NUCLEAR ANALYTICAL TECHNIQUE-PIXE, A SUITABLE METHOD FOR ELEMENTAL ANALYSIS OF CONVENTIONAL TO ADVANCED MATERIALS - AN OVERVIEW

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Abstract: Particle induced X-ray emission (PIXE) technique has been used for a variety of analytical problems with MeV accelerator. In general, the sample has to be kept inside a high vacuum chamber for analysis by PIXE. To overcome the difficulties arising for analysis of large archaeological and incompatible biological specimens, there is a need to take the beam outside the chamber and the technique is known as external PIXE technique. The paper presents some of the preliminary experimental results along with various aspects on which studies could be extended.

Keywords: PIXE, Nuclear Analytical Techniques, External PIXE, Biological Specimens.

1. Introduction: A brief description of the various available analytical techniques that are adopted to characterize various materials, for getting the information about the elemental concentrations, all major, minor and trace, is presented. Various analytical techniques are available for characterization of materials and the determination of elemental composition in conventional samples to advanced materials. Widely used methods are INAA, XRF, ICP-MS, EMPA, AAS and PIXE. Some of these methods are destructive, therefore the samples cannot be used for further studies; some of the techniques give information on one element only or require sample preparation before the analysis. In comparison, PIXE has some distinctive features of being non destructive and quick, allowing multi elemental determination and requiring little or no sample preparation.

PIXE is a unique technique for performing non-destructive analysis using characteristic X- rays induced by a proton beam of few MeV directed towards the surface of the specimen. This technique has been successfully used for analysis in various fields, including environmental, biological, archeological, biomedical, archeometrical, geological and also in the field of nanotechnology. Since PIXE is one of the most promising methods of multi elemental analysis introduced in recent years. Many applications and examples which show that PIXE method can be used for geological applications have been described.

PIXE is a relatively simple, yet powerful analytical technique that can be used to identify and quantify trace elements in a sample. It is a nuclear analytical technique for rapid analysis of a wide range of trace elements with ppm sensitivity. Due to its high sensitivity, it is used in the fields of geology and geophysics for investigating trace elements in minerals of geological specimens. Use of protons offers a good sensitivity even at lower atomic numbers due to the fact that bremsstralung caused by protons is low compared to the electron excitation. PIXE technique has rapidly gained acceptance as a valuable analytical tool because of the ever-increasing need for the elemental analysis of very small amounts of sample, as in the case of geological materials such as gemstones, precious minerals etc.

Proton induced x-ray emission (PIXE) is a unique technique for performing non-destructive analysis using measurements of characteristic x-rays induced by a millimetre of proton beam (MeV energy scale) directed onto the surface of specimen[1]. This technique has successfully been used for analysis of various types including environmental [2], biological [3], archaeological [4] and geological [5] materials for almost three decades. The reasons that the PIXE technique is a method of choice for analysis of samples are its well-known features like: multi elemental capabilities, small sample mass requirement, high sensitivity, dynamic range, and simple sample or virtually no need for sample preparation. While dealing with specific archaeological (manuscripts etc), biological (fishes, teeth etc.) and geological materials (big rocks etc.), the vacuum PIXE technique is destructive due to the requirement for either sub-sample or a part for compatible that destroy the original material at the sample preparation stage itself. Elements of anthropogenic origin like sulphur, potassium, lead, bromine, etc. are present in environmental samples mainly as micron to sub-micron fractions. They often adhere on the larger particles of natural origin (silicates, carbonates, sea spray, etc.) and form a thin film over these particles. This technique has several other advantages apart from easy sample handling and positioning in air. Objects of large size and complex structure and shape can be analysed in-situ without the need of sampling, a fact particularly valuable for art objects. Also, it is possible to study materials containing volatile compounds, which could otherwise not stand vacuum. The risk of damage due to heating is considerably reduced because of efficient cooling by the air or the helium flow. However, charging of insulating materials is totally avoided without the need of a thin conducting coating required when operating under vacuum [7-9]. Indeed charging effects can produce potentials up to several tens of keV, accelerate accordingly secondary electrons with a subsequent extension of the bremsstralung background to higher energy. The detection limit would then be marked increased [11]. The PIXE technique of ion beam analysis is first used in 1992 at Ion Beam Laboratory, Institute of Physics (IOP), Bhubaneswar. Since then, PIXE analysis of various environmental, geological [10] and biological [6] samples including fly ash, pond ash, plant soil, water, iron & chromium ores, human & animal tissue, blood, kidney stone, gallstone, hair, etc. has been analysed at IOP and were reported from time to time. The PIXE technique employs 3 MV Tandem pelletron accelerator and samples are kept in vacuum and irradiated. This paper describes the recent development of PIXE set-up at Institute of Physics and some of the preliminary experiments that were carried out [12].

