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Poly zirconium chloride (PZC) as an efficient adsorbent for ^{99}Mo

H. Salehi[1*], M. Zoghi[1] , M. Alimoradi[2], A. Niazi[2] and H. Abbasi[1]

1. Nuclear Science Research School, NSTRI, AEOI, Iran
2. Department of Chemistry, Faculty of Science, Arak Branch, Islamic Azad University, Arak, Iran

ABSTRACT

This radionuclide is almost exclusively produced from the decay of its parent ^{99}Mo. ^{99m}Tc is the most available and important nuclide used as a diagnostic agent in nuclear medicine. To develop a newly practical ^{99m}Tc generator using (n,γ) ^{99}Mo, an inorganic polymer adsorbent has been developed framed with oxygen-zirconium-chlorine bonds with a high adsorption of Mo, The polyzirconium adsorbent was named PZC (poly zirconium chloride) which made clear that PZC has an ability and high performance as a practical adsorbent for ^{99m}Tc generator using (n, γ) ^{99}Mo. The amount of ^{99}Mo adsorbed to PZC reached about 250mg/g PZC. ^{99m}Tc was eluted with a small volume of saline solution in the yield of about 76.43 %. The breakthrough of ^{99}Mo in the elution of ^{99m}Tc was less than 0.015%.

Keywords: ^{99}Mo / ^{99m}Tc generator; poly zirconium chloride; radiopharmaceutical; ^{99}Mo adsorbent

INTRODUCTION

^{99m}Tc labeled radiopharmaceuticals have extensive applications in diagnostic nuclear medicine. [1] This radionuclide is supplied in the form of a generator in which it is separated from the parent ^{99}Mo, most commonly by column chromatography, over alumina and less widely by solvent extraction or sublimation.[2,3] Generators based on column chromatography have several advantages over the other methods for use in hospital radiopharmacy. But chromatographic generators based on adsorption of molybdate on substrates like alumina have a limited capacity for molybdate. [4,5]

Gel generator of zirconium molybdate[6,7] which is applicable to ^{99}Mo obtained easily by (n, γ) reaction of natural Mo. Also the gel has been applied to a ^{188}W/^{188}Re generator. [8,9,10] The gel generator for ^{99}Mo has been investigated on the preparation and on the elution of ^{99m}Tc. Through these studies, several difficulties of the gel preparation have been presented, [11,12] the gel preparations depend on many complex factors[6,7] such as concentrations of Mo and Zr, and the reaction temperature of the gel, and eventually the eluted ^{99m}Tc is influenced by the preparations.

* . Corresponding author: E-mail: hsalehi@aeoi.org .ir

In the present study, the adsorbent PZC was synthesized [13] and performance of the PZC for a 99mTc generator has been examined mainly on two essential items; adsorbility of 99Mo obtained by (n,γ) reaction of natural Mo and elution of 99mTc from the generator prepared with 99Mo.

EXPERIMENTAL

Materials and methods

All chemicals were obtained from commercial sources and used without further purification.
Zirconium chloride (Merck), Isopropyl alcohol (Merck), Sodium hydroxide (Merck), Hydrochloric Acid (Merck), Arsenaso III (Merck), 99MoO3 obtained from Jaber Labs (NSTRI), Inductivity coupled plasma(ICP) Perkin-Elmer Optima 3000, Multichannel analyzer with HPGe detector (silena 2000). Beta counting system with GM detector (Nuclear Enterprises).
Synthesis of the adsorbent
PZC was prepared from Zirconium chloride and isopropyl alcohol;

$ZrCl_4$ + iso-$(CH_3)_2CHOH$ → $ZrCl_X$-{$OCH(CH_3)_2$}$_y$ --heating--> PZC

35 grams of Zirconium chloride were dissolved in 18.5 grams of isopropyl alcohol and solution was heated up to 90 c gradually and kept for 3hr. The water-soluble precursor was converted to the insoluble polymer by heating for 1 hr. at 160 c.

