Nuclear Science and Techniques **20** (2009) 22–26

Targetry of MoO_3 on a copper substrate for the no-carrier-added ^{94m}Tc production *via* $^{94}Mo(p,n)^{94m}Tc$ reaction

SADEGHI Mahdi[1,*] KAKAVAND Tayeb[2] AREF Morteza[2] SARABADANI Parvin[1]

[1] *Agricultural, Medical & Industrial Research School, P.O. Box: 31485-498, Gohardasht, Karaj, Iran*

[2] *Physics Department, Zanjan University, P.O. Box: 451-313, Zanjan, Iran*

Abstract ^{94m}Tc was produced *via* $^{nat}Mo(p, xn)^{94m}Tc$ reaction. Deposition of MoO_3 on Cu substrate was carried out *via* two special sedimentation methods for the production of ^{94m}Tc. The 533 mg of MoO_3, 600 μL of collodium (nitrocellulose) and 3 mL of acetone were used to prepare a MoO_3 layer of 11.69 cm^2 and 45.81 $mg \cdot cm^{-2}$. Also, a MoO_3 layer was prepared by 533 mg of MoO_3, 71.188 mg of methylcellulose and 4 mL of water. The targets were checked by SEM and thermal shock test.

Key words ^{94m}Tc, Targetry, Sedimentation, Radionuclide production, Molybdenum oxide

1 Introduction

^{99m}Tc is the most important radionuclide for diagnose in nuclear medicine and widely used in SPECT. To quantify [^{99m}Tc]-radiopharmaceutical biodistribution in human, it would be meaningful to use PET and a positron-emitting Tc isotope. The chemistry and metabolic methods of ^{99m}Tc may be used for the positron emitter ^{94m}Tc in tomography imaging[1,2].

Four of the neutron-deficient isotopes of Tc, i.e. ^{92}Tc ($T_{1/2}$=4.4 min), ^{93g}Tc ($T_{1/2}$=2.75 h), ^{94g}Tc ($T_{1/2}$=4.9 h), and ^{94m}Tc ($T_{1/2}$=52.5 min), decay by β^+ emission. The ^{94m}Tc, having relatively high positron branching ratio (72%), positron end-point energy (2.47 MeV), and suitable half-life, makes it desirable for this purpose[2,3]. And the suitability of ^{94m}Tc for PET has been shown by phantom measurements, animal experiments, and patient studies[4,5].

The highest yield of ^{94m}Tc is given by the $^{94}Mo(p,n)^{94m}Tc$ reaction, with E_p=7–13 MeV protons and an acceptable level of impurity. This is the suitable method for large-scale ^{94m}Tc production using small cyclotrons[2,6-8]. Different targets were investigated to produce ^{94m}Tc. A vertical beam target for simultaneous irradiation of molten-enriched $^{94}MoO_3$ and sublimation of ^{94m}Tc was reported, but it was not very successful[8]. The ^{94m}Tc can be produced by irradiating metallic ^{92}Mo deposited on Ni foil with α particles of E_α=18–26 MeV at 0.2–1.0 μA[2]. Similarly, thin samples were prepared on Cu foil by adding small amounts of NC (nitrocellulose) suspensions of very fine ^{94}Mo or $^{94}MoO_3$ powder in water-free acetone. The samples were irradiated by protons of up to 18.4 MeV and beam current of 100–200 nA[7].

A conventional target preparation method that a $^{94}MoO_3$ pellet is placed into a depressed Al holder or Pt disc and covered by thin Al or Ta foil has been reported to study the ^{99m}Tc production using proton beams of (14.7 MeV, 4 μA)[9], (5–13MeV, 5–8 μA)[2,8], and (13.8 MeV, 10–15 μA)[10]. The aim of this work was to prepare a target by coating MoO_3 on a pure copper substrate via two sedimentation methods with sufficient stability at high-power beam bombardments.

2 Materials and methods

High purity (>99%) MoO_3 powder (10–20 μm particle size) 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%), from Aldrich Chemical Co., was used as a target material for irradiation.

