model 671(ORTEC) for our PIXE experiments. It magnifies the milli-volts type output signals from pre-amplifier into volt range. Pulse shaping, energy spectrometry, linear, and noise removing functions are provided by this amplifier, needed for performance optimization of analog electronics. It facilitates the accurate pulse amplitude measurement with Analog to Digital Converter (ADC), and Multi Channel Analyzer (MCA). The amplifier shapes the pulse to optimize the energy resolution, and to minimize the risk of overlap between successive pulses. We used the spectroscopy amplifier for shaping the pulses as our requirement using its various options like course & fine gain, shaping time etc. As well as the spectroscopy amplifier provides dc power to the pre-amplifier.

V. Multi Channel Analyzer

In this research works, Ether-NIM Model 919E (ORTEC) Multi Channel Buffer has been used, which is one of the basic components of the MCA consisting of microprocessor and a memory for the supports of data acquisition. A software package is installed in the computer used for data acquisition, control and display the data saved by MCA Buffer. This NIM module MCA Buffer is designed for high performance data acquisition in nuclear spectrometry applications. Accordingly, X-ray photons as the characteristics of elements of the sample are detected by the [Si (Li)] detector and fit them as electrical signals to the spectroscopy amplifier using pre-amplifier then to the Multi Channel Analyzer for acquisition and analysis purposes. The MCA is a digital instrument that measures rapidly the spectrum of pulse heights emerging from a nuclear pulse amplifier.

VI. Particle induced x-ray emission spectroscopy

Atomic fluorescence based PIXE spectroscopy is one of the most common and widely used analytical techniques at MeV energy accelerators and the analysis is performed with characteristic X-rays. When charged particles with sufficient energy hit a sample, a vacancy in the inner shells of an atom may be created. The probability of creating a vacancy is higher when the velocity of the incoming ions matches the velocity of the inner shell electrons. For MeV ions Atomic fluorescence based PIXE spectroscopy is one of the most common and widely used analytical techniques at MeV energy accelerators and the analysis is performed with characteristic X-rays. When charged particles with sufficient this probability (cross-section) for ejecting inner shell electrons is quite high. Such a vacancy can be filled in a number of ways and one of the processes may emit X-rays with energy characteristic of that particular atomic number. In the PIXE-technique these characteristic X-rays are detected using semiconductor detectors. An energy dispersive analysis of the detected signals can reveal the identity of different elements present in the sample and more importantly, by measuring the charge, i.e. the number of incoming particles, the concentrations of the elements can be accurately quantified. PIXE is a truly multi-elemental technique and can identify elements from Na and up through the rest of the periodic table [3]. Applications of this technique are wide-ranging and diverse. In the Accelerator Facilities Laboratory of Atomic Energy Centre Dhaka, PIXE is routinely being used for the analysis of environmental, biological and geological samples.

The whole system is calibrated by determining the sensitivity factors. The sensitivity factors are deduced theoretically or in a semi empirical way from the calculated cross-sections of X-ray excitation and from the absorption data. The detector solid angle and the energy dependent detector efficiency play important role in the calibration procedure.

To analyse complex PIXE spectra, the GUPIX with DAN-32 interface software have been used. GUPIX uses all the updated and best databases available (cross-sections, fluorescence and Coster-Kronig probabilities, stopping powers and attenuation coefficients). These measures greatly improve the accuracy of the quantification of the PIXE analysis. The PIXE analysis process is mainly based upon the following equation [4, 5]:

$$Y(Z, M) = Y_i(Z, M) \cdot Q \cdot C_z \cdot T(Z) \cdot \epsilon_z \cdot H \text{-----(i)}$$

where,

$Y(Z,M)$ is the measured X-ray yield computed by the fitting program.

$Y_1(Z,M)$ is the theoretical X-ray yield per unit beam charge, per unit solid angle and per unit concentration computed from the GUPIX database, which includes the matrix correction and secondary fluorescence for the thick targets.

Q is the measured beam charge or some value proportional to the charge.

C_z is the concentration of the element quoted by the manufacturer or measured by some other method.

$T(Z)$ is the fractional transmission of X-rays through absorbers.

