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AN EXPERIMENTAL SETUP FOR PIXE ANALYSIS IN A MEDICAL CYCLOTRON AT TENMAK-NUKEN

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Abstract

A 30 MeV cyclotron is operated at TENMAK-NUKEN for producing medical radioisotopes with three beamlines and a fourth beamline is dedicated for research purposes. The minimum energy of extracted proton beam from cyclotron is 15 MeV. There is no facility in Türkiye for applying ion beam analysis techniques (IBA) currently. These techniques generally require 1-5 MeV proton beam energy. An energy degrader system was designed and installed on the R&D beamline for this purpose. The degrader system is capable of decreasing the energy down to 1 MeV with pA to µA current levels. A high vacuum irradiation chamber is designed and installed at the end of the beamline. The chamber has ports to install several types of detectors for different IBA techniques. This work includes the description of the setup and preliminary PIXE measurements.

INTRODUCTION

Cyclotrons are compact particle accelerator systems used for scientific research, medical and industrial applications. Turkish Energy, Nuclear and Minerals Research Agency (former Turkish Atomic Energy Authority) acquired a 30 MeV energy proton cyclotron with associated systems dedicated to the production of medical radioisotopes and scientific research purposes in 2011. The accelerator facility (TENMAK-PAF) has three beamlines for production of medical isotopes and another beamline for scientific research purposes. The research vault comprises a five-port electromagnet which can deflect the proton beam up to an angle of ∓40° with respect to the beam axis. The minimum beam energy and current provided by the cyclotron is 15 MeV and 0.1 µA respectively. This constraint on energy and current excludes certain applications. One of these applications is ion beam analysis (IBA) which finds use in a wide range of disciplines such as chemistry, biology, solid state physics, materials science, archaeology, anthropology etc. Non-destructive IBA techniques in general and certain irradiation requests from internal/external researchers require relatively low particle energies and low beam currents, i.e. 1-5 MeV energy with pA-nA current levels. A way to decrease the energy and the current is to let the beam pass through a thin foil during which the particles interact with the foil medium causing a decrease in energy and current. In this work, development and tests of the irradiation system and preliminary studies with PIXE method are described.

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IRRADIATION SETUP

Low-energy and low-current irradiations for the required absorbed doses demand additional effort and equipment in medical cyclotrons due to the design priorities for this type of accelerators. Therefore an energy degrader system is developed to decrease the energy of the cyclotron extracted beam. The cyclotron's original beam current measurement system is capable of measuring the current down to 0.1 µA which is convenient for radioisotope production purposes but not suitable for material irradiations and non-destructive analysis. Thus, a beam current measurement system is designed to measure the beam current down to tens of pA levels. Also, an irradiation chamber, an energy degrader system and various collimators were manufactured to irradiate assorted types of samples while controlling the beam energy, size and position (see [1] for details). The irradiation system requires additional conditions for shielding, positioning of the particle/radiation detectors and data acquisition systems to perform non-destructive analysis (Fig. 1). The secondary gamma and neutron radiation generated from the energy degraders is the primary concern due to increased background radiation in X-ray and gamma analysis. A water-cooled degrader system is installed in the cyclotron room, just after the extraction of the beam, to decrease the radiation background in the R&D room.

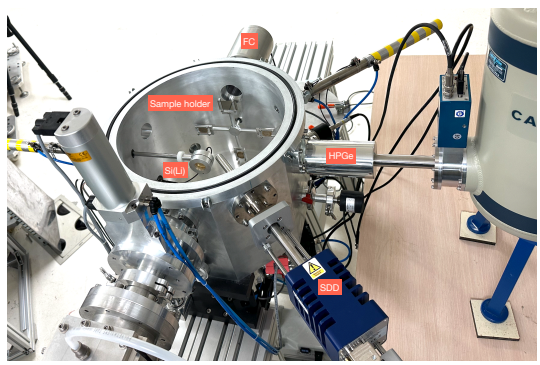

Figure 1: Irradiation system and detector placement.

