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Application of AnaLig resin for ^{99m}Tc separation from ^{100}Mo target irradiated in cyclotron

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HIGHLIGHTS

- Dissolution of metallic molybdenum in hydrogen peroxide.
- ^{99m}Tc production in cyclotron.
- Technetium adsorption on AnaLig Tc-02 resin.
- Technetium separation from access of molybdenum.

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ABSTRACT

The purpose of this study was the development of procedure for molybdenum metallic target processing after its irradiation in a cyclotron. As a first step the dissolution of molybdenum in various physical forms was investigated. The concentrations of NaOH and $(NH_4)_2CO_3$ allowing the highest sorption of Tc on AnaLig Tc-02 resin had been found. Based on these results the sintered irradiated Mo pellet was processed. The radionuclidic and radiochemical purities of separated Tc product were evaluated.

1. Introduction

The crisis of ^{99}Mo supply in 2008 prompted the search for alternative methods of ^{99m}Tc production. The use of particle accelerators widely used for PET isotopes production was a rather obvious choice. The ^{99m}Tc can be obtained in ^{100}Mo (p,2n) reaction with a moderate cross section for proton energy around 16 MeV (Khandaker et al., 2007; Lebeda and Pruszynski, 2010; Gagnon et al., 2011; Qaim et al., 2014). The ^{100}Mo with enrichment close to 100% is commercially available. The extensive research and developments works have been carried out in the last years which confirmed the suitability of this method for production of ^{99m}Tc at high radioactivity level. The works performed at Edmonton and Vancouver centers in Canada should be mentioned (Morley et al., 2012; Gagnon et al., 2012; McEwan et al., 2012; Schaffer et al., 2015; Bénard et al., 2014). International Atomic Energy Agency established the coordinated research program IAEA No 17419 (2012–2015) "Accelerator-based alternatives to non-HEU production of Mo-99/Tc-99m" in order to promote this technology in member states. We participated in this project among a number of teams from other countries performing studies on the molybdenum dissolution and ^{99m}Tc separation from molybdenum. In extraction of technetium from molybdenum target irradiated with a proton beam the time is a very important factor. The dissolution of molybdenum target followed by the technetium separation must be done in the shortest possible time. There is a large number of Tc separation methods from Mo as a macro component. Extensive review of these method can be found in (Dash et al., 2013). The most important ones are: sublimation, solvent extraction using MEK (methyl ethyl ketone), chromatography with variety of adsorbents and extraction chromatography with technetium specific resins (TEVA, ABEC, AnaLig Tc-02). The use of AnaLig Tc-02 resin for ^{99}Tc determination was described earlier by Paučová et al. (2012), giving the equivalent results to those when TEVA resin was applied. We performed earlier the separation of ^{99m}Tc from Mo using 3 different chromatographic methods: C-18 coated with PEG, Dowex-1 and AnaLig Tc-02 resins obtaining the highest separation yield with the last one (Wojdowska et al., 2015). The aim of our

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studies was the development of separation procedure giving the highest Tc recovery in the shortest time. In separation experiments we used ^{99}Mo, obtained from fission of ^{235}U in nuclear reactor, used for the production of ^{99}Mo/^{99m}Tc radionuclide generators. In order to dissolve the irradiated target in a short time and with relatively low volume of solution we performed the detailed studies using molybdenum targets of different physical form (metal foil, powder, pressed or pressed and sintered pellet). Finally we irradiated a sintered pellet of Mo enriched to 99.82% in ^{100}Mo and processed this pellet to obtain the purified ^{99m}Tc solution.

2. Experimental

2.1. Dissolution of Mo metal

Extensive studies were carried out on dissolution of natural molybdenum metal in various physical forms, as presented in Table 1. The experimental set-up consisted of a magnetic stirrer drive with a water bath on top of it. Dissolutions were carried out in the flat bottom flask of 100 mL volume, with a magnetic mixing element in it, which was immersed in water bath. The 30% hydrogen peroxide at ambient temperature was added first in 1 mL portions (up to about 3 mL) and then in 0.5 mL ones. A turbulent exothermic reaction with a gas release started. The set-up was heated to required temperature together with Mo material to be dissolved. The temperature inside the flask rose quickly above that of water bath. The solution was colorless at the beginning, changing to orange, orange-brown and a light yellow after complete dissolution. The color disappeared after addition of NaOH or $(NH_4)_2CO_3$ solutions. The physical form of Mo metal, mass of material used, volume of H_2O_2, starting temperature and time for complete dissolution are given in Table 1.

