Overview of ion beam analysis applications at the Lebanese accelerator

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Abstract: A 5SDH NEC tandem Pelletron accelerator facility was established, years ago, at the Lebanese Atomic Energy Commission within the IAEA TC project LEB 1003. Since its installation in 1999, Ion Beam Analysis techniques IBA are performed, in order to provide elemental analysis in a wide range of applications. In this paper, some selected examples on IBA applications in materials science, environment and archeology are shown as to summarize the different activities of the accelerator during the past few years.

1. Introduction

In 1999, the Lebanese Atomic Energy Commission LAEC acquired, through a national technical cooperation project with the International Atomic Energy Agency (IAEA), the first and unique ion beam facility in Lebanon. It is fully operational since 2001 and certain research activities, related to ion beam analysis techniques IBA, are undertaken. The facility is based on a 1.7 MV tandem Pelletron model 5SDH from NEC [1]. It offers to the Lebanese researchers, from different research centers and universities, the possibility to carry out locally their research projects by taking advantages of the use of IBA techniques.
The main strength of IBA is to be multi-elemental, fast and non destructive analysis and could be performed simultaneously. Hence, they could determine the elemental composition of different kind of samples, covering nearly the whole periodic table, with sensitivity for some elements of ppm level (part per million). Furthermore, they can measure the stoichiometry and thickness of thin films, or multilayer, elaborated on bulk materials with a possibility to perform elemental depth profile. The analytical capabilities available on the facility include particle-induced X-ray emission PIXE [2], proton induced gamma-ray emission PIGE, Rutherford backscattering spectrometry RBS [3], nuclear reaction analysis NRA and elastic recoil detection analysis ERDA. The most undertaken research activities are related to IBA applications in materials science and archeology. However, other applications in aerosols, pharmaceutical science and biomedicine are in growing interest. As we were not able to describe in this short paper all the development and application aspects of the facility, only some selected applications will be exposed, in order to show capabilities and applicability of IBA in Lebanon.

2. The accelerator facility

National Electrostatic Corporation, USA, is the manufacturer of the 5SDH Tandem Pelletron electrostatic accelerator which was installed in 1999 and it is fully operational since 2001 (Fig.1). The accelerator is composed of Radio Frequency ion source (RF) to produce charged particles from H_2 and He gases, the accelerator itself where a potential difference and an electric field is created to accelerate the produced ions, focusing magnet, switcher magnet and vacuum chamber where the target to be analyzed is placed. The accelerated tube placed in the accelerator axis has a vacuum inside of 10^{-8} torr and high pressure outside of 8 bars of SF6 insulating gas. Two turbo-molecular pumps with fore pump assure the vacuum quality of the ion beam path. A nitrogen stripper gas is available at the terminal electrode in the mid accelerator to convert the positive ions to negative ones.

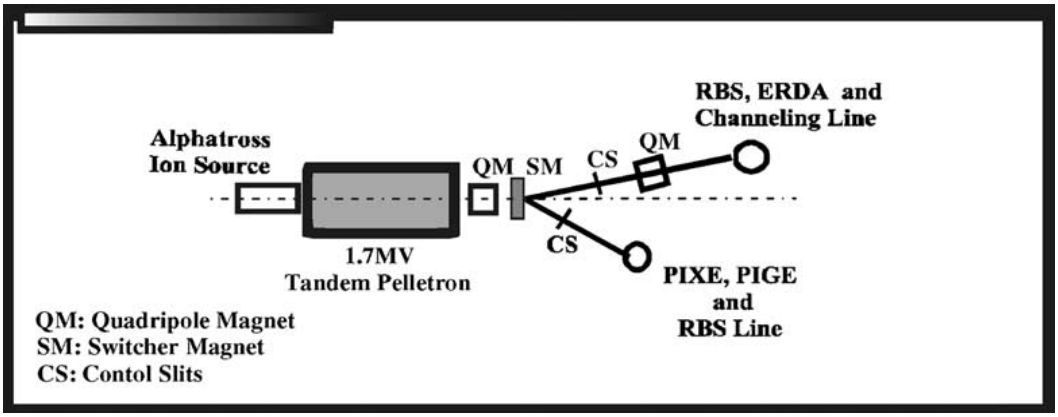

Fig.1: Schematic layout of the accelerator laboratory with the two existing beam lines: (1) RBS, ERDA and Channeling line at –15° and (2) PIXE, PIGE and RBS line at +30°.

