

$^{99}\text{Mo}/^{99m}\text{Tc}$ Radioisotope Generator Based on Zirconium Selenomolybdate as Column Matrix

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Abstract—Zirconium selenomolybdate gel matrices were prepared by mixing solutions of zirconium oxychloride, selenous acid, and sodium molybdate in different manners. The elution of the generated ^{99m}Tc radioactivity from the obtained zirconium selenomolybdate- ^{99}Mo matrices was studied using small chromatographic columns of the matrix. 6 g of zirconium selenomolybdate- ^{99}Mo matrix dried at 50°C, packed in a glass column (1.1 cm i.d.), was eluted by passing 10 mL of 0.9% NaCl at a flow rate of 1 mL min⁻¹ and ambient temperature (~25°C). The ^{99m}Tc elution yield of 77.1% was obtained. Radionuclidic, radiochemical, and chemical purity of the eluate proved to be suitable for use in medical applications.

Keywords: $^{99}\text{Mo}/^{99m}\text{Tc}$ generators, zirconium selenomolybdate matrix

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In nuclear medicine, ^{99m}Tc is the most frequently used radiotracer because of its suitable physical and nuclear properties. It emits monoenergetic (140 keV) γ -photon suitable for γ -cameras. In addition, its half-life of 6 h is suitable for performing the required tests without exposing the patient to unacceptable radiation dose. Planar cameras are used for bone scanning with ^{99m}Tc -MDP, and SPECT cameras are used for cardiac scanning with ^{99m}Tc -Sestamibi [1–7]. Cyclotron-produced ^{99m}Tc is not practical because of the high cost and short half-life [8–10]. The main source of ^{99m}Tc is the β^- -decay (87.5%) of ^{99}Mo ($T_{1/2} = 65.94$ h). Technetium-99m of high purity, suitable for medical applications, is obtained by elution of radioisotope generators based on alumina columns loaded with fission ^{99}Mo of high specific activity [11]. Other sorbents were also tested for preparing $^{99}\text{Mo}/^{99m}\text{Tc}$ generators, such as silica gel, hydrous zirconium oxide, manganese dioxide, and hydrous titanium oxide [12]. The main disadvantage of these generators is the limited adsorption capacity of the base material for ^{99}Mo , which, in turn, requires the use of ^{99}Mo of very high specific activity (of the order of tens thousands Ci g⁻¹), produced from nuclear reactors via fission route. Generation of high amounts of intermediate and long-lived fission product waste as a result of complex multistep separation and purification of fission ^{99}Mo in expensive hot cells with sophisticated equipment and infrastructure are the

main disadvantages of fission ^{99}Mo radioisotope generators [13]. Alternative technologies are required to overcome these drawbacks. These alternative technologies are based on incorporating neutron activation ^{99}Mo of low and medium specific activity (0.3–3.5 Ci g⁻¹ Mo) into insoluble precipitates of high Mo content such as Ce, Zr, Mg, Ti, Fe, Th, Sn, and Te compounds, which are then used as column matrices [14–28]. Post- and pre-irradiation-formed gels are the two common techniques used for preparing gel generators. Isotope exchange allows overcoming the radiation degradation resulting from irradiation of the prepared gel material and the multistep procedure in pre- and post-irradiation-formed gel techniques, respectively [16, 29]. Solvent extraction of $^{99m}\text{TcO}_4^-$ or sublimation of $^{99m}\text{Tc}_2\text{O}_7$ has also been used in some generator systems for extraction of ^{99m}Tc from ^{99}Mo of relatively low specific activity, produced by direct neutron activation of molybdenum targets [12].

This study was aimed at preparing $^{99}\text{Mo}/^{99m}\text{Tc}$ generator based on zirconium selenomolybdate as a column matrix by post-irradiation-formed gel technique.

EXPERIMENTAL

All chemicals used were of A. R. grade. Distilled water was used for preparing and washing the solutions.

