



Original Article

# Evaluation of Thick Target PIXE Analysis in the 5SDH-2 Pelletron Accelerator Facility at VNU University of Science

Bui Thi Hoa<sup>\*</sup>, Nguyen The Nghia, Vi Ho Phong, Tran The Anh

*VNU University of Science, 334 Nguyen Trai, Thanh Xuan, Hanoi, Vietnam*

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**Abstract:** This paper presents the evaluation of the Thick Target Particle Induced X-rays Emission (TTPIXE) technique using standard samples. The element-dependent standardization factor,  $H$ , as a function of X-ray energies is calibrated using standard sample NIST-611 and validated with two standard samples: IAEA-Soil7 and NIES-Pepperbush. The obtained results are in good agreement with the reference data.

**Keywords:** PIXE, thick target, standard sample, calibration, 5SDH-2 pelletron

## 1. Introduction

Particle-induced X-rays Emission (PIXE) is the elemental analysis techniques using ion beam accelerated by small-sized accelerators [1-3]. This method is capable of analyzing quantitatively and simultaneously trace elements from sodium to uranium with many powerful features such as fast and non-destructive analysis, part-per-million sensitivity and low backgrounds [4].

The analysis approaches for PIXE analysis technique can be classified into two types based on the thickness of the target with respect to the energy loss of the incident ion beam: thin and thick target. Among these, the formulation for PIXE analysis is rather simple in the case of thin target. However, sample preparation is complicated, expensive and even impossible for several kinds of materials [5]. In the approach of thick target PIXE (TTPIXE) [6], sample preparation is easier and simpler than that for a thin target, but the spectrum analysis is more complicated due to additional considerations such as the slowing down of the incident ion and the absorption of the emergent X-rays inside the sample [7]. All the physical parameters relating to these effects, such as ion stopping power, X-ray attenuation factor,

<sup>\*</sup> Corresponding author.

*E-mail address:* [buiithoa.k55@hus.edu.vn](mailto:buiithoa.k55@hus.edu.vn)

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need to be taken into account in a proper and consistent manner for a quantitative analysis. Therefore, experimental conditions as well as analysis steps can easily affect the result [5, 8]. To take this challenge, the method should be well calibrated and validated using standard reference material (SRM).

The 5SDH-2 (two-stage) Pelletron Accelerator at VNU University of Science is designed with a maximum electrostatic accelerating voltage of 1.7 Megavolt. The accelerator is equipped with two ion sources, which can produce and accelerate a wide range of ion beam species from  $^1\text{H}$  (proton) up to  $^{92}\text{U}$  [9]. Currently, two beamlines are installed: The analytical beamline and the implantation beamline. The analytical beamline is equipped with the NEC RC43 end station, which is designed to perform ion beam analysis techniques including Rutherford Backscattering Spectrometry (RBS) and Channeling analysis, Elastic Recoil Detection Analysis (ERDA), Nuclear Reaction Analysis (NRA) and PIXE. The implantation beamline is dedicated to the application of MeV-ion implantation in materials science and industry. This study is carried out on the analytical beamline.

In this work, we evaluate TPIXE analysis technique, which is routinely performed in the 5SDH-2 Pelletron Accelerator at VNU University of Science using a set of standard samples. The results obtained from these procedures are presented and discussed.

## 2. Experiment

### 2.1. Sample Preparation

The thick standard sample NIST-611 [10] is utilized to calibrate the experimental constant  $H$ . The other two standard samples (IAEA-Soil7 [11] and NIES-Pepperbush [12]) are used to evaluate the precision and the sensitivity of the system. Each sample is analyzed three times on three different days to evaluate the stability of the analysis system.

The NIST-611 sample contains 61 elements presented at nominal abundances of about 500 ppm. The sample is the form of a circular-cross-section wafer with a diameter of about 12 mm and a thickness of 1 mm. It has a nominal composition of 72 %  $\text{SiO}_2$ ; 14 %  $\text{Na}_2\text{O}$ ; 12 %  $\text{CaO}$  and 2 %  $\text{Al}_2\text{O}_3$  in mass fractions. The samples are thick enough to be able to fully stop the incoming proton beam.

IAEA-Soil7 and NIES-Pepperbush samples are in fine powder form. Polyvinyl alcohol adhesive is added to bind powder particles together to avoid powder particle falling into analysis chamber under the high vacuum condition. The sample is pasted into a wafer as shown in Figure 1. Finally, the wafer which contains the sample is placed at target holder for irradiating.

