

# Preparation and characterization of cerium(IV) tellurium molybdate gel and its application as a bed for chromatographic $^{99}\text{Mo}/^{99\text{m}}\text{Tc}$ generator

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Cerium(IV) tellurium molybdate was prepared as a new gel bed for chromatographic  $^{99}\text{Mo}/^{99\text{m}}\text{Tc}$  generator. The gel material prepared with the optimum conditions was characterized with IR spectroscopy, thermal analysis, XRD, EDX and FESEM. The highest precipitation yield of  $^{99}\text{Mo}$  within the formed gel was found to be 97.6 % at a Ce:Te:Mo molar ratio of 1:0.2:1. The prepared  $^{99}\text{Mo}/^{99\text{m}}\text{Tc}$  chromatographic column based on Ce(IV) tellurium molybdate gel bed showed a good performance. The  $^{99\text{m}}\text{Tc}$  elution yield was  $77.8 \pm 3.0$  % with a radionuclidic purity of  $\geq 99.99$  %, radiochemical purity of  $96.5 \pm 1.3$  % (as  $^{99\text{m}}\text{TcO}_4^-$ ) and pH-value in the range of 5-7. Molybdenum and cerium levels in  $^{99\text{m}}\text{Tc}$  eluates did not exceed 1 and 0.3 ppm, respectively, while no tellurium was detected.

## 1. Introduction

Chromatographic column generators of  $(n,\gamma)^{99}\text{Mo}/^{99\text{m}}\text{Tc}$  gel have low-specific activity  $^{99}\text{Mo}$ , but they represent an alternative option to those of alumina loaded with higher specific-activity fission- $^{99}\text{Mo}$ . Gel generators can participate in the solution of the problem of the increasing demand of  $^{99\text{m}}\text{Tc}$ , which is used in more than 80 % of nuclear medicine applications, all over the world. Among the advantages of gel generators are the easy and quick preparation of  $(n,\gamma)^{99}\text{Mo}$  solution, low cost and minimal radioactive waste.<sup>1</sup> Criteria for good gel generators include high-content of  $^{99}\text{Mo}$  in the gel bed, high-elution yield and high radionuclidic, radiochemical and chemical purities of the  $^{99\text{m}}\text{Tc}$  eluates. Many gel materials, including acidic salts of polyvalent metals and salts of heteropolyacids, have been used as beds for  $^{99}\text{Mo}/^{99\text{m}}\text{Tc}$  generators, e.g., zirconium molybdate,<sup>2-6</sup> stannic molybdate,<sup>7</sup> titanium molybdate,<sup>8</sup> cerium(IV) molybdate,<sup>9</sup> 12-molybdocerate(IV),<sup>10</sup> 6-tungstocerate(IV)<sup>11</sup> magnesium molybdate<sup>12</sup> and zirconium molybdophosphate.<sup>13</sup> Zirconium molybdate is the most famous material used as a bed for  $^{99}\text{Mo}/^{99\text{m}}\text{Tc}$  generators, but the synthesis and study of other gel materials constitute a renewable research filed in literature to obtain more alternatives.

For zirconium molybdate gel, replacement of 5 % (molar) of zirconium with Ce(IV) enhances the  $^{99\text{m}}\text{Tc}$  elution yield due to overcoming the reduction of  $^{99\text{m}}\text{Tc}$  species, because of radiolysis, and oxidation of its species to  $^{99\text{m}}\text{TcO}_4^-$ .<sup>14</sup> Thus, in the present work, using such high molar ratio of cerium (IV) will be advantageous especially in case of high- $^{99}\text{Mo}$  activity generators in which the radiolysis effect will be higher. In addition, adding a small ratio of tellurium (IV) was found to increase the  $^{99}\text{Mo}$  uptake by the formed gel comparing with the case of CeMo gel.<sup>9</sup>

This work aims at the preparation of cerium(IV) tellurium molybdate gel material containing a high content of molybdenum which can be used as a new bed material for a chromatographic  $^{99}\text{Mo}/^{99\text{m}}\text{Tc}$  radioisotope generator of a good performance.

