



ISSN: 2319-5967

ISO 9001:2008 Certified

International Journal of Engineering Science and Innovative Technology (IJESIT)

Volume 4, Issue 3, May 2015

The powdered molybdenum target preparation technology for ^{99m}Tc production on C18 cyclotron

A. Avetisyan, R. Dallakyan, R. Sargsyan, A. Melkonyan, M. Mkrtchyan, G. Harutyunyan, N. Dobrovolsky

A. I. Alikhanyan National Science Laboratory (Yerevan Physics Institute)
Foundation, Yerevan, Armenia

Abstract: An alternative way of accelerator production of ^{99m}Tc on cyclotrons for medical purposes is under active consideration in recent years in the world. At the same time, powdered molybdenum is being used as target material. One of the problems with that is the following requirement: the target must have high mechanical strength and high thermal conductivity. To meet these requirements, there has been developed a target preparation technique using laser treatment.

Keywords: Medical isotope, ^{99}Mo , target laser treatment, ^{99m}Tc production

I. INTRODUCTION

^{99m}Tc is the most widely used isotope in nuclear medicine today with over 30 million diagnostic medical imaging scans every year around the world [1]. According to the Center for Radiology and Burns, the ^{99m}Tc isotope's demand of Armenia is about 5000 doses per year.

The technique of direct production (bypassing the intermediate phase of the parent ^{99}Mo isotope) of ^{99m}Tc on cyclotron proton beams is actively developed in many scientific centers in recent years [2-7].

The C18 cyclotron (produced by IBA, Belgium, Fig. 1), which can produce ^{99m}Tc on its proton beam, is in the stage of installing in the Isotope Production Center in Armenia. An appropriate technology has been developed at the National Science Laboratory after A. Alikhanyan (Yerevan Physics Institute).

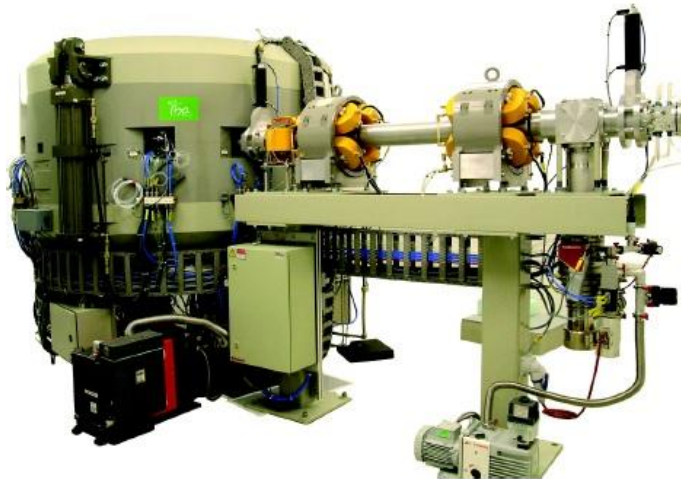


Fig. 1. General view of the C18 cyclotron with extracted beam pipe

For irradiation of solid targets in the standard specification of the C18 cyclotron a target module is used (Fig. 2), in which the target itself is fixed by pneumatic clamps. During the irradiation, the target is cooled by a helium flow on the front side and by a water stream under a pressure of about 8 bars on the rear side.

