

# Recent developments in IBA analysis at CENTA, Bratislava

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**Abstract.** An experimental setup used at the CENTA (Centre for Nuclear and Accelerator Technologies) laboratory to carry out IBA (Ion Beam Analysis) techniques is described. PIXE (Particle Induced X-ray Emission) technique offers a unique way of non-destructive elemental analysis using accelerated ion beams. The calibration of instrumental constants (H-values) of SDD and BEGe detectors, used in the setup, is described in detail. The H-value was determined as a function of X-ray energy by using thin MicroMatter standards of chosen elements. The calibrated H-values were then used in GUPIXWIN for the determination of elemental concentrations in two IAEA reference materials, which were compared with reference values. A reasonable agreement was achieved between the measured and reference values, while the calibrated H-values will be used for further analyses.

## 1 Introduction

Particle induced X-ray emission is a non-destructive analytical technique used for studying surface elemental composition, with high sensitivity for multiple elements. It is frequently applied as a complementary technique to other IBA techniques, such as Rutherford backscattering spectrometry (RBS), nuclear reaction analysis (NRA), and elastic recoil detection analysis (ERDA), to obtain more complex information about the analysed sample. Possibilities of PIXE techniques to determine the elemental composition of thin and thick samples were demonstrated in many applications, e.g. [1-7]. In the case of thin films, the intensity of measured X-rays depends on the accumulated beam charge, the ionisation cross-section, the fluorescence yield, the transmission through filters (present between a sample and the detector), and the detector efficiency [8]. For the thick samples, also stopping power of the ion beam, and attenuation of the X-rays in the sample complicate the calculation of elemental concentration [9].

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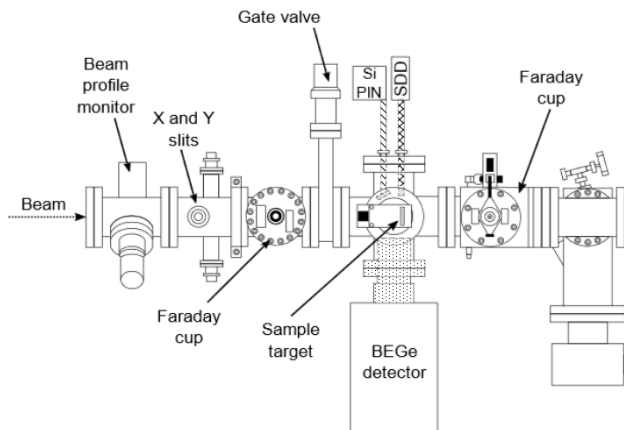
The calibration of the PIXE system is needed when quantitative analysis is involved. The instrumental constant H-value was proposed to include the geometry of the setup [10], and it has been frequently used for the determination of elemental concentrations [11-13].

## 2 Experimental setup

CENTA laboratory was established in 2013 at the Faculty of Mathematics, Physics and Informatics of the Comenius University. The ion beam facility has a 3 MV Pelletron tandem accelerator dedicated to IBA techniques and the accelerator mass spectrometry (scheduled for 2022). A detailed description of the laboratory and its accelerator system can be found in [14]. IAEA funded the installation of the PIXE/PIGE (Particle Induced Gamma-ray Emission) beamline at the CENTA in 2015 [15], and a new detection system was incorporated into the chamber used for IBA analysis in 2019. The current experimental setup allows simultaneous PIXE, PIGE and RBS analysis.

### 2.1 IBA detection system

The detection system includes three separate detectors. Silicon drifted detector of 70 mm<sup>2</sup> FAST SDD (Amptek) in X-123 configuration with C2 (Si<sub>3</sub>N<sub>4</sub>) entrance window is used for PIXE analysis of light elements (from carbon to yttrium). It is suitable for X-ray detection in the range of 0.1-15 keV with an energy resolution of 122 eV for 5.9 keV (<sup>55</sup>Fe). Germanium BE2825 BEGe detector (Mirion, former Canberra) with 0.6 mm carbon window and 2 mm PE cover with the 5 mm hole in diameter in the centre is used for PIGE analysis as well as for PIXE analysis of elements heavier than titanium. The energy resolution of 390 eV for 5.9 keV and 1.8 keV for 1.3 MeV (<sup>60</sup>Co) makes it suitable for operation in energy regions of 4-75 keV and 50-1600 keV, respectively. The Si PIN photodiode Hamamatsu S3590 is used to register backscattered ions in front of the sample. A schematic view of the IBA chamber with detector positions is shown in Fig. 1.



**Fig. 1.** Scheme of the IBA chamber in the CENTA laboratory.

3 MeV proton beams are frequently used for PIXE analysis with intensities ranging from 0.5 to 1.5 nA. For thin samples, the FC (Faraday cup) behind the sample is used with the current integrator Ortec 439 for charge determination. For thick samples, the charge can be estimated directly by its collection from the sample by insulated sample holder and current

integrator, or by periodic insertion of FC in front of the sample. Beam position on the sample is adjusted by the in-chamber camera and by recording a fluorescence on the scintillating surface. At least three spots on the surface of the sample were irradiated by protons during the PIXE measurements to estimate the homogeneity of the sample material.

