PRELIMINARY RESULTS OF SAMPLE ACTIVATION MEASUREMENT USING A HPGE DETECTOR FOR THE NANO PARTICLE FABRICATION BY PROTON BEAM*

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Abstract

The sample activation during proton beam irradiation sometimes interrupts the instant measurement and investigation of the changes inside the samples after irradiation process. During the experiments for heavy metal nano-size particle fabrications with about 35MeV and a few micro ampere proton beam, we found that the samples were highly activated after proton beam irradiations. To investigate the radiation source from the samples, we measured the gamma-ray spectra using HPGe spectroscopy system. The results were compared to the calculated results by the MCNP code simulation. The samples were small amount of heavy metal, such as platinum and cobalt, dispersed in the water mixed by ethanol with 0.6% mole concentration in the Pyrex beaker. In addition, we studied how we can shorten the cooling time by controlling incident proton energy and container material.

INTRODUCTION

High energy proton beam can activate target material radiologically by the nuclear reaction, such as (p,n). Through this primary nuclear reaction between high energy proton and target material, neutron can be produced secondary, which also contribute to the activation of the sample additionally. The reactions are too complicate to estimate and analyze by code simulation. For this reason, measurements of the cross-sections of proton-induced reactions using HPGe gamma-ray spectroscopy system have been conducted continuously for many kinds of target materials and wide energy range of proton [1-2].

Radioactivation of target materials by proton beam is very useful phenomena for radioisotope production but not for other experiments, such as nano particle fabrication, etc. To reduce the activation level in nano particle fabrication experiments, we investigated the radiation source and activation level according to the proton beam energy, current, and container materials. We are showing the preliminary results in this paper.

EXPERIMENT

Code Simulation

For the first step, we evaluated the radioactivities of the proton-irradiated samples in nano particle fabrication by code simulations. The simulations were performed with computer codes Monte Carlo Neutral Particle eXtended (MCNPX), Particle and Heavy Ion Transport (PHIT), and induced radioactivity calculation code DCHAIN-SP to evaluate the amounts of radio nuclides produced by protons and secondary produced neutrons in the aqueous solution of the H_2PtCl_6 contained in the Pyrex beaker. The schematic diagram of model applied to the code simulation is shown in Fig. 1, and the results are displayed in Fig. 2. As shown in Fig. 2, activation level is dependent on the irradiation time and decreased after 100 minutes rapidly. It means that the radiation sources for high activities are very short half life time. According to the calculation results, 511-keV gamma ray is mainly contributed to the increase of the activation level of samples. The 511-keV gamma ray can be produced by the annihilation processes.

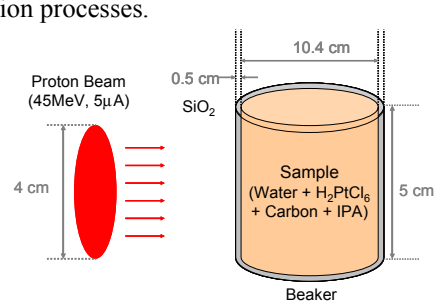

Figure 1: Modeling for MCNPX code simulation for Pt nano particle fabrication.

Nano Particle Fabrication Experiment

Nano particle fabrication experiments were conducted by using 42-MeV proton beam. The 10 ml vial filled with H_2PtCl_6 aqueous solution with 0.5% concentration was

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irradiated until the colour of solution was changed from light yellow to black, which means that the Pt nano particle produced. It takes 18 sec with 1 μA and 300 sec with 10nA beam current. Fig. 3 shows the pictures of the vial before and after beam irradiation.