2. Experimental Facility: The proton beam energy of 3 MeV obtained from the 3 MV tandem type pelletron accelerator collimated by a graphite collimator to a beam of 3 mm diameter and beam was extracted using a Kapton foil at exit point of the chamber (8 microns thickness). The Kapton™ foil is used as exit window due to its special characteristics like low beam-induced background emission, minimal energy loss and radiation damage resistance [3, 13-16]. The beam was allowed to travel about 3 cm in air and about 2.4 MeV proton beam was used to irradiate the targets and the external PIXE set-up as shown in Fig.1. The targets were kept in air over a sample stand (capacity: 5kg) making an angle 45^o to the beam direction. The samples are irradiated with maximum beam current of 20 nA that exits through 8 μm thick Kapton™ window designed to withstand the atmospheric pressure. A Si(Li) detector (active area 30 mm^2) of CANBERRA with a full-width at half maximum (FWHM) of 165 eV at 5.9 keV (active area $30mm^2$, beryllium window thickness 8 μm, placed at 90^o with respect to the beam direction) was used to detect characteristic X-rays emitted from the targets. A 25 μm thick aluminised Mylar absorber (with 6% hole) 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 S-100 multi channel analyser, which was calibrated with ^{241}Am X-ray source.

3. Performance and Applications of the PIXE Set-Up: PIXE technique has been successfully used for analysis in various fields, such as environment, biology, biomedicine, geology and nanotechnology. But here in this paper we discussed very few applications of this technique in Environment, biomedicine and geology.

3.1 Environment: Nowadays, many laboratories are involved in environmental research using the PIXE technique. The characterization of fly ash, which is an environmental material of importance, mostly due to its potential for utilization by plants and animals, was studied extensively in IOP, Bhubaneswar, laboratory using this technique. Fly ash is associated with various useful constituents such as Ca, Mg, Mn, Fe, Cu, Zn, Ba, S and P, along with appreciable amounts of toxic elements like Cr, Pb, Hg, Ni, V, As and Ba. The concentration of trace elements in ash is extremely variable and depends on the composition of the parent coal, conditions during coal combustion, efficiency of emission control devices, etc. Figure 1 is a fly ash spectrum taken with the external PIXE set-up. The study indicates that the PIXE technique with external proton beam could effectively be used for characterization of fly ash with good sample turnover time. Aerosols are another frequently studied environmental material using vacuum PIXE technique. But this technique causes the loss of volatile elements like mercury. Another practical obstacle being the beam current used for aerosol samples in conventional PIXE is usually less, but could be increased by about four times in the case of external PIXE.

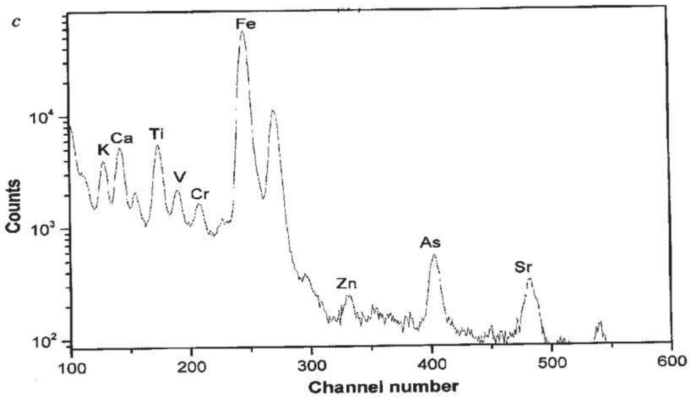

Figure 1: External PIXE Spectrum of Fly Ash, An Environmental Sample.

3.2 Geology: In geochemical explorations, PIXE has been widely used by several workers, which includes pioneering works of CSIRO (Australia) and Schonland research (South Africa) groups on various materials, including diamond. But due to the requirement of large number of samples to be irradiated in order to achieve better statistics, the conventional PIXE technique requires more sample turnover time. Another aspect, which should not be ignored, is the characterization of particular spots of a geological material followed by identification through ore microscopy. Vijayan and his group has reported the use of PIXE and other X-ray techniques on geological materials in finding out the trace elemental ratios, which were used in interpreting the environment of a depositional basin. It is felt that the problem of analyzing big samples could be overcome successfully by the external PIXE technique. Figure 2 is an external PIXE spectrum of soapstone taken using the present set-up at IOP, Bhubaneswar, with no sample preparation procedure. Among other elements, Fe is found to be present dominantly in this sample. We are planning to use the external PIXE set-up to analyze different spots and inclusions observed in geological materials, followed by cross-sectional cutting in order to investigate the distribution of trace elements.