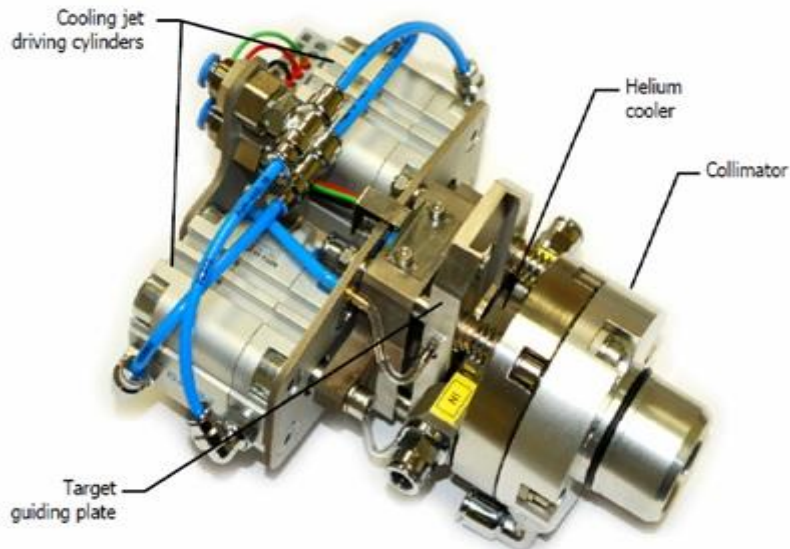


Fig. 2. The general view of the Nitra Solid Compact TS06 target module

The target itself is a metallic disk with a central deepening in which the material for irradiation is located. In Fig. 3, that area is marked as “target area”.

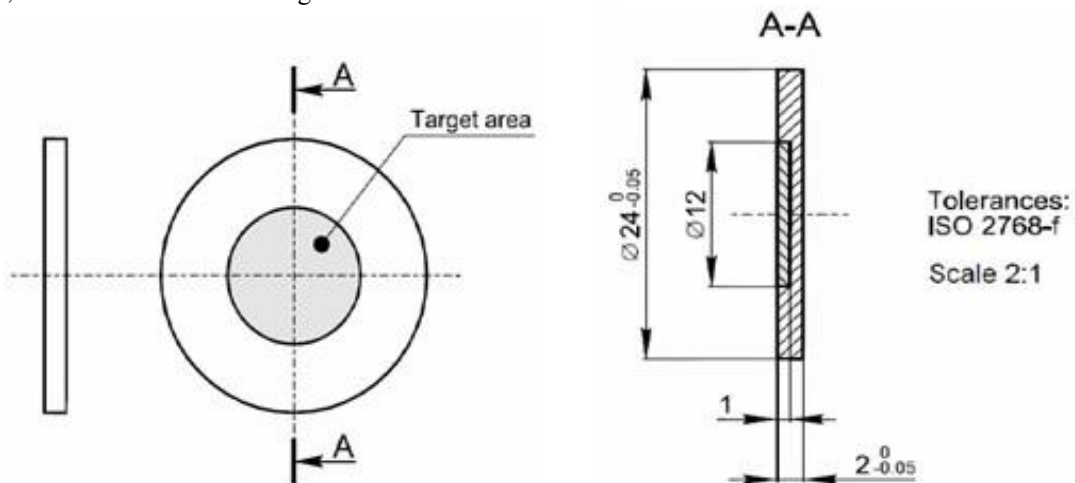


Fig. 3. Solid target disk

II. REGULAR TARGETS PREPARATION METHOD

The target consists of a metallic disk (see Fig. 3) [3] (page 132-137) and the material for irradiation placed in the target area.

The requirements to the target disk are as follows: high mechanical strength and high thermal conductivity for effective removal of the heat produced during irradiation and chemical inactivity.

In terms of durability and chemical inactivity, niobium and titanium are a good fit. The thermal conductivity of niobium is twice as better as that of titanium.

^{100}Mo or natural molybdenum $^{\text{nat}}\text{Mo}$ fine powder is used as material for irradiation in order to produce $^{99\text{m}}\text{Tc}$. It is required to make a solid disk from the powder to place it in the target area. There has been usually used the method of powder metallurgy, where the fine powder is first compressed then roasted at high temperatures. The edges of powder grains melt and join each other, creating a quasi-solid structure. The different-scale images of the compressed powder taken by an electron microscope are shown in Fig. 4 ([8])

