ACTIVATION AND DISCOLORATION OF POLYMER BY PROTON BEAM*

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Abstract

During the beam irradiation experiments with more than a few MeV energetic protons, nuclear reactions are occurred in sample materials. Because of nuclear reactions, the samples are activated so many kinds of additional problems for the post-processing of the samples are caused; such as time-loss, inconvenience of sample handling, personal radiation safety, etc. For in-vitro experiments, we observe 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 are activated and how the level of the activation can be reduced. In our proton beam irradiation experiments, the target materials can 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), and PMMA (Poly methacrylate). We used 45 MeV proton beams (MC-50 Cyclotron, KIRAM) with 10 nA.

INTRODUCTION

When more than a few MeV energetic protons interact with matters, many nuclear reactions are occurred and a large number of stable and radioactive nuclides are produced in sample materials. Sample activation problem can be very severe because it causes many kinds of additional problems for the post-processing of the samples, such as time-loss, inconvenience of sample handling, personal radiation safety, etc. Most serious problem is that immediate treatment of the sample is impossible in some experiments such as in-vitro experiments. To solve these problems, we studied why the samples are activated and how the level of the activation can be reduced. It is known that the main reasons of activation are the nuclear reactions with elements of the target material by primary protons and secondary produced neutrons. Even though the irradiation conditions are same, the level of the activation can be different depending on the target materials.

The purpose of this study is to reduce sample activation. The sample activation can be defined as the container activation and activation of sample itself. Therefore, we tried to reduce the activation level by changing the container materials so we prepared some candidates of container materials, such as, PS (Polystyrene), PMP (Polymethypenten), and PMMA (Polymethyl methacrylate). They are base materials of bottles or beakers used in laboratory frequently.

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EXPERIMENTS

Sample Preparation

To measure activation of container, we prepared PS, PMP, and PMMA by 3×3 cm^2 which are base material of bottles or beakers and are usually used for container (Table 1). We designed that protons deposit all their energy inside of front part so we calculated proton beam range (thickness) of each material by using the SRIM (The Stopping and Range of Ions in Matter) code [1]. The calculated proton range of PS, PMP, and PMMA are about 13.5 mm, 15.5 mm, and 11.0 mm, respectively then we piled up each plate by the calculated depth (Fig. 1).

Table 1: About Samples

	Molecular Formula	Calculated Depth	Sample Geometry
PS	C_8H_8	13.5 mm	~1.2 mm ×12 sh.
PMP	C_6H_{12}	14.25 mm	~1.3 mm ×13 sh.
PMMA	$C_5H_8O_2$	11.57 mm	~1.2 mm ×10 sh.

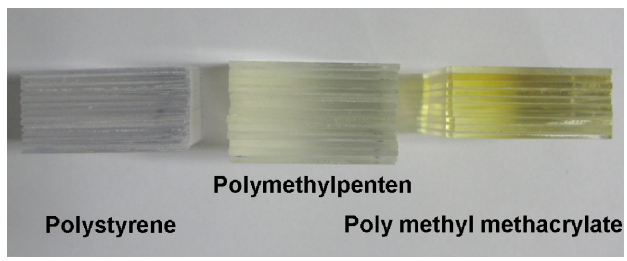

Figure 1: Photograph of target samples.

Proton Beam Irradiation

The 45 MeV beam line (Fig. 2), which was installed at KIRAMS (MC-50 cyclotron, Korea Institute of Radiological and Medical Science) [2, 3], was used for this experiment with 10 nA current and flux of 1.274 ×10^{10} cm^2/sec. We put each sample 30 cm away from 2mm aluminium window so the protons lose their energy about 6 MeV by penetrating beam window and air gap. The actual incident proton beam energy was about 39 MeV.

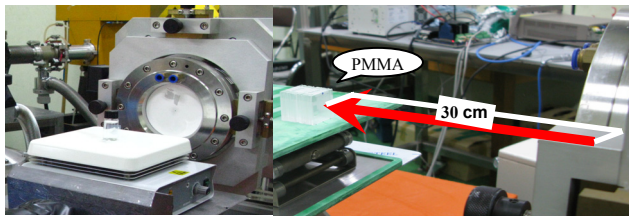

Figure 2: 45 MeV beam line of MC-50 cyclotron at KIRAMS.

