III. 4. Experimental Investigations on the Proton-induced Activation Reactions on Tantalum and Molybdenum

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**Introduction**

Tantalum and molybdenum are the important structural materials that are used in various parts of accelerator. Tantalum is important as target for the production of medically important radioisotopes such as $^{178}$W/$^{178}$Ta, $^{172}$Hf/$^{172}$Lu, $^{177}$Lu etc. Tantalum is a monoisotopic element, so it has special interest to test model calculations.

Molybdenum is used as target material for the production of medical radioisotopes like as $^{99m}$Tc/$^{99}$Mo, $^{96}$Tc etc. $^{99}$Mo is the parent nuclide of the daughter $^{99m}$Tc, which is widely used in diagnostic nuclear medicine. Due to their nuclear decay properties, this radioactive pair is well suited for the production and distribution of the $^{99}$Mo $\rightarrow$ $^{99m}$Tc generator over the world. The amount of $^{99}$Mo consumed for radiopharmaceuticals increases year by year. The $^{99}$Mo is produced commercially by using nuclear reactors and these are based on the (n,$\gamma$) and (n,fission) nuclear reactions. Currently, there is no supply of accelerator-made $^{99m}$Tc or $^{99}$Mo anywhere. But it is possible to make production of $^{99m}$Tc and $^{99}$Mo by the proton bombardment via the most suitable reactions $^{100}$Mo(p,2n)$^{99m}$Tc and $^{100}$Mo(p,pn)$^{99}$Mo respectively. Molybdenum is also useful as a refractory and corrosion resistant material in accelerator applications.

Activation cross-section for Ta+p and Mo+p reactions are of interest in various fields, like accelerator technology, charged particle activation analysis, thin layer activation, medical isotope production, model calculations etc. The present work was undertaken to give new and reliable excitation functions and thick target yields for Ta+p and Mo+p reactions in the energy range 20-70 MeV.
Experimental techniques

The excitation functions of the proton-induced activation reactions on tantalum and molybdenum were measured using a conventional stacked foil technique. The stacked samples were irradiated with 70 MeV collimated proton beam of 100 nA and 200 nA using a k= 90 MeV AVF Cyclotron at Cyclotron and Radioisotope Center (CYRIC) of Tohoku University. Cu and Al foils were inserted into stack as monitor, beam energy degraders and to check the beam parameters. We have to mention that separate irradiations were performed for tantalum and molybdenum.

The activity of the residual nuclei was measured nondestructively by HPGe gamma-ray detector. For short-lived radionuclides, measurements were done about 10 minutes after end of irradiation. To minimize the relative errors of the calibration curve, several gamma lines were used to determine the activity for a given radionuclides, where it was possible. The detector efficiency versus energy curve was determined experimentally using the standard gamma-ray point sources, $^{152}$Eu, $^{133}$Ba, $^{241}$Am, $^{60}$Co, $^{137}$Cs etc.

The proton energy degradation along the stack was determined using the computer program SRIM-2003$^1)$. The proton beam intensity was determined via the monitor reactions$^2)$, $^{27}$Al(p,x)$^{22,24}$Na and $^{nat}$Cu(p,x)$^{62,63}$Zn taken place at the front radioactive monitor foils of the stack considering that the monitor foils were irradiated simultaneously and measured with the same detector and in a comparable geometry as the targets. From the decay rates of the radioactive products and the measured beam current, the cross-sections were deduced for the productions of the radionuclides. It has been observed that two or more radionuclides emitted gamma-rays having very close energies, which cannot be resolved by HPGe-detector. We have separated the contribution to the peak area from different processes using their another independent gamma-rays and by establishing decay curve. The data were corrected for the sum-coincidence effect caused by the coincidental detection of two or more gamma-rays in cascade by using the SUMECC code$^3)$.

Results and discussion

Activation cross-sections were measured for the production of $^{90,93m,99}$Mo, $^{93m,93,94m,94,95m,95,96,97}$Tc, $^{89,89m,90,92m,95,96,97}$Nb, $^{86,88,89,90,92}$Zr and $^{174,175,176,177,178}$Y from molybdenum target and $^{174,175,176,177,178}$Ta, $^{173,174,175,176,177,178}$W, $^{173,175}$Hf and $^{179}$Lu from tantalum target. The thick target integral yields were derived by using measured excitation functions. This work has given new data for all of the investigated radionuclides. We have also remeasured the excitation functions for monitor reactions and obtained good consistency
with recommendation. Although, it has been found that the recommended cross-section data for $^{27}$Al(p,x)$^{22}$Na reaction are not in agreement with this work below 50 MeV.

The main problem in the stacked foil technique is energy degradation calculation along sample layers. We observed that the reliability of SRIM calculation decreases with the increasing depth of stack. It has been corrected by comparing with recommendation data of monitors.

The efficiencies obtained by using $^{133}$Ba standard source were deviated from the common trend of other sources. The established efficiency versus energy curve was consistent with EGS4 Monte Carlo Code$^4$.

We observed that two or more radionuclides emitted gamma-rays having very close energies which were difficult to separate by the graphical analysis of the multicomponent decay curve. We analyzed data using the difference in half life of the nuclides by plotting the photon emission rate as a function of time. An example on the radioactive decay curve for 140.5 keV peak energy is shown in Fig.1. From Fig.1, we see that the daughter, $^{99m}$Tc activity grows to a maximum and then decreases at a constant rate which depends on the decay rate of the parent, $^{99}$Mo and that fact indicates the parent-daughter transient equilibrium, $^{99}$Mo $\rightarrow$ $^{99m}$Tc. Actually the directly produced $^{99m}$Tc completely decayed out before the measurement. The measured activity was the sum of 140.5 keV gamma from the daughter, $^{99m}$Tc and 141.2 keV from $^{90}$Nb, because these gamma lines could not be resolved by the HPGe-detector. We deduced the activities of 140.5 keV and 141.2 keV gamma lines from the independent gamma lines of $^{99}$Mo and $^{90}$Nb respectively and compared with the results of radioactive decay curve and obtained excellent agreement.

The cross-sections and integral yields obtained in the present work would be useful to upgrade theoretical codes, for estimation of the activity for future accelerator developments and other radiation safety problems, for thin layer activation technique and for checking the yield on enriched target for medical isotope production. The results of this work have been submitted to publish in the J. Nucl. Scien. and Tech.$^5$ and in the J. Appl. Radiat. and Isotopes$^6$.

References

Fig. 1. Separation of complex decay of a mixture of activities for 140 keV gamma line.