Adsorption of Mo to PZC

The Mo adsorption characteristics of PZC were examined with non-radioactive Mo. In this way aqueous solution of Mo (the pH was about 7) [14] was prepared from Na_2MoO_4 solution. One gram of PZC was added to the Mo solution of more than 10ml kept at a specific temperature (90c) [15] in an oil bath. The amounts of Mo adsorbed and Zr detached from PZC were determined by measuring the amounts of Mo remained in the solution using an inductive-coupled plasma emission spectrometer. To study the influence of the pH of the solution to the Mo adsorption capacity of PZC, the pH value of Mo solution before adsorption procedure was controlled by addition of 3.5 M NaOH solution or 3.5 M HCl solution. [Table.1]

Adsorption of ^{99}Mo to PZC

Adsorption of ^{99}Mo to PZC was examined using ^{99}Mo produced by (n,γ) reaction of ^{98}Mo, the obtained sample of $^{99}MoO_3$ was dissolved in 10ml of 3.5M NaOH solution , then diluted with water up to about 20ml. From the original solution, some portions including 150 to 300 mg of ^{99}Mo were taken, the acidities of the solutions neutralized to pH=7 with 3.5 M HCl, and the volumes adjusted to 10ml with water. One gram of PZC was added to each solution and the mixture kept standing at 90° C for 3hr in an oil bath. The amount of adsorbed ^{99}Mo was measured by comparing the 0.7395 MeV γ-ray peak of ^{99}Mo remaining in the solution with that of the original ^{99}Mo solution. A Gamma-XHPGe photon detector system with 2 KeV resolution from 1.8 KeV to 2 MeV, was used for radioactivity measurement.

Performance of the PZC for a ^{99m}Tc generator

After ^{99}Mo was adsorbed, each PZC was transferred and packed into a glass column of 10 mm inner diameter and 70 mm long equipped with a glass filter. The packed PZC was washed out with about 30ml of saline solution to remove the excess Mo and a slight amount of Zr released from PZC. With 10 ml of saline solution, ^{99m}Tc converted from ^{99}Mo was eluted from the column at one day after the PZC was packed in the glass column. Elution yield of ^{99m}Tc (Table.2) and the amount of ^{99}Mo detached were determined by measuring 0.1405 MeV γ-ray for ^{99m}Tc and 0.7395 MeV γ-ray for ^{99}Mo of both the effluents and the original ^{99}Mo solution. Notice that ^{99m}Tc elution yield was determined by comparing the generated amounts of ^{99m}Tc based on the radioactive equilibrium curve for ^{99m}Tc and ^{99}Mo, because ^{99m}Tc and ^{99}Mo in the column did not attain to the equilibrium in 1 day after the packing.

RESULTS AND DISCUSSION

According to the preliminary experiment on the adsorption of Mo to PZC, it is clear that ~250 mg of Mo can be adsorbed on 1 gr of PZC by keeping the mixture for 3hr at 90c. Based on these results, the following adsorption experiments were done under the conditions described above. Table.1 shows that the PZC can adsorb Mo almost quantitatively when about 250 mg of Mo was loaded on 1 gr of PZC and that the amount of Zr released from PZC was below the detection limit of 1×10^{-3} ppm, except in the case of less than100 mg of Mo in 10ml which amount of Zr released from PZC was higher than detection limit.

Table 1. Adsorption of Mo to PZC

Experiment No.	Mo loaded (mg)	Mo adsorbed (mg/g-PZC)	Zr released (ppm)
1	50	49.9	117
2	100	99.1	2.63
3	106	106	0.94
4	150	149.9	<0.001
5	205	204.8	<0.001
6	252	252	<0.001
7	256	252.1	<0.001

The amounts of ^{99}Mo adsorbed to 1 gram of PZC ranged from 136mg to 261.9mg, with an average of 230mg of ^{99}Mo loaded.

For the elution experiments one day after loading ^{99}Mo, the ^{99m}Tc yields are shown in the last column of Table2.

Figure 1 shows the ^{99m}Tc elution efficiency one day later and average elution yield of ^{99m}Tc was 76.43 %.

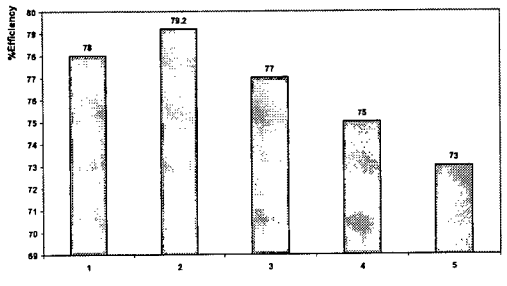

Fig. 1. ^{99m}Tc Efficiency in 5 elution days (Generator No. 4 of Table.2).