The nominal voltage attainable at the terminal electrode, 1.7 MV, allows protons (H^+) and alpha-particles (He^{2+}) to have 3.4 MeV and 5.1 MeV respectively, as maximum energy. The beam will then be directed onto the surface of various materials for analysis. The accelerated energetic ions interact, under vacuum of 10^{-6} Torr, with atoms and nuclei of the studied sample, also called target. The X-rays or scattered beam particles that result from such a bombardment can be used to determine properties of the "target" materials. Several detectors around and inside the reaction chamber are placed to detect the interaction products between the ion beam and the target. A silicon drifted lithium Si(Li) detector is used to detect the emitted characteristic X-rays, high purity germanium detects the prompt gamma and partially depleted PIPS detector measures the energy of the backscattered particles, allowing simultaneous measurements of PIXE, RBS and PIGE techniques when needed. The scattering chamber is equipped with a wheel target holder (up to16 samples) monitored by PC through a stepping motor.

3. Examples

3.1. Archeology

In Lebanon, study of ancient ceramics is of interest for the geographical and historical context which makes it a meeting point of several civilizations and cultures during many centuries. In view of the considerable Lebanese heritage, a special attention was focused on the PIXE applications in archeology, where its capability in the field was proven many times. In this perspective, our laboratory is trying, through the participation in archeological research projects, to contribute to a better understanding of the national heritage. The opportunity of our work was to establish locally, in Lebanon, a powerful analytical technique that archeologists need sometimes to resolve some problems. After the civil war (1975-1990) and following up the project of reconstruction of Beirut city center, which was destroyed, large-scale campaigns of archeological excavations were undertaken that unearthed vestiges dated from the Bronze Age to the Ottoman period.
The first study [4] concerned a roman kiln site of amphorae and common wares. Primary, the aim was to characterize chemically the output of this workshop and thus provide reference data for Beirut's ceramic production. These references would constitute the first step of a database of chemical analyses focusing on archaeological ceramics in Lebanon. The second aim was to test the attribution to Beirut of several other categories of amphorae and to get

some insight into the diffusion of Beirut's production in the Mediterranean area through the study of examples found in Gaul. Both the constitution of reference groups and the tentative attribution of exported ceramics to their origin relied on the use of multivariate statistical techniques. In fact, the elemental analysis done by PIXE showed that some of the 'Carrot' amphorae from southern France were exported from Beirut to the western Mediterranean (Fig.2).

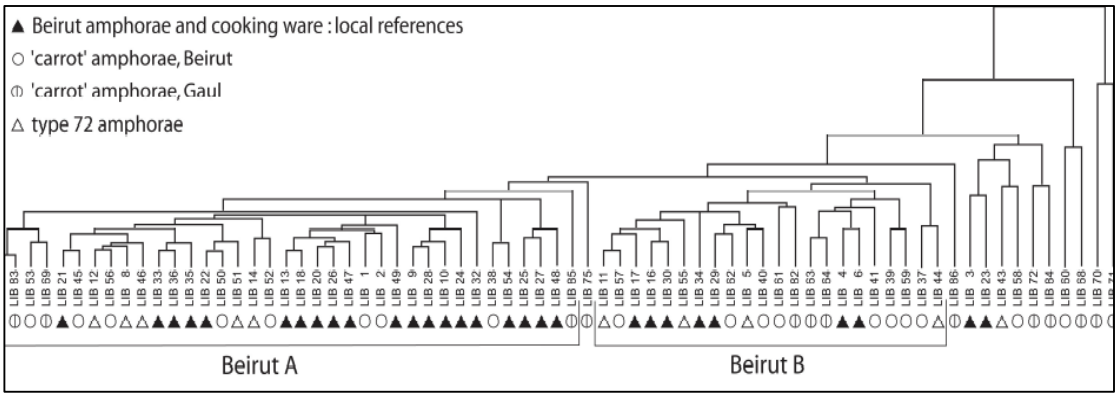

Fig.2: Classification of Roman amphorae from Beirut and from Gaul: Beirut reference samples for local production, 'carrot' and Reynolds type 72 amphorae (see symbols chart). The hierarchical cluster analysis was applied to standardized concentrations of 12 elements. The compositional groups are underlined and the reference samples are indicated by black triangles.