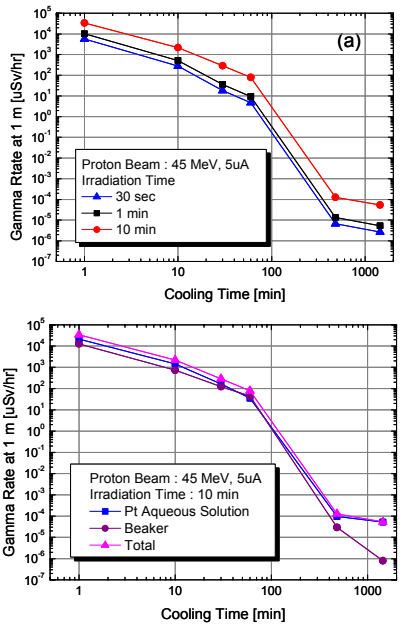

Figure 2: Results of MCNP code simulation for Pt nano particle fabrication. Cooling down time after irradiation of 45-MeV proton with 5μA according to (a) irradiation time and (b) samples.

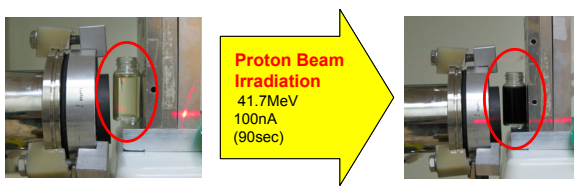

Figure 3: Photograph of the nano particle fabrication.

RESULTS

Gamma-Ray Spectra

We measured gamma-ray spectra from samples after 45-MeV proton beam irradiation with 5 μA beam current for 5 minutes. In the gamma-ray spectra shown in Fig. 4, 511keV gamma-ray peak can be identified.

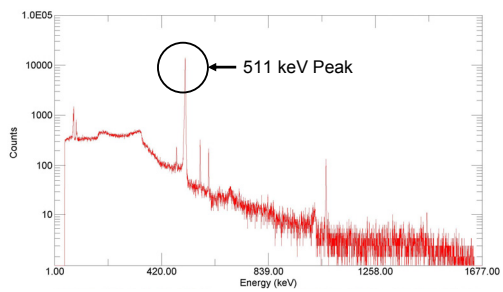

Figure 4: Gamma-ray spectrum for proton beam irradiated sample.

The gamma-ray spectra for pyrex are shown in Fig. 5. The peak height of 511-keV gamma ray peak was reduced to 1/5 after 400 seconds. It is well agreed with calculated results. But it is not impossible to measure radioactivity and sources reaction because the nuclear reactions too complicated. In the results of the measurements for kapton, PMMA and quartz, etc. we can identified that activation level for kapton film is the least among these various kinds of materials.

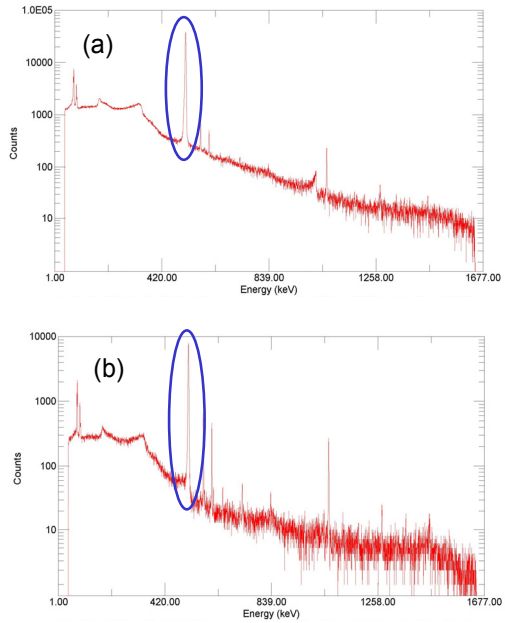

Figure 5: Gamma-ray spectra for pyrex beaker (a) 30 sec later, (b) 430 sec later.

Metal elements contained in the solution can be transferred to radioactive isotopes by proton beam irradiation. For the Cobalt nano particle fabrication using $CoSO_4$ aqueous solution, radioactive isotopes, such as ^{57}Co ($t_{1/2}$: 271.79 days), ^{56}Co ($t_{1/2}$: 77.27 days), ^{58}Co ($t_{1/2}$: 70.86 days), and ^{60}Co ($t_{1/2}$: 5.27 yr.), can be produced. The spectra measured HPGe gamma-ray spectroscopy for $CoSO_4$ aqueous solution and Cobalt nano particle powder produced by proton beam irradiation were displayed in Fig. 6. Radioactive isotopes produced by nuclear reaction, such as ^{57}Co-57, ^{58}Co, and ^{60}Co, were detected.