Measurement and Analysis

During the beam irradiation experiments, we could observe discoloration of samples by installed CCTV and we assumed that the color changes were caused by proton bombardments. Optical transmittance of samples was measured by UV/VIS-Spectrophotometer (UV-2550, Shimadzu Corp.) and the results were compared with the calculated ionization of each sample. Activation of samples was analyzed by using an HPGe detector (GR1518, Canberra, Inc., relative efficiency: 15%, Full width half maximum @1.33 MeV: 1.8 keV).

RESULTS

Transmittance

The unirradiated samples are transparent but the color of proton-irradiated PMP and PMMA was changed to yellow (Fig. 1). We supposed that the color changes were caused by proton bombardments so we measured transmittance of each plate with UV/VIS Spectro photometer.

For PMP, the transmittance was decreasing from the 1st plate to the 11th plate then rapidly rose at the 12th plate. The thickness of colored plates was about 14.3 mm and an average calculated result was about 13.9 mm. In case of PMMA, the transmittances of the 1st to the 9th plate are not much different but the transmittance of the 10th plate rose to about 1. The results showed similar tendency to the calculated ionization values, and the locations having a maximum transmittance value (the 11th plate for PMP and the 9th for PMMA) were same as calculated peak ionization value (Fig. 3). Also we could see that protons deposited all their energy inside of samples.

Some kinds of carbon/glass fiber plastics show an LET effect by proton or neutron irradiation [4, 5]. Also, B. A. Briskman reported that the proton effect on optical transmission spectra in the visible region is highly more pronounced than the effect by neutron [6].

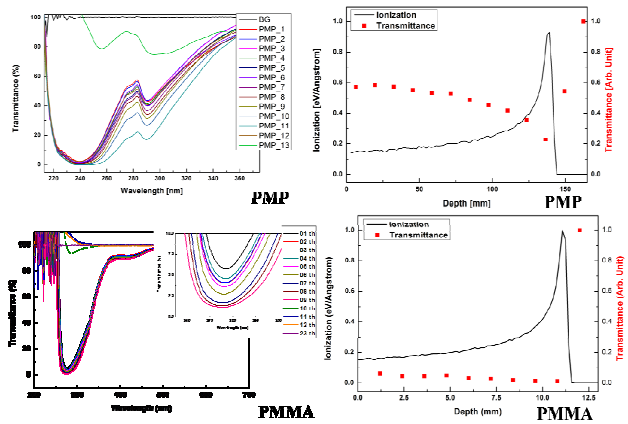

Figure 3: Ionization and transmittance of PMP and PMMA.

Gamma Ray Spectrum

Fig. 4 shows gamma ray spectra of samples. Dominant gamma energy is 511 keV by annihilation radiation. In β^+decay, positrons emit in the primary decay process and they combine with normal negative electrons near the end of their range. When the positron and electron disappear, they are replaced by two oppositely directed 511 keV photons [7]. In our experiments, lots of produced radio nuclides undergone β^+decay and have short decay time so we could have observed 511 keV peak until 2 days after proton beam irradiation. After that we could see ^{7}Be (477 keV) peak from all samples. (Fig.4)

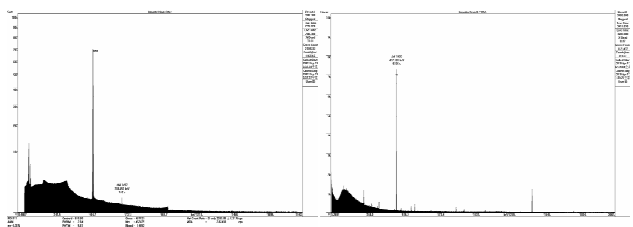

Figure 4: Gamma ray spectra of PS. The left figure is for one day after proton beam irradiation and main peak is 511 keV (annihilation). The right figure is for 5 days after beam irradiation and main peak is ^{7}Be (477 keV).