ϵ_z is the intrinsic detector efficiency.

H is the product of the detector solid angle and any correction factor for the charge measurement. So,

$$H = \Omega \cdot \delta \text{-----(ii)}$$

Where,

Ω is the solid angle and δ is the calibration factor for the charge measurement and the detection system.

VII. Sampling And Sample Preparation

Surrounding the Jamuna Fertilizer area of Sarishabari was chosen for the study of soil pollution level. Samples have been collected in order to following all the precautions to protect the contamination probability. The soil samples were dried in an oven at 70° C and some of them were dried using Freeze Drying System at the temperature of -85° C to make them moisture free. 0.289gm dried powder of soil are pressed into 7 mm diameter and 1 mm thickness pellet with a graduated stainless steel hand press pellet maker and Hydraulic pellet maker respectively. The pellets were mounted on 35 mm slide frames with adhesive tape and preserved in desiccators until irradiation. The irradiations were done using the proton beam energy 2.3 MeV and finally data has been analyzed with the help of GUPIX software. For the determination of toxicity as well as degree of pollution, finding the concentration of different trace and major elements within the sample were the priority.

VIII. Results And Discussion

A number of samples have been irradiated using the proton beam of current range 7 to 15 nA.

Table 1: Concentration Range and Average Concentration of Different Elements Found Within the Soil Samples Irradiated.

Elements	Concentration range (ppm)	Average concentration (ppm)
Aluminum (Al)	6382.9 - 6455.3	5081.76
Silicon (Si)	144.5 - 7546.8	1798.58
Phosphorus (P)	0 - 146.8	146.8
Sulfur (S)	51.1 - 70.7	40.6
Chlorine(Cl)	26.6 - 84.2	49.1
Potassium (K)	4826.3 - 10485.2	9176.8
Calcium (Ca)	1397.7 - 8040.9	4169.98
Titanium (Ti)	1479.7 - 3587.3	2975.92
Vanadium (V)	36.6 - 145.7	107.35
Chromium (Cr)	73.1 -99.7	82.74
Manganese (Mn)	349.1 - 928.9	536.9
Iron (Fe)	10976.8 - 23451.7	11264.78
Nickel (Ni)	19.2 - 70.1	46.875
Copper (Cu)	1.6 - 44.2	16.66
Zinc (Zn)	31.5 - 108.3	67.8
Germanium (Ge)	2.6 - 25.6	14.9
Rubidium (Rb)	88.5 - 202.2	141
Strontium (Sr)	60.3 - 195.2	128.78
Zirconium (Zr)	126.3 - 731.9	298.42

Elements detected in the irradiated samples are Al, Si, P, Cl, S, K, Ca, Ti, Cr, Mn, Fe, Ni, Cu, Zn, Rb, Sr, and Zr, their concentration range, average concentration have been presented in the Table 1.

According to the measured elemental concentration shown in the data Table 1, are not the same as the concentration of different elements of standard soil sample “Soil-7 (IAEA). Many of the elements detected in the irradiated samples are having higher concentration than of the standard Soil-7. Significant amount of increase of Cr, Ti, Zn, and Mn is seen in the soil samples studied. Extremely high concentration of Al and Fe is observed in maximum soil samples. The obtained elemental concentrations found in different soil samples are illustrated through bar diagram in Fig. 1.

