Investigation of the proton-induced activation reactions on natural molybdenum.

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Excitation functions of the proton-induced activation reactions on a natural molybdenum target were measured using the stacked foil activation technique in the energy range 22-67 MeV at the Tohoku Unuversity cyclotron laboratory. In addition the thick target integral yield was desired using the measured cross-section data.

1. Introduction

Molybdenum is important as an accelerator structural material and the measurement of cross-section data on this element is of interest to apply to the thin layer activation (TLA) technique to determine the rate of wear, corrosion and erosion processes of Mo. The ^{99m}Tc ($T_{1/2}$= 6.02 h) isotope is the single most important radioisotope for diagnostic nuclear medicine. The ^{99m}Tc labeled radio pharmaceuticals account for over 80 % of all diagnostic nuclear medicines used over the world and is provided through a ^{99}Mo→^{99m}Tc generator system.

In every day practice ^{99}Mo is produced by use of research reactors. There are two ways of ^{99}Mo production, i.e., the (n,γ) and (n, fission) nuclear reactions. Production of ^{99m}Tc and ^{99}Mo by charged particle bombardment is also possible. The most suitable reactions for the production of ^{99}Mo is ^{100}Mo(p,pn)^{99}Mo and ^{100}Mo(p,2p)^{99}Nb(15 s) → ^{99}Mo. For production of ^{99m}Tc the ^{100}Mo(p,2n)^{99m}Tc reaction is preferable.

Several authors have reported a variety of data of proton induced reactions on the molybdenum but most of these studies are limited to a maximum proton energy of 40 MeV. A few of them did measurement at higher energy, but large discrepancies are found among them [1,2]. Therefore, the data of isotope production from Mo target in medium proton energy are not sufficient yet. The present work was performed to give reliable excitation functions and thick target integral yields of Mo+p reactions in the energy range 22-67 MeV using the 90 MeV AVF Cyclotron at Cyclotron and Radioisotope Center (CYRIC) of Tohoku University.

The cross-sections were also calculated theoretically by using the Monte Carlo code, PHITS (Particle and Heavy Ion Transport code System) [3], which is a new version of NMTC/JAM code based on INC and GEM models, to compare with experimental values.

2. Experimental technique

The independent and "cumulative cross-sections" of the proton-induced reactions on molybdenum were measured as a function of proton energy in the range 22-67 MeV using the conventional stacked foil activation technique. Special care was taken in preparation of uniform targets with known thickness, in determination of the proton energy degradation and of the intensity of the bombarding beam along the target stack and in determination of the activities of the samples. High purity (> 99%) Mo foil (49.58 μm thick) of natural isotopic composition (^{92}Mo 14.8%; ^{94}Mo 9.3%; ^{95}Mo 15.9%; ^{96}Mo 16.7%; ^{97}Mo 9.6%; ^{98}Mo 24.1%; and ^{100}Mo 9.6%) was used as a target material for the irradiation. The stack was assembled from Mo, Cu and Al like as Cu–Al–Mo and 10 groups were set together in an aluminum holder for irradiation. The size of the target was about 1 cm ×1 cm which is sufficiently larger than the proton beam collimator diameter, 8 mm. The stacked samples were irradiated by a 70 MeV collimated proton beam (8 mm in diameter) of ~25 nA for 1 hr and 17 minutes using the AVF Cyclotron at CYRIC, Tohoku University. A proton beam accelerated by the AVF Cyclotron was transported to the target room. It was necessary to ensure that equal areas of the monitor and the target foils intercepted the beam. The irradiation geometry used guaranteed that practically the whole entering beam passed through every foil. Reactions induced on aluminum (102 μm thick) and copper (104.2 μm thick) foils were used to monitor the parameters of the bombarding beam. The complete excitation functions of the monitor reactions were measured simultaneously with the reactions induced on molybdenum to confirm the beam intensity and the energy, and also to check the relative behavior of the recommended data.

