Activation of Polymethyl-Methacrylate by Proton Beam

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1. Introduction

Activation has been used for many kinds of useful applications, such as use of the radioisotopes for diagnosis and treatment of cancer, and wear (or corrosion) investigation for using thin layer activation (TLA) technique [1], etc. But activation also has severe problems for the post-processing of the samples; such as time-loss, inconvenience of sample handling, personal radiation safety, etc. For in-vitro experiments, we observed death of tumor cells by proton irradiation. The use of large activated container material can cause erroneous results in this case. To solve these problems, we studied why the samples were activated and how the level of the activation could be reduced. In proton beam irradiation experiments, the target materials could be defined as the container and sample itself. We could easily reduce activation of container material comparing to activation of sample itself. Therefore, we tried to find less activated container material by irradiating proton beam in PS (polystyrene), PMP (polymethypenten), PMMA (poly methacrylate), PVC (polyvinyl chloride), and PP (polypropylene), which are the base materials of the bottles and Petri dishes, frequently used in laboratories. We used 45 MeV proton beams (MC-50 Cyclotron, KIRAMS) [2] with 10 nA. In this study, we presented characteristics of proton beam irradiated PMMA.

2. Experiments

2.1. Sample Preparation

To measure activation of container, we prepared 3 sets of PMMA (C5H8O2) sample. The first, we needed to observe that protons deposit all their energy inside of sample so we calculated proton beam range (thickness) by using the SRIM (The Stopping and Range of Ions in Matter) code [3]. The calculated proton range of PMMA was about 11.9 mm, and we piled up each plate by the calculated depth (Table I). And then we used Qvalue calculator [4] to expect produced stable or radioisotopes and their Q-value. Also we use TALYS code [5] to confirm major produced radio-isotopes. Table II shows nuclear reactions, Q-values, cross-sections, gamma-ray energys, and their half lifes of produced radio-isotopes. To verify most influential radio-isotope for dose rate, we prepared 2 sets of sample and separated each sample by 2 part (Table II); 39 MeV -23.6 MeV and 23.6 MeV - 0 MeV (the Q-value of ^{11}C is about 23.6 MeV.), and 39 MeV - 13.4 MeV and 13.4 MeV - 0 MeV (the Q-value of 15 O is about 13.4 MeV.).

Table I: About sample						
SET	Molecular	Calculate	Sample			
	Formula	Depth	Geometry			
1	39 MeV - 0 MeV	11.9 mm	1.2 mm $ imes$ 11 sh.			
2	39 MeV - 23.6 MeV	7.1 mm	6 sh. + 5 sh.			
	23.6 MeV - 0 MeV	4.8 mm				
3	39 MeV - 13.4 MeV	10.2 mm	9 -1 · 2 -1			
	13.4 MeV - 0 MeV	1.7 mm	8 sn. + 3 sn.			

Table II: Possible nuclear reactions and their properties when protons irradiate in PMMA.

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Nuclear	Q-value	XS	E	Half			
Reaction	[keV]	[Barn]	[keV]	life			
12C(p, d)11C	-16497.0	7.242E+01	511	20.4m			
12C(p, n+p)11C	-18721.6						
	-23359.5	2.629E+00	511				
12C(p, t)10C			718	19.2 s			
			1021				
$^{16}O(p, d)^{15}O$	-13439.3	6.625E+01	511	122 s			
${}^{16}O(p, n+p){}^{15}O$	-15663.9						
$^{16}O(p, t)^{14}O$	-204051	3.350E+00	511 2312	70.6 s			
${}^{16}O(p, n+d){}^{14}O$	-26662.4						
${}^{16}O(p, 2n+p){}^{14}O$	-28886.9						
${}^{16}O(p, a){}^{13}N$	5218 /	1.311E+01	511	9.97m			
${}^{16}O(p, p+t){}^{13}N$	-3210.4						
${}^{16}\text{O}(\text{p, n}+{}^{3}\text{He}){}^{13}\text{N}$	-23032.3						
${}^{16}O(p, 2d){}^{13}N$	-23790.0						
$^{16}O(p,$	-29003.0						
$(n+p+d)^{13}N$	-31289.5						
$^{16}O(p, 2n+2p)^{13}N$	-55514.1						
${}^{16}O(p, d+a){}^{11}C$	-23659.0	1.117E+01	511	20.4m			
$^{16}O(p, n+p+a)^{11}C$	-25883.5						

2.2. Proton Beam Irradiation

The 45 MeV beam line (Fig. 1), which was installed at KIRAMS (MC-50 cyclotron, Korea Institute of Radiological and Medical Science), was used for this experiment with 10 nA current and flux of 1.274×10^{10} cm²/sec. We put each sample 30 cm away from 2mm aluminum window so the protons lost their energy about 6 MeV by penetrating beam window and air gap. The actual incident proton beam energy was about 39 MeV. In our experiments, irradiation time of proton beam varied from 0.5 to 5 minutes which is to observe dose rates depending on irradiation time and radioisotopes having short life time; ¹⁰C (T_{1/2} : 19.2 s) and ¹⁴O (T_{1/2} : 70.6 s).



Fig. 1, 45 MeV beam line of MC-50 cyclotron at KIRAMS.

2.3. Measurement and Analysis

After proton beam irradiation, dose rate of the each sample was measured by radiation survey meter (FH40GL-10, THERMO) and we fitted the results by exponential decay function to verify dominant decay components. Activation of samples was analyzed by an HPGe detector (GR1518, Canberra, Inc., relative efficiency: 15%, Full width half maximum @1.33 MeV: 1.8 keV).

3. Results

After proton beam irradiation of different conditions, we measured the radiation dose rate for about an hour. Fig 2 shows dose rates of different irradiated times, and we fitted these results by exponential decay function. The dominant decay times was about 18 s, 119 s, and 1126 s, so we assumed that those were mixed ¹⁰C ($T_{1/2}$: 19.2 s), ¹⁵O ($T_{1/2}$: 122 s), ¹³N ($T_{1/2}$: 9.97 m), and ¹¹C ($T_{1/2}$: 20.4 m).



Fig. 2, Dose rates of different beam conditions (39 MeV - 0 MeV)



Fig. 3, Dose rate of last 3 sheets (the sample set of 13.4 MeV - 0 MeV)

To confirm this, we analyzed the sample of 13.4 MeV - 0 MeV (Fig. 3). We had expected it only had a decay component of ^{13}N and the result was well fitted. In case of the sample set of 23.6 MeV - 0 MeV, we could observed decay components of ^{13}N , and ^{15}O , except ^{11}C .

It is too difficult to observe radio-isotopes having short decay time. To observe peaks of ¹⁰C, and ¹⁴O, we analyzed the sample immediately after proton beam irradiation. We could observe the peaks (Fig. 4) when proton beam was irradiated for one minute. The major gamma ray peak was 511 keV (annihilation) because proton-rich nuclides usually decay by emitting positive electrons and neutrinos [6].



Fig. 4 Gamm-ray spectrum of PMMA (the first circle is 511 keV, the second is 10 C (715 keV), and the last is 14 O (2312 keV).

4. Conclusion

The major activation components of PMMA are ${}^{10}C$ (T_{1/2}: 19.2 s), ${}^{15}O$ (T_{1/2}: 122 s), ${}^{13}N$ (T_{1/2}: 9.97 m), and ${}^{11}C$ (T_{1/2}: 20.4 m). We could observe peaks of ${}^{10}C$, and ${}^{14}O$, but we could not quantify them. Also we could observe 511 keV from PMMA

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