Figure 2 shows that molybdenum content was 0.005% which according to British Pharmacopoeia, [14] is less than 0.015%. So optimal parameters for ^{99}Mo adsorption and ^{99m}Tc generator column preparation were reaching the requirements of the international pharmacopoeia.

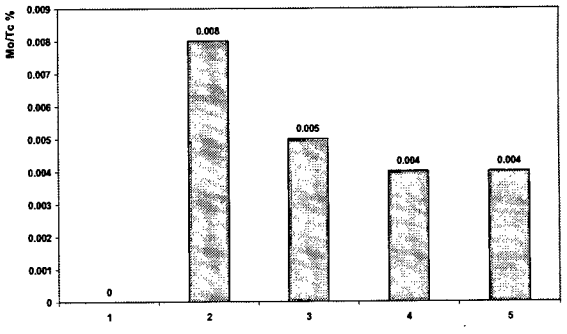

Fig.2. ^{99}Mo breakthrough in 5 elution days (Generator No. 4 of Table.1).

Figure 3 shows the elution volumes profile of ^{99m}Tc obtained from the generator. According to this figure 71.57% of ^{99m}Tc could be collected in the first 5ml of the effluent. The activity of technetium-99m can be eluted in a sterile vial.

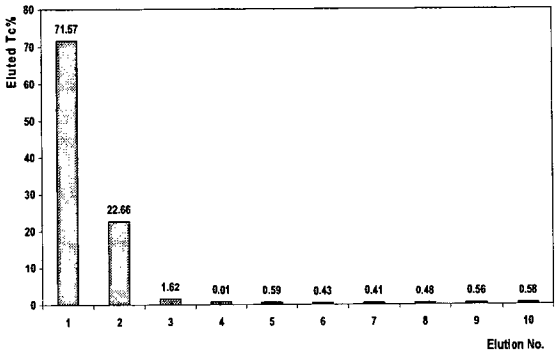

Fig.3. Elution curve of the generator in 5ml elution volume.

The radiochemical assays were performed by paper chromatography in saline. In this way radiochemical purity of the eluent was 95.78%. Also chemical purity was determined by colorimetery and radionuclide purity was measured by gamma-XHPGe. (Table 3)

Table 2. The amount of 99Mo adsorbed to 1 gr PZC

Experiment No.	^{99}Mo loaded (mg)	^{99}Mo Radioactivity (Bq)	^{99}Mo adsorbed (mg)	^{99m}Tc eluted (%)
1	150	4.81×10^{8}	136	80
2	180	4.81×10^{8}	169.2	80
3	200	5.07×10^{8}	187.9	79.5
4	250	5.07×10^{8}	241	76.4
5	260	5.07×10^{8}	232	78
6	290	6.58×10^{8}	256	72
7	300	6.58×10^{8}	261.9	69

Table 3. Quality control of 99mTc

parameters		**standard**	**measuring method**	**1**	**2**	**3**	**4**	**5**
	pH	5 to 7	pHmeter paper	5.5	5.5	5.5	5.5	5.5
Radionuclide Purity	^{99}Mo/^{99m}Tc%	$\leq1.5\times10^{-2}$%	curimeter	accept	accept	2×10^{-2}	accept	accept
	β Emmiters	$\leq1\times10^{-6}$%	scentilation counter	accept	accept	accept	accept	accept
	γ Emmiters except ^{99}Mo	$\leq1\times10^{-2}$%	γ spectrometer	accept	accept	accept	accept	accept
radiochemical purity	TcO_4^-	≥95%	chromatography	93.2	96.4	95.8	96.6	96.9
Chemical purity	Al	≤10 ppm	colorimetery	-	-	-	-	-
	Zr	≤5 ppm	colorimetery	-	-	-	-	-

CONCLUSION

The PZC offers an alternative to the gel technology. It is apparent that ^{99m}Tc generator can be prepared using a new inorganic polymer adsorbent and ^{99}Mo which produced by neutron irradiation of natural molybdenum trioxide powder.

Elution profile shows efficient elution and ^{99}Mo breakthrough when compared with commercially available Tc generators was acceptable.

The main parameters of PZC based ^{99m}Tc generator were stated as follows: Mo-adsorption capacity about 250 mg/g PZC and ^{99m}Tc elution yield higher than 76.43 % were found with PZC adsorbent and ^{99m}Tc can be eluted and collected in the first 5ml of the effluent. Radiochemical purity of ^{99m}Tc was 95.78% , and chemical purity and radionuclide purity were acceptable.

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