Radiometric identification and measurements were made with a multichannel analyzer (MCA) of Inspec-2000 model, Canberra series (the United States), coupled with a high-purity germanium (HPGe) coaxial detector of GX2518 model. A UV-160 A UV-VIS spectrometer (Shimadzu, Japan) was used for determining Zr and Mo. X-ray diffraction (XRD) analysis was performed with an 18 kV diffractometer (D8 Advance, Bruker, the United States) with monochromated CuK_α radiation ($\lambda = 1.54178 \text{ \AA}$). Elemental analysis was carried out with a BW-1200 X-ray fluorescence spectrometer (Philips, Netherlands). The morphology was examined with a JSM-6510A field emission scanning electron microscope (FESEM) (Japan). The IR spectrum was recorded with an MB157S FT-IR spectrometer (Bomem, Canada) in the range from 4000 to 400 cm^{-1} at room temperature. Thermal analysis, including simultaneous thermal gravimetric (TGA) and differential thermal (DTA) analysis, was carried out with a DTG-60H device (Shimadzu, Japan).

Radioactive ^{99}Mo solution. ^{99}Mo was obtained by eluting a $^{99}\text{Mo}/^{99m}\text{Tc}$ generator with alumina-packed columns (Mon-Tek Generator, Monrol Nuclear Products Industry and Trade, Turkey), loaded with 20 GBq on calibration date. Each column was eluted with a 2 M NH_3 solution. The ^{99}Mo eluate was passed through a $0.45\text{-}\mu\text{m}$ Millipore filter to retain any possible alumina particles. The ammonium molybdate- ^{99}Mo solution was gently heated to dryness and then redissolved in 0.1 M NaOH solution. Alternatively, for preparing $^{99}\text{Mo}/^{99m}\text{Tc}$ generators, solutions of fission ^{99}Mo ($\sim 100 \text{ mCi}$) in 0.1 M NaOH solution were obtained from the ETRR-II Radioisotope Production Facility (RPF). Then, ^{99}Mo solution was diluted with 1.0 M HCl to the desired pH value.

Preparation of zirconium selenomolybdate gel matrix. 0.25 M Mo(VI) solution was prepared by dissolving 3.025 g of $\text{Na}_2\text{MoO}_4 \cdot 2\text{H}_2\text{O}$ in a small portion of distilled water with the addition of 5 mL of a ^{99}Mo solution along with a few drops of 30% H_2O_2 . The resulting sodium molybdate solution was warmed to destroy excess H_2O_2 and then cooled to room temperature before adjusting to pH about 4.5 with 2 M HNO_3 . The volume of the Mo(VI) solution was completed to 50 mL with distilled H_2O . 0.25 M Zr(IV) solution was prepared by dissolving 5.062 g of $\text{ZrOCl}_2 \cdot 8\text{H}_2\text{O}$ in a small portion of distilled water and then was completed to 62.5 mL, after which it was heated to 60°C and then left to cool to room temperature ($\sim 25^\circ\text{C}$).

0.2 M Se(IV) solution was prepared by dissolving 3.224 g of H_2SeO_3 in a small portion of distilled water and then was completed to 50 mL. Mixtures with different Zr(IV) : Se(IV) : Mo(VI) molar ratios were prepared by dropwise addition of the Mo(VI) solution to the stirred Se(IV) solution, followed by dropwise addition of the Zr(IV) solution. Varied volumes of the three solutions were used to obtain different molar ratios. The pH of the solution for preparing the ZSM gel was adjusted to ~ 5.0 with 2 M Na_2CO_3 . The precipitates formed were digested in the mother solution for 24 h at room temperature before phase separation by centrifugation. After separation, the precipitate was washed several times with distilled water until the washing filtrates were free from chloride ions, dried at 50°C for 24 h, and pulverized to the desired particle size. The precipitation yield Y [% ^{99}Mo in the form of Zr(IV) selenomolybdate gel] was radiometrically determined using the formula

$$Y = 100\% \times (A_0 - A_F)/A_0,$$

where A_0 and A_F are the radioactivities of ^{99}Mo in the initial mixed solution and supernatant after separating the precipitated ZSM gel, respectively, measured under identical geometric and counting conditions.