3. Data analysis

The activities of the radioactive products in the target and monitors were measured nondestructively using HPGe-detector gamma ray spectroscopy at 5 cm and 19 cm from the detector surface. The efficiency versus

energy curve of the detector was determined experimentally using the standard gamma-ray point sources with known strength, ^{152}Eu, ^{133}Ba, ^{241}Am, ^{60}Co and ^{137}Cs at both 5 cm and 19 cm from the endcap of the detector. The proton beam intensity was determined via the monitor reactions [4], ^{27}Al(p,x)^{22,24}Na and ^{nat}Cu(p,x)^{56}Co,^{62,65}Zn taken place at the top radioactive monitor foils of the stack considering that the monitor foils were irradiated simultaneously and measured at the same detector and in a comparable geometry as the Mo targets. The proton energy degradation along the stack was determined using the computer program SRIM-2002 [5].

The cross-sections were deduced for the production of ^{93m,99}Mo, ^{94,95,95m,96}Tc, ^{90,92m,95,96}Nb, ^{86,88,89}Zr and ^{86,87,88}Y in the proton energy range 22-67 MeV by using the well-known activation formula. The decay data for monitors and molybdenum were taken from ENSDF (1996) . The cross–section data of the monitors were taken from EXFOR [2003]. The thick target integral yields were determined by using the measured cross-sections and stopping power via integration from the respective threshold up to 67 MeV. The following errors were considered to derive total uncertainty on cross-section values: statistical error (0.3-5%), error in proton flux (5.8%) and the error in the energy dependence of efficiency calibration (~3%). The overall uncertainty of the cross-sections is around 10 %.

The activities of the140 keV, 776 keV and 765 keV gamma rays emitted from different radinuclides were also corrected by using the independent gamma rays of the corresponding radionuclides and the establishing decay curve.

The activity of 511 keV and 909.10 keV gamma lines were corrected for subtracting the background counts from total measuring counts. The data were corrected for the coincidence-summing effect caused by the coincidence detection of two or more gamma-rays.

We have observed good agreement between the measured results at short and long distances from the detector surface after the coincidence-summing correction. We have also obtained excellent agreement in the numerical values of proton fluxes determined individually using the monitor reactions after coincidence-summing correction

The proton flux determined was considered constant through the foil stacks to avoid the effects of the energy broadening and mean energy on the cross-section measurements. There is a very small loss in the number of protons and not possible to measure it practically.

4. Results and discussion

The measured excitation functions with other experiments and theoretical calculations are shown in Figs. 1-11. The thick target integral yields determined in the present work are given in Figs. 12-15. The obtained results are discussed in the following subsequent sections.

4.1. *Excitation functions of the proton-induced reactions on Mo*

4.1.1 *^{100}Mo(p,x)^{99}Mo and ^{nat}Mo(p,x)^{93m}Mo*

The measured excitation function of ^{99}Mo production is shown in Fig. 3. ^{99}Mo produced in directly or through the decay of the parent isotope ^{99}Nb (15 s) by proton activation on ^{100}Mo target isotope. To determine the production cross-section of ^{99}Mo, the isotopic aboundance of ^{100}Mo (9.6 %) was normalized to 100% enriched ^{100}Mo isotope. The hump at around 41 MeV in Fig.1 indicates two reaction channels leading to the production of ^{99}Mo are ^{100}Mo(p,pn)^{99}Mo (Q = –8.3 MeV) and ^{100}Mo(p,2p)^{99}Nb (15 s) $\rightarrow$ ^{99}Mo (Q = – 11.14 MeV). Takacs et al. [1] reported cross-section data up to 37 MeV and Levkovskij [6] reported up to 29 MeV for ^{99}Mo production on enriched ^{100}Mo isotope that are shown in Fig.1. Our measured values are consistent with Scholten et al.[7] at the two points, 22 and 35 MeV, but his results at other energies are scattered. Our measured values showed excellent agreement with Takacs et al. and Levkovskij in lower energy region and this fact confirms the reliability of our measured data at the energies above 37 MeV. Lagunas-solar et al.[8] reported numerical cross-section data shown in Fig.1 that are much lower than the recently published data in lower energy region.