According to the ALICE-91[11] and TALYS-1.0[12] codes and published data[7,13], the proton entrance energy should be less than 15 MeV to get full benefit

* Corresponding author. *E-mail address:* msadeghi@nrcam.org

Received date: 2008-09-17

of excitation function and to avoid the formation of radionuclide impurities. For a given beam/target angle geometry, the MoO_3 coating thickness was chosen to provide a light-particle exit energy of about 5 MeV. According to SRIM code[14], the thickness is 929.6 μm for 90° geometry. To minimize the MoO_3 thickness, a 6° geometry is preferred, in which a 97.2-μm MoO_3 layer is recommended.

To prepare the target, a Teflon (PTFE) device was made. It consists of two plates of 19cm×10cm×3cm. The upper plate has an elliptical window of 11.69 cm^2, which is the same as the copper substrate placed between the two plates. The upper part is fitted on it with six supporting pins and it is sealed by an O-ring fitted-window. The geometrical shape of the window determines the actual target coating area. For the thickness of 97.2 μm, 533 mg of MoO_3 is required.

A MoO_3 layer was deposited on the Cu substrate by collodium (NC) or methylcellulose (MC) methods.

The NC method (Type 1 samples): A suspension of very fine MoO_3 powder in water-free acetone was obtained by mixing and stirring. Collodium (2% nitrocellulose in amyl acetate solution) was added to the MoO_3 suspension, stirred for 1–2 min and loaded into the cylinder of the upper disk immediately. A Teflon plate with a small hole covered the window. The solution evaporated slowly at room temperature through the hole after about 7 h. This refinement procedure is a result of experiments with different amounts of collodium and acetone.

The MC method (Type 2 samples): Methylcellulose powder dissolved in the water (not hot) was added to the mixture of MoO_3 and water. Stirring it for 3–4 min, a homogenous suspension was obtained. It was loaded into the cylinder of the upper disk at once. The water was evaporated slowly at room temperature in about 24 h.

For the Type 1 samples, the system window must be covered to prevent the solvent (acetone) from quick evaporation and poor adhesion of the target material on the copper substrate; whereas for the Type 2 samples, covering the window was not needed.

An unclean surface of the Cu backing may cause blistering, cracking, gas pits and peeling off of the MoO_3 layer. The substrate was cleaned with sandpaper (grade 1000), and washed by water, and then a mixture of the alkali cleaning powders to remove oil contaminators. Finally, the surface was cleaned with acetone.

Thermal shock test was performed to check adhesion of the samples. They were annealed at different temperatures (Type 1 samples: 100, 150 and 200℃ for 30 min; Type 2 samples: 200, 300 and 350℃ for 1 h) followed by quenching them in 8℃ water immediately. The targets were examined by SEM (JEOL model JSM 6400, operated at 20 kV).

3 Results and discussion

3.1 Adhesion agent amount

Adhesion is an important factor for both the samples. Insufficient amount of NC or MC causes poor adhesiveness of the deposited MoO_3 layer, but their overabundance reduces thermal conductivity of the target. To optimize the quantity for maximum adhesion and thermostability of the MoO_3 layer, the two types of samples were examined with different quantities of NC and MC with respect to the MoO_3 (Tables 1 and 2). The optimum NC and MC amounts are respectively 2.00% and 13.36% of the MoO_3.

Table 1 Influence of amount of cellulose nitrate with 3 mL acetone for 533 mg MoO_3

Collodion* / μL	NC / mg	NC/MoO_3 / wt%	T / mg·cm^{-2}	Adhesion	Comments
60	1.068	0.20	42.86	Unfavorable	Rough, porous
100	1.780	0.33	43.57	Unfavorable	Rough, porous
120	2.136	0.40	43.80	Unfavorable	Rough, porous
150	2.670	0.50	43.62	Tolerable	Rough
300	5.340	1.00	44.28	Tolerable	Rough
600	10.680	2.00	45.81	Excellent	Smooth

* 2% cellulose nitrate solution in amyl acetate; density 0.89 g/cm^3.

Table 2 Influence of amount of methylcellulose for 533 mg MoO_3

Water / mL	MC / mg	MC/MoO_3 / wt%	T / mg·cm^{-2}	Adhesion	Comments
1	17.797	3.34	46.50	Unfavorable	Rough, porous
2	35.594	6.68	46.47	Tolerable	Smooth
3	53.391	10.02	48.94	Tolerable	Smooth
4	71.188	13.36	50.98	Excellent	Reflective, smooth
6	106.800	20.04	52.94	Excellent	Reflective, smooth

3.2 Solvent amount

Solvent volume affects adhesion of the target, too. Fewer amount of solvent results in fast evaporating, hence undesired adhesion. In order to achieve the required tenacity, different amounts of acetone were applied to prepare the suspension (Table 3).