Elution of ^{99m}Tc from ^{99}Mo - ^{99m}Tc chromatographic columns. Unless otherwise stated, small chromatographic columns consisting of glass burettes of different internal diameter and containing different amounts of zirconium selenomolybdate- ^{99}Mo matrix were used. The columns were conditioned for ^{99m}Tc elution by passing 20 mL of 0.9% NaCl solutions through the bed matrix and left for 24 h before elution. The columns were eluted 10 times with different flow rates (0.25, 0.5, 0.75 and 1.0 mL min^{-1}) over the course of 15 days (shelf life). The time between each two successive elution runs was 23 h. The ^{99m}Tc elution yield was determined by comparing the counting rates of the 140 keV peak in the γ -ray spectrum of the column directly before and after elution, taking the contribution of ^{99}Mo to this peak into account [15]. The elution profiles were drawn for various elution flow rates. pH values of the eluate solutions were determined with a pH meter.

The effect of the bed size was studied using chromatographic columns (0.6 cm i.d.) packed with 1.0, 3.0, and 6.0 g of zirconium selenomolybdate- ^{99}Mo matrix. Alternatively, equal amounts of the zirconium selenomolybdate- ^{99}Mo gel (1.0 g each) were packed in

Preparation conditions and the properties of ZSM gel matrix samples (ZSM-1, ZSM-2, ZSM-3).

Sample no.	Preparation conditions							Characterization of the gel matrix								
	initial concentration			mixing ratio (v/v)			pH	weight loss at T , °C			XRF analysis, wt %			radiometric analysis		
	Zr	Se	Mo	Zr	Se	Mo		100	150	200	Zr	Se	Mo	Zr	Se	Mo
ZSM-1	0.2	0.2	0.2	1.55	0.5	1.25	5 ± 0.1	6.3	11.58	12.5	29.82	8.7	24.18	29.51	8.75	24.52
ZSM-2	0.2	0.2	0.2	1.55	1.25	1.25	5 ± 0.1	6.4	8.85	10.97	26.11	14.88	21.047	26.72	15.10	21.12
ZSM-3	0.2	0.2	0.2	1.55	0.25	1.25	5 ± 0.1	7.8	9.5	10.4	25.42	28.18	9.99	24.91	28.32	9.79

glass burettes with different internal diameters of 0.4, 0.6 and 1.1 cm to study the effect of the internal diameter on the elution profile of ^{99m}Tc from the chromatographic column.

For dry elution studies, a stream of air was passed through the molybdate bed to keep it free from the interstitial saline solution for the next elution.

Preparation of ^{99m}Tc generator based on zirconium selenomolybdate- ^{99}Mo gel matrix. A glass column (1.1 cm i.d.) was packed with 6 g of the zirconium selenomolybdate- ^{99}Mo gel. The generator was conditioned for elution of ^{99m}Tc by passing 20 mL of 0.9% NaCl solution through the column bed and then left overnight to be ready for the elution of the generated ^{99m}Tc radionuclide.

The ^{99}Mo content in the eluate (^{99}Mo breakthrough) was determined, in the case of its presence, as the contribution of the count rate of 181 or 739 keV peak in the γ -ray spectrum to the total count rate. The γ -ray spectrum of the eluate was measured directly after elution and after 3 days. The radionuclidic purity of the eluted ^{99m}Tc was determined as the contribution of the count rate of the 140 keV peak in the γ -ray spectrum.

The radiochemical purity defined as the percentage of ^{99m}Tc radioactivity present in the $^{99m}\text{TcO}_4^-$ form was determined by ascending paper chromatography using Whatman no. 1 chromatographic paper and 85% methanol as a developing solvent. The activity distribution along the chromatogram was determined with a γ -ray spectrometer, and R_f was calculated as the ratio of the distance (cm) traveled by ^{99m}Tc species from the starting line to the peak position to the distance (cm) traveled by the solvent from the starting line to the solvent front.

The possible Zr, Mo, and Se impurities in ^{99m}Tc eluate were determined by ICP and/or UV-VIS spectrometry. The zirconium selenomolybdate gel prepared under optimum conditions was characterized by IR spectroscopy, thermal analysis (TGA, DTA), XRD, XRF, and FESEM.