The measured ^{93m}Mo production cross-section shown in Fig.2 increases almost linearly. Various reaction channels are opened at different stages depending on threshold energy in the overall investigated energy range contributed to the direct formation of ^{93m}Mo.

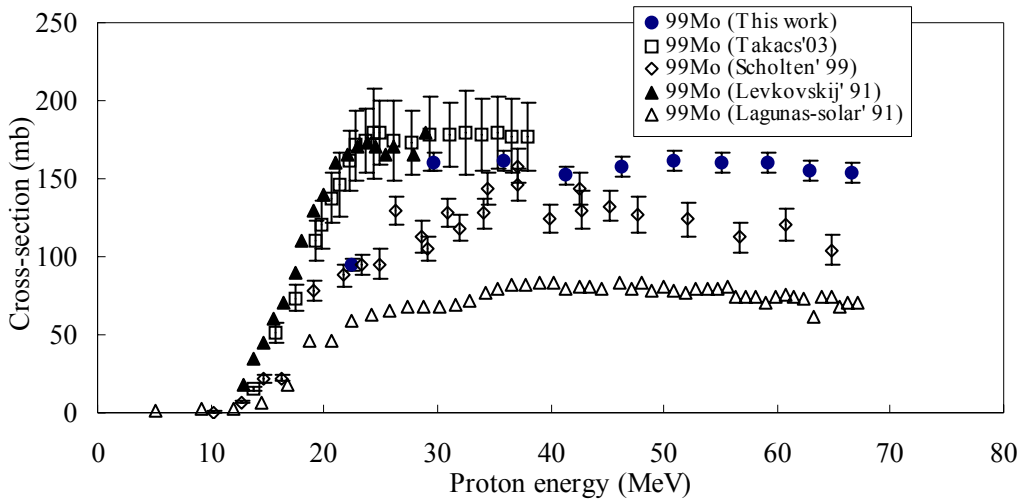

Fig.1 Excitation function of the ^{100}Mo(p,x)^{99}Mo reaction.

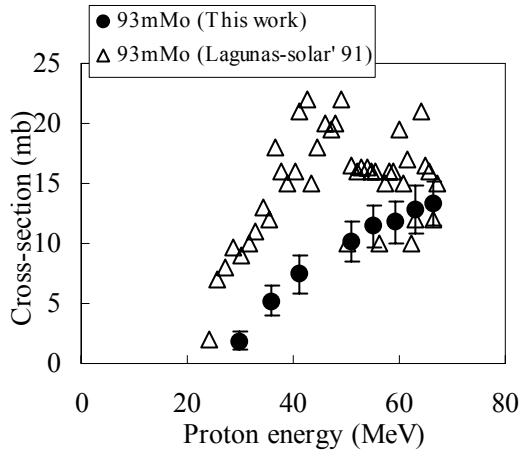

Fig.2 Excitation function of the ^{nat}Mo(p,x)^{93m}Mo reaction

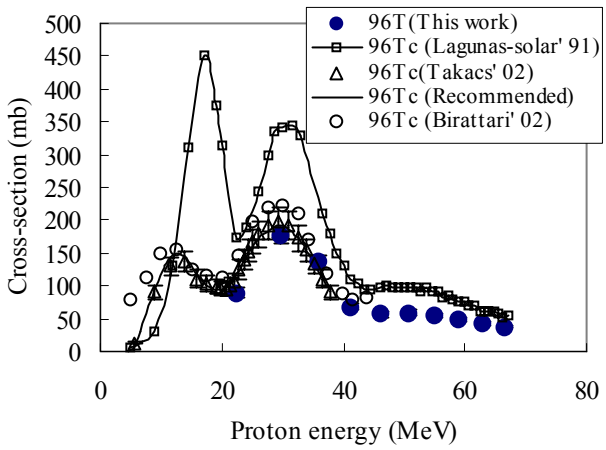

Fig.3 Excitation function of the ^{nat}Mo(p,x)^{96}Tc reaction.