Energy Degrader System

A water-cooled degrader (Fig. 2) system degrades the beam energy down to 3 MeV from 15.6 MeV using three stacked pyrolytic graphite foils with a total thickness of 1.3 mm. The foil stack is sandwiched between copper blocks and aluminum flanges for adequate cooling and collimation. The cooling system of the degrader assembly utilizes deionized water from the cyclotron cooling loop with a flow of

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6 L/min. The water circulates in welded copper tube and the system is tested up to 350 W beam power with inconsiderable temperature increase. The collimators are manufactured from aluminum to avoid persisting induced radioactivity. A safety interlock in the PLC software is put into effect to avoid overheating of the degrader with a PT100 sensor.

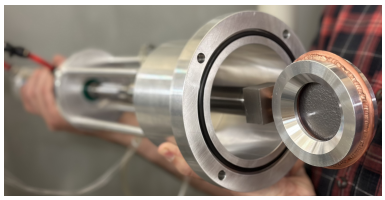

Figure 2: Water-cooled energy degrader.

Irradiation Chamber

An in-vacuum irradiation chamber is installed at the end of the R&D beamline. The chamber has openings for various devices to measure the beam profile, current, energy and detectors to detect secondary particles and prompt radiation. A port has a pneumatic cylinder which moves a 40 mm laser marked alumina beam screen in front of the beam at 45° to the beam direction. A port with a glass window allows the observation of the beam profile on the screen and on the samples. The other ports are reserved for radiation detectors for other ion beam applications. The chamber has a four-arm motorized carousel sample holder to irradiate different samples in a single run. A Faraday cup (FC) is installed at the far end of the chamber to measure and monitor the beam current. The beam from the 5-port magnet enters the chamber through a conical collimator and a replaceable circular aluminum collimator along the beam axis. The former has four isolated electrical connections to monitor the beam position and size. The vacuum is maintained at around $1x10^{-6}$ mbars with a two-stage pump system.

Radiation Detection System

A Si(Li) detector (RT-500-5-E/T-M/B, Mirion Technologies) is used for measuring the beam energy and energy spread. This detector has 5 mm thickness, a resolution of 30 keV at 5.5 MeV energy and is capable of measuring beam energies up to 30 MeV. The energy calibration of this detector is done in vacuum with a planar source of mixed alpha emitters (Americium-241, Plutonium-239, Uranium-234/238). A pneumatic cylinder moves the Si(Li) detector in front of the beam perpendicular to the beam axis. A vacuum operable silicon drift detector (SDD-EDS, Rayspec) for PIXE analysis is installed at an angle of 45 degrees with the beam axis. This detector has a thickness of 8 µm and a resolution of 133 eV at 5.9 keV. These specifications are convenient for distinguishing X-rays of different elements up to 20 keV energy with sufficient counting efficiency.

BEAM SHAPING AND MEASUREMENTS

IBA techniques require low energy and low current ion beams compared to a beam from a medical cyclotron. The extracted beam from the cyclotron has a minimum energy of 15 MeV and minimum measurable current of 0.1 µA. A degrader system is used to lower the beam energy down to the energies required for IBA techniques. The beam diffused by the carbon foil is refocused with the quadrupole doublets and shaped with collimators. The beam energy is degraded to 3.7 MeV with an energy spread of 0.97 MeV FWHM (measured with Si(Li) detector) for an incoming beam energy of 15.9 MeV. Monte Carlo [2] simulations and actual measurement results of the energy and the spread of the beam after the degrader system is shown in Fig.3. The measured peak shape is close to the simulation results except for a lower FWHM. The reason for higher FWHM in the simulations may be not taking into account the two quadrupole doublets in the code.

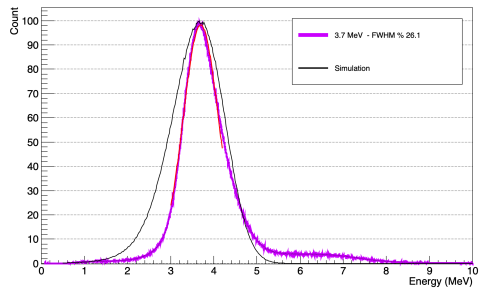

Figure 3: Si(Li) measurement and FLUKA simulation.