RESULTS AND DISCUSSION

The chemical composition, particularly the Mo content of the zirconium selenomolybdate gel, is a significant factor for its use as a column matrix in preparation of gel-type $^{99}\text{Mo}/^{99m}\text{Tc}$ radioisotope generators. The chemical composition of the obtained zirconium selenomolybdate materials was studied radiometrically by dissolving the gel materials prepared from the radioactive solution of ^{95}Zr , ^{99}Mo , and ^{75}Se in a hot concentrated HCl solution. The table compiles the data on the Mo(VI) precipitation yield, the corresponding equilibrium pH value, and some properties of the formed zirconium selenomolybdate- ^{99}Mo products at the given Zr(IV) : Se(IV) : Mo(VI) molar ratios. As can be seen, the precipitation yield of Mo(VI) from the Mo(VI)–Se(IV)–Zr(IV) mixed solutions decreased with increasing Se concentration. The Mo content decreased with increasing Se(IV) content of the gel matrices, ZSM-1 > ZSM-2 > ZSM-3. This means that the selectivity of the positively charged zirconia surface to molybdate and/or selenite anionic species depends on their concentration in the mixed solution. At low selenite concentrations, the molybdate anions show higher affinity for the positively charged zirconia surface than the selenite anions do. The SeO_3^{2-} anions appreciably contribute to the ZSM structure at low Mo(VI) content to keep the zirconia surface neutral. The relative content of Zr(IV) in the gel matrices decreased as the initial concentration of Se(IV) in the mixed solutions increased: ZSM-1 > ZSM-2 > ZSM-3. In addition, the weight loss of the precipitates dried at 150 and 200°C to constant weight decreased with increasing Se(IV) content of the gel matrix. Therefore, ZSM-1 sample of zirconium selenomolybdate was chosen for characterization (via IR, thermal analysis, XRD, and FESEM) and further preparation and quality control of $^{99}\text{Mo}/^{99m}\text{Tc}$ generator. The suggested formula of the formed gel matrices may be written as $\text{ZrO}(\text{OH})^+ (\text{HSeO}_3)_{1-x} (\text{HMoO}_4)_x \cdot n\text{H}_2\text{O}$ [21].

The FT-IR spectrum of zirconium selenomolybdate

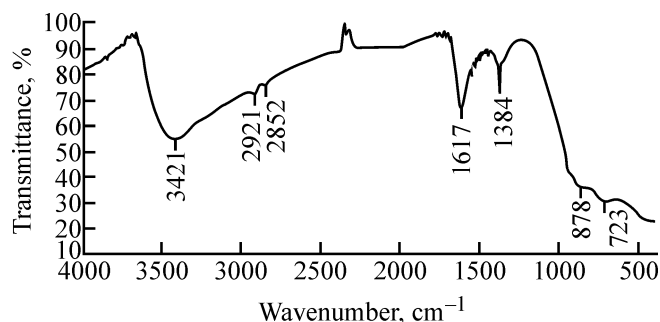


Fig. 1. IR spectrum of ZSM gel matrix.

gel matrix (ZSM-1) shows the presence of extra water molecules in addition to $-\text{OH}$ groups and metal oxides present in the material (Fig. 1). The bands observed at 878 and 722 cm^{-1} may be due to metal–oxygen bonds, which are manifested in the range 650–1000 cm^{-1} . The band at 1380 cm^{-1} belongs to bending vibrations of metal-bonded hydroxyl groups. The strong and sharp peak at 1617 cm^{-1} may be due to HOH bending vibration. The absorption bands at 2900 and 3421 cm^{-1} may be assigned to stretching vibrations of coordinated water [30, 31].

Figure 2 shows the TG and DTA curves of the zirconium selenomolybdate gel. The first step in the TG curve between 40–200°C with the weight loss of 11.58% corresponds to the loss of physically adsorbed water and water of crystallization with an endothermic peak in the DTA curve at 125.44°C. The weight loss in the range 200–440°C was small (2.6%). An endothermic peak observed at 500.6°C with the weight loss of 11.165% in the range 435–530°C can be assigned to the evaporation of SeO_2 and coordinated water and to decomposition of hydroxyl groups. The weight loss in the range 600–900°C was 4.51%. The presence of suitable amounts of water in the gel material is important to enhance the ^{99m}Tc diffusion through the gel material during the elution [32].

Figure 3 shows the X-ray diffraction pattern of the dried zirconium selenomolybdate gel, which shows that it is amorphous. The amorphous structure is much better than the crystalline structure because of more efficient diffusion of the eluent through the amorphous material, increasing the elution yield.

The SEM images of the prepared ZSM material at different magnifications are shown in Fig. 4. As can be seen, the gel matrix has a porous basic structure, which makes it good material for diffusion of ^{99m}Tc during elution [33].