4.1.2 *^{nat}Mo(p,x)^{96,95,95m,94}Tc*

To obtain high reliability, we measured excitation function of ^{96g}Tc production by using the intense independent gamma line, 812.5 keV. The measured excitation function of ^{96}Tc production shown in Fig. 5 is due to a sum of three single processes, ^{96}Mo(p,n)^{96}Tc (Q = – 3.75 MeV), ^{97}Mo(p,2n)^{96}Tc (Q = – 10.57 MeV) and ^{98}Mo(p,3n)^{96}Tc (Q = – 19.21 MeV). It is interesting to note that the recommended values and large number of recently published data for ^{96}Tc production are available only up to 38 MeV. Our measured values show very good agreement with the latest data reported by Takacs et al. and the recommended values shown in Fig.3 and that fact confirms the high reliability of the measured cross-section values of ^{96}Tc production in the whole investigated energy range of the present measurement. Only Lagunas-solar et al. reported ^{96}Tc production cross-sections in the energy range 5-67 MeV and it is so much higher than recommendation in lower energy region that it can not be believed and no explanation for such a high value was found in his work. Therefore our measured values may be considered as a first one from 40 MeV to 67 MeV.

Birattari et al.[9], Kormali et al. and Levkovskij indicate the reliability of the measured values of ^{94}Tc in lower energy region that are shown in Fig.4. Because of the short half-life and the existence of numerous reaction channels are available for ^{94}Tc production from a natural Mo target, this radionuclide was produced abundantly in the 22-67 MeV energy region studied in the present work. The measured excitation function of ^{95}Tc (20 h) radionuclide production via the numerous direct reaction channels and the IT decay of ^{95m}Tc (61 d) is compatible with Levkovskij (1991) and Birattari et al. (2002) both in shape and magnitudes. In the present work, the cross-sections for ^{95m}Tc production are also measured and shown in Fig. 5. In the cases of the investigated Tc-radionuclides productions, PHITS calculation does not provide cross-sections.

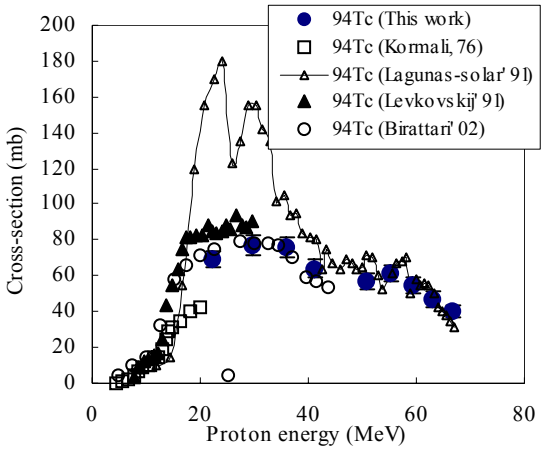

Fig.4 Excitation function of the ^{nat}Mo(p,x)^{94}Tc reaction.

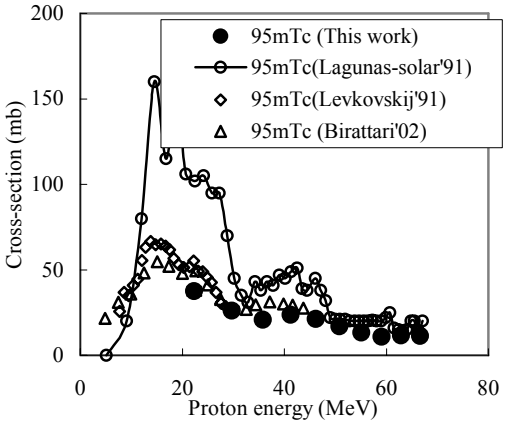

Fig.5 Excitation function of the ^{nat}Mo(p,x)^{95m}Tc reaction.