## 2. Experimental

All the chemicals used were of AR grade. Distilled water was used for preparing solutions.

A  $\gamma$ -ray spectrometer composed of a multichannel analyzer (MCA) of Inspector 2000 model (Canberra Series, USA), coupled with a high-purity germanium (HPGe) coaxial detector of GX2518 model, was used for  $\gamma$ -radioactivity identification and measurement.

A pH-meter with a microprocessor (Hanna Instruments pH211 model, Portugal) was used for measuring pH-values of solutions. An analytical balance (A&D Engineering Inc., AND HR-202 model, USA) having dual range (42 g/0.01 mg, 210 g/0.1 mg) was used for weighing. A centrifuge (REMI Laboratory, R32A model, India) was used for centrifugation of the mixtures to efficiently separate the precipitated gel materials from the supernatants.

A UV-VIS spectrophotometer (Shimadzu, UV-160 A model, Japan) was used for detection of Ce and Mo in  $^{99\text{m}}\text{Tc}$  eluted from the Ce(IV) tellurium molybdate- $^{99}\text{Mo}$  column.

The IR spectrum was recorded with an FT-IR spectrometer (Bomem, model MB157S, Canada) in the range from 4000 to  $400\text{ cm}^{-1}$  at room temperature.

Thermal analysis, including simultaneous TGA and DTA, was carried out with a thermal analyzer (Shimadzu, model DTG-60H, Japan). X-ray diffraction was performed with an 18 kV diffractometer (Bruker, model D8 Advance, USA) with monochromated  $\text{CuK}\alpha$  radiation ( $\lambda = 1.54178\text{ \AA}$ ).

Elemental analysis was performed using energy dispersive X-ray spectrometer (ISIS-300, Oxford EDX). FESEM images were taken using field emission scanning electron microscope (FESEM) (JEOL JSM-6500F, USA).

**2.1. Molybdenum-99 radiotracer.**  $^{99}\text{Mo}$  was obtained by eluting commercial  $^{99}\text{Mo}/^{99\text{m}}\text{Tc}$  generators with alumina-packed columns (Mon-Tek Generator, Monrol Nuclear Products Industry and Trade Inc., Turkey; loaded with 20 GBq on calibration date). Each column was eluted with 20 ml of 2 M  $\text{NH}_3$  solution 20 days after the calibration date. Then the  $\text{NH}_3$  solution was evaporated to dryness, and the residual  $^{99}\text{Mo}$  was redissolved in 5 ml of 0.1 M NaOH.

**2.2. Preparation of cerium(IV) tellurium molybdate (CeTeMo) gel.** A 0.1 M Mo(VI) solution was prepared by dissolving 2.42 g of  $\text{Na}_2\text{MoO}_4 \cdot 2\text{H}_2\text{O}$  in 20 ml of distilled  $\text{H}_2\text{O}$

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with the addition of 2.5 ml of  $^{99}\text{Mo}$  solution along with few drops of 30%  $\text{H}_2\text{O}_2$ . The volume of the  $\text{Mo(VI)}$  solution was completed to 100 ml with distilled  $\text{H}_2\text{O}$ . A 0.1 M  $\text{Ce(IV)}$  solution was prepared by dissolving 6.32 g of  $(\text{NH}_4)_4\text{Ce}(\text{SO}_4)_4 \cdot 2\text{H}_2\text{O}$  in 100 ml of dilute  $\text{H}_2\text{SO}_4$  to obtain a solution of pH1. A 0.1 M  $\text{Te(IV)}$  solution was prepared by dissolving 1.60 g of  $\text{TeO}_2$  in 100 ml of 5 M  $\text{NaOH}$ . Mixtures with different  $\text{Ce(IV)}:\text{Te(IV)}:\text{Mo(VI)}$  molar ratios were prepared by adding  $\text{Mo(VI)}$  solution dropwise to  $\text{Ce(IV)}$  solution and, then,  $\text{Te(IV)}$  solution was added dropwise to the  $\text{Mo(VI)}-\text{Ce(IV)}$  mixture. Varied volumes of the three solutions were used to obtain different molar ratios. Thereafter, pH of the final mixture was adjusted to 5.5 by using  $\text{H}_2\text{SO}_4/\text{NaOH}$  solutions. The final volume of each mixture was 50 ml. The mixtures were left for 10 h to allow the formed precipitates to settle and then were centrifuged. The supernatant of each mixture was checked radio-metrically to determine the precipitation yield of  $^{99}\text{Mo}$  within the formed gel material.