In a similar way, another study, using PIXE cluster analysis, was done on imported ceramics found in Beirut and suspected to belong to north Syrian products, which will provide key evidence for the roles in trade of the various postulated centers in northern Syria and northern Lebanon [5]. A protocol analysis setup, including 3 MeV proton beam and Al funny filter, as x-ray absorber, was used [6]. The elemental composition provided by PIXE and based on 12 most abundant elements, ranging from Mg to Zr, was used in a multivariate statistical program to identify and classify the studied objects into different groups, and the results showed clearly an attribution of the studied samples to "Amrit" region (Fig.3).

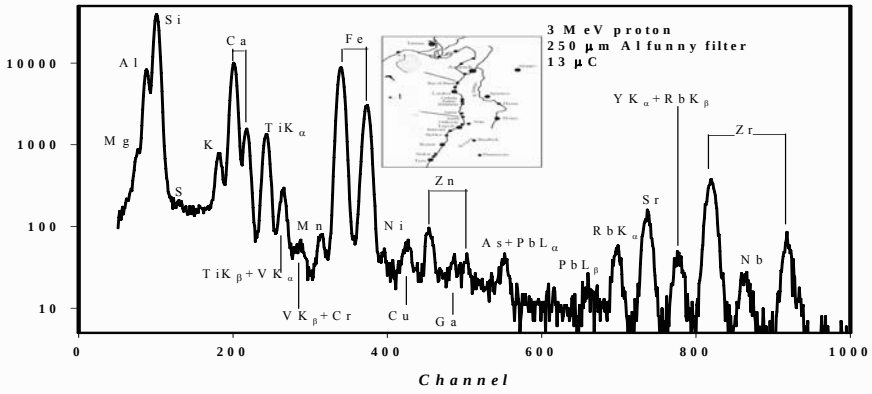

Fig. 3: PIXE spectrum of typical pottery, obtained with 3 MeV protons and a 250 µm Al funny filter. The PIXE protocol allowed the determination in one run measurement of almost 30 elements from Na to Pb; (inside) a map of the classical sites of Levant where the two locations of Ras-al-basit and Amrit are the suspected provenance of the studied excavated pottery from Beirut.

3.2. Materials science

Since 2001, many IBA applications related to the characterization of new materials have been performed at the accelerator laboratory. The first paper of the group appeared in 2002 [7], where the phosphorous signal from thin silicon oxide layer, on silicon substrate, was extremely difficult to determine, when using a typical PIXE analysis with 3 MeV protons. In fact, the huge Si signal overlapped the P one, however, the use of an appropriate filter, low energy PIXE and a tilting angle was very successful and an obvious quantification of P was obtained (Fig. 4).