The following measurements were made for two different settings in the experiments: In the first case, beam shaping collimator aperture at the entrance of the irradiation chamber is 1 mm. The recofused beam is measured as 235 pA in the irradiation chamber Faraday cup for a beam current of 10 µA at the cyclotron stripper. In the second case, for a collimator aperture of 10 mm, the beam current is measured as 7 nA in the irradiation chamber Faraday cup for a beam current of 4 µA at the cyclotron stripper. This corresponds to a 0.002% and 0.175% current reduction for 1 mm and 10 mm final collimator aperture, respectively. The current stability at pA levels is shown in Fig.4. It is possible to lower the current down to 5 pA level with the ∓2.5 pA uncertainty by optimizing the ion source parameters and applying data manipulation methods such as noise filtering. The stability of the current is important for charge integration which directly affects the uncertainty analysis, especially for non-conducting samples in IBA techniques.

The beam size is measured as 11.4 mm on the alumina screen with a 10 mm collimator. Homogeneity of the beam is measured as 90% for 8 mm beam size using image processing techniques (Fig.5). This homogeneity ensures irradiating the samples as evenly as possible thus allowing bulk PIXE analysis of inhomogeneous samples with higher accuracy.

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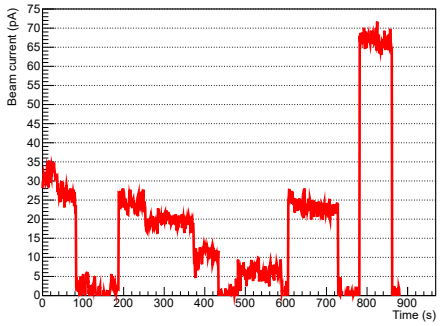

Figure 4: Current stability.

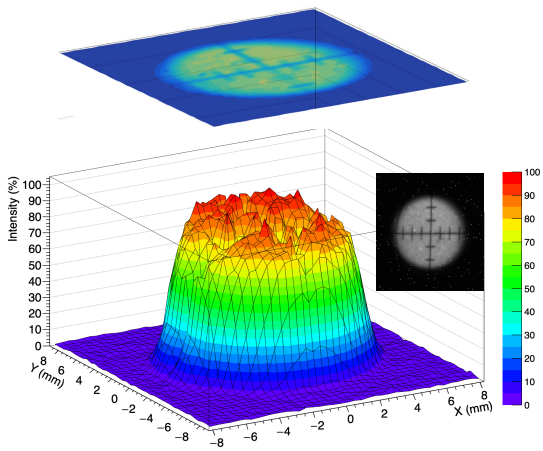

Figure 5: Beam homogeneity measurement.

PRELIMINARY PIXE ANALYSES

Several preliminary experiments are done with the non-destructive analysis system constructed in TENMAK PAF R&D irradiation vault. GUPIXWIN [3] software is used in the analyses. This software offers several PIXE analysis modes. One of these methods, analysis with H value, uses a certified reference material to calculate a value which is expected to remain almost constant for a well-defined geometry, surface roughness and sample composition for the X-ray energies of interest (1-20 keV). The instrumental value, H incorporates detector solid angle and calibration factor of charge integration [4]. It also includes corrections for geometry, detector crystal thickness and any imprecise data used in the software [5]. The H value method is selected in the experiments as the initial choice of analysis technique due to ease of use, less dependence on energy resolution and time consuming precise geometric parameter measurements of the setup required for standardless analysis. The reference concentration values of the element(s) in the standard foil is used to calculate the H value of the analysis setup.