The precipitated gel obtained from the mixture with the optimum  $\text{Ce}:\text{Te}:\text{Mo}$  molar ratio (i.e., the mixture with which the highest precipitation yield of  $^{99}\text{Mo}$  was achieved) was found to be that with 1  $\text{Ce}:$  0.2  $\text{Te}:$  1  $\text{Mo}$ . The mixture with the optimum molar ratio was prepared again by adding 50 ml of 0.1 M  $\text{Mo(VI)}$  solution, containing  $^{99}\text{Mo}$  radiotracer, dropwise to 50 ml of 0.1 M  $\text{Ce(IV)}$  and then 10 ml of 0.1 M  $\text{Te(IV)}$  was added dropwise to the  $\text{Mo(VI)}-\text{Ce(IV)}$  mixture. The pH-value was thereafter adjusted to 5.5 by using  $\text{H}_2\text{SO}_4/\text{NaOH}$  solutions. The mixture was left for 10 h to allow the precipitate to settle. The mixture was then centrifuged, the supernatant was removed by decantation while the precipitate was spread as a thin layer on a watch glass and dried for 12 h at  $50^\circ\text{C}$ . The dried gel was pulverized to  $\sim 0.1\text{--}0.2$  mm particle size and, then, used as a column bed for a  $^{99}\text{Mo}/^{99\text{m}}\text{Tc}$  chromatographic generator. The latter mixture was prepared again by the same method, but without adding  $^{99}\text{Mo}$  radiotracer and with washing the dried gel with excess distilled water followed by re-drying overnight at  $50^\circ\text{C}$ , to characterize the prepared gel.

**2.3. Characterization of CeTeMo gel.** The inactive prepared CeTeMo gel (with the optimum molar ratio) was characterized by IR spectroscopy, thermal analysis (TGA and DTA), XRD, EDX and FESEM.

**2.4. Preparation of  $^{99}\text{Mo}/^{99\text{m}}\text{Tc}$  chromatographic column generator.** A 0.7 cm i.d. glass column was used as a bed for the  $^{99}\text{Mo}/^{99\text{m}}\text{Tc}$  generator. The column was provided with a small piece of glass wool, as a support for the gel bed, and a bottom stopcock. 1 g of the pulverized CeTeMo gel bed was packed into the column by settling from distilled water. The column was excessively washed with distilled water and subsequently conditioned with 50 ml of 0.9 %  $\text{NaCl}$  for further elution of  $^{99\text{m}}\text{Tc}$ .

**2.5. Elution of  $^{99}\text{Mo}/^{99\text{m}}\text{Tc}$  generator and quality control of the eluted  $^{99\text{m}}\text{Tc}$ .** Technetium-99m was firstly eluted 23 h after the conditioning time by passing 10 ml of 0.9 %  $\text{NaCl}$  solution through the CeTeMo gel column. The column was eluted 14 times with different flow rates (0.25, 0.5, 0.75 and 1 ml/min) over the course of 25 days. The time between each two successive elutions was  $\geq 23$  h. The  $^{99\text{m}}\text{Tc}$  elution yield was determined by comparing the counting rates of the 140-keV peak in the  $\gamma$ -ray spectrum of the column directly before and after elution, taking the contribution of  $^{99}\text{Mo}$  to this peak into account.<sup>15</sup> The elution profiles were drawn for various elution flow rates. pH-values of the eluate solutions were determined using the pH-meter.