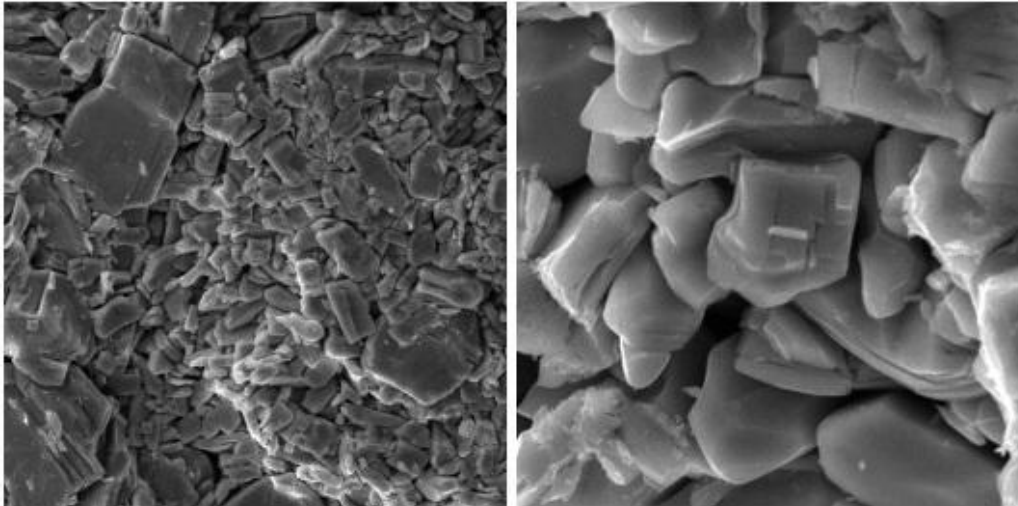


Fig. 4. Molybdenum powder grains images of different scale after thermal processing taken by an electron microscope.

We have developed a natural molybdenum powder compression method using a press (Fig. 5) that can exert a force of ~40000 N.



Fig. 5. A press with a matrix and a punch for pressing of molybdenum into a disk.

The view of the tablet after press treatment is shown in Fig. 6.

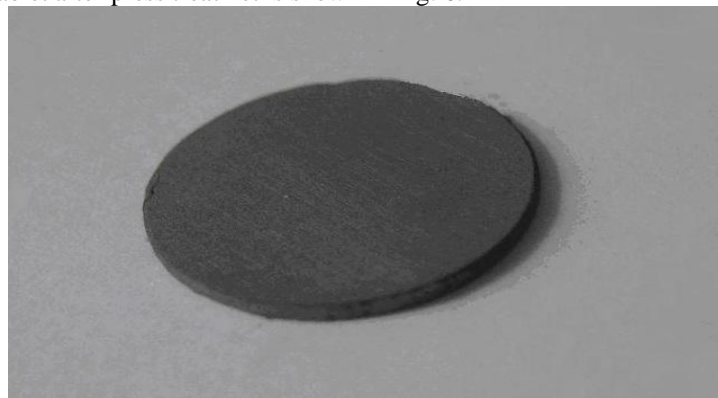


Fig. 6. A tablet of natural molybdenum obtained by the compression method.

However, without additional processing the tablet does not have enough durability and can crash while set up in the target area or its evacuation after the irradiation?

To increase the mechanical durability, we have developed a tablet surface burning method using a focused laser beam. A solid-state laser with the following specifications has been used:

Wave length	1.6 μm
Impulse energy	250 MJ
Impulse repetition frequency	40 Hz
Impulse duration	200 μs

A special device for burning has been developed. The laser beam was held through a beam expander and then was focused by a lens having a focal distance of $F = 150 \text{ mm}$. The light spot's diameter in the focus varied within the range of 150-300 μm .