The angle between the sample surface normal and the detector normal is approximately 10°. The X-ray spectra are acquired with a digital analyser (CAEN). 99.99% gold with a thickness of 50 µm is irradiated to determine the H value and its variation with X-ray energy by a proton beam

Table 1: Comparison of XRF and PIXE Analyses of the Standard Mix Foil

Element	XRF(%)	Unc.(∓)	PIXE(%)	Unc.(∓)
Copper	56.17	N.A	56.16	0.22
Nickel	41.66	N.A	41.95	0.17
Manganese	1.29	N.A	1.23	0.008
Iron	0.68	N.A	0.57	0.005

with an energy of 3.2 MeV and an average current of 7 nA for 155 seconds. The H value calculated with the irradiated gold standard foil is 0.000489. This H value is used for the analysis of 99.99% copper and mix standard foil (composition: 55% Cu, 45% Ni, thickness: 100 µm) irradiated under the same experimental conditions. The elemental composition is measured as 100% for copper foil and the analysis results of the mix standard foil are given in Table 1. PIXE spectrum of the mix standard foil is shown in Fig. 6. The analysis results indicate negligible dependence of the H value on X-ray energy in the spectral region of interest. The elemental concentrations of the mix sample is compatible with measurements done with a portable XRF spectrometer (no uncertainties are reported with this detector). The software reports the counting statistics uncertainty and peak fitting uncertainty for the PIXE analysis. The reported uncertainty values in Table 1 is the combined uncertainty of these two components. The major component of the combined uncertainty is due to the peak fitting process. Reducing the background in the setup and setting the peak fitting parameters more precisely are expected to decrease the uncertainty and the limit of detection considerably.

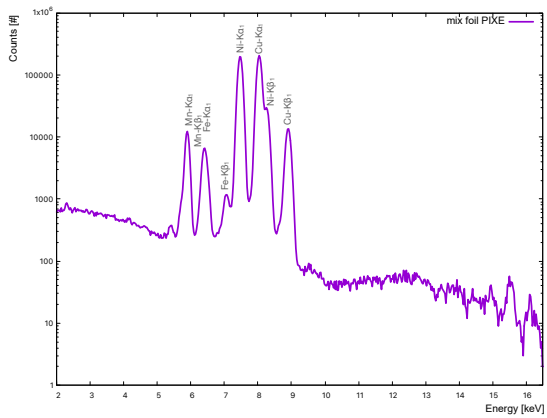

Figure 6: PIXE spectrum of the mix standard foil.

As for real life samples, the setup is used to determine the elemental content of 1 TL (Turkish Lira) metal coins. Coins minted on different dates are placed in the experimental chamber with a mix standard foil. This foil is used to determine the H value. Table 2 shows a comparison of PIXE and portable XRF analysis results. We believe that the main reason for the differences is the changes in the sample position due to the holder mechanism in the PIXE test system

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Table 2: Comparison of XRF and PIXE Analysis Results of 1 TL Metal Coins

Element	1 TL(2009)		1 TL(2015)		1 TL(2022)	
	PIXE	XRF	PIXE	XRF	PIXE	XRF
	%		%		%	
Copper	73.2	71.78	71.22	70.56	76.70	72.63
Nickel	14.37	15.63	14.78	16.34	15.02	15.96
Zinc	9.12	9.53	8.74	9.13	8.57	8.50
Manganese	0.14	0.13	0.33	0.262	0.15	0.12
Iron	0.17	0.17	0.46	0.44	0.07	0.05

and the background X-rays produced by scattered protons inside the chamber. Therefore, several improvements are planned in this direction such as using a collimator around the detector, an absorber on the detector endcap and voltage suppressors around the sample for stray electrons and coating the chamber walls.

CONCLUSION AND OUTLOOK

A non-destructive analysis setup at TENMAK-PAF medical cyclotron facility is designed and assembled in the R&D vault. This setup is consists of a number of additional beam shaping equipment and measurement systems required by IBA methods. PIXE method is applied for the first time in Türkiye and the preliminary results are satisfactory. Detectors for IBA techniques such as PIGE (HPGe) and RBS (PIPS) detectors are installed and measurements are ongoing. Further modifications are needed to improve the accuracy of the measurements through the use of electron suppressors, adjusting the measurement geometry (i.e. moving the sample closer to the detector), shielding and/or collimating the detectors. Future plans also include moving the setup to the 40° port (rightmost port of the 5-port magnet) and utilizing a water-cooled slit to achieve a better energy resolution. These changes will have a positive impact on the reliability of the measurements in the application of complementary ion beam analysis methods.

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