The percent  $^{99}\text{Mo}$  content in the eluate was determined, in the case of its presence, as the contribution of the count rate of 181- or 739-keV peak in the  $\gamma$ -ray spectrum to the total count rate. The  $\gamma$ -ray spectrum of the eluate was measured directly after elution and after 5 days. The radionuclidic purity of the eluted  $^{99\text{m}}\text{Tc}$  was determined as the contribution of the count

rate of 140-keV peak in the  $\gamma$ -ray spectrum of the eluate to the total count rate.

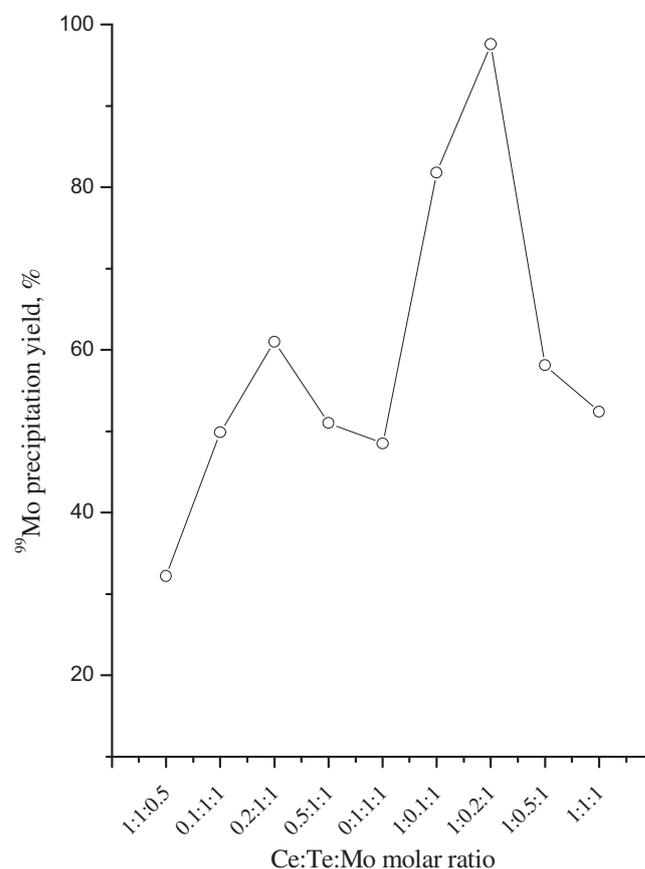
The radiochemical purity of the eluted  $^{99\text{m}}\text{Tc}$  (contribution of  $^{99\text{m}}\text{TcO}_4^-$  count rate to the total count rate the eluted  $^{99\text{m}}\text{Tc}$ ) was determined by ascending paper chromatography using Whatman no. 1 chromatographic paper and a mixture of 85% methanol + 15%  $\text{H}_2\text{O}$  as a developing solvent.<sup>16</sup> The activity distribution along the chromatogram was traced using the  $\gamma$ -ray spectrometer to determine  $R_f$ , where:

$$R_f = \frac{\text{traveled distance of the radioactivity from the start point to the peak position}}{\text{traveled distance of the solvent from the start point to the solvent front}}$$

The possible impurities of molybdenum, cerium and tellurium originated from the CeTeMo gel matrix, were determined in  $^{99\text{m}}\text{Tc}$  eluates spectrophotometrically by using thiocyanate method (maximum absorption at 470 nm), persulfate method (maximum absorption at 320 nm), Bismuthiol II method (maximum absorption at 330 nm), respectively.<sup>17</sup>

