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Depth profile of production yields of $^{nat}Pb(p, xn)\ ^{206,205,204,203,202,201}Bi$ nuclear reactions

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ABSTRACT

Experimental and simulation studies on the depth profiles of production yields of $^{nat}Pb(p, xn)\ ^{206,205,204,203,202,201}Bi$ nuclear reactions were carried out. Irradiation experiments were performed at the high-intensity proton linac facility (KOMAC) in Korea. The targets, irradiated by 100-MeV protons, were arranged in a stack consisting of natural Pb, Al, Au foils and Pb plates. The proton beam intensity was determined by activation analysis method using $^{27}Al(p, 3p1n)^{24}Na$, $^{197}Au(p, p1n)^{196}Au$, and $^{197}Au(p, p3n)^{194}Au$ monitor reactions and also by Gafchromic film dosimetry method. The yields of produced radio-nuclei in the ^{nat}Pb activation foils and monitor foils were measured by HPGe spectroscopy system. Monte Carlo simulations were performed by FLUKA, PHITS/DCHAIN-SP, and MCNPX/FISPACT codes and the calculated data were compared with the experimental results. A satisfactory agreement was observed between the present experimental data and the simulations.

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1. Introduction

In recent years, several large accelerator facilities have been constructed in Korea. Rare Isotope Science Project (RISP) [1] and Korea Heavy Ion Medical Accelerator (KHIMA) [2] facilities have been designed and constructed. Pohang Accelerator Laboratory X-ray Free Electron Laser (PAL-XFEL) [3] is in commissioning stage. Korea Multi-purpose Accelerator Complex (KOMAC) [4], Korea National Cancer Center (KNCC) [5] and Samsung Medical Center (SMC) [6] have serviced their hadron beam for various applications. In the frame of above projects, for the safety issue and shielding analysis of the accelerator facilities, extensive studies including experiments and simulations on the production yields of residual nuclei induced in the accelerator materials such as Pb, Cu and Bi by protons and heavy ions are in progress.

Lead (Pb) is important as a shield material at accelerator facilities and is widely used in different nuclear technologies as a target material. Many experimental studies have been carried out on the measurement of production yields and cross-sections of radio-nuclei induced in natural Pb target by a wide range of proton energies in recent years [7–21]. However, around 100-MeV proton range, only a few experiments have been conducted so far [20,21].

In this work, we extended experimentally and theoretically earlier works. Production yields of $^{nat}Pb(p, xn)\ ^{206,205,204,203,202,201}Bi$ nuclear reactions at different depth points of the natural Pb target irradiated by the 100-MeV protons were measured. The measured data were compared with the results calculated by using the FLUKA [22], PHITS [23], and MCNPX [24] codes. The (p, xn) nuclear reactions were selected owing to their independent cross-sections which caused independent production yields. Cross-sections for the production of a nuclide is denoted as independent if the nuclide can only be produced directly through the nuclear reaction between the projectile and the target nucleus and not via subsequent β^-, β^+, EC or α-decays [20]. Therefore, such yields are more valuable for verification of the nuclear models implemented in the codes.

2. Experiments and methods

2.1. Target and irradiation conditions

In this paper, results of two irradiation experiments were reported. A layout of the one of experimental sets-up, described by SimpleGeo4.3 software [25], is illustrated in Fig. 1. High-purity natural Pb as the activation foils and Al and Au as the monitor foils were used to avoid contamination production resulting from interaction of protons with elemental impurities. The weight and thickness of each foil and plate were measured before irradiations.

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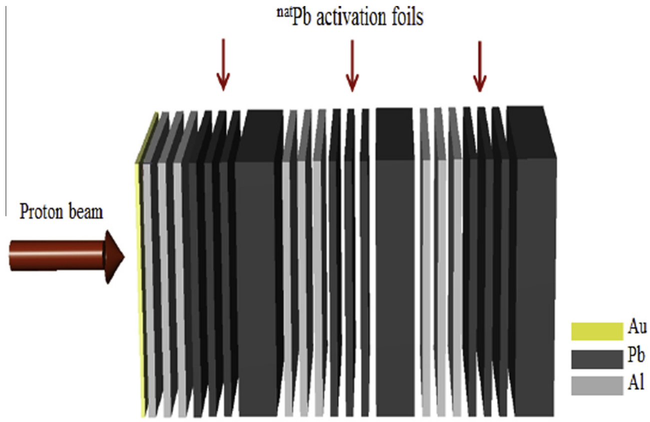

Fig. 1. Experimental set-up described by simpeGeo4.3 software.