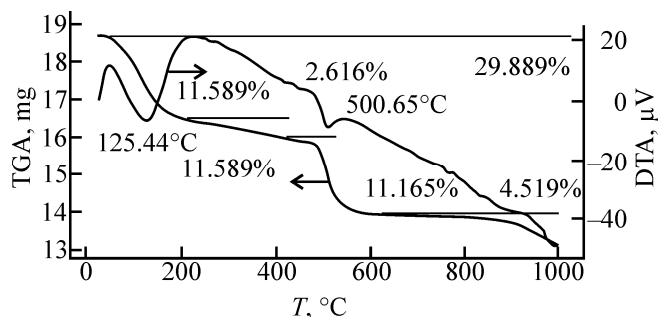


Fig. 2. TGA and DTA curves of ZSM gel matrix.

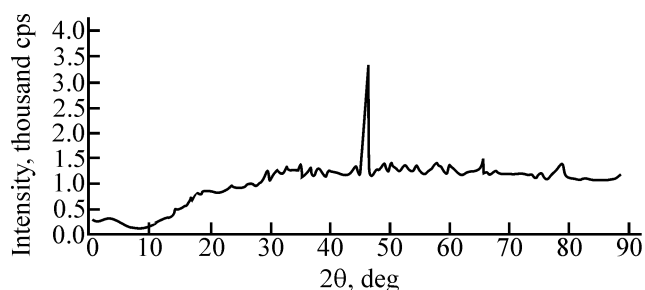


Fig. 3. X-ray diffraction pattern of ZSM gel matrix.

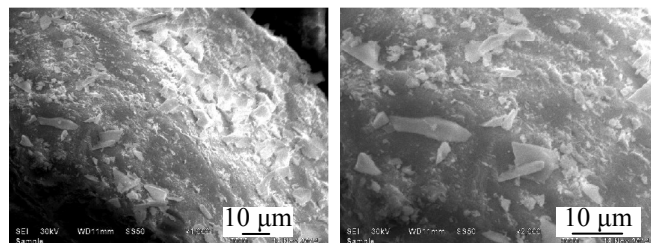


Fig. 4. FESEM images of ZSM gel matrix.

Elution of ^{99m}Tc . Figure 5 shows how the internal diameter of the column and the amount of the matrix influence the ^{99m}Tc elution profile. As can be seen, large size of the bed matrix is the major factor influencing the ^{99m}Tc elution performance. The position of the maximum elution is displaced to higher eluate volumes with increasing inside diameter of the column (0.46, 0.6, and 1.1 cm i.d.), i.e., with decreasing height of the zirconium selenomolybdate bed. About 97, 86.2, and 69.9% of the eluted ^{99m}Tc radioactivity was eluted with the first 3 mL. The elution yield was found to be 83.3, 81.7, and 76.8% for 0.46, 0.6, and 1.1 cm inside diameter, respectively. The elution profiles of ^{99m}Tc from the 1.1 cm i.d. column containing different amounts of the molybdate matrix (1, 3, and 6 g) are shown in Fig. 6. As can be seen, the position of the maximum elution remains unchanged (3 mL) with in-

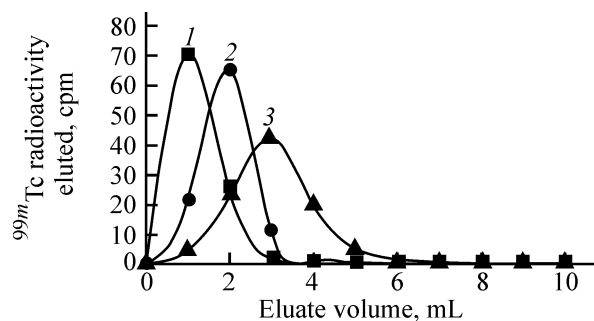


Fig. 5. Elution profile of ^{99m}Tc from column (1) 0.46, (2) 0.6 and (3) 1.1 cm i.d., packed with 1 g of ZSM, with 10 mL of 0.9% NaCl solution at a flow rate of 1 mL min^{-1} and room temperature ($\sim 25^\circ\text{C}$).