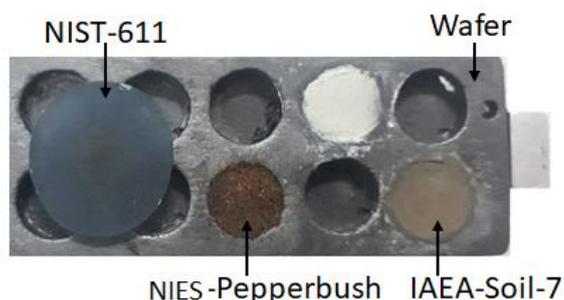


Figure 1. Prepared samples on the wafer.

## 2.2. Experimental Setup

The experimental setup for PIXE analysis is described in Figure 2. A proton beam accelerated by 5SDH-2 Pelletron Accelerator strikes into the target, resulting in electromagnetic radiation in the form of characteristic X-rays. The X-rays are detected by a Silicon Drift Detector (SDD) with a resolution of 139 eV at 5.9 keV, located at  $32.8^\circ$  to the direction of the incoming beam. The detector is connected to a multi-channel analyzer through electronic processing modules [12]. The angle between incoming beam and target plane is  $90^\circ$ . The target holder is connected with a current integrator to measure the number of incoming protons. A Mylar filter is placed between the sample and detector in order to filter out low energy X-ray in spectrum and back-scattered proton [13]. All the equipment and samples are placed in a scattering chamber with a vacuum of  $10^{-6}$  Torr.

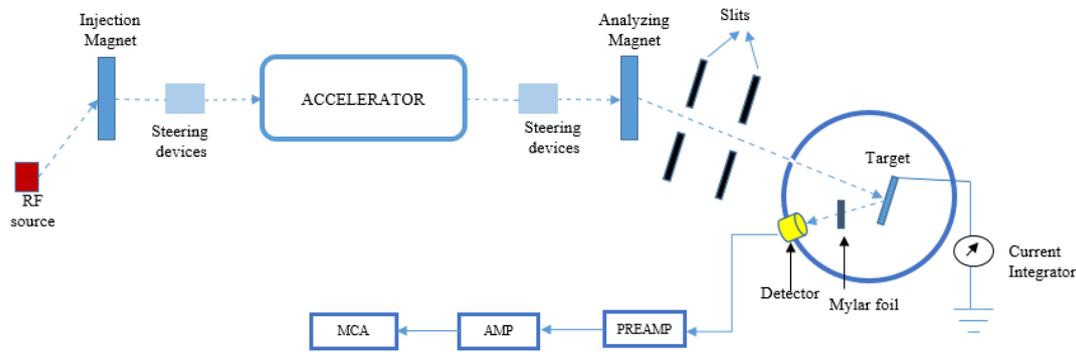


Figure 2. Experimental setup for PIXE analysis at 5SDH-2 Pelletron Accelerator.

Each sample is bombarded by two proton beams with different energy in order to identify all the elements: the low energy proton beam with energy of 830 keV only excites light elements from  $^{11}\text{Na}$  to  $^{26}\text{Fe}$ , thus suitable for determining the major elemental composition, which made up of the sample matrix; The high energy beam with energy of 2615 keV is used to analyze elements from  $^{26}\text{Fe}$  to  $^{92}\text{U}$ . A Mylar filter with a thickness of 12 (100)  $\mu\text{m}$  is used for the case of low (high) energy beam.

## 3. Data Analysis

The intensity of a particular X-ray line  $Y(Z)$  in thick target PIXE is described by the following formula [5]:

$$Y(Z) = \frac{N_{av} \omega_z b_z \Omega \varepsilon_z^i t_z N_p C_z}{A_z} \int_{E_0}^0 \frac{\sigma_z(E) \exp\left\{-\left(\frac{\mu}{\rho}\right) \frac{\cos \alpha}{\sin \theta_{TO}} \int_{E_0}^0 \frac{dE}{S(E)}\right\}}{S(E)}, \quad (1)$$

where  $N_{av}$  is Avogadro's number,  $\omega_z$  is the fluorescence yield,  $b_z$  is the fraction of the X-rays in the selected line,  $\Omega$  and  $\varepsilon_z^i$  are solid angle and the intrinsic efficiency of the X-rays detector, respectively,  $t_z$  is X-rays attenuation factor for any absorber placed between sample and detector,  $N_p$  is the number of incident protons with energy  $E_0$  at angle  $\alpha$  to the target normal,  $A_z$  is the atomic mass,  $\sigma_z(E)$  is the ionization cross-section for the corresponding shell,  $\frac{\mu}{\rho}$  is the mass attenuation coefficient within the sample matrix for the X-ray line of interest,  $\theta_{TO}$  is the X-ray take-off angle with respect to the target

normal. Elemental matrix of the sample is crucial in thick target PIXE analysis. It strongly affects the estimation of the attenuation of the beam energy and X-rays intensity in the sample.

