# 2.16 PREPARATION OF CHROMATOGRAPHIC AND SOLID-SOLVENT EXTRACTION $^{99m}$Tc GENERATORS USING GEL-TYPE TARGETS

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## Abstract

We have studied two types of targets Zirconium-Molybdate (ZrMo) and Titanium-Molybdate (TiMo) prepared by precipitating reaction between Ammonium-Molybdate and Zirconium-Chloride or Titanium-Chloride solutions, respectively. Other types of targets were also prepared by co-precipitating ZrMo or TiMo with hydrous Manganese-Dioxide, hydrous Silica, and hydrous Titanium-Dioxide or by impregnated ZrMo or TiMo with Iodate anions.

The results of our investigations on extraction of Tc-99m from neutron irradiated TiMo solid phase using solvents such as MEK, Aceton, Ethylic Ether, Chloroform, etc showed that separation yield (SY) of Tc-99m in case of Aceton extraction was from 70% to 80% and in other cases not higher than 40%. The Tc-99m elution curves and column kinetic in case of Aceton extraction (after evaporation of Aceton and recovery of Tc-99m in 0,9% NaCl solution) was superior than in case chromatographic generator using saline eluant.

As a result obtained, two types of generators were successfully prepared and put into use:

- Chromatographic generator using Titanium-Molybdate target as column packing material and saline as eluant.
- Solid-Solvent extraction $^{99m}$Tc generator using Titanium-Molybdate target (as solid phase) and aceton as extracting solvent.

## INTRODUCTION

After the reconstruction of our reactor of power of 500 KW in late 1983, the production of $^{99m}$Tc nuclide in our institute has been based on MEK extraction method. The inherent disadvantages of this method compelled us to start our studies on gel type generator in late 1984 /5/. We first aimed to develop Titanium- and Zirconium- Molybdate targets giving high separation yield (SY) of $^{99m}$Tc with suitable quality for present and future application in nuclear medicine. As a result obtained, two types of generators were successfully prepared and put into use/6-9/:

- Chromatographic generator using Titanium-Molybdate target as column packing material and saline as eluant.
- Solid-Solvent extraction $^{99m}$Tc generator using Titanium-Molybdate target (as solid phase) and aceton as extracting solvent.

## CHROMATOGRAPHIC $^{99m}$Tc GENERATOR

We have studied two types of targets Zirconium-Molybdate (ZrMo) and Titanium-Molybdate (TiMo) prepared by precipitating reaction between Ammonium-Molybdate and Zirconium-Chloride or Titanium-Chloride solutions, respectively. Other types of targets were also prepared by co-precipitating ZrMo or TiMo with hydrous Manganese-Dioxide, hydrous Silica, and hydrous Titanium-Dioxide or by impregnated ZrMo or TiMo with Iodate anions.

After being irradiated in our reactor of thermal neutron flux of 1.8 $x10^{13}$ $n/cm^2$.sec for different periods of time, the targets were packing in the column and directly (or after passing different oxidizing solutions through it) eluted with 0,9% NaCl solution.

The results obtained showed that all types of ZrMo targets with even shorter time of irradiation gave SY of $^{99m}$Tc not higher than 12%. The treatment with oxidizing solution did not improve the SY significantly.

The TiMo target also gave low SY of $^{99m}$Tc (about 25%) but after treatment with oxidizing solution SY rose to higher than 90%. In case of TiMo target our results were in agreement with Mani's {1} although the preparation methods were different.

TiMo target co-precipitated with Manganese-Dioxide (TiMoMn) gave SY of from 70% to 80% (without treatment with oxidizing solution after irradiation for an irradiation time of one hour. For longer irradiation SY varied with the content of Manganese-Dioxide, but the treatment with oxidizing solution after irradiation gave a yield of higher than 90%.

TiMo targets coprecipitated with hydrous silica and Titanium-Dioxide gave SY significantly lower than TiMo.

TiMo target impregnated with Iodate gave good SY, higher than 85% either in one or a hundred hours of irradiation time (without treatment with oxidizing solution).