Gamma Rate Measurement

The gamma rates from the irradiated samples were measured by using the survey meter, ESM FH 40 G-L (Thermo Electron Co.) for the various irradiation conditions. The proton energies were 42 MeV and 20 MeV, beam current were 10, 50, 100, 500, 1000, 2000 nA. Gamma rate of instant measurement is dependent on the proton energy and beam current. It was increased with the increase of proton beam energy and current. The results are summarized in Fig. 7. The gamma rates were decreased rapidly as the lapse of time. To reduce the activation level of the samples for nano particle fabrication, we have to choose proper proton energy below 20 MeV.

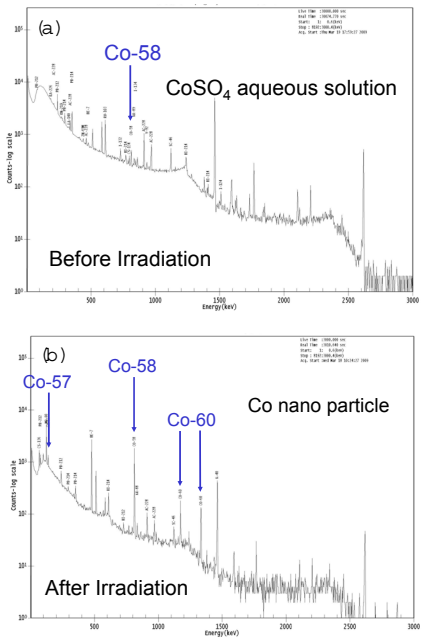

Figure 6: Gamma-ray spectra from (a) $CoSO_4$ aqueous solution and (b) cobalt nano particles fabricated by proton beam irradiations.

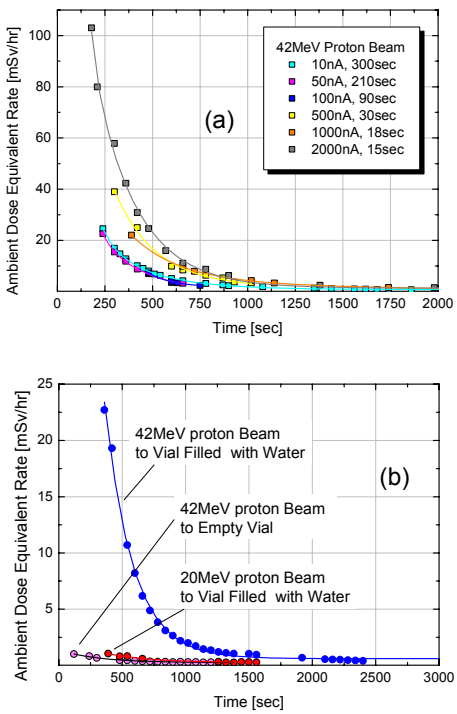

Figure 7: Ambient dose equivalent rate for 10 mL glass vial filled with water according to (a) the irradiation time and beam current, (b) proton energies.

CONCLUSION

We investigated the activation levels of various materials concerned with nano particle fabrication process using high-energy proton beam irradiation. By using HPGe detector system, gamma-ray spectra from activated samples were also measured. The 511keV annihilation radiation is the main reason for the high activation level of samples irradiated by high-energy proton beam. The results was agreed well to the calculation results by MCNPX. For the results, we recognized that to minimize the activation problem, we have to reduce the incident proton energy below the Q-value and choose proper materials for the fabrication process.

REFERENCES

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[2] M. U. Khandaker, et al., "Measurements of the proton-induced reaction cross-sections of ^{nat}Mo by using MC50 cyclotron at the KIRAMS" J. Korea Phys. Soc., Vol. 48, No. 4, pp. 821~826, 2006.