2.2. Sorption and recovery of pertechnetate

The Tc sorption yield on Analig Tc-02 resin was studied as a function of NaOH and $(NH_4)_2CO_3$ concentration of 1.05, 1.4, 2.8 and 4.2 M for the former and 0.5, 1.5 and 2.5 M for the latter. The solution containing sodium molybdate and about 1 GBq of $Na^{99m}TcO_4$ obtained from ^{99}Mo/^{99m}Tc radionuclide generator (POLATOM, Poland) was introduced on the polypropylene (PP) column of 5 mm diameter and 15 mm height filled with AnaLig Tc-02 resin, which was prior activated using the respective NaOH or $(NH_4)_2CO_3$ solution. The concentration of Na_2MoO_4 was 80 mg Mo/mL for NaOH and 5 mg Mo/mL for $(NH_4)_2CO_3$, respectively. The 5 mg Mo/mL concentration in $(NH_4)_2CO_3$ solution was selected due to the limited solubility of molybdate in this solution. After sorption, the columns were rinsed with 3 mL of sodium hydroxide or ammonium carbonate solutions of the concentration as used for sorption and then with 1.5 mL of water. The flow rate was 0.4 mL/min. This flow rate was selected as a result of a number of trials which gave the highest Tc retention in the shortest time. The effluents from the columns were collected separately: during the loading of Mo solution, after rinsing columns in turn with NaOH or $(NH_4)_2CO_3$ solution and after rinsing the columns with water. The activity of ^{99m}Tc in the collected solutions was measured with the dose calibrator Capintec CRC-55tR (Capintec Inc., US). For each concentration of NaOH or $(NH_4)_2CO_3$ the separations and measurements were carried out in triplicate. All solutions were fed into the columns with peristaltic pumps and delivered for measurements in penicillin vials. The results are shown in Tables 2 and 3 as mean value with standard deviation calculated from 3 measurements. In the first row the loss of ^{99m}Tc due to incomplete adsorption of Tc on the column is shown. Then losses in washing with respective solutions and water are presented followed by the percentage of ^{99m}Tc not recovered and recovered from the columns.

Table 1
Characteristics of dissolution process of various Mo materials.

Mo form	Mo mass [g]	Volume of 30% H_2O_2 [mL]	Temperature [C]	Time to complete dissolution [min]
Powder	0.50	4.5	RT	5
Powder	1.12	10	RT	2
Foil 1 mm	0.51	6	60	15
Foil 1 mm	0.50	6	80	5
Foil 1 mm	0.53	6	90	2
Pellet	0.72	10	80	3
Pellet	0.76	10	90	2
Pellet sintered	0.72	10	80	60
Pellet sintered	0.74	10	90	45

2.3. Separation of cyclotron-produced ^{99m}Tc

Molybdenum target was prepared by pressing metal powder containing 99.82% of ^{100}Mo into the pellet of 12 mm diameter and 0.723 g mass. After sintering in hydrogen atmosphere at 1600 °C for 60 min, the pellet was mounted into aluminum holder which was fixed in GE PET-trace 840 cyclotron (at Heavy Ion Laboratory of University of Warsaw) proton path and irradiated for 2.5 h in 2.5 μA current to the total activity of 1.6 GBq at EOB (Table 4). After irradiation the target in holder was automatically released into the transportation container and shipped to our laboratory at POLATOM for chemical processing.

After releasing from holder the target was dissolved in 15 mL of 30% H_2O_2 at 90 °C in 60 min and alkalized with 10 M NaOH to final NaOH concentration of not less than 2 M in the total volume of about 20 mL. The ^{99m}Tc was separated in 3 columns connected in series containing AnaLig Tc-02, Dionex in H^+ form (On-Guard II H; Dionex) and Alumina A cartridge (Waters) (Fig. 1). The latter two minicolumns were activated with 5 mL of H_2O before use. The solution consisting of ^{100}Mo and ^{99m}Tc in 2 M NaOH was loaded on the first column containing 100 mg of AnaLig-TC-02 resin at the flow rate 0.4 mL/min. The non-adsorbed ^{100}Mo and radioactive impurities other than Tc were collected from this column in the waste vessel. The AnaLig-TC-02 column was then rinsed with 3 mL of 2 M NaOH and 2 mL of water. Finally, the ^{99m}Tc was eluted in 5 mL of water and was further passed through the cation exchange column where it was neutralized and then the ^{99m}Tc was trapped on a small Alumina A cartridge. The

Table 2
Distribution of ^{99m}Tc activity in sorption, rinsing and elution processes.

^{99m}Tc in fraction [%]	NaOH concentration [M]			
	1.05	1.4	2.8	4.2
Loading	1.59 ± 0.46	1.41 ± 0.27	1.25 ± 1.16	0.48 ± 0.18
Rinsing with NaOH	1.53 ± 0.32	2.09 ± 0.90	0.64 ± 0.37	0.25 ± 0.11
Rinsing with water	5.61 ± 2.12	4.57 ± 0.85	1.56 ± 0.64	1.29 ± 0.62
Elution with water	90.75 ± 1.18	91.31 ± 1.77	95.95 ± 2.00	96.72 ± 0.81
Residue on column	0.52 ± 0.18	0.62 ± 0.09	0.60 ± 0.20	1.26 ± 0.55

Table 3
Distribution of ^{99m}Tc activity in sorption, rinsing and elution processes.

^{99m}Tc in fraction [%]	$(NH_4)_2CO_3$ concentration [M]		
	0.5	1.5	2.5
Loading	3.21 ± 2.20	0.29 ± 0.14	0.20 ± 0.07
Rinsing with $(NH_4)_2CO_3$	2.41 ± 0.36	0.19 ± 0.03	0.19 ± 0.09
Rinsing with water	6.06 ± 3.64	1.15 ± 0.68	10.04 ± 1.82
Elution with water	86.17 ± 4.25	94.47 ± 0.37	88.36 ± 2.06
Residue on column	2.16 ± 0.20	3.90 ± 0.77	1.21 ± 0.35