^{7}Be is the radioisotope with 477.60 keV ($T_{1/2}$: 53.22d) and its possible nuclear reactions are ^{12}C(p, d+α)^{7}Be, ^{12}C(p, n+p+α)^{7}Be, ^{13}C(p, t+α)^{7}Be, ^{13}C(p, n+d+α)^{7}Be, ^{13}C(p, 2n+p+α)^{7}Be, ^{17}O(p, n+d+2α)^{7}Be, and ^{17}O(p, t+2α)^{7}Be [8]. Among these reactions, the cross-section of ^{12}C (about 41.0 mb at 39 MeV) is the largest (Fig. 5). The Q-value of ^{7}Be is about 28 MeV for ^{12}C so only the 1st to the 5th ~ the 6th plates were emitted gamma-ray of 7Be. Table 2 and Fig. 5 (right bottom) shows measured activities of each plate. The total activities in order are

PS, PMP, and PMMA and it is similar to number of carbon in sample materials.

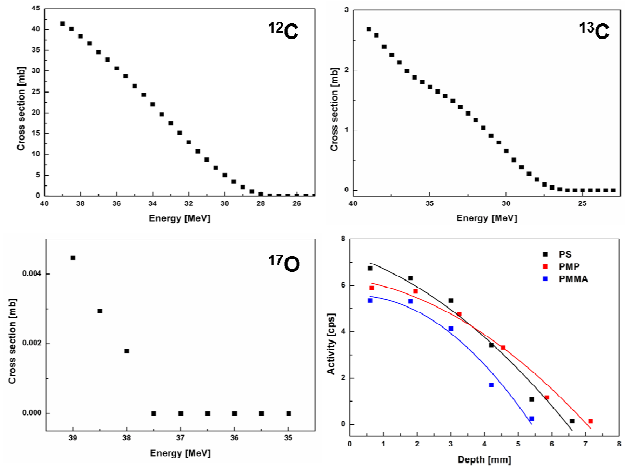

Figure 5: Cross-section of ^{12}C, ^{13}C, and ^{17}O [9] and activity of samples.

Table 2: Measured Activities of Samples [CPS]

	PS	PMP	PMMA
1st	6.71340	5.88869	5.34122
2nd	6.31667	5.73306	5.30833
3rd	5.34361	4.75562	4.12479
4th	3.41618	3.31292	1.69681
5th	1.08392	1.16486	0.24986
6th	0.13882	0.13590	-
Total	23.01259	20.99105	16.72101

CONCLUSION

The transmittance was decreasing from the 1st plate to the 11th plate then rapidly dropped at the 12th plate for PMP. The results show similar tendency to the calculated ionization values, and the locations having a maximum transmittance value (the 11th plate). In gamma ray spectra of PS, PMP, and PMMA, dominant gamma energy was 511 keV (annihilation). Lots of produced radio nuclides undergone β^+decay but they have short decay time so we could have observed 511 keV peak until 2 days after proton beam irradiation. We could also observe ^{7}Be and we assumed that the number of carbon in sample materials decides production of ^{7}Be.

REFERENCES

[1] James F. Ziegler, "SRIM 2003", (2003). [Online]; http://www.srim.org/.
[2] B. H. Choi, "Status of Proton Engineering Frontier Project", Proc. of the PAC 06, (2006).
[3] K. R. Kim, et al., "Proton beam energy measurement using semiconductor detectors at the 45 MeV test beam line of PEFP", Proc. of the PAC 07, (2007).
[4] B. Bodmann, et al., "LET effects of neutron irradiated plastic scintillators", Nucl. Instr. and Meth. in Phys. Res., B 208, p. 495–499, (2003).
[5] R. L. Clough, et al,." Discoloration and subsequent recovery of optical polymers exposed to ionizing radiation", Polym. Degrad. Stab. 49, p. 305-313, (1995).
[6] B.A. Briskman, "Specificity of proton irradiation effects on polymers", Nucl. Instr. and Meth. in Phys. Res., B 265, p. 72–75, (2007).
[7] Glenn F. knoll, "Radiation Detection and Measurement", John Wiley & Sons, Inc., (2000).
[8] "National Nuclear Data Center (NNDC)", Brookhaven National Laboratory, [Online] ; http://nndc.bnl.gov/
[9] A. J. Koning, et al., "TALYS-1.0", Proc. Of ND2007, EDP Science, p. 211-214, (2008).