Concerning treatments with oxidizing solutions, three solutions were investigated: 0.1M $KMnO_4$, 0.1M $KBrO_3$ and 0.1M $K_2CrO_4$ solutions. Among these treatments that with 0.1M $KMnO_4$ solution was highvalued. This treatment composes of two processes subsequently occurring one after another: First washing column with 0.1M $KMnO_4$ solution and subsequently with distilled water, then converting the small amount of $MnO_4^-$ ions remained on the column to Manganese-Dioxide. This process assured the elimination of trace amounts of $KMnO_4$ which could be eluted with pertechnetate ions and would interfer with the chemical reactions involved in the $Sn^{2+}$ reduction of $TcO_4^-$ ions in the preparation of radiopharmaceuticals. Besides, Manganese-Dioxide particles adhered on the surface of TiMo particles play a role as stabilizer assuring consistently good SY and good adsorbent for trace of many metal ions.

The kinetic of TiMo bed was also studied. In fig.1 the effect of total radioactivity of Tc-99m on eluate volume was illustrated.

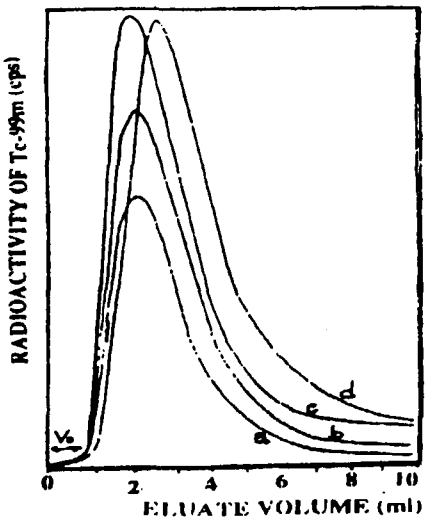



Fig.1 : $^{99m}$Tc elution profiles of TiMo beds of various specific radioactivity.

TiMo bed weight: 7,0 g,
Column diameter: 10mm,
a. 50 mCi
b. 70 mCi Eluant: 0,9% NaCl
c. 100 mCi
d. 100 mCi Aceton eluant

As seen the eluate volume was not effected by total radioactivity. Fig.2 showed the effect of bed weight of TiMo target packing in generator on elution (or retention) volume ($V_R$). $V_R$ increased non-linearly with bed weight. This means that if $V_R$ was chosen for elution, then the larger the bed weight the higher the radioactive concentration of $^{99m}$Tc in eluate is. The radioactive concentration of $^{99m}$Tc increased from 3% in case of column of 3g of TiMo to 17% in case of 7g packing.

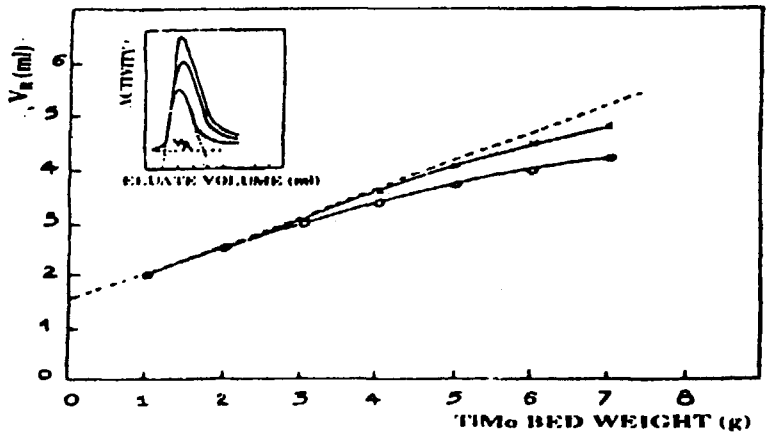



Fig.2: Retention volume vs. TiMo bed weight.
-o-o-o- 0,9% NaCl solution as eluant
-x-x-x- Aceton as eluant
(TiMo particle size: 100-200 mesh, Column size: 10 mm in diameter, Specific radioactivity of TiMo : 10 mCi/g)