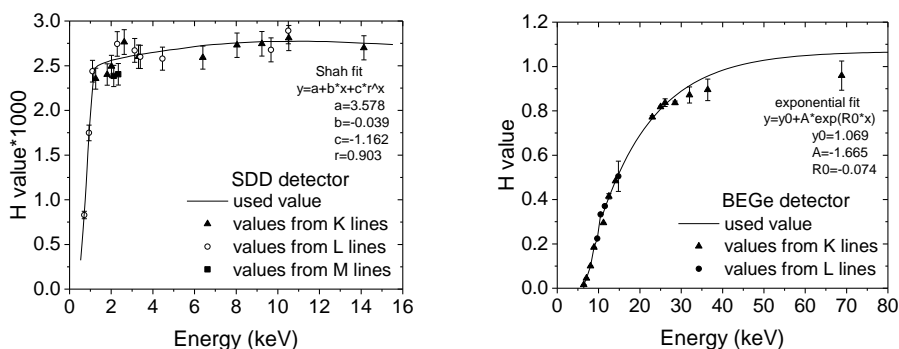
## 2.2 Sample preparation

For the determination of the elemental concentration in the samples, the instrumental constant is needed. The so-called H-value was determined using thin MicroMatter standards in defined geometry. Due to different energy ranges, standard materials Fe, Cu, GaAs, CdSe, SrF<sub>2</sub>, MoO<sub>3</sub>, Sn, BaF<sub>2</sub>, Au, Pb and MgF<sub>2</sub>, GaP, KCl, were used for calibration of BEGe and SDD detectors, respectively.

Reference materials IAEA-158 Marine sediment and IAEA-359 Cabbage were used to validate the constructed H-values calibration. About 140 mg of reference material was mixed at the ratio of 80:20 with the Hoechst wax micro powder (Supelco) and pulverized by mortar and pestle. After thorough mixing, the samples were pelletized by KBr die with a diameter of 13mm and installed in the IBA chamber for evacuation.

## 3 Results and discussion

X-ray spectra from irradiated MicroMatter standards were processed by GUPIXWIN software, where the H-values were set as 1. Afterwards, the experimental H-value was calculated as the ratio between the calculated concentration from measured spectra and the nominal value. The energy dependence of H-values for both detectors is shown in Fig. 2. Because of complicated shapes, values were fitted by analytical functions in the range from 1.5 keV and 10 keV, for SDD and BEGe detectors, respectively. As can be seen, the H-value for the SDD detector is almost constant with a strongly decreasing trend below 1.5 keV, where the floating average was used instead of the fit. The same approach was used for values below 10 keV for the BEGe detector. This is the result of using C2 window transmissions for low Z elements (supplied by the manufacturer) during the calculation of thin standards concentrations. The trend of H-value energy dependency for the BEGe detector is different because we do not know the X-ray transmission factors through the detector window and the absorber for this detector. Therefore, together with geometry and efficiency, they were included in the final H-value with strong energy dependence.



**Fig. 2.** Comparison of used and measured H-values for SDD (left) and BEGe (right) detectors.

To validate the H-value calibration of both detectors, the prepared thick samples of IAEA reference materials were measured in the same geometry. Elemental composition of samples was used for calculation of attenuation coefficient of each reference sample in GUPIXWIN

with no additional correction. Calculated elemental concentrations of both reference materials are compared in Table 1 with their nominal values. The concentrations of elements below the detection limits of PIXE spectra were not compared.

As can be seen in Table 1, elemental concentrations of the IAEA-158 reference material are in good agreement with measured values, except for chromium, where a higher value was measured with SDD detector. The iron level in IAEA-359 was higher when compared with the reference value. The observed higher chromium and iron levels may be due to a contamination introduced by using a stainless-steel spoon during the sample removal from the mortar after its pulverizing. The iron contamination is not seen in the IAEA-158 sample due to its high iron content. Lower values of potassium and calcium observed in IAEA-359 may be due to inhomogeneities in the pressed samples.

**Table 1.** Elemental concentrations (mg kg<sup>-1</sup>) with standard deviations for used reference materials

		Reference value	SDD	BEGe
IAEA-158 Marine sediment	Na	23800 ± 1000	26200 ± 2400	
	Al	51800 ± 3400	52400 ± 5200	
	K	20000 ± 1600	19400 ± 1400	
	Ca	64900 ± 5800	78600 ± 7900	
	V	73 ± 3.7	69 ± 49	
	Cr	74.4 ± 5.8	514 ± 36	
	Mn	356 ± 24	385 ± 35	
	Fe	26300 ± 1400	25800 ± 2000	25800 ± 2100
	Ni	30.3 ± 2.9	24 ± 7.6	
	Cu	48.3 ± 4.2	47 ± 23	
	Zn	140.6 ± 9.5	145.5 ± 19	174 ± 14
	Sr	473 ± 25	432 ± 86	502 ± 15
	Mg*	10390 ± 960	11040 ± 900	
	Ti*	3490 ± 170	3430 ± 270	
	Br*	224 ± 15	199 ± 50	214 ± 11
IAEA-359 Cabbage	Rb	82 ± 10		123 ± 11
	Ba	1028 ± 46		905 ± 64
	Na*	580 ± 21	790 ± 60	
	Mg	2160 ± 50	1960 ± 170	
	K	32500 ± 690	27800 ± 2700	
	Ca*	18500 ± 510	15900 ± 1500	
	Mn	31.9 ± 0.6	43 ± 17	
	Fe	148 ± 3.9	243 ± 10	338 ± 14
Zn	38.6 ± 0.7	34.1 ± 10.2	41.5 ± 2	
Sr	49.2 ± 1.4		36.4 ± 2	

\*IAEA referred to as information values

## 4 Conclusion

For the successful use of the PIXE method in the CENTA laboratory, the energy dependence of the H-value was determined. The set of thin MicroMatter standards was used for calibration of SDD and BEGe detectors used for PIXE analysis. IAEA reference materials 158 (marine sediment) and 359 (cabbage) were used for validation of this calibration. Reasonable agreement was achieved between the reference and measured values for these

reference materials. Higher concentrations of iron and chromium were determined in both samples, which probably resulted from contamination during the sample preparation. Therefore, the preparation method needs further improvements. Presented H-value dependence will be used for PIXE analysis of various samples analysed in the CENTA laboratory.

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