GUPIX software is employed to process recorded spectra [14]. The software utilizes non-linear least-squares fit method with Marquardt algorithm for fitting. Yield of an X-ray line  $Y(Z, M)$  for an element  $Z$  in a matrix of elements  $M$  is described as follows:

$$Y(Z, M) = Y_t(Z, M) \cdot Q \cdot C_z \cdot H \cdot T_z \cdot \varepsilon_z \quad (2)$$

where  $Y_t(Z, M)$  is the theoretical yield,  $Q$  is the measured beam charge,  $C_z$  is the actual concentration of element  $Z$  in  $M$ ,  $T_z$  is the transmission of the X-rays through Mylar filter,  $\varepsilon_z$  is the intrinsic efficiency.  $H$  is an element-dependent standardization factor, which is initially assigned as the detector solid angle  $\Omega$ . The  $H$  value is used to correct systematic uncertainty of the analysis parameters such as the imperfect knowledge in calculating of  $Y_t(Z, M)$ , the thickness of the filter, imperfection of detector efficiency model and systematic bias of the measured beam charge. In this study,  $H$  value is calibrated using NIST-611 standard sample by comparing the elemental concentrations obtained with  $H = \Omega$  with certified concentrations to determine the respective correction factor. The calibrated  $H$  value is stored and used to analyze other spectra which are recorded with the same conditions. Ideally, standard sample should have a similar elemental matrix and density with samples intended to be analyzed.

For the low energy run, the concentrations of major and light elements can be analyzed using the option: “Interactive matrix solution” of GUPIX software. Since the effect of X-ray attenuation is more prominent for low energy X-rays of light elements, energy-dependent corrections for  $H$  values are applied to all elements emerged in the X-ray spectra. In the high energy run, a thicker filter is used to reduce excessive counts of low energy X-rays emerged from the major light elements. On this run, the primary focus on higher- $Z$  trace elements with higher energy X-rays allows us to use a single (energy-independent)  $H$  value.

#### 4. Results

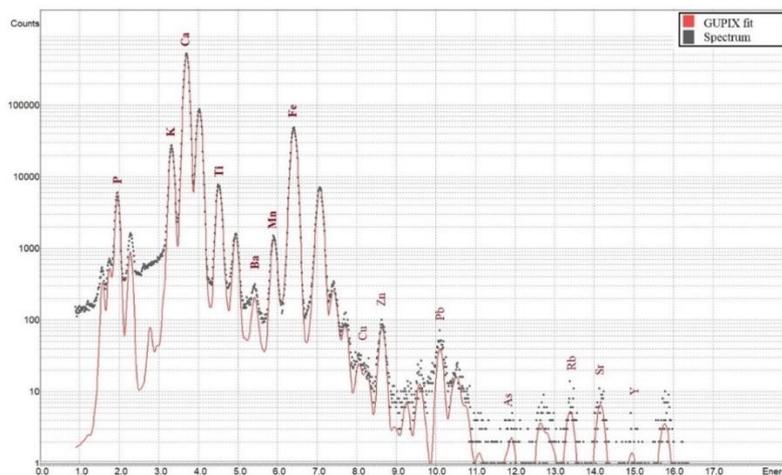


Figure 3. The PIXE spectrum (gray points) and fitted line (red) of IAEA-Soil7 sample bombarded with 2615 keV proton beam.

Table 1. Analysis results and the certified concentration of the IAEA- Soil7 sample.

Element	Analysis result (ppm)				Certified Concentration (ppm)
	Day 1	Day 2	Day 3	Average	
Na	2433 ± 553	2384 ± 483	2337 ± 490	2381 ± 292	2400 ± 48
Mg	11986 ± 304	11259 ± 285	11207 ± 284	11461 ± 168	11300 ± 194
Al	53302 ± 554	50724 ± 553	49856 ± 509	51208 ± 310	47000 ± 1696
Si	175316 ± 877	162886 ± 814	164027 ± 787	166960 ± 476	180000 ± 7752
P	481 ± 48	437 ± 43	469 ± 45	461 ± 26	460 ± 10
K	12474 ± 122	11934 ± 117	11820 ± 116	12063 ± 68	12100 ± 339
Ca	175246 ± 561	170267 ± 528	167346 ± 485	170578 ± 301	163000 ± 4118
Ti	2441 ± 78	3047 ± 84	3160 ± 99	2832 ± 50	3000 ± 266
Mn	661 ± 8	621 ± 8	657 ± 9	645 ± 5	631 ± 11
Fe	26615 ± 53	25155 ± 91	26786 ± 99	26336 ± 42	25700 ± 266
Cu	7 ± 3	12 ± 3	6 ± 3	9 ± 2	11 ± 1
Zn	102 ± 4	95 ± 4	103 ± 4	100 ± 2	104 ± 3
Ga	12 ± 3	9 ± 3	9 ± 3	10 ± 2	10 ± 1
As	23 ± 6	17 ± 6	16 ± 6	18 ± 4	13 ± 0
Rb	65 ± 10	49 ± 10	38 ± 10	50 ± 6	51 ± 2
Sr	105 ± 15	94 ± 14	102 ± 14	100 ± 8	108 ± 3
Y	16 ± 12	14 ± 11	20 ± 11	17 ± 7	21 ± 3
Ba	153 ± 44	164 ± 42	160 ± 46	159 ± 25	159 ± 16
Pb	43 ± 15	57 ± 14	51 ± 16	50 ± 9	60 ± 4