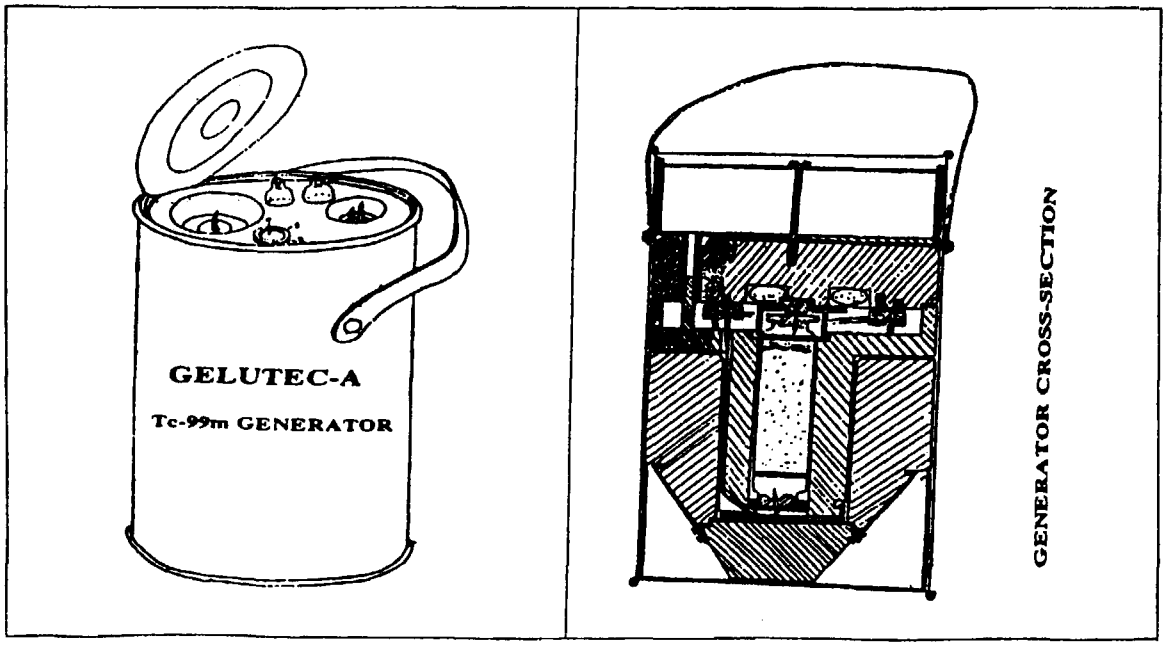
Based on the results obtained we prepared the generators of 150 mCi of Tc-99m. (Bed weight of TiMo: 5g, Irradiation time: 100hours, Cooling time: 48 hours, Thermal neutron flux: 1.8 x $10^{13}$ n/cm$^3$.sec). The quality of pertechnetate solution of our generators was found to meet all the requirements of $^{99m}$Tc pertechnetate injection as specified in various pharmacopeia {2,3}. These generators were successfully used in hospitals in Vietnam.

