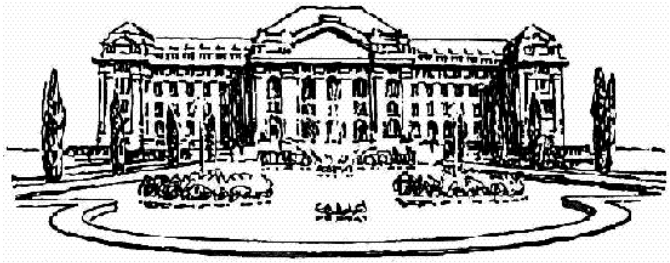

Developements in Nuclear Reaction Analysis and interdisciplinary investigations with ion beam methods

PhD thesis

Szíki Gusztáv Áron

Debreceni Egyetem TTK -ATOMKI
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Preface

In the analytical laboratory, built on the 5 MV Van de Graaff accelerator of ATOMKI we apply a wide range of ion beam analytical techniques /PIXE (Particle Induced X-ray Emission), NRA (Nuclear Reaction Analysis), RBS (Rutherford Back Scattering)/, most of them both in micro and macro ion beam investigations. Regarding PIXE, with a recently developed μ-PIXE set-up [1], we can analyse the Z=6-92 atomic number range with excellent sensitivity for most of the elements. Quantitative PIXE analysis can be routinely performed with the PIXEKLM program package [2] without standards (Fundamental Parameter Method).
For certain NRA techniques /NRA with particle detection, DIGE (Deuteron Induced Gamma Ray Emission)/ – which are effective complementary methods of PIXE for the analysis of light elements and also effectively applicable both in micro and macro ion beam investigations – there has not been an optimal set-up so far. Moreover, for DIGE – which is effectively applicable for the simultaneous analysis of the $3 \leq Z \leq 8$ atomic number range – a Fundamental Parameter Method was not worked out. Thus the quantitative analysis was performed with standards or approximations.

In the current thesis I present instrumental and methodical developments in the field of Nuclear Reaction Analysis which give solution for a great proportion of the above mentioned deficiences, making the methods more sensitive and accurate. Following this I present interdisciplinary investigations which I performed with DIGE and PIXE techniques using the above mentioned developments.

Experimental methods and arrangements

The experiments were performed at the 5 MV Van de Graaff accelerator of ATOMKI, partly on the OM (Oxford Microbeam) type microprobe working on the 0° beam line

and on a macro beam set-up on the right 30° beam line. The applied analytical methods were Proton Induced X-ray Emission (PIXE), Proton Induced Alfa Emission and Deuteron Induced Gamma Ray Emission (DIGE).

The PIXE measurements were performed with the μ-PIXE setup [1] working on the OM type microprobe. For the investigations a proton beam with an energy of 2 MeV was used. The beam current varried between 20 and 200 pA, the spot size was 2×2 μm^2. The PIXE spectra were evaluated with the PIXEKLM program package [2].

The macro and micro-DIGE measurements were performed on self-developed experimental set-ups. For the macro and micro-DIGE measurements deuteron beams of 0.6-1.8 MeV and 1.8 MeV energy and of 0.2-45 nA and 0.1-0.6 nA current were applied respectively. Gamma rays were detected by a high purity germanium detector with 40% relative efficiency related to a 3×3 inch NaI detector. The gamma spectra were evaluated with the FORGAMMA [3] program package.

Analysis with Proton Induced Alpha Emission was performed on the nuclear microprobe. For the measurements a proton beam with an energy of 0.675 MeV and a current of 0.2-0.3 nA was used. The spot size of the beam was 2×2 μm^2. The alfa particles were detected with a recently developed detector array built of PIN silicon photodiodes.

