Predictive Modeling of Radioisotope Production in Intermediate Energy Proton Irradiation Using PHITS-DCHAIN

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1. Introduction

Proton irradiation is a useful method for simulating the effects of neutron irradiation on materials, providing advantages in terms of both time and cost [1]. Typically, a 3 MeV proton beam is utilized due to its minimal risk of inducing proton-related nuclear reactions, facilitating straightforward post-irradiation analysis, as inferred from Figure 1. Nevertheless, the availability of high-current 3 MeV proton beams in the Republic of Korea is notably limited, posing challenges for the local research community in employing this technique. While Tandem-type accelerators can reliably deliver such low-energy proton beams, the predominant types of proton accelerators within the country are either linear accelerators or cyclotrons, both of which generally operate at energies exceeding 10 MeV.

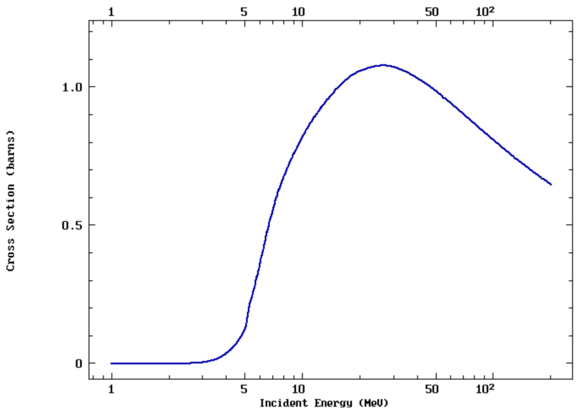

Fig. 1. Production cross-section of residual nuclei, $\sigma(p, X)$ of Fe-56 in JENDL-5 library [2] used in current PHITS-DCHAIN calculation.

Meanwhile, the concept of Intermediate Energy Proton Irradiation (IEPI), denoting irradiation with protons above 10 MeV, is gathering interest [3]. Despite the potential for nuclear reactions that could result in sample radioactivity, the method is recognized for its significant advantages. While concerns about radioactivity exist, IEPI's ability to closely mimic nuclear fusion conditions and to facilitate comprehensive testing of bulk samples—owing to the deep penetration capabilities of intermediate energy protons, reaching depths of 100 μm and potentially up to 200-300 μm in metals—makes it an attractive option. In this context, our study aims to explore the feasibility of IEPI research using domestic facilities, initially by identifying radioactive nuclides in samples irradiated with 12 MeV protons.

2. Method and Result

2.1. IEPI experiment

Proton irradiation experiments were conducted at the Korea Multi-purpose Accelerator Complex (KOMAC) in Gyeongju, Korea, utilizing a 20 MeV proton beamline (TR23) with proton energies set at 12.1 MeV. Figure 2 illustrates the experimental setup. The material selected for the irradiation experiments was commercial grade SS316, commonly chosen for proton irradiation research because of its widespread application. The chemical composition of SS316 sample is analyzed by inductively coupled plasma mass spectrometer (ICP-MS), as shown in Table 1.

Table 1: Chemical composition of commercial grade SS316 sample used in IEPI experiment.

	^{nat}Cr	^{nat}Mn	^{nat}Fe	^{nat}Ni	^{nat}Cu	^{nat}Mo
Comp. (wt%)	18.31	0.94	67.39	10.87	0.38	2.12

Prior to irradiation, the specimen surfaces were meticulously polished, starting with SiC sheets ranging from 800 to 2000 grits, followed by a SiC suspension from 6 μm to 0.25 μm, and concluding with an Al_2O_3 suspension of 0.05 μm. The total proton fluence reached was 2.0 × 10^{14} p/cm^2, achieved through a pulse current delivering 1.16 × 10^{10} p/cm^2 per pulse. Using this setup, achieving the total fluence required a duration of 40 minutes, with the beam precisely collimated to a 1.5 cm radius circle. Throughout the experiment, the temperature increase in the sample was carefully controlled, not exceeding +12 °C, as verified by a thermocouple attached to the sample's rear.

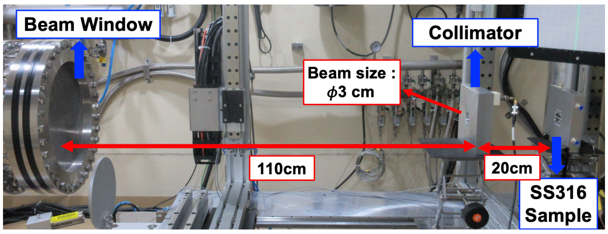

Fig. 2. Photo of the experimental setup at the 20-MeV beamline of KOMAC (TR23).

2.2. HPGe measurement analysis

Gamma-ray spectra of the samples were obtained following cooling periods of 3 and 42 days to ensure decay of short-lived radionuclides. Measurements were performed using a p-type coaxial high-purity germanium (HPGe) detector, interfaced with an 8k channel PC-based analyzer. The HPGe detector showed an energy resolution of 0.875 keV at the full-width at half-maximum (FWHM) and a relative efficiency of 30% at 122 keV. To mitigate the effects of Compton scattering and signal pile-up, the samples were positioned at a considerable distance from the detector's surface. Gamma-ray spectral analysis was conducted using Genie-2000 software. Figure 3 illustrates the HPGe detection results 3 days post-irradiation, highlighting the decay of short-lived nuclides like ^{95}Tc and ^{52}Mn. This finding is further confirmed by a subsequent HPGe analysis of 42 days post-irradiation (not shown in this abstract). Table 2 lists the identified radionuclides detected through HPGe measurements, along with their respective half-lives. There, the abundance of parent nuclide is also presented considering the natural abundance of each element.