The target was arranged in a stack consisting of an Au (19.3 gcm^{-3}, 99.95% purity), nine Al (2.69 gcm^{-3}, 99.999% purity) and nine Pb (11.35 gcm^{-3}, 99.99 + % purity) foils together with three 5-mm thick Pb plates in the form of Au-three Al-four Pb-three Al-four Pb-three Al-four Pb array. The typical thickness of individual Au, inner Al, outer Al and Pb foils was 30, 200, 100, and 125 µm, respectively. Natural Pb activation foils were placed between two Pb foils to avoid cross contamination and recoil effects and only inner Pb foils were used for radio-nuclei measurements. Similar to the Pb activation foils discussed above, Al monitor foils also were placed between two Al foils. The cross-sectional size of the target was about 5 cm × 5 cm. Natural Pb plates were placed between Pb activation foils to degrade proton beam energy twice. The total thickness of Pb plates was calculated using the computer program SRIM-2008 [26], assuming that the incident proton energy into the target was 103 MeV. Condition of another irradiation experiment was similar to the discussed irradiation except for the thickness of natural Pb activation foils and beam monitor foils. In this irradiation, the thickness of Pb activation foils was 50, 500 and 500 µm, respectively and also only Au foils were used to measure proton beam intensity. In each irradiation, the total thickness of targets was considered to be larger than the range of proton beam. In addition, a vacuum window composed of Al_4Be_6 (500 µm thickness, 2.1 gcm^{-3}) was placed at the end of the vacuum beam pipe at a distance of 1.7 m from target. Au foil at the beginning of the target stack, and Al foils were placed to determine proton beam intensity. Besides, a Gafchromic film with a dimension of 7 cm × 7 cm was placed in front of the target to measure beam intensity and two dimensional profile.

Both experiments were performed at the high-intensity proton linac facility (KOMAC). The targets were irradiated for 10 and 3.4 min with 103-MeV protons (1 Hz repetition rate). The beam shape at the target was described by Gaussian distribution in both x and y coordinates (the z-coordinate is the beam direction) based on the beam profile on the Gafchromic film. The Full Width at Half Maximum (FWHM) of the distribution was 1.9 and 1.8 cm in x- and y-direction, respectively. The energy of incident proton beam was calibrated to be 103 ± 0.13 MeV using multi-energy degrading techniques [27].

The decay data were taken from National Nuclear Data Center (NNDC) [28] (see Tables 1 and 2). The cross-sections for the monitor reactions ^{27}Al(p, 3p1n)^{24}Na, ^{197}Au(p, p1n)^{196}Au, ^{197}Au(p, p3n)^{194}Au were taken from Refs. [21,29].

2.2. Gamma-ray spectrum measurement

After irradiation, the gamma-ray spectra of Au, Al, and Pb foils were measured using an HPGe detector with a relative efficiency of 15% at 5 cm from the surface of detector. In each experiment, measuring the production yield started about half an hour after the end of the irradiation due to measure the production yields of short-lived radio-nuclei. The efficiency of the HPGe detector was experimentally determined using standard multiple gamma-ray sources, ^{152}Eu, ^{137}Cs, and ^{133}Ba at 5 cm from the surface of detector. The resolution of the HPGe detector was 1.94 keV for gamma ray of 1.33-MeV. The spectrum analysis was done by the Canberra's Genie2000 gamma analysis software package (version 3.2) [30].

3. Simulation of experiment

Monte Carlo simulations were performed by FLUKA (version 2011.2c.0), PHITS (version 2.64) and MCNPX (version 2.7) codes and compared with the measurements to verify validity of physical models and nuclear data libraries in the Monte Carlo codes. The measured and calculated production yields of ^{206,205,204,203,202,201}Bi radio-nuclei were compared at the end of irradiations. The input model of simulations was quite similar to experiments, consisting of the proton beam, Al_4Be_6 window and target.

We used the default setting for precision simulations "PRECISIO" in FLUKA. The production yields of radio-nuclei in the ^{nat}Pb activation foils were calculated using RESNUCLEi scoring. FLUKA physical models are described in Refs. [22,31]. Additionally, the energy distributions of proton beam onto the front surface of Pb, Au, and Al foils were calculated by FLUKA code. The production yields of Bi radio-nuclei were estimated using MCNPX code with an inventory code, FISPACT (version 2007) [32], and PHITS code with an inventory code, DCHAIN-SP (version 2001) [33]. In this work, Bertini [34] and INCL4.6 [35] models were used in MCNPX and PHITS, respectively to calculate neutron flux spectrum. Consequently, the calculated differential neutron flux was used as an input of the inventory codes, FISPACT and DCHAIN-SP to determine yields of products. The inventory codes require neutron energy spectrum, total neutron flux, mass of each element, irradiation scenario such as irradiation and cooling time as the input data to provide production yields of radio-nuclei as an output.

Table 1
Decay characteristics of monitor reactions and contributing reactions [28].

Nuclide	Half-life	E_γ [keV]	I_γ [%]	Contributing reactions	Q-value [MeV]
^{196}Au	6.17 d	333.03	22.9	^{197}Au(p, p1n)	−8.07
ε: 93% β^-:7.0%		355.73	87.0		
^{194}Au	38.02 h	293.54	10.6	^{197}Au(p, p3n)	−23.29
ε: 100%		328.50	60.4		
^{24}Na	14.99 h	1368.62	99.9	^{27}Al(p, 3p1n)	−31.43
β^-:100%		2754.0	99.8		

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