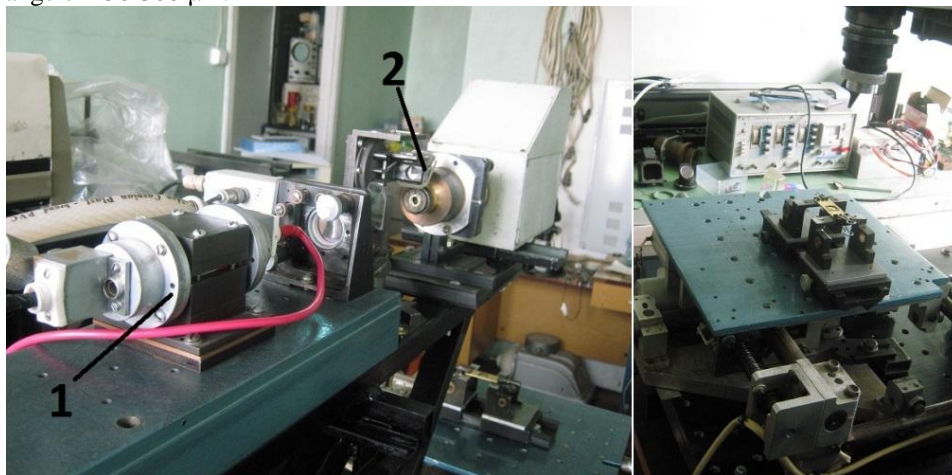
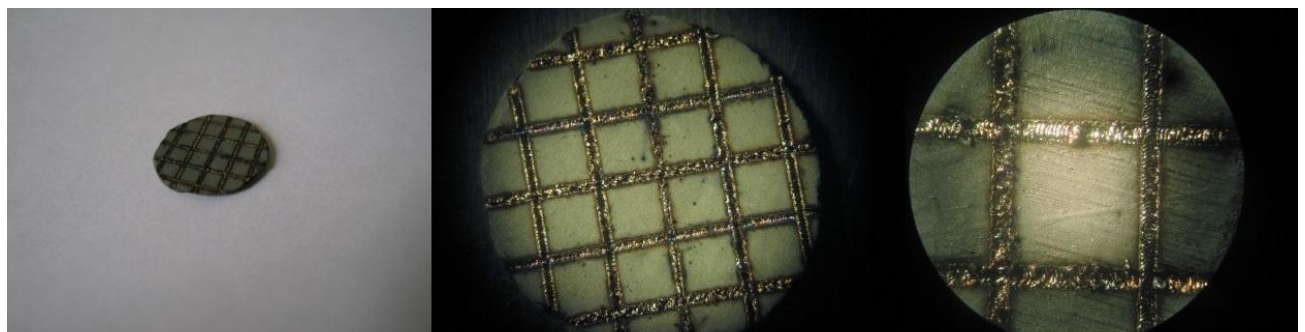


Fig. 7. The automatic tablet processing device for molybdenum target laser processing: the solid-state laser (left) and the 2-dimensional coordinate table with remote computer control (right). 1- solid state laser, 2- beam expander.

The molybdenum powder is melted in trace of laser, creating a solid strip of molybdenum with a width of a few hundred micrometers. The images of a tablet after orthogonal processing with a laser beam are shown in Fig. 8.



(a)

(b)

(c)

Fig. 8. Molybdenum tablet processed by a laser beam at different magnifications: a) general view, b) x20 times zoomed under optical microscope c) x42 times zoomed.

It is well known that Mo oxidizes in the air at $T > 600^\circ\text{C}$. During laser processing the temperature of molybdenum tablet surface is more than 2700°C for a very short time. That creates an anxiety that a part of metallic Mo could be transformed to MoO_3 . To check the possibility, the laser processing was performed in a special airtight box filled with inert gas neon. Three identical tablets were prepared to check the possibility of

oxidizing. One was simply pressed without any laser processing. The second tablet was processed by a laser in air atmosphere, and the third one underwent the same laser processing in a neon atmosphere. As a calibration point, the pure MoO₃ powder has been tested. All four samples were investigated under X-ray phase analysis. The results are presented in Fig. 9.