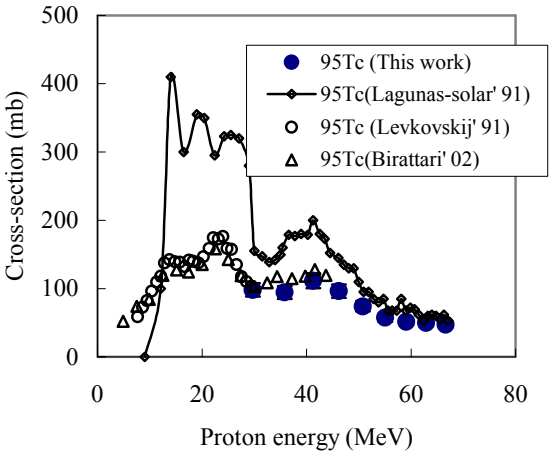

Fig.6 Excitation function of the ^{nat}Mo(p,x)^{95}Tc reaction

4.1.3 *^{nat}Mo(p,x)^{90,92m,95,96}Nb*

The production of Nb radioactivities must be considered as a potential sources of radionuclide contamination in the production of ^{99}Mo and /or ^{99m}Tc. The production cross-sections of Nb radionuclides in the investigated energy range are not reported yet anywhere. In the present work, ^{90}Nb, ^{92m}Nb, ^{95}Nb and ^{96}Nb radionuclides were identified as the reaction products and the obtained results are shown in Figs. 7 -9. The production of Nb radionuclides is most likely due to (p,α)-type reactions on Mo target, the Coulomb barrier in both the entrance and exit channels largely inhibits the probability of these reactions. In the case of ^{90}Nb, its production through the decay of the parent ^{90}Mo also contributed on the direct formation cross-sections from below 30 MeV as shown in Fig.7.

4.1.4 *^{nat}Mo(p,x)^{86,88,89}Zr*

Our measured excitation functions of Zr radionuclides along with the values of model calculation using the PHITS code are shown in Fig. 10. The theoretically calculated values of ^{89}Zr production are completely supporting the experimental values, both in shape and magnitudes. The PHITS calculation for ^{88}Zr radionuclide is also consistent with the measured excitation function. By considering the good agreement between the experiment and theoretical calculations, it could be mentioned that the productions of ^{89}Zr and ^{88}Zr radionuclides are most likely direct formations from Mo targets. The shape of excitation functions for ^{86}Zr is similar with model calculations.

4.1.5 *^{nat}Mo(p,x)^{86,87,88}Y*

The experimentally measured cross-sections for the productions of yttrium radionuclides from Mo target are not available in literature. The measured cross-sections are shown in Fig. 11 with the PHITS calculations. The measured excitation functions for all of the identified Y-radionuclides are larger than that of theoretical calculations using PHITS code. Because, all of the Y-radionuclides are produced through the decay of parent radionuclides, i.e., the experimental values are "commulative" while PHITS calculation shows the cross-section of direct reaction.

4.2. *Thick target integral yields as a function of proton energy*

Mo is an impotant material in nuclear technology. For different possible practical applications we have deduced the thick target integral yields by using the recently measured excitation functions and the available literature data at low energies, which was not covered by us. In most of the cases, the thick target integral yields are linearly rising with the increase of bombarding proton energy that are shown in Fig.12-15. It has been found from Fig. 15 that the contribution of the investigated energy range to the total integral yields (from threshold) is not very significance for ^{88}Y radionuclide.

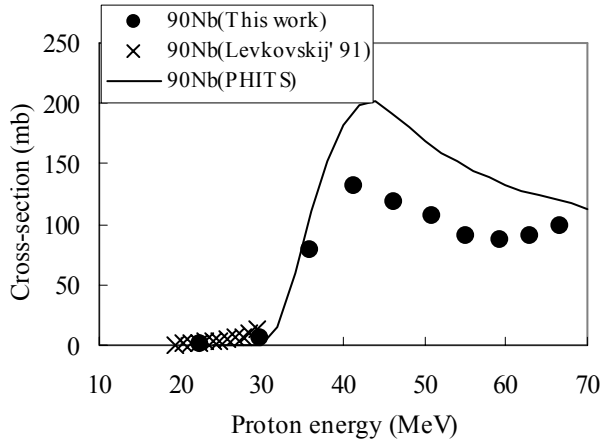

Fig.7 Excitation function of the $^{nat}Mo(p,x)^{90}Nb$ reaction.