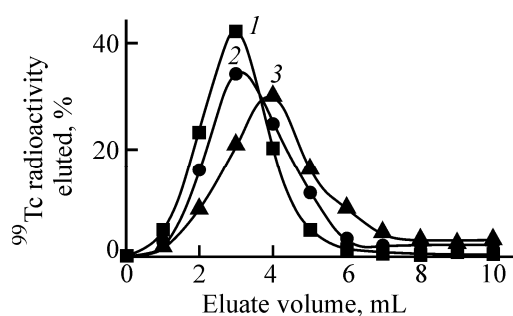


Fig. 6. Elution profile of ^{99m}Tc from columns (1.1 cm i.d.) containing (1) 1, (2) 3, and (3) 6 g of ZSM with 10 mL of 0.9% NaCl solution at a flow rate of 1 mL min^{-1} and room temperature ($\sim 25^\circ\text{C}$).

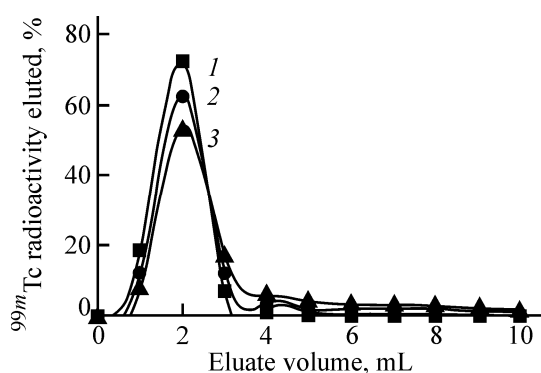


Fig. 7. Elution profile of ^{99m}Tc from a column (0.6 cm i.d.) containing 1 g of ZSM with 10 mL of 0.9% NaCl solution at flow rates of (1) 0.5, (2) 1, and (3) 2 mL min^{-1} and room temperature ($\sim 25^\circ\text{C}$).

creasing amount of the matrix from 1 to 3 g but is shifted to larger eluate volumes (4 mL) with increasing amount of the matrix from 3 to 6 g. About 89.9, 77.3, and 48% of the generated ^{99m}Tc activity was eluted

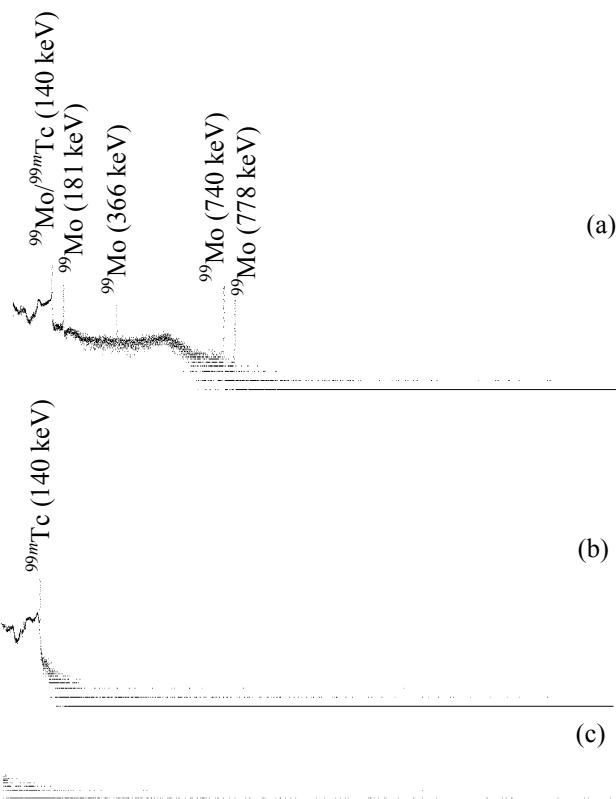


Fig. 8. γ -Ray spectra of (a) ZSM column (1.1 cm i.d.) loaded with ^{99}Mo , (b) ^{99m}Tc eluate measured immediately after elution, and (c) ^{99m}Tc eluate measured 7 days after elution.

with the first 4 mL. The elution yield was found to be 81.7, 79.2, and 77.1% for 1, 3 and 6 g, respectively.