Table 2. Analysis results and the certified concentration of the NIES-Pepperbush sample.

Element	Analysis result (ppm)				Certified Concentration (ppm)
	Day 1	Day 2	Day 3	Average	
Mg	4716 ± 144	5566 ± 150	5228 ± 144	5161 ± 84	4080 ± 2
K	13850 ± 101	16052 ± 111	14981 ± 105	14895 ± 61	15100 ± 6
Ca	12200 ± 127	14155 ± 136	13058 ± 131	13093 ± 76	13800 ± 7
Mn	1700 ± 111	2360 ± 123	1997 ± 116	1995 ± 67	2030 ± 17
Fe	193 ± 6	183 ± 6	193 ± 6	190 ± 3	205 ± 17
Co	32 ± 3	30 ± 3	29 ± 3	30 ± 2	23 ± 3
Ni	10 ± 2	10 ± 2	11 ± 2	10 ± 1	9 ± 1
Cu	8 ± 2	8 ± 2	9 ± 2	8 ± 1	12 ± 1
Zn	392 ± 8	393 ± 9	429 ± 9	404 ± 5	340 ± 20
Rb	68 ± 13	77 ± 14	93 ± 15	79 ± 8	75 ± 4
Ba	182 ± 8	161 ± 8	187 ± 8	176 ± 4	165 ± 10

The fitted PIXE spectrum of sample IAEA-Soil7 is shown in Figure 3 as an example. The analysis results and certified concentrations are summarized in Table 1 and Table 2 for sample IAEA-Soil7, NIES-Pepperbush, respectively. The results obtained on different days and their averaged concentrations are shown separately in each column. The average is calculated from the measurement results and weighted by their errors and used to compare with certified concentration. The results of all elements in the two samples are consistent with certified concentration. The ratio of the measured and certified concentration is shown in Figure 4 for both samples IAEA-Soil7 and NIES-Pepperbush. The deviations from certified concentration are almost less than 20%. It is just slightly larger for elements whose concentrations are small (just around 10 ppm) such as, Y in sample IAEA-Soil7 and Cu in sample NIES-Pepperbush, so the analysis errors of these elements are large. The significant difference in composition of NIST-611 and the two other samples also effects the results.

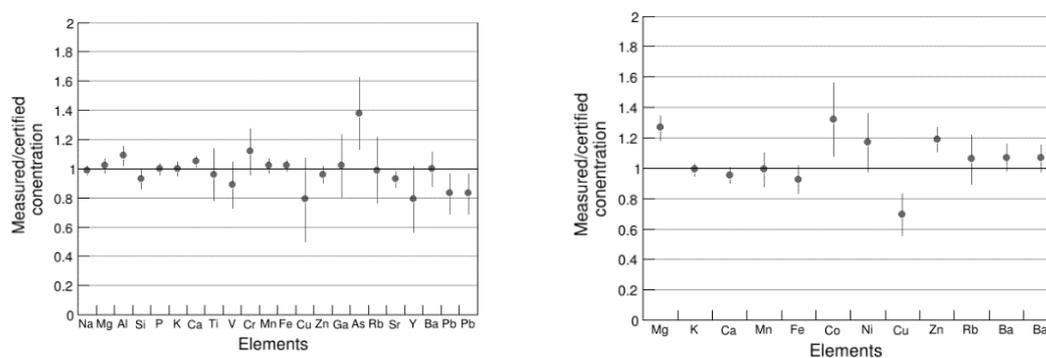


Figure 4. The ratio of measured and certified concentration of IAEA-Soil7 (left) and NIES-Pepperbush (right).

## 5. Conclusion

We have evaluated the thick target PIXE analysis system on the 5SDH-2 tandem accelerator at VNU University of Science using standard samples and GUPIX software. The instrument constant H was calibrated using NIST-611 sample. The calibrated system was validated with two standard samples from IAEA and NIES. The stability of the system was evaluated by irradiating and analyzing the samples on three different days. The results obtained on different days were consistent and in good agreement with certified concentrations. This shows that the system is well calibrated and stable.

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