3.3 Target quality control

Homogeneity of the MoO3 layer, which may affect the production rate of ^{94m}Tc, was determined by standard deviation of the layer thickness measured at several spots by micrometer, while the morphology by an SEM (see Figs.1 and 2 for the best coating).

Results of the thermal shock tests are given in Tables 4 and 5. No crack formation or peeling off was observed with the MoO_3 layers at 150℃ for 30 min (NC) and 300℃ for 60 min (MC), indicating a good adhesion for the purpose. The results also indicate that the Type 1 and Type 2 targets can resist 150℃ and 300℃, respectively, without any crack or peeling off.

Table 3 Influence of amount of acetone for 533 mg MoO_3

Collodion / μL	Acetone / mL	NC/Acetone /g·L^{-1}	Adhesion
600	3	3.56	Excellent
	4.5	2.37	Excellent
	6	1.78	Excellent
300	3	1.78	Tolerable
	4.5	1.187	Tolerable
	6	0.89	Tolerable
150	3	0.89	Tolerable
	4.5	0.59	Tolerable
	6	0.45	Tolerable

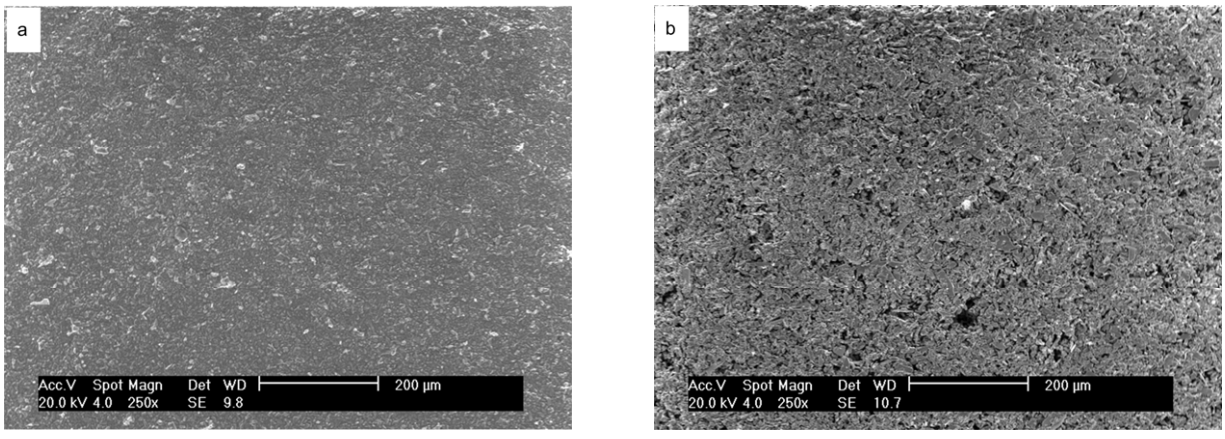

Fig.1 SEM images of molybdenum oxide deposit on the Cu backing 533 mg MoO_3, 600 μL Collodion, and 3 mL acetone suspension. (a) 45.81 mg·cm^{-2} thickness, (b) 42.86 mg·cm^{-2} thickness.