Figure 7 shows the elution profiles of ^{99m}Tc from 1 g of zirconium selenomolybdate (0.6 cm i.d.) with 10 mL of 0.9% NaCl solution at flow rates of 0.5, 1, and 2 mL min^{-1} . The broad and tailed elution profiles with increasing flow rate may be attributed to slow sorption/desorption kinetics of ^{99m}Tc . The elution yield was found to be 82.2, 81.7, and 78.3% at flow rates of 0.5, 1, and 2 mL min^{-1} , respectively. The same behavior was reported for the elution of ^{99m}Tc from cerium molybdate gel matrix.

$^{99}\text{Mo}/^{99m}\text{Tc}$ radioisotope generator. The above data on the $^{99m}\text{Tc}(\text{VII})$ elution under different conditions are promising for constructing a $^{99}\text{Mo}/^{99m}\text{Tc}$ radioisotope generator of the chromatographic column type, based on precipitated zirconium selenomolybdate- ^{99}Mo . A glass column (1.1 cm i.d.) was packed with 6 g of zirconium selenomolybdate- ^{99}Mo gel, conditioned, and eluted with 10 mL of 0.9% NaCl solution.

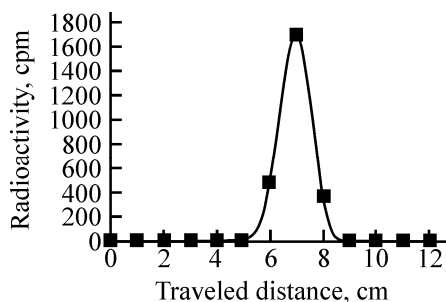


Fig. 9. Radiochromatogram of ^{99m}Tc eluate from a column (0.6 cm.i.d.) containing 6 g of ZSM with 10 mL of 0.9% NaCl solution at a flow rate of 1 mL min⁻¹.

Figures 8a–8c show the γ -ray spectra of the column containing 6 g of zirconium selenomolybdate- ^{99}Mo (1.1 cm i.d.) and of ^{99m}Tc eluates immediately after elution and 7 days later, respectively. Only the ^{99m}Tc peak was detected in the eluates. The radionuclidic purity of the eluates was verified by γ -ray spectrometry and radioactive decay measurements using an NaI(Tl) scintillation counter. The half-life of ~ 6 h, corresponding to ^{99m}Tc , was obtained. As judged from the residual radioactivity belonging to long-lived impurities (Fig. 8c), the radionuclidic purity was $\geq 99.99\%$.

Figure 9 shows the radiochromatograms of ^{99m}Tc eluates obtained from 6 g of zirconium selenomolybdate- ^{99}Mo with 0.9% NaCl at a flow rate 1 mL min⁻¹. The peaks in the chromatograms were identified by γ -ray spectrometry and radioactive decay measurements. In all chromatograms, there was only one peak at ~ 7 cm due to $^{99m}\text{TcO}_4^-$. The radiochemical purity was found to be 98.3%. This value complies with the recommended specifications for ^{99m}Tc -labeled radiopharmaceuticals.

The ^{99m}Tc eluates were chemically analyzed for the presence of Zr, Mo, and Se as chemically stable impurities from the zirconium selenomolybdate matrix. The Mo concentration in the eluates was found to be within 5 $\mu\text{g mL}^{-1}$. Such Mo content of ^{99m}Tc eluates is below the permitted limit for medical applications.

The pH of ^{99m}Tc eluates was found to be about 5.