### 3. Results and discussion

CeTeMo gels were prepared with different  $\text{Ce}:\text{Te}:\text{Mo}$  molar ratios. As indicated by Figure 1, precipitation yield of  $^{99}\text{Mo}$  increased gradually from 32.2 % at  $\text{Ce}:\text{Te}:\text{Mo}$  molar ratio of 1:1:0.5 till reaching a first lower maximum value of 61 % at the molar ratio of 0.2:1:1 and then, it slightly decreased to 48.5 % at the molar ratio of 0:1:1:1. Thereafter, the precipitation yield sharply increased to a second higher maximum value of 97.6 % at the molar ratio of 1:0.2:1 and then sharply decreased to 58.1 % at the molar ratio of 1:0.5:1. Further change of the molar ratio to 1:1:1 yielded a precipitation yield of 52.4 %. Hence,  $\text{Ce}:\text{Te}:\text{Mo}$  molar ratio of 1:0.2:1 was chosen for characterization of the prepared CeTeMo gel (via IR, thermal analysis, XRD, EDX and FESEM) and further preparation and quality



**Figure 1.** Precipitation yield of  $^{99}\text{Mo}$  as a function of  $\text{Ce}:\text{Te}:\text{Mo}$  molar ratio used for preparing CeTeMo gel.

control of  $^{99}\text{Mo}/^{99\text{m}}\text{Tc}$  generator. Thus, the addition of tellurium during gel preparation, with a Ce:Te:Mo molar ratio of 1:0.2:1, increased the precipitation yield of  $^{99}\text{Mo}$  to 97.6 %, which is markedly higher than the yields of 35.8 and 63 %  $^{99}\text{Mo}$  in the case of CeMo gel with Mo:Ce molar ratios of 1:1 and 2:1, respectively.<sup>9</sup>

Figure 2 shows IR spectrum of CeTeMo gel. The IR spectrum includes peaks at  $501\text{ cm}^{-1}$  (due to Te-O-Te bending mode),  $874\text{ cm}^{-1}$  (due to Mo-O stretching mode),  $1401\text{ cm}^{-1}$  (due to Ce-O-H bending mode),  $1614$  and  $3411\text{ cm}^{-1}$  (due to O-H bending and stretching modes of lattice water, respectively).<sup>9,18-21</sup>

Figure 3 shows the TGA and DTA curves of the CeTeMo gel. There was a 8.3 % weight loss on TGA curve in the temperature range from 16 to  $200^\circ\text{C}$  due to the loss of lattice water with an endothermic peak on DTA curve at  $71^\circ\text{C}$ . The presence of lattice water in a suitable amount is important to enhance the diffusion of  $^{99\text{m}}\text{TcO}_4^-$  through the gel material during elution.<sup>22</sup> In the range of  $200\text{--}800^\circ\text{C}$ , there was another weight loss of 4.1% due to decomposition of the gel material to

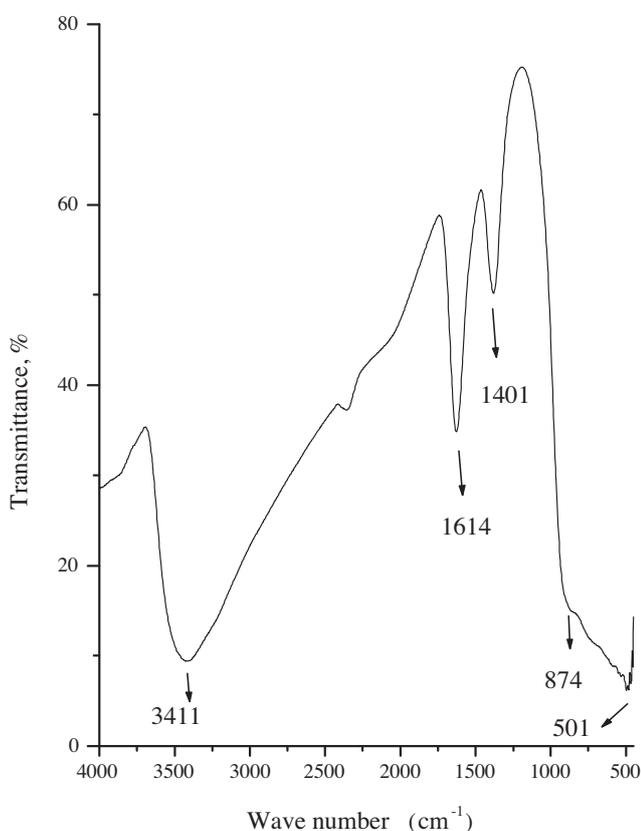


Figure 2. IR spectrum of CeTeMo gel.