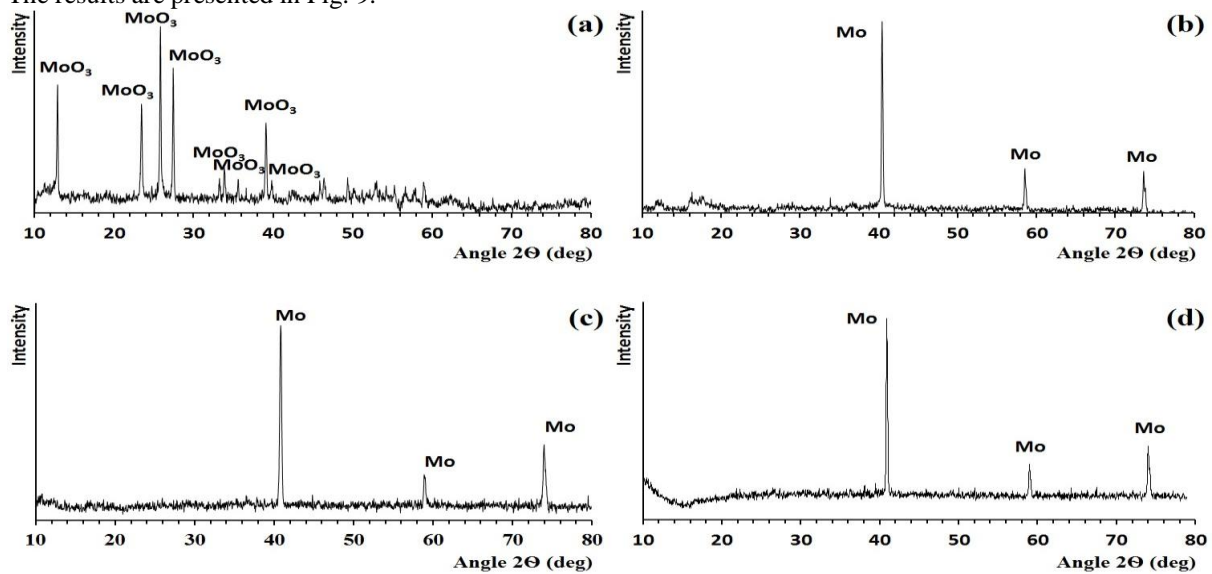


Fig. 9. X-ray phase analysis spectra a) for MoO₃ powder, b) metallic Mo pressed to tablet without any additional processing, c) metallic Mo pressed to tablet and processed under laser beam in air atmosphere, d) metallic Mo pressed to tablet and processed under laser beam in neon atmosphere.

These results show that during laser processing Mo did not transform to MoO₃ neither in the air nor in the neon atmosphere. That could be due to very short time of high temperature presence, which is not enough for the slow chemical process of oxidizing. After such processing, the mechanical strength of tablets should increase. The reason for such an increase is that the melted stripes of metallic molybdenum on the surface of the tablet play the role of steel fittings. For objective estimation of such assumption, a device for the measurement of the relative strength of tables is made. The strength of tablets was measured with and without processing.

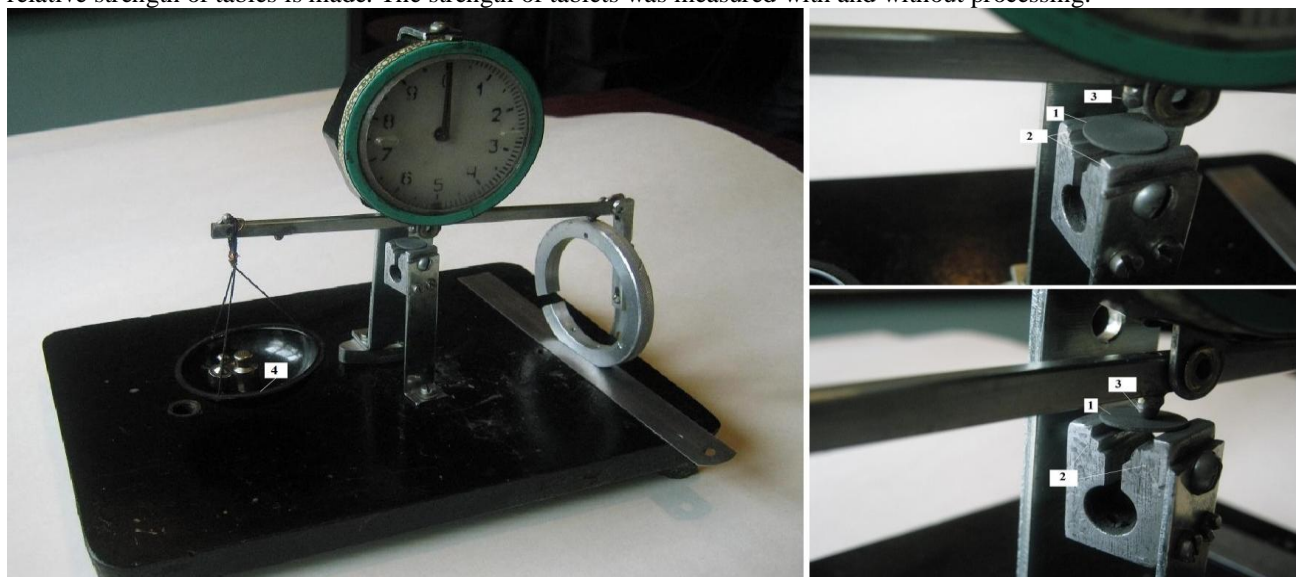


Fig. 10. Tablets mechanical strength measurement device. The molybdenum tablet (1) is set on support (2), the steel ball (3) presses against the tablet with a force determined by the mass of the weights in the scale dish (4). Upper right picture - before strength measurement, bottom right picture - the ball presses against the tablet surface.