## SOLID-SOLVENT EXTRACTION GENERATOR

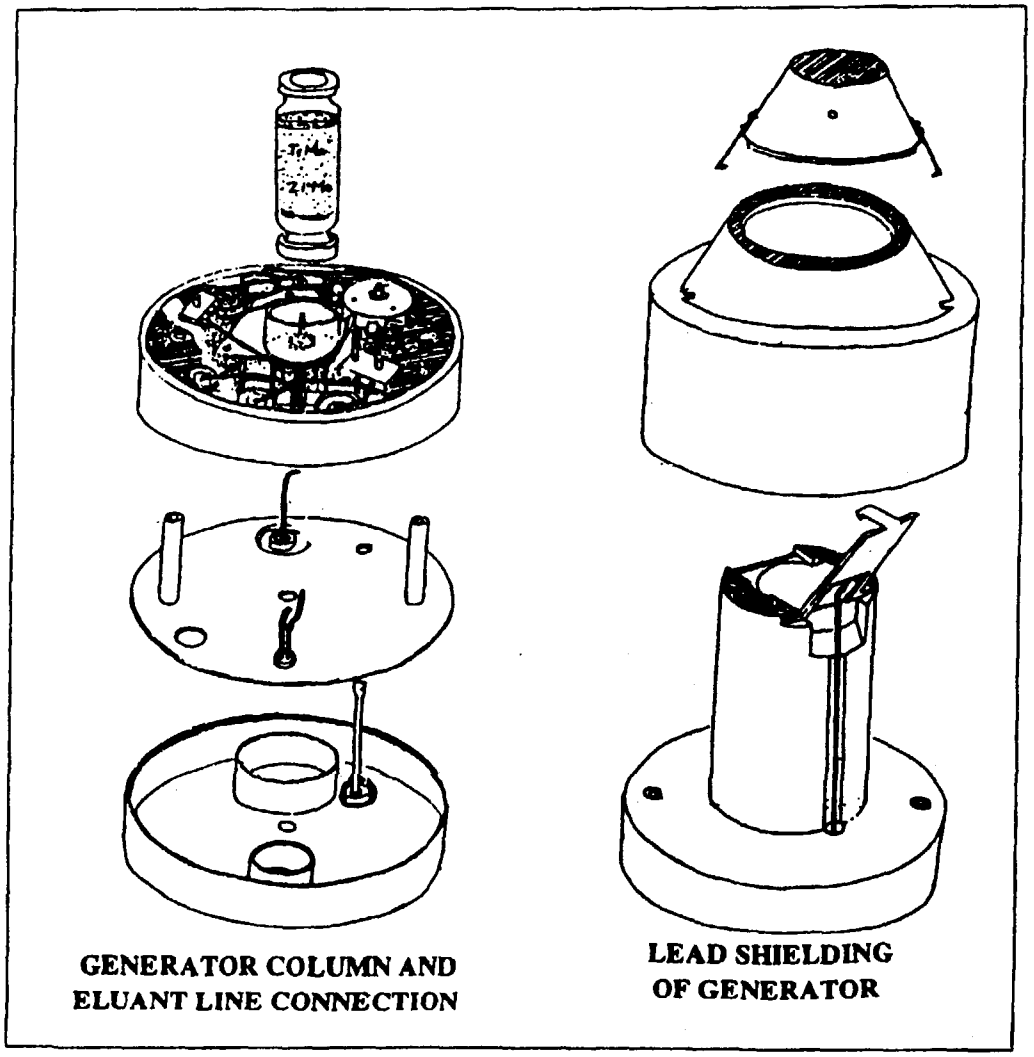
·Chromatographic gel-type generator may give a $^{99m}$Tc pertechnetate solution of limited radioactive concentration as foreseen. In case of bolus injection, Tc-99m solution obtained from extraction generator, the solid-solvent extraction generator developed by us using TiMo target as solid phase offers to users the advantages of an easy-to-use, economical process and non-elaborated apparatus. This generator gives a sterile pertechnetate solution of high and invariable quality.

The results of our investigations on extraction of Tc-99m from neutron irradiated TiMo solid phase using solvents such as MEK, Aceton, Ethylic Ether, Chloroform, etc showed that separation yield (SY) of Tc-99m in case of Aceton extraction was from 70% to 80% and in other cases not higher than 40%. The Tc-99m elution curves and column kinetic in case of Aceton extraction (after evaporation of Aceton and recovery of Tc-99m in 0,9% NaCl solution) was superior than in case chromatographic generator using saline eluant.

The great advantages of the solid-solvent extration generator mentioned above-small eluate volume, high SY, excellent quality of pertechnetate solution – and useful chemical and physico-chemical properties of aceton were found to meet the requirements of an easy-to-use and effective production method of $^{99m}$Tc from $^{99}$Mo of low specific radioactivity.




**Figure**




**Figure**

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