A $^{99}$Mo-$^{99m}$Tc GENERATOR BASED ON THE USE OF ZIRCONIUM MOLYBDOPHOSPHATE-$^{99}$Mo GEL

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A modified $^{99}$Mo-$^{99m}$Tc gel generator is described. The present generator uses an insoluble zirconium molybdophosphate (ZrMP) gel tagged with $^{99}$Mo. Molybdenum-99 is chemically combined in the gel structure and cannot be eluted from the matrix. The presence of phosphate increases the chemical stability of the gel and decreases the molybdenum breakthrough. The prepared gel is sufficiently porous to permit ready diffusion of $^{99m}$TcO$_4^-$ which can be eluted with saline in yields of up to 90%. The gel was found to contain 25.1% Mo, 21.9% Zr, and 0.7% P in a molar ratio of 1.09:1.0:0.09, respectively. The high molybdenum content of the gel allows the use of cheap, non-polluting (n, $\gamma$)$^{99}$Mo. The eluted $^{99m}$Tc was of high purity and can be used for medical and pharmaceutical applications.

Technetium-99m and $^{99m}$Tc labeled radiopharmaceuticals are still the most widely used radiodiagnostic agents in nuclear medicine.1-3 Technetium-99m is readily available in the form of a generator in which it is separated from the parent $^{99}$Mo most commonly by alumina column chromatography.4 The chromatographic generator, containing fission $^{99}$Mo adsorbed onto alumina, remains the most common source of $^{99m}$Tc for nuclear medicine, however, its use is sufficiently difficult to justify searching for an alternative generator.1,5,6 The development of new techniques for production of $^{99m}$Tc is of great importance especially for those countries having low power research reactors. Both solvent extraction and sublimation generators have been used with varying degrees of success.1,5

More recent work has illustrated that the low and medium specific activity of (n, $\gamma$)$^{99}$Mo can be incorporated in an insoluble gel structure, which after granulation efficiently releases $^{99m}$Tc on elution.1,6,7

This study describes the preparation and use of zirconium molybdophosphate gel tagged with tracer level of (n, $\gamma$)$^{99}$Mo as a generator for $^{99m}$Tc.

Experimental

Materials: all chemicals used were of analytical grade. Molybdenum-99 was obtained by neutron irradiation of MoO$_3$ and was used as a tracer for Mo in the form of ZrMP.
Preparation of ZrMP-\textsuperscript{99}Mo gel: 2 g of neutron irradiated MoO\textsubscript{3} was dissolved in 5M NaOH by warming with addition of a few drops of H\textsubscript{2}O\textsubscript{2} according to the reaction:

\[
\text{MoO}_3 + 2\text{NaOH} \rightarrow \text{Na}_2\text{MoO}_4 + \text{H}_2\text{O}
\] (1)

The molybdate-\textsuperscript{99}Mo solution formed was converted into molybdophosphate-\textsuperscript{99}Mo by reacting with the calculated amount of Na\textsubscript{2}H\textsubscript{2}P\textsubscript{2}O\textsubscript{7} (160 mg) according to the following reaction:

\[
12\text{Na}_2\text{MoO}_4 + \text{NaH}_2\text{PO}_4 + 25\text{HNO}_3 \rightarrow \text{H}_3\text{PMo}_{12}\text{O}_{40} + 25\text{NaNO}_3 + 12\text{H}_2\text{O}
\] (2)

H\textsubscript{2}O\textsubscript{3} was added to adjust the pH to 4.5. The molybdophosphate-\textsuperscript{99}Mo solution formed was added slowly to a zirconium oxy-nitrate (4.19 g) solution during stirring. The temperature was raised to 55–66 °C. A zirconium molybdophosphate-\textsuperscript{99}Mo gel was formed according to the reaction:

\[
\text{H}_3\text{PMo}_{12}\text{O}_{40} + \text{ZrO(NO}_3)_2 \cdot x\text{H}_2\text{O} \rightarrow \frac{1}{2}\text{ZrMP} \cdot x\text{H}_2\text{O} \text{gel}
\] (3)

The mixture was kept stirring for 30 minutes at 55–66 °C. The precipitate was then vacuum filtered on a Buchner funnel and air-dried in an oven at 55–60 °C for 12–15 hours.

**Gel-analysis:** the ZrMP gel prepared was analyzed for its contents of Zr, Mo and P. The gel was decomposed by dissolving in 3M NaOH to bring P and Mo into solution possibly in the form of Na\textsubscript{2}H\textsubscript{2}P\textsubscript{2}O\textsubscript{7} and Na\textsubscript{2}MoO\textsubscript{4}, respectively. Zirconium is precipitated as solid zirconium hydroxide, which is filtered off and washed with H\textsubscript{2}O and concentrated NH\textsubscript{4}OH, then ignited to ZrO\textsubscript{2} at 1000 °C or dissolved in 9M HCl and measured by spectrophotometry as the Arsenazo-III complex.\textsuperscript{8} Molybdenum and phosphorus contents were measured by spectrophotometry as the thiocyanate and molybdenum blue complexes.\textsuperscript{9,10}

**Preparation and elution of generator:** 2 g of ZrMP gel was added to a glass column (10 mm x 50 mm) fitted with sintered glass frit and containing nearly boiling distilled water. The column was washed with 100 ml of boiling distilled water followed by 100 ml saline. The column generator was now ready for elution of \textsuperscript{99m}Tc every 24 hours using 0.9% NaCl.

**Eluate analysis:** the \textsuperscript{99}Mo activity in the column was measured in a dose calibrator CRC-2N Capintec (C II) on the first day and calculated on subsequent days using decay