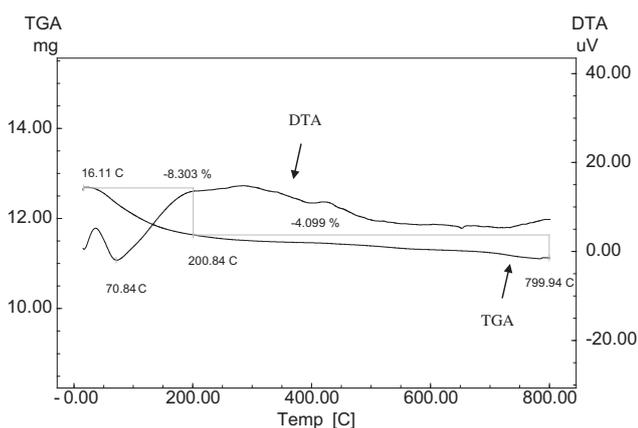


Figure 3. TGA and DTA curves of CeTeMo gel.

cerium, molybdenum and tellurium oxides, in addition to subsequent sublimation of molybdenum oxide.<sup>21,23</sup> The thermal gravimetric behavior of CeTeMo gel resembles to a great extent that of cerium(IV) molybdate (CeMo) gel.<sup>9</sup>

Energy dispersive X-ray (EDX) spectrometric elemental analysis indicated that the prepared gel material incorporated ~27 % (wt./wt.) of molybdenum. This Mo content is considered high (where gel matrices used in  $^{99}\text{Mo}/^{99\text{m}}\text{Tc}$  generators should contain high Mo content; > 25 %) and comparable with Mo content in zirconium molybdate gels (30 %).<sup>24</sup>

Figure 4 shows the XRD pattern of the CeTeMo gel, which indicates a nearly amorphous structure (low-crystallinity structure) of the prepared CeTeMo gel. The amorphous structure is usually preferred to the rigid crystalline structure, where the latter one hinders the mobility of  $^{99\text{m}}\text{Tc}$  leading to a decrease in the elution yield. In addition, the amorphous structure has a more mechanical strength than the crystalline one, i.e., it is more resistant to abrasion and dissolution.<sup>25</sup>

Figure 5 shows FESEM images of the CeTeMo gel. Image (a) indicates the gel particles with a size range of ~100-200  $\mu\text{m}$  (~0.1-0.2 mm), while the more magnified images (b) and (c) indicate many meanders and cavities, the presence of which facilitates the diffusion of saline solution through the gel bed during  $^{99\text{m}}\text{Tc}$  elution.

Figure 6 compiles the elution profiles of the  $^{99}\text{Mo}/^{99\text{m}}\text{Tc}$  generator at flow rates of 0.25, 0.5, 0.75, and  $1\text{ ml min}^{-1}$ . The elution yield of  $^{99\text{m}}\text{Tc}$  was  $77.8 \pm 3.0\%$ . Thus, the average  $^{99\text{m}}\text{Tc}$  elution yield for CeTeMo gel is slightly higher than that for CeMo gel (75.4 %).<sup>9</sup> Sharp elution peaks were obtained with the flow rates of 0.25, 0.5 and  $0.75\text{ ml min}^{-1}$  (the most sharp one was obtained with the flow rate of  $0.75\text{ ml min}^{-1}$ ), while a relatively broad one was obtained with the flow rate of  $1\text{ ml min}^{-1}$ . A maximum percent fraction of  $^{99\text{m}}\text{Tc}$  activity was obtained at the 2<sup>nd</sup> ml of eluate for the flow rates of 0.25, 0.5 and  $1\text{ ml min}^{-1}$  (49.7, 54.1 and 33.1 %, respectively), while it was obtained at the 1<sup>st</sup> ml of eluate for the flow rate of  $0.75\text{ ml min}^{-1}$  (82.5 %).