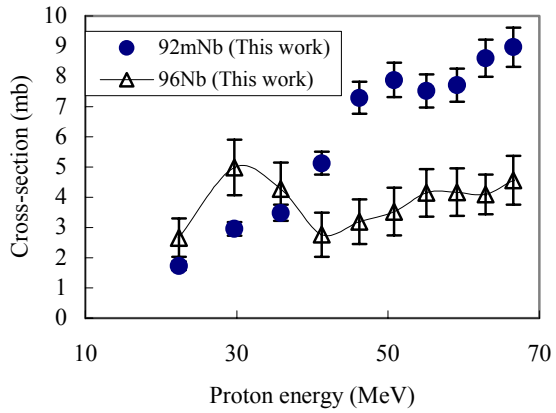

Fig.8 Excitation function of the $^{nat}Mo(p,x)^{92m,96}Nb$ reaction.

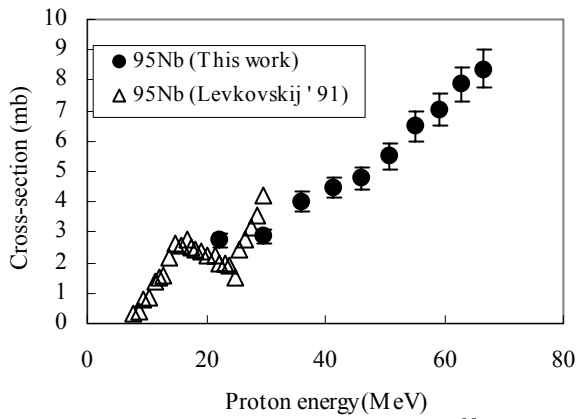

Fig.9 Excitation function of the $^{nat}Mo(p,x)^{95}Nb$ reaction.

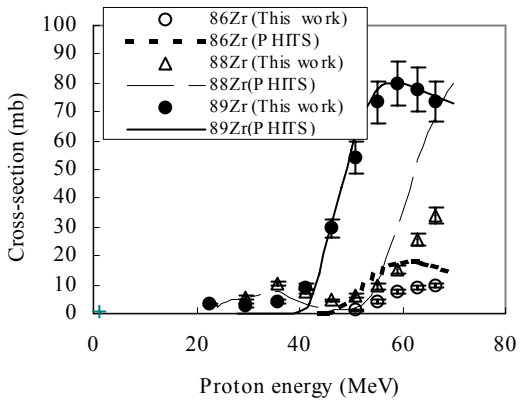

Fig.10 Excitation function of the $^{nat}Mo(p,x)^{86,88,89}Zr$ reaction.

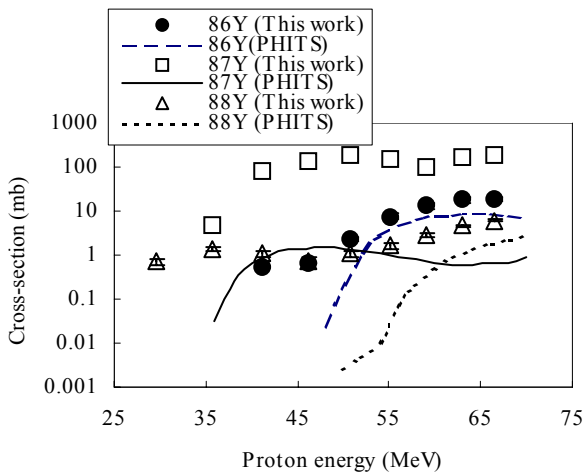

Fig.11 Excitation function of the $^{nat}Mo(p,x)^{86,87,88}Y$ reaction.

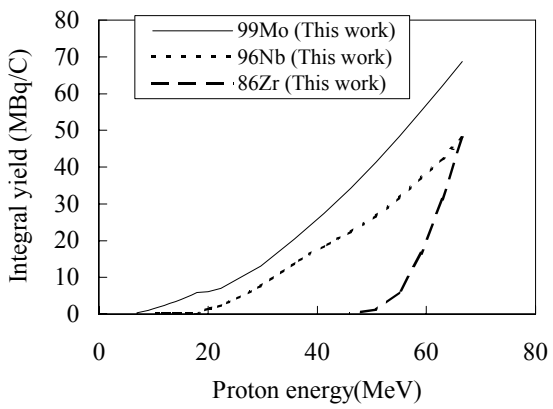

Fig. 12 Thick target integral yields for the production of ^{99}Mo, ^{96}Nb and ^{86}Zr radionuclides.