REFERENCES

1. Bryan, R.N., *Introduction to the Science of Medical Imaging*, New York: Cambridge Univ. Press, 2010.
2. Eckelman, W.C. and Coursay, B.M., *Int. J. Appl. Radiat. Isot.*, 1982, vol. 33, p. 10.
3. Evans, J.V. and Shying, M.E., Zirconium molybdate gel as a generator for technetium-99m, *AAEC/E59*, 1984.
4. Tubis, M. and Wolf, W., *Radiopharmacy*, New York: Wiley, 1976, p. 263.
5. Moraes, V., Marczewski, B., Dias, C.R., and Junior, J.A.O., *Braz. Arch. Biol. Technol.*, 2005, vol. 48, pp. 51–56.
6. Monroy-Guzman, F., Díaz-Archundia, L.V., and Ramírez, A.C., *Int. J. Appl. Radiat. Isot.*, 2003, vol. 95, pp. 27–34.
7. Zolle, I., *Technetium-99m Radiopharmaceuticals: Preparation and Quality Control in Nuclear Medicine*, Berlin: Springer, 2007.
8. Uddin, M.S., Hagiwara, M., Tarkani, F. and Babs, M., *Appl. Radiat. Isot.*, 2004, vol. 60, pp. 911–920.
9. Jalilian, A.R., Targholizadeh, H., Raisali, G.R., et al., *DARU*, 2001, vol. 19, no. 3, pp. 187–192.
10. Guerin, B., Tremblay, S., Rodrigue, S., et al., *J. Nucl. Med.*, 2010, vol. 51, no. 4, pp. 13N–16N.
11. Arino, H. and Kramer, H.H., *Int. J. Appl. Radiat. Isot.*, 1975, vol. 26, p. 301.
12. Dash, A., Knapp, F.R., and Pillai, M., *Nucl. Med. Biol.*, 2013, vol. 40, p. 167.
13. Fission molybdenum for medical use, *IAEA-TECDOC-515*, Vienna: IAEA, 1989.
14. Al-Absy, M. and El-Bayoumy, S., *Isotopenpraxis*, 1990, vol. 26, p. 60.
15. El-Kolaly, M., *J. Radioanal. Nucl. Chem.*, 1993, vol. 170, p. 293.
16. El-Absy, M., El-Enein, M.A., Raieh, M., and Aly, H., *J. Radioanal. Nucl. Chem.*, 1997, vol. 218, p. 157.
17. Monroy-Guzman, F., Romero, O.C., and Velazquez, H.D., *J. Nucl. Radiochem. Sci.*, 2007, vol. 8, p. 11.
18. Monroy-Guzman, F., Díaz-Archundia, L.V., and Hernandez-Cortes, S., *J. Braz. Chem. Soc.*, 2008, vol. 19, p. 380.
19. Mostafa, M., El-Sadek, A., El-Said, H., and El-Amir, M., *J. Nucl. Radiochem. Sci.*, 2009, vol. 10, p. 1.
20. Mostafa, M., Ramadan, H.E., El-Amir, M., and El-Said, H., *Radiochemistry*, 2013, vol. 55, p. 332.
21. El-Absy, M., El-Amir, M., Fasih, T., et al., *J. Radioanal. Nucl. Chem.*, 2014, vol. 299, p. 1859.
22. El-Amir, M., Mostafa, M., and Ramadan, H.E., *J. Nucl. Radiochem. Sci.*, 2014, vol. 14, p. 1.
23. Boyd, R.E., *Radiochim. Acta*, 1982, vol. 30, pp. 123–146.
24. El-Absy, M.A., El-Naggar, I.M., and Audah, A.I., *J. Radioanal. Nucl. Chem.*, 1994, vol. 183, no. 2, p. 339.

25. Narasimhan, D.V.S., Vanaja, P., and Mani, R.S., *J. Radioanal. Nucl. Chem.*, 1984, vol. 85, pp. 345–355.
26. Vanaja, P., Ramamoorthy, N., Iyer, S.P., and Mani, R.S., *Radiochim. Acta*, 1987, vol. 42, pp. 49–52.
27. Maoliang Li, Production of gel-type Tc-99m generator for nuclear medicine, *Lecture of IAEA Expert Mission for Brazil*, Sao Paulo (Brazil), March 8–22, 1996.
28. Mostafa, M., Saber, H.M., El-Sadek, A.A., and Nassar, M.Y., *Radiochim. Acta*, 2015, vol. 104, no. 4, pp. 257–265.
29. Mostafa, M., Saber, H.M., El-Sadek, A.A., et al., *Radiochemistry*, 2016, vol. 58, no. 4, p. 409.
30. Davis, M., *Infrared Spectroscopy and Molecular Structure*, Amsterdam: Elsevier, 1963, p. 318.
31. Semagne, B., Diaz, I., Kebede, T., and Taddesse, A.M., *React. Funct. Polym.*, 2016, vol. 98, no. 2, pp. 17–23.
32. Davarpanah, M.R., Nosrati, S.A., Fazlali, M., et al., *Appl. Radiat. Isot.*, 2009, vol. 67, pp. 1796–1801.
33. Nabi, S.A. and Shalla, A.H., *J. Hazard. Mater.*, 2009, vol. 163, pp. 657–664.