Comparison of Code Simulation and Measurements: Radio-activation of ZrO₂ by Proton Beam Irradiation

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1. Introduction

In proton irradiation experiments with more than a few MeV in proton energy, radio-activation of targets is essentially caused by proton-induced nuclear reactions. In this paper, we irradiated 20 MeV proton beam to ZrO_2 target with average current of 400 nA. We measured emitted gamma ray energy to detect residual isotopes in the ZrO_2 target by using HPGe detector, and we also measured gamma dose rate by using radiation survey meter. Then we calculated radio-activation of ZrO_2 target by Monte Carlo code simulation to compare measured and calculated results.

2. Experiments

2.1. Proton beam irradiation

To measure activation of ZrO₂, we irradiated proton beam to $ZrO_2 + Y$ (3mol%) compound (Table 1). The 20 MeV pulsed proton beam line (Fig. 1), which was installed at KAERI (Korea Atomic Energy Research Institute), was used for this experiment with 4 mA current, pulse width of 100 µsec, repetition rate of 1 Hz, and flux of 7.95 ×10¹¹ proton/cm²·sec. We put ZrO₂ target in front of 0.5 mm aluminum window so the protons lost their energy about 2.8 MeV by penetrating beam window. The actual incident proton beam energy to ZrO₂ target was about 17.2 MeV. We irradiated 1257 pulses, so total proton beam flux was about 1.00×10¹⁵ proton/cm²·sec.



Fig 1: 20 MeV beam line at KAERI (left), and proton beam irradiated ZrO_2 target.

Table 1: Characteristics of ZrO ₂ sample					
Molecular	Density	Sample			
Formula	$[g/cm^3]$	Geometry			
$ZrO_2 + Y$ (3 mol%)	6.05	$\phi 10 \text{ mm} imes 1 \text{ mmt}$			

2.2. Code simulation

First, we calculated secondary neutron produced by proton beam with MCNPX v. 2.5 code [1] and production rate of radioisotopes in ZrO_2 target with PHITS v. 2.23 code [2]. With these results of MCNPX and PHITS code simulation, we calculated radioactivity and gamma-ray energy intensity of equipments with various proton beam operation condition by using DCHAIN-SP code [3]. Finally we evaluated gamma dose rate from the previously calculated gamma-ray energy intensity with MCNPX code [4]. The schematic diagram of the model applied to the code simulation is shown in Figure 2.



Fig. 2: The schematic diagram for code simulation

2.3. Measurement and Comparison

After proton beam irradiation to the target, dose rate of the ZrO_2 target was measured by radiation survey meter (FH40GL-10, THERMO), and residual radioisotopes of target were analyzed by a high purity germanium detector (GR1518, Canberra, Inc., relative efficiency: 15%, Full width half maximum @1.33 MeV: 1.8 keV). Before we measured residual radio-isotopes of target, we had measured gamma ray background for a day to subtract environmental gamma ray background [5]. In this method, we can verify only gamma emitted radio-isotopes, but pure beta emitter. Finally, we compared calculated residual radio-isotopes and dose rate with measured data.

3. Results

In this experiment, we could measure residual radioisotopes and dose rate immediately after proton beam irradiation. Because calculated dose rate of the ZrO_2 target at 0 day in figure 4 was about 30 mSv/h. It was excess dose rate for handling the ZrO_2 target for person, so we waited 20 days to cool down the radioactivity of the ZrO_2 target. Then we measured residual radioisotopes and dose rate.



Fig. 3: Measured gamma ray energy spectrum.

Radio- Isotope	T _{1/2} [d]	Energy [keV]	Intensity	Activity [nCi]	
			[%]	Measured	Calculated
^{95m} Nb	3.61	235.69	24.8	-	0.063
⁸⁷ Y	3.33	388.53	82.2	-	0.61
		484.80	89.8	-	
⁹⁵ Zr	64.0	724.19	44.27	45.60	44.45
		756.72	54.38	45.43	
⁸⁸ Zr	83.4	392.9	97.30	190.2	186.86
⁹⁵ Nb	34.9	765.8	99.81	1447.0	1399.67
⁸⁸ Y	107	989.04	93.7	34.63	31.80
		1836.06	99.2	35.03	
⁸⁹ Zr	3.27	909.15	99.04	2086.0	1672.16
		1657.3	0.106	2080.0	
		1713.0	0.745	2044.0	
		1744.5	0.123	2043.0	
^{92m} Nb	10.2	912.93	1.78	6412.0	5743.79
		934.50	99.07	6454.0	
		1847.5	0.85	6658.0	

Table 2: Major residual radio-isotopes in ZrO₂ target.

Figure 3 show background subtracted gamma ray energy spectrum of the proton beam irradiated ZrO₂ target. We compared calculated and measured radioactivity of the proton beam irradiated ZrO₂ target (Table 2). The major residual radio-isotopes of the proton irradiated ZrO₂ target are ⁹⁵Nb, ⁸⁹Zr, ^{92m}Nb, and ⁸⁸Zr. The difference was about maximum 10 %. In this result, we excluded radio-isotopes having a long half life, because their production was too small to detect by HPGe detector.



Fig. 4: Measured and Calculated of dose rate for ZrO₂ target.

As the results, we might think that code simulation of the proton beam irradiated ZrO_2 target was well fitted to real results. For radiation safety of person who experiments proton beam irradiation to some targets, we need to simulate radio-activation of targets to estimate level of dose rate before proton beam irradiation.

4. Conclusion

The major residual radio-isotopes of the proton irradiated ZrO_2 target are ⁹⁵Nb, ⁸⁹Zr, ^{92m}Nb, and ⁸⁸Zr. The difference of measured and calculated activity is about maximum 10 %. The dose rate of measured and calculated for the proton irradiated ZrO_2 target was 3.0 μ Sv/h and 3.51 μ Sv/h respectively.

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