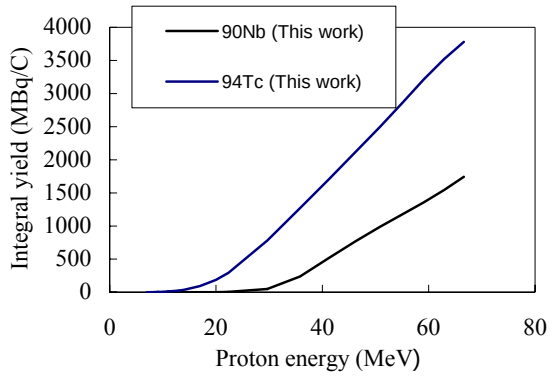

Fig. 13 Thick target integral yields for the production of ^{90}Nb, and ^{94}Tc radionuclides.

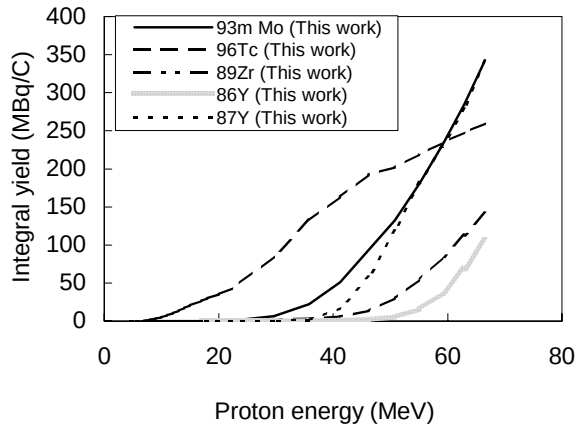

Fig. 15 Thick target integral yields for the production of ^{93m}Mo,^{96}Tc, ^{89}Zr, ^{86}Y and ^{87}Y radionuclides.

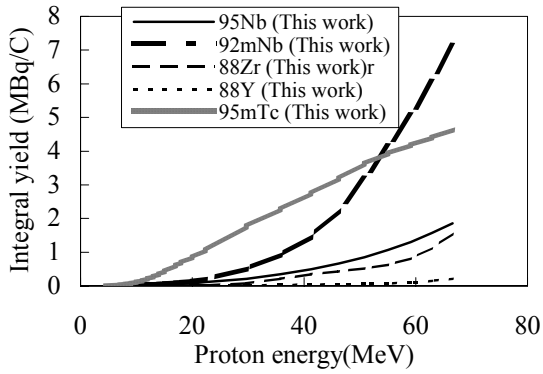

Fig. 14 Thick target integral yields for the production of ^{95}Nb, ^{92m}Nb, ^{88}Zr, ^{88}Y and ^{95m}Tc radionuclides.

5. Conclusion

We have measured the excitation functions for the production of the radionuclides with >4 h half lives through the proton-induced activation reactions on molybdenum in the energy range 22-67 MeV using the stacked foil technique. In the cases of Mo- and Tc-radionuclides, in view of the excellent consistency with the available literature and recommended data of lower energy region it may be considered our measured excitation functions as only one reliable above 38 MeV (upto 67 MeV). No cross-section data for Nb, Zr and Y radionuclides productions exist in our investigated energy range. The present experiment has given new data for all of the investigated proton-induced reactions on Mo in the energy range 22-67 MeV. In cases of the investigated Mo- and Tc-radionuclides and some of Nb-radionuclides, results were not obtained by PHITS due to its limitation.

From the new cross-sections, first integral yields are reported at higher energies (up to 67 MeV). We have to mention that in the investigated energy range no directly measured yield values exist .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.

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