Results

1. With the modification of the sample chamber of the Oxford type microprobe we developed an efficient set-up for μ-DIGE measurements. We were the first in the world to position the focus of the microprobe outside the chamber under vacuum. With this modification we significantly increased the solid angle attainable with gamma ray detectors, and the sensitivity of μ-DIGE technique this way. At the new position of the beam focus I determined the geometrical beam size theoretically (12.5×12.5 μm^2) and the real beam size experimentally (15×17μm^2). The difference between the two values is not significant, which indicates that the set-up works optimally *[3]*.
2. I was the first to measure the yield of the $^{14}N(d,p\gamma)^{12}N$ nuclear reaction for the 7299 and 8310 keV gamma lines on a thin sample in the analytically important 0.6-1.9 MeV

deuteron energy range. Using the measured yield functions I worked out a Fundamental Parameter Method for the DIGE analysis of nitrogen at 1.8 MeV deuteron energy. The method enables the determination of nitrogen in samples with arbitrary thickness, and with known main constituent composition without any standards. I tested the accuracy of the method on compounds with well known composition and on special nitrogen standards with a poly-ethylene-glycol ($HO-(CH_2-CH_2-O)_n-H$) matrix. The results of the tests have shown that the method is capable of the analysis of the >0.2wt% concentration range using 200-400 nA beam current with 5-10% accuracy. Consequently it is applicable both in micro and macro ion beam analysis, in most cases without damaging the sample *[5]*.

3. We were the first to construct a highly effective detector array from PIN silicon photodiodes for μ-NRA measurements. The main advantage of PIN diodes over annular surface barrier detectors, which are widely used for the above purpose, is their lower price. The obtained solid angle with the PIN diode array (1.87 sr) is slightly larger than the largest reported value for an annular surface barrier detector (1.63 sr, [4]) *[4]*.

4. On the basis of the $^{11}B(p,\alpha)^8Be$ nuclear reaction we applied the PIN diode array for the nuclear microprobe analysis of boron. I determined the minimum detection limit for boron attainable with the new detector array. The value of 5 μg/g is significantly lower than the reported ones (10-100 μg/g) for annular surface barrier detectors, consequently it is unique in the field of nuclear microprobe analysis. The PIN diode array is excellently applicable for producing boron elemental maps with 2×2 μm^2 lateral resolution. With the new detector array the microbeam study of the 20-160 μg/g boron content of geological obsidians became possible, which had been unsuccessful with μ-PIGE technique before [5] *[4]*.

5. In connection with the investigation of the 10-1000 μg/g nitrogen content of CVD (chemical vapour deposited) diamonds I was the first to employ DIGE technique for the determination of nitrogen in carbon matrix. I studied the factors affecting the attainable minimum detection limit (MDL) in detail. With the proper choice of the experimental conditions and evaluation procedure I managed to reduce the value of MDL from ~3000 μg/g to 130 μg/g. This reduced value is only partly suitable for the investigation of CVD diamonds, but could be useful for the study of other materials with high carbon content (e.g. organic materials) *[1]*.

6. I was the first to apply μ-PIXE and μ-DIGE methods for the investigation of prehistoric incrusted ceramics which were escavated in the territory of Hungary. The ceramics derived from four different prehistoric cultures. Determining the elemental composition of the ornaments I managed to get information about the materials which were used in the different cultures to prepare them. Producing elemental maps I gained further information about the structure of the ornaments. With μ-PIXE and μ-DIGE measurements I verified the archaeological hypothesis that the ornaments of the ceramics from *Vörs-Máriaasszonysziget* (Late Copper Age and Early Bronz Age) contain bone grit. On the basis of the μ-PIXE study of the ceramics from *Balatonfűzfő* and *Papkeszi* (Middle Bronz Age) I confirmed that their ornaments consist of a mineral form of calcium carbonate. I showed that the composition of the ornaments of the ceramics from Baradla cave is very similar to the composition of the bulk material of them. The high SiO_2 és Al_2O_3 content indicates that the main constituents of the ornaments are kaolin and quartz *[2]*.

Publications related to the thesis

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