Radionuclidic purity of the obtained  $^{99\text{m}}\text{Tc}$  eluates was  $\geq 99.99\%$ , i.e., in all cases  $^{99}\text{Mo}$  content in the eluate did not

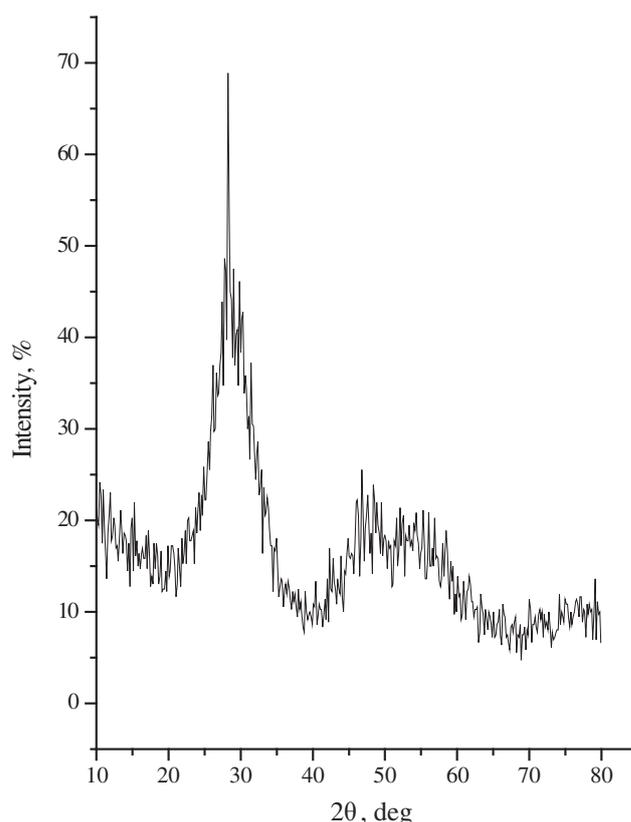
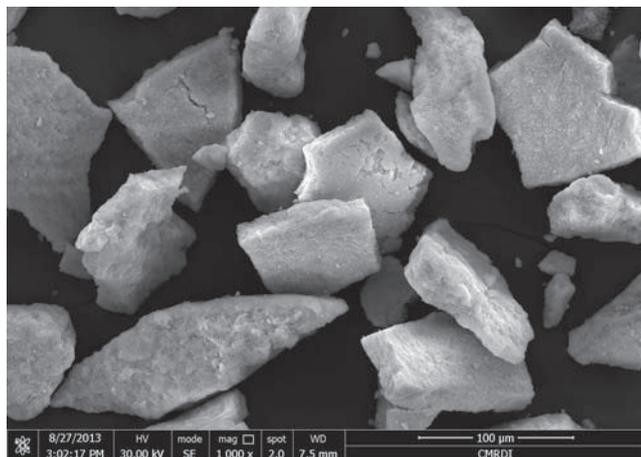
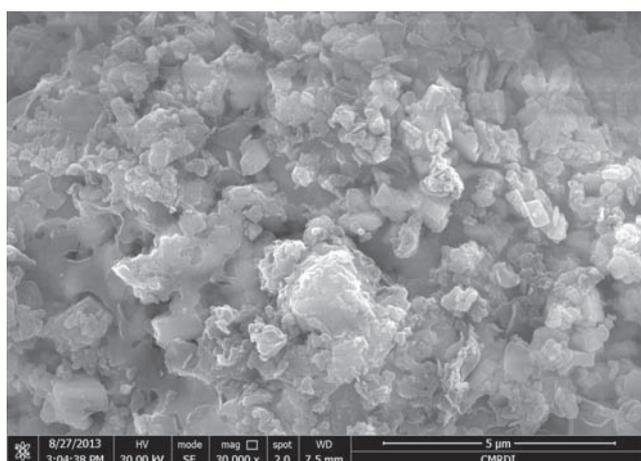