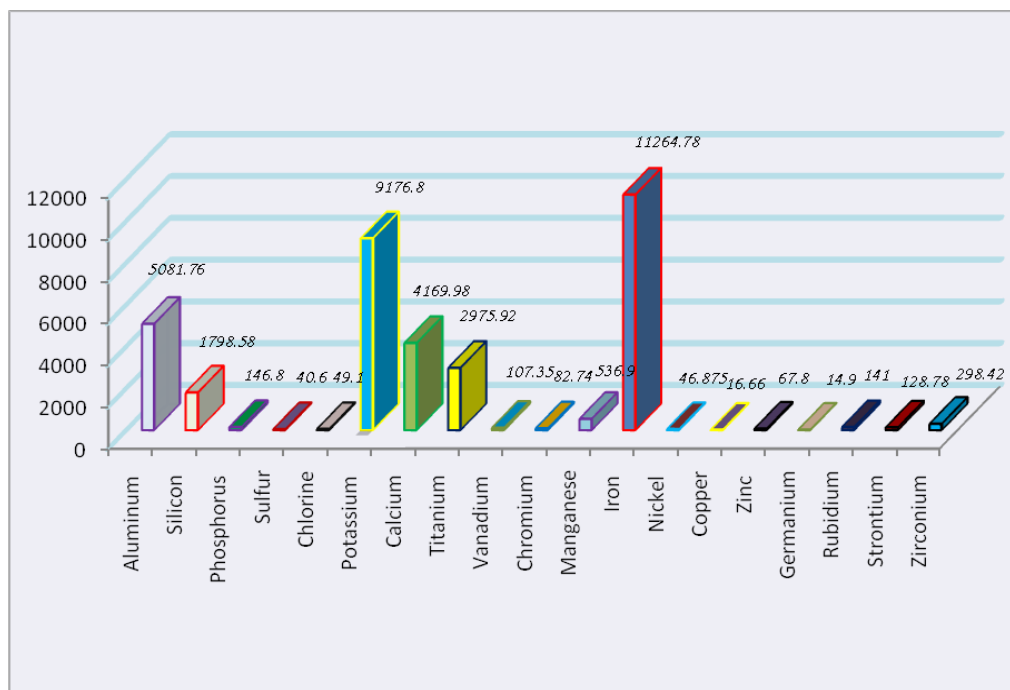


Figure 1: Bar chart of obtained elements and their average concentrations (mg/kg).

If the concentration of any element in the soil exceeds the essential limit, it becomes toxic and harmful for environment and as well as living being [6, 7]. It is therefore, essential to have as detailed information on the nature and the extent of the actual and potential toxicities of the soil so that proper precaution can be taken to improve the environment of the country.

Deficiency and excess of some of the trace elements like Cr, Mo, Zn, Cu, Mn, etc are playing the important roles in human health as well as environmental degradation. Excessively high intake for a long time of most of the essential trace elements shows the evidence of toxicity in living beings [8].

IX. Conclusion

The present study suggests that the soils around the Jamuna Fertilizer area of Sarishabari, where the aluminum, chlorine, iron, titanium, zirconium, zinc, manganese, silicon, and chromium concentrations are higher than the permissible level that may deteriorate the environment and may pose threat to human health. The significance of the problem associated with the elemental toxicity of soil has to be measured and confirmed. Such study will provide sufficient knowledge to evaluate the significance of the problem related to especially environment and human health.

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References

- [1] M. Govil, Proton Induced X-ray Emission – A tool for non-destructive trace element analysis, *Current Science*, 80(12), 2001, 1542-1549.
- [2] Md. Lokman Hossen, S. M. Azharul Islam, M. J. Abedin, S. Akter, O. F. Rasel, M. M. Ahsan, R. Khatun, A. N. Monica, Elemental profile analysis of some traditional medicinal plants of Bangladesh using PIXE technique, *Journal of Nuclear and Particle Physics*, 4(5), 2014, 137-141.
- [3] S. A. E. Johanson and J. L. Campbell, *PIXE: A novel technique for elemental analysis* (New York, John Wiley & Sons, 1988).
- [4] J. L. Campbell, D. Higuchi, J. A. Maxwell, W.J. Teesdale, *Nucl. Instr. Meth. (B77)*, 1993, 95).
- [5] Z. Nejedly, J. L. Campbell, W. J. Teesdale, C. Gielen, *Nucl. Instr. Meth.*, (B132, 1997, 489).
- [6] Eric J. Underwood, *Trace elements in human and animal nutrition* (New York, Academic Press, 1997).
- [7] Ana Pantelicca, Antoaneta Ene, M. Gugiu, C. Ciortea, O. Constantinescu, PIXE analysis of some vegetable species, *Romanian Reports in Physics*, 63(4), 2011, 997-1008.
- [8] Md. Joynal Abedin, Study of the effects of industrial pollutants using ion beam analytical techniques, doctoral thesis, Jahangirnagar University, Savar, Dhaka, 2012.