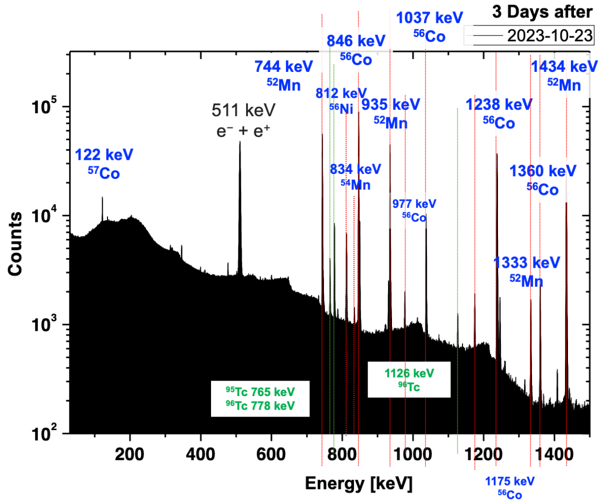

Fig. 3. Gamma-Ray Spectral Analysis 3 Days Post-Irradiation Using High-Purity Germanium Detector

Table 2: Identified Radioisotopes in SS316 Samples After 12.1 MeV Proton Irradiation

Radio Isotope	Half-life[d]	Decay constant [/h]	Abundance of Parent nuclide in the sample[wt%]
^{95}Tc	0.83	3.47×10^{-2}	^{95}Mo(0.34%)
^{96}Tc	4.28	6.75×10^{-3}	^{96}Mo(0.35%)
^{52}Mn	5.59	5.17×10^{-3}	^{52}Cr(15.34%)
^{56}Ni	6.08	4.75×10^{-3}	^{56}Fe(61.84%)
^{56}Co	77.24	3.74×10^{-4}	
^{57}Co	271.74	1.06×10^{-4}	^{57}Fe(1.43%)
^{54}Mn	312.20	9.25×10^{-5}	^{54}Cr(2.4%)

2.3. PHITS-DCHAIN modeling of IEPI experiment

Isotope production and resulting radioactivity from irradiation were quantified using the PHITS (ver 3.33) and DCHAIN codes [4], based on the experiments carried out at KOMAC. The PHITS simulations, involving Monte Carlo transport calculations for the proton beam used in the experiments, indicated a statistical error of less than 10^{-2}/source. Following this, the DCHAIN code was employed to solve the Bateman equations and estimate the sample's radioactivity. These estimations are depicted in Figure 4, which also includes the experimentally measured radioactivity counts obtained using a NaI detector. Due to the lack of calibration for the NaI detector setup, only the relative intensities of the radioactivity are shown. From these results, we can tentatively validate that the decay trend of radioactivity is reasonably predicted by the PHITS-DCHAIN simulation. The discrepancies observed are likely attributed to the limitations in the current nuclear reaction cross-section library. Efforts to rectify this include cross-comparison with other libraries, such as TENDL, to refine the accuracy of the decay trends observed in proton-irradiated samples.

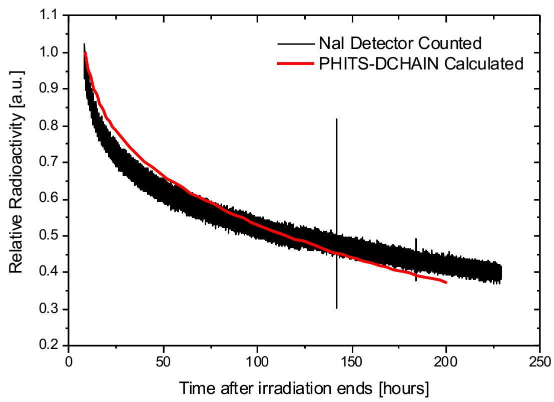

Fig. 4. Comparison of Relative Radioactivity Over Time: NaI Detector Measurements Versus PHITS-DCHAIN Simulations

3. Conclusion

This research aimed to evaluate the practicality of conducting Intermediate Energy Proton Irradiation (IEPI) experiments using a domestic proton irradiation facility. Before conducting the experiment, it is important to predict the types of radioactive nuclides produced, for which the PHITS-DCHAIN computational approach was employed. The subsequent comparison of these predictions with experimental results has validated that the modeling can reliably identify the types of radioactive nuclides generated. Furthermore, an attempt was made to predict post-experimental radioactivity levels. Due to limitations in the experimental setup for measuring absolute radioactivity, a comparison of relative radioactivity values between the experiment and the model was performed. This comparison indicated that the current model can reasonably forecast the decay trends of the sample, which could inform researchers of the necessary cooling period for the sample before further analysis. As indicated, future work will include enhancing the calculation results by investigating the impact of the nuclear reaction cross-section library. It is anticipated that refining the modeling will eventually permit the estimation of absolute radioactivity in samples following IEPI experiments.

Acknowledgement

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