The tablets relative strength measuring device is shown in Fig. 10. The pressure of steel ball (3) on the tablet (1) is determined by the mass of weights in the scale dish (4). At a certain point, the tablet breaks under the pressure. Ten tablets with approximately the same characteristics were made. The durability of five tablets having not undergone and five tablets having undergone laser processing has been measured. The results of measurements are shown in Table I and Table II.

Table I. Relative strength of tablets without laser processing.

№	Diameter (mm)	Thickness (mm)	Weight (mg)	Mechanical strength (a.u.)
1	9	0.8	580	445
2	9	0.76	560	476
3	9	0.7	570	560
4	9	0.72	540	426
5	9	0.74	535	436

Table II. Relative mechanical strength of tablets after laser processing.

№	Diameter (mm)	Thickness (mm)	Weight (mg)	Mechanical strength (a.u.)
1	9	0.75	560	884
2	9	0.78	570	844
3	9	0.79	570	724
4	9	0.80	580	604
5	9	0.76	535	644

The mechanical strength increased more than 1.5 times after laser treatment as is seen from the tables above. The developed technique of laser treatment of the surface of compressed molybdenum powder tablets can be used to make real solid targets for irradiation by proton beam of the cyclotron.

The tablet (disc) target preparation technique by laser treatment

Using the above-mentioned technique the disk targets were made in accordance with the drawings in Fig. 3. The fine powder of natural molybdenum was compressed in the central part of the disc (Fig. 11).



Fig.11. The disc target with the compressed natural molybdenum powder (the central circle).

Further, to increase the surface strength of the molybdenum pressed in the center of the target disk, its surface was treated similarly as in case of individual tablets. The processing parameters remained the same. The result is shown in Fig. 12.



Fig.12. The target disk with a pressed molybdenum therein after the laser treatment (left), with x40 magnification (right).

However, the molybdenum powder pressed into the center of the target disk has insufficient adhesion to the disk and may fall out during transportation after irradiation. Therefore, additional treatment with a laser beam was performed at four diametrically opposite points, providing sufficient adhesion, as is shown in Fig. 13.

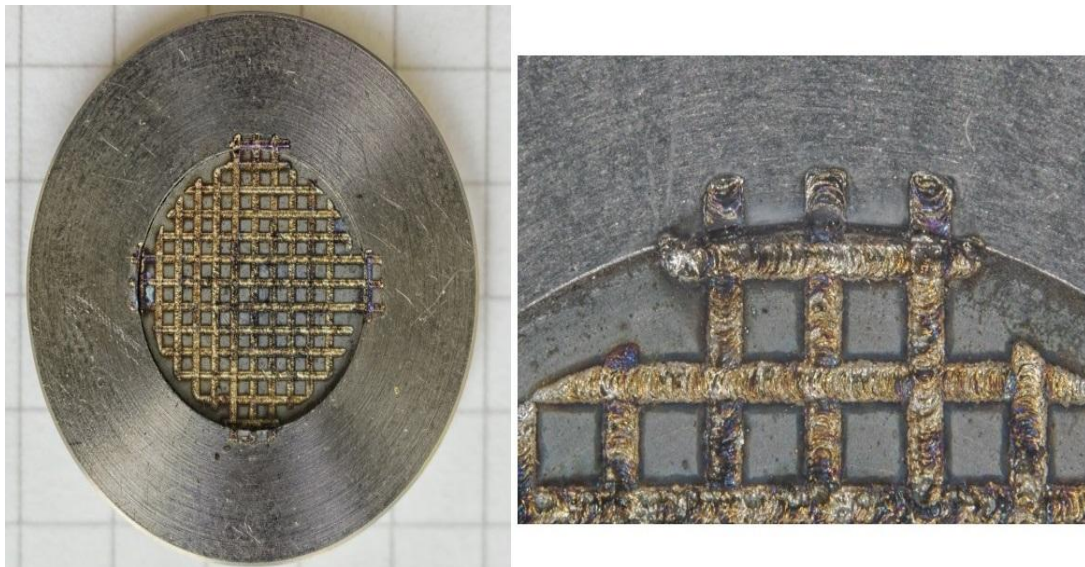


Fig. 13. The target disc with molybdenum after the additional strengthened adhesion at four points (left side), at x30 magnification (right).