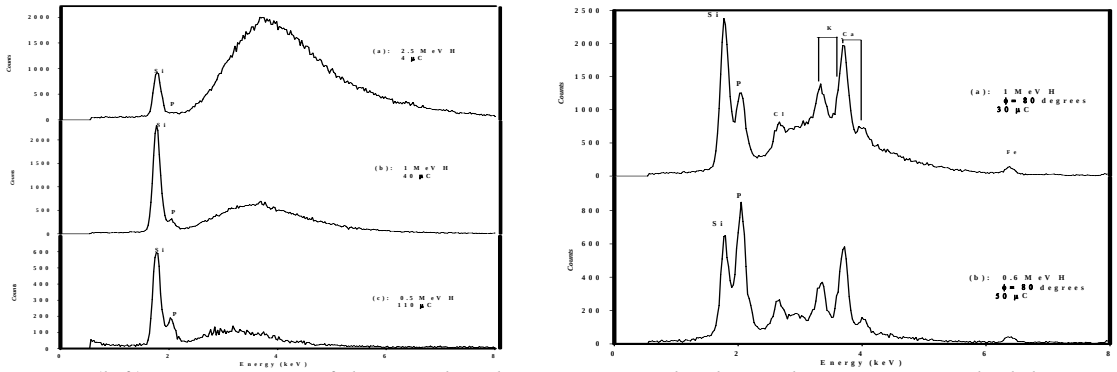

Fig. 4: (left) PIXE spectra of the 2% phosphine CVD sample obtained using 146 μm thick kapton X-rays filter and proton beam at (a) 2.5 MeV, (b) 1 MeV and (c) 0.5 MeV; (right) a drastically change was occurred when using PIXE measurements under 80° tilting angle at (a) 1 MeV and (b) 0.6 MeV.

RBS technique was used to characterize metal contact, such as Pt, Au or Ag, deposited by electroless process on II-VI semi-conductor (SC) material, namely CdTe [8]. Thickness, stoichiometry and elemental composition of the new-formed surface layers were determined as function of experimental conditions (dilution, pH), as well as the related concentration depth profile (Fig. 5)

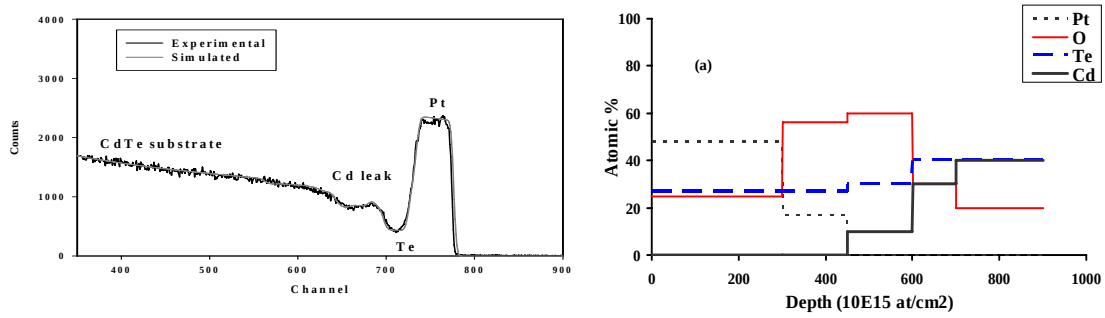

Fig. 5: (left) fitting of an experimental RBS spectrum, using 2 MeV He^{2+}, where multi-layers of different stoichiometry and thickness, were formed following platinum electroless deposition on cadmium telluride substrate; (right) depth profile determination of the components, according to RBS results.

Sometimes, typical RBS with 2 MeV was not helpful to determine light elements such as the determination of carbon, oxygen and nitrogen in nitrided stainless steel. However, the elastic backscattering spectroscopy at 5 MeV He^{2+} beam (Fig. 6), was used successfully for the investigation of the steel samples [9]. The non-Rutherford backscattering, using resonant energies and cross sections, was performed to determine the thickness and accurate

stoichiometry of silicon carbide layers deposited on silicon substrate by pulsed laser deposition [10]. In addition, the combination of PIXE and RBS analysis using 3 MeV protons, determined accurately the elemental stoichiometry of Tl-1223 superconducting samples substituted by scandium ($TlBa_2Ca_{2-x}Sc_xCu_3O_{9-\delta}$,, with $0 \leq x \leq 0.6$) and prepared via solid-state reaction technique [11].