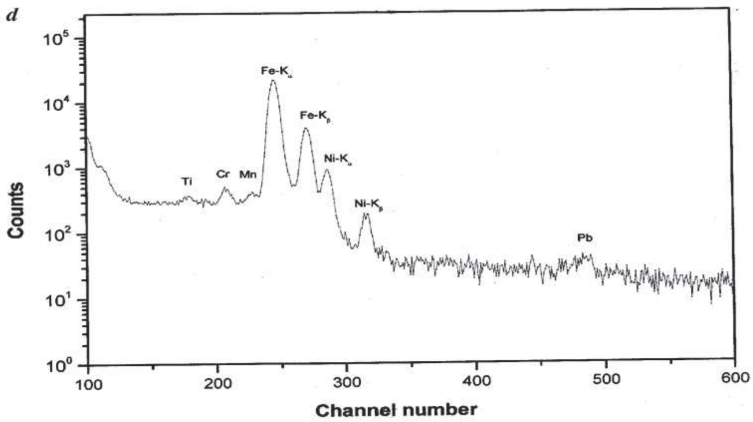

Figure 2: The spectrum of Soapstone studied using External PIXE

A selected number of Eastern Ghats moonstone gems have been studied by our group using complementary and non-destructive PIXE (Proton Induced X-ray Emission) technique. Thirteen elements including V, Co, Ni, Zn, Ga, Ba and Pb were established in these moonstones, which may be useful to interpret the various geochemical conditions and the probable cause of their inceptions in moonstone gemstones matrix. The chemical constituents of moonstones from parts of the Eastern Ghats geological formations of Andhra Pradesh, India and their gemological studies were carried out.

3.3 Biology & Biomedicine:

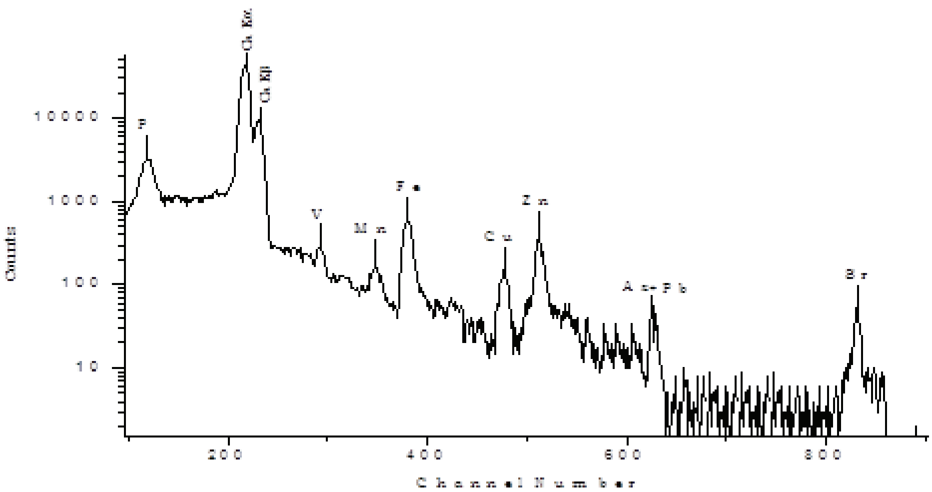

Figure 3: The External PIXE Spectrum of the Dental Cementum Part of a Tooth

The current study is an attempt for the specific detection of trace elemental role on different parts of human teeth and their result on caries regions. In the present study, 12 elements together with trace namely P, V, Ca, Fe, Cu, Mn, Sr, Zn, Ba, As, Pb and Ni were determined. Among the detected elements, Calcium and Phosphorous were found to be the major elements whereas all other elements were found at trace level It is examined that the respective concentrations of Phosphorous, Calcium, Iron, Zinc and Lead elements in enamel are high in quantity than those in cementum and dentine. From the nondestructive elemental analysis on various parts of the teeth, it was clear that the elemental profile in the carious part of the teeth are badly affected compared with other portions of teeth. The excess and deficiency of these identified elements may also cause the dental carries.

Conclusion: A brief description of the various available analytical techniques that are adopted in georesearch and mineral industry, for getting the information about the elemental concentrations, all major, minor and trace, is presented. Widely used methods are Instrumental Neutron Activation Analysis (INAA), X-ray Fluorescence (XRF), Inductively Coupled Plasma Mass Spectroscopy (ICP-MS), Electron Micro Probe Analysis (EMPA), Atomic Absorption Spectrometry (AAS) and Particle Induced X-ray Emission (PIXE). Some of these methods are destructive, therefore the samples cannot be used for further studies; some of the techniques give information on one element only or require sample preparation before the analysis. In comparison, PIXE has some distinctive features of being non destructive and quick, allowing multi elemental determination and requiring little or no sample preparation.
PIXE is a relatively simple, yet powerful analytical technique that can be used to identify and quantify trace elements in a sample. It is a nuclear analytical technique for rapid analysis of a wide range of trace elements with ppm sensitivity. Due to its high sensitivity, it is used in the fields of geology and geophysics for investigating trace elements in minerals of geological specimens. Use of protons offers a good sensitivity even at lower atomic numbers due to the fact that bremsstralung caused by protons is low compared to the electron excitation. PIXE technique has rapidly gained acceptance as a valuable analytical tool because of the ever-increasing need for the elemental analysis of very small amounts of sample, as in the case of geological materials such as gemstones, precious minerals etc. In the present work we reviewed the various mineral deposits and geological applications of PIXE technique in and around Andhra Pradesh, India. The present study, thus establishes usefulness and versatility of the PIXE technique for research in Geo-scientific methodology.

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