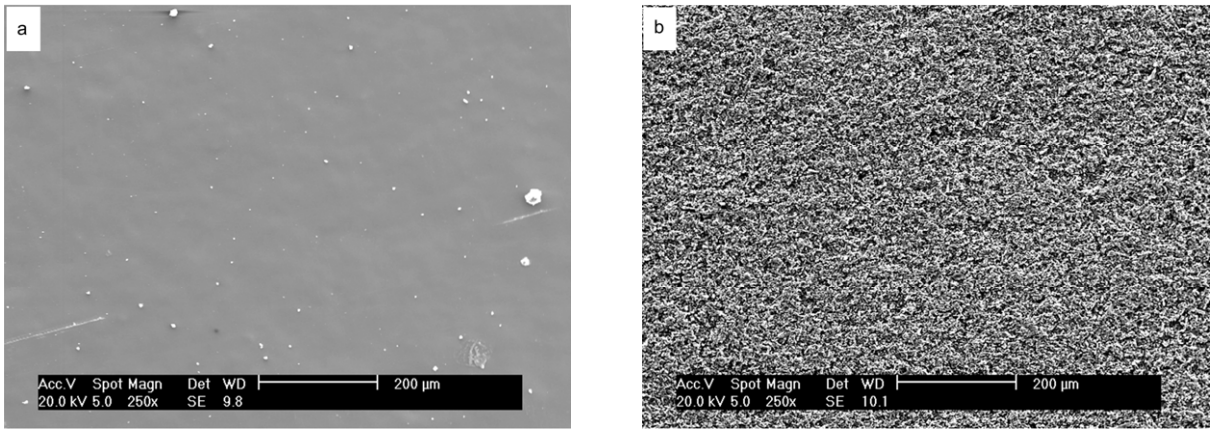

Fig.2 SEM images of a molybdenum oxide deposit on the Cu backing. (a) 533 mg MoO_3, 71.188 mg methylcellulose, and 4 mL water suspension, 50.98 mg·cm^{-2} thickness and (b)533 mg MoO_3, 17.797 mg methylcellulose, and 1 mL water suspension, 46.50 mg·cm^{-2} thickness.

Table 4 Thermal shock test for cellulose nitrate*

W(NC) / mg	*W*(NC)/W(MoO_3) / %	Adhesion**	100℃	150℃	200℃
2.67	0.50	Tolerable	Unstable	Unstable	Unstable, peeling off
5.34	1.00	Tolerable	Stable	Unstable	Unstable, peeling off
10.68	2.00	Excellent	Stable	Stable	Unstable, peeling off

* The heating of the target for 30 min followed by submersion of the hot target in cold water (8℃)

** Adhesion at the normal temperature

Table 5 Thermal shock test for methylcellulose*

W(MC) / mg	*W*(MC)/W(MoO_3) / %	Adhesion**	200℃	300℃	350℃
35.594	6.68	Tolerable	Stable	Unstable	Unstable
53.391	10.02	Tolerable	Stable	Unstable	Unstable
71.188	13.36	Excellent	Stable	Stable	Unstable
106.800	20.04	Excellent	Stable	Stable	Unstable

*The heating of the target for 60 min followed by submersion of the hot target in cold water (8℃)

** Adhesion at the normal temperature

4 Conclusion

In summary, ^{94m}Tc was produced by irradiation of MoO_3 thick deposit target that was prepared by means of the sedimentation method and the optimum parameters were obtained. The target was irradiated at 30 μA current and no degradation was observed. Improvement of cooling system that contains a jet of water-cooling and a circulating flow of chilled helium in front of the target would improve the thermal conductivity.

References

1 Zolle I. Technetium-99m pharmaceuticals: preparation and quality control in nuclear medicine. Berlin, Heidelberg: Springer, 2006.

2 Qaim S M. Nucl Med Biol, 2000, **27:** 323–328.

3 Firestone R B, Shirley V S, Baglin C M, *et al.* Table of isotopes. New York: Wiley, 1996.

4 Nickles R J, Nunn A D, Stone C K, *et al.* J Nucl Med, 1993, **34:** 1058–1066.

5 Stone C K, Christian B T, Nickles R J, *et al.* J Nucl Cardiol, 1994, **1:** 425–433.

6 Strohmaier B, Faßbender M, Qaim S M. Phys Rev C, 1997, **56:** 2654–2665.

7 Rösch F, Qaim S M. Radiochim Acta, 1993, **62:** 115–121.

8 Christian B T, Nickles R J, Stone C K, *et al.* Appl Radiat Isot, 1995, **46:** 69–73.

9 Rogers B E, Parry J J, Andrews R, *et al.* J Nucl Med, 2005, **46:** 1889–1897.

10 Smith M F, Daube-Witherspoon M E, Plascjak P S, *et al.* Med Phys, 2001, **28:** 36–45.

11 Blann M. RSIC Code Package, PSR-146, 1991.

12 Koning A J, Hilaire S, Duijvestijn M C. AIP Conference Proceedings, 2004, **769:** 1154–1159.

13 Skakun E A, Batij V G, Rakivnenko Yu N, *et al.* Yadernaya Fizika, 1987, **46:** 28–32 (in Russion).

14 Ziegler J F, Biersack J P, Littmark U. IBM-Research, New York (USA), 2003.