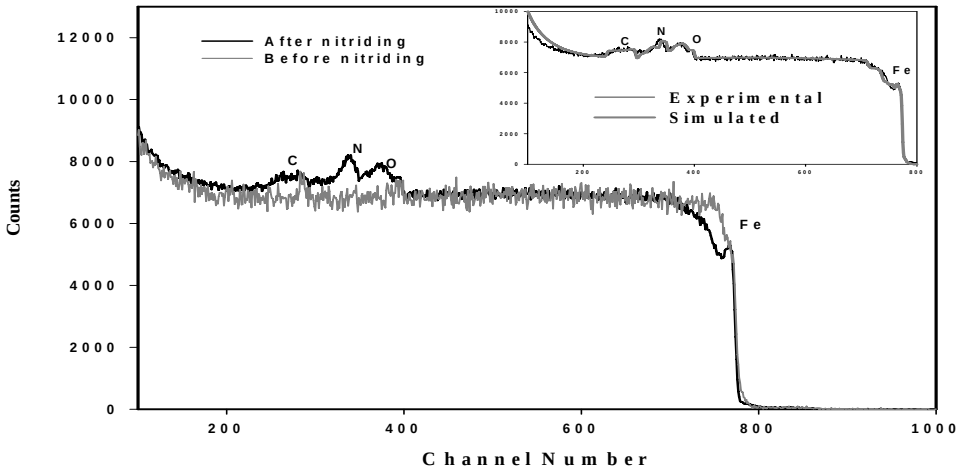

Fig. 6: Comparison of the experimental spectra of AISI-304 sample, bombarded under normal incidence with 5 MeV of alpha-particles: (a) as received before nitriding (gray line) and (b) after plasma nitriding (black line). The inner sketch represents the simulated spectrum (gray line) that fit the experimental one (black line). Several simulated layers were necessary to fit the experimental spectrum.

3.3 Miscellaneous

Other diverse applications were also undertaken, such as the characterization and quantification of active ingredients of some commercial drugs, where IBA techniques showed their capabilities in this field of applications [12]. In fact, the complex molecule based of the active ingredient is recognized by its sulfur and fluorine signals, which are detected by combining PIXE and PIGE techniques (Fig. 7). Environmental studies using IBA were performed such as lebanese soil and aerosols [13,14].

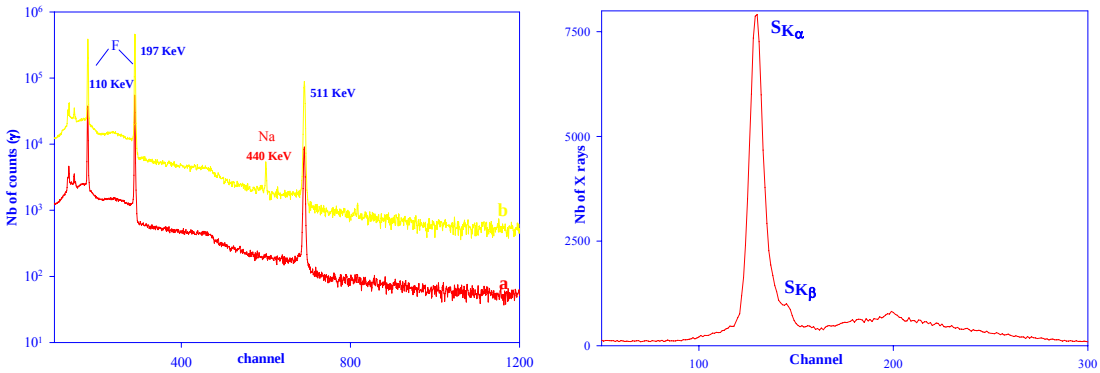

Fig. 7: (left) PIGE spectrum of ultra pure celecoxib standard and of one commercial celecoxib drugs (right) Typical X-ray spectrum of a commercial celecoxib drug obtained by using 3 MeV H^+ with a 131 µm kapton filter

4. Conclusion

This paper highlighted the different research activities, IBA techniques related, undertaken at the Lebanese accelerator facility. It showed their ability to resolve some problematic in different fields of applications, at the national level, as well as at the regional or international one, through bilateral or multipartite collaborations. This facility opened the possibility to local researchers and students from the different Lebanese universities to perform partly or totally their research work in proximity and to get benefit of IBA advantages and capabilities. Thus, more than 40 papers in peer reviewed international journals are produced, beside BS and MS thesis. The accelerator group is pursuing the effort to upgrade the accelerator facility, in order to enlarge the end-users community, by introducing the possibility of new applications. In this issue, an IAEA TC project is starting this year where an external beam with a possible microprobe is planned.

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