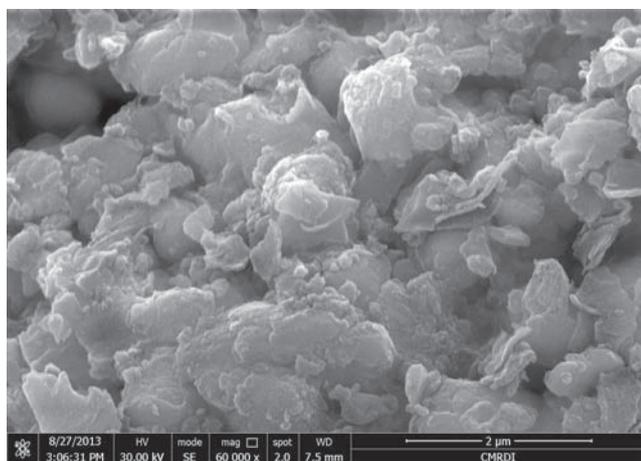
Figure 4. XRD pattern of CeTeMo gel.



(a)



(b)



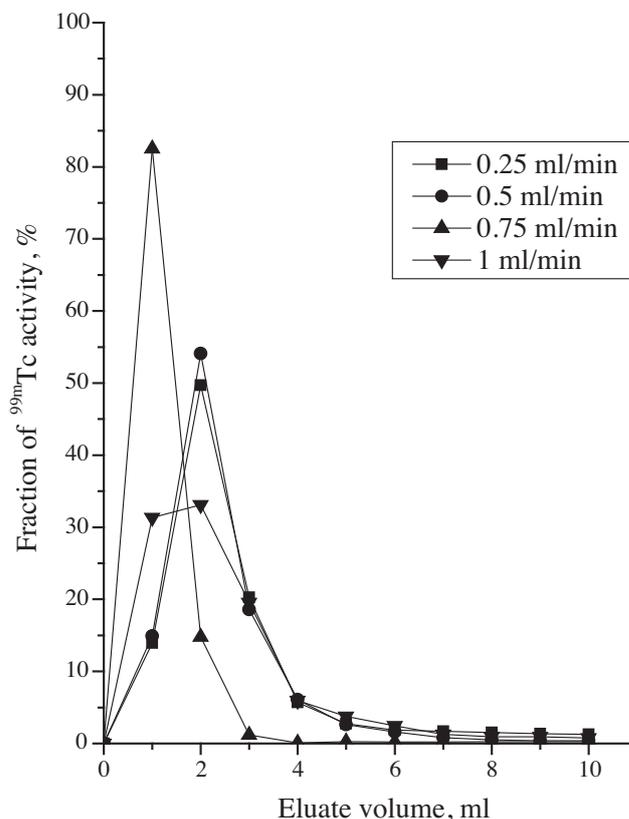
(c)

**Figure 5.** FESEM images of CeTeMo gel.

exceed  $1 \times 10^{-3}$  %. pH-values of  $^{99m}\text{Tc}$  eluates ranged from 5-7. Radiochemical purity was  $96.5 \pm 1.3$  % as  $^{99m}\text{TcO}_4^-$ . Chemical impurities of molybdenum and cerium in  $^{99m}\text{Tc}$  eluates did not exceed 1 and 0.3 ppm, respectively, while no tellurium was detected.

#### 4. Conclusion

Cerium(IV) tellurium molybdate (CeTeMo) gel was successfully prepared as a high-Mo content material with a high  $^{99}\text{Mo}$  precipitation yield. CeTeMo gel was used as a new bed material for a chromatographic  $^{99}\text{Mo}/^{99m}\text{Tc}$  radioisotope generator showing a good performance; suitable  $^{99m}\text{Tc}$  elution yield, high radionuclidic, radiochemical and chemical purities and pH-



**Figure 6.** Elution profiles of  $^{99m}\text{Tc}$  from CeTeMo gel column as a function of flow rate.

value in the range of 5-7.

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