III. CONCLUSION

The developed technology allows significantly increasing the mechanical strength of compressed fine molybdenum powder targets by way of laser treatment of their surfaces. At the same time, the melted parts form solid-state molybdenum metal strips. It is experimentally shown that the strength after such treatment grows more than 1.5 times. This method may be also useful for other powder-state targets. The targets prepared by this technique are ready for irradiation by the proton beam of C18 cyclotron.



ISSN: 2319-5967

ISO 9001:2008 Certified

International Journal of Engineering Science and Innovative Technology (IJESIT)

Volume 4, Issue 3, May 2015

ACKNOWLEDGMENT

This work was performed under the financial support of Armenian State scientific budget and IAEA CRP Contract 18029. The authors gratefully thank Dr. Suren Kharatyan (Institute of Physical Chemistry of NAS of Armenia) for his kind help by X-ray analysis.

REFERENCES

- [1] R. Avagyan, A. Avetisyan, I. Kerobyan, R. Dallakyan. Photo-production of $^{99}\text{Mo}/^{99\text{m}}\text{Tc}$ with electron linear accelerator beam. *Nuclear Medicine and Biology*, 41 (2014) 705–709.
- [2] Brigitte Gue´rin, Se´bastien Tremblay, Serge Rodrigue et al. Cyclotron Production of $^{99\text{m}}\text{Tc}$: An Approach to the Medical Isotope Crisis. *THE JOURNAL OF NUCLEAR MEDICINE* Vol. 51, No. 4, April 2010.
- [3] Katherine Gagnon, Fran¸ois B´enard, Michael Kovacs, Thomas J. Ruth, Paul Schaffer, John S. Wilson. Cyclotron production of $^{99\text{m}}\text{Tc}$: Experimental measurement of the $^{100}\text{Mo}(p,x)^{99}\text{Mo}$, $^{99\text{m}}\text{Tc}$ and $^{99\text{g}}\text{Tc}$ excitation functions from 8 to 18 MeV. *Nuclear Medicine and Biology* 38 (2011) 907–916.
- [4] F. T´arkányi, F. Ditr´oi, A. Hermanne, S. Takács, A.V. Ignatyuk. Investigation of activation cross-sections of proton induced nuclear reactions on natMo up to 40 MeV: New data and evaluation. *Nuclear Instruments and Methods in Physics Research B* 280 (2012) 45–73.
- [5] Hossain Targholizadeh, Gholamreza Raisali, Amir R. Jalilian, Nima Rostampour, Mohammadreza Ensaf, Mohsen K. Dehghan. Cyclotron production of technetium radionuclide using a natural metallic molybdenum thick target and consequent preparation of [Tc]-BRIDA as a radio-labeled kit sample. *Nukleonika* 2010; 55(1):113–118.
- [6] Lagunas-Solar MC. Accelerator production of $^{99\text{m}}\text{Tc}$ with proton beams and enriched ^{100}Mo targets. IAEA TECDOC series No.1065. 1999; Feb 25:87-112.
- [7] B. Scholten, R. M. Lambrecht, M. Cogneau, H. Vera Ruiz, S. M. Qaim. Excitation functions for the cyclotron production of $^{99\text{m}}\text{Tc}$ and ^{99}Mo . *Appl Radiat Isotopes*. 1999; 51:69-80.
- [8] REPORT on the 2nd Research Coordination Meeting on “Accelerator-based Alternatives to Non-HEU Production of $^{99}\text{Mo}/^{99\text{m}}\text{Tc}$ ” 7-11 October 2013 Legnaro, Italy. http://www.naweb.iaea.org/naweb/iachem/working_materials/TR-RCM2-Final-Report-Draft-oct-22nd.pdf.