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RADIONUCLIDIC GENERATORS FOR THE PRODUCTION OF TECHNETIUM-99M AND RHENIUM-188

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Abstract. *The paper deals with production and application of open radioactive sources in modern diagnostic and therapeutic nuclear medicine. In principle, besides adequate physical, chemical and biological properties a reliable route for the routine production of both radionuclide and radiopharmaceutical should be on disposal. The favorite route are radionuclidic generators as they enable simple production of shorter-lived radionuclides at relatively low costs. The main radionuclide in diagnosis is technetium-99m. Its advantages are favorable nuclear properties, availability of large activities of high quality ^{99m}Tc at reasonable prices by using (n,f)^{99}Mo/^{99m}Tc generators and versatile chemistry. It is used in the investigation of practically all organs and tissues in the human body. Among the beta-emitters the radionuclides of rhenium have been recently proposed as suitable candidates for therapeutic applications. The most important is ^{188}Re which is obtained from ^{188}W/^{188}Re generator. Some of the important radiopharmaceuticals labeled with rhenium radionuclides used for certain specific indications are also shown. The activities performed in the Laboratory for Radioisotopes (Vinča Institute of Nuclear Sciences) are also enclosed.*

Key words: *technetium-99m, rhenium-186, rhenium-188, diagnosis, therapy, nuclear medicine*

1. INTRODUCTION

In the last century radioactivity found its application in life sciences, especially in medicine. Despite the increasing number of non-radioactive tracer methods developed in the last decades, radionuclides and radiopharmaceuticals remain an indispensable tool in many procedures in nuclear medical diagnosis and therapy. They exhibit high sensitivity combined with the ease of detection from outside of a closed system, such as the human body. An additional advantage is that only extremely small concentrations are needed which do not cause any farmacodynamic effect.

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In diagnosis the dominant radionuclide is ^{99m}Tc. It is estimated that it is involved in about 80% of all imaging procedures in nuclear medicine. Due to its relatively short half-life (6.02 hours) it is routinely produced in ^{99}Mo/^{99m}Tc generators. The main is the chromatographic type based on ^{99}Mo produced by uranium fission. Due to the inherent disadvantages of the production of (n,f)^{99}Mo, alternative technologies based on ^{99}Mo obtained by (n,γ) nuclear reaction are investigated.

Once separated from its parent ^{99}Mo, technetium can be, in the form of sodium pertechnetate ($Na^{99m}TcO_4$) directly applied. However, it is mainly used for the preparation of radiopharmaceuticals targeting different organs and tissues in the human body. For the routine use the so-called instant kit technology of the production of radiopharmaceuticals was developed. Numerous reviews covering particular aspects of the production of ^{99m}Tc and its use in nuclear medicine can be found in literature [1].

Generally, the endotherapy is a matter of compromise. There is no ideal radionuclide like ^{99m}Tc in diagnosis. The therapeutic effect depends not only on the nuclear, chemical and biological properties of a given radionuclide or radiopharmaceutical. Very significant factors are also the nature and the localization of the pathological process. Although it is unlikely that such an ideal radinuclide like ^{99m}Tc will emerge, the application of radionuclides in therapy gained renewed interest in the decade. A significant progress has been achieved in tumor targeting and therapy control. Besides the well known ^{32}P, ^{89}Sr and ^{131}I, in consideration are new radionuclides obtained either in reactor or in cyclotron.

Rhenium radionuclides, particularly ^{188}Re, are very promising candidates. Rhenium-188 is sometimes denoted as the radionuclide of the future due to its favorable physical properties and relatively easy production from ^{188}W/^{188}Re generator. A further advantage is chemical similarity of rhenium and ^{99m}Tc. As the chemistry of technetium is well elaborated it can be expected that many of general approaches available for labeling agents with ^{99m}Tc can be adapted for use with rhenium.

The paper covers the activieties and the results gained during the develoment of ^{99}Mo/^{99m}Tc generator in the Laboratory for Radioisotopes (Vinča Institute of Nuclear Sciences). Shown are some of the radiopharmaceuticals labeled with ^{99m}Tc. In nuclear medicine there is a significant interest in the application of ^{188}Re in therapy. Preliminary experiences with ^{188}W/^{188}Re generator system are mentioned. Some of the ^{186,188}Re-radiopharmaceuticals for specific applications are also shown.

2. TECHNETIUM-99m IN DIAGNOSIS

2.1. Production of molybdenum-99

To meet the ever-growing demand for ^{99m}Tc a procedure for the production of sufficient activities of the parent radionuclide ^{99}Mo should be available. Molybdenum-99 is produced in nuclear reactor. By nuclear reaction ^{98}Mo(n,γ)^{99}Mo the so-called (n,γ)^{99}Mo is obtained. The advantages are relatively cheap target material (metallic molybdenum or oxide in natural abundance or enriched in ^{98}Mo), simple radiochemical treatment of the irradiated target and production of low quantities of low-activity waste. The main disadvantage is low specific activity (≤ 400 GBq/g Mo). By neutron-induced uranium fission in nuclear reactor the so-called (n,f)^{99}Mo is produced. The main advantages of this nuclear reaction are large activities and very high specific activities of the obtained ^{99}Mo. However, there are some principal dis-

advantages, which have to be carefully considered. They are: economical (high costs of the target material- uranium enriched up to nearly 100% in ^{235}U, complex and expensive separation of ^{99}Mo and recovery of the not reacted uranium), ecological (generation of large quantities of high activity waste) and political (target material has the potential for the clandestine use in nuclear weapons).

2.2. Molybdenum-99/technetium-99m generators

In principle, the separation of ^{99m}Tc from its parent ^{99}Mo can be carried out by two aqueous methods – sorption and extraction – and by one dry method – sublimation. The kinetics of the pair ^{99}Mo-^{99m}Tc enables the accumulation of new activity of the daughter after each separation. The devices in which the separation is achieved are named generators.

The procedures for the separation of ^{99m}Tc and ^{99}Mo are shown in Table 1. The chromatographic type, based on $(n,f)^{99}Mo$ adsorbed on alumina, is the main source of ^{99m}Tc. It provides high quality ^{99m}Tc-eluates of large activities and radioactive concentrations. They are also simple and safe for handling.

The inherent problems with the production of $(n,f)^{99}Mo$ prompted the search for alternatives. Many attempts were made to develop the so-called alternative technologies based on $(n,\gamma)^{99}Mo$. They exploit differences in the volatility of technetium and molybdenum oxides (sublimation generators). The separation can be achieved by extraction, usually by using methyl ethyl ketone (extraction generator). The gel generator combines the performances of the chromatographic generator and the use of $(n,\gamma)^{99}Mo$. The gel is zirconium or titanium molybdate, which is, after the activation in the nuclear reactor, packed into the generator column. The content of molybdenum in the column is thus raised up to 40 wt%, in comparison with only 0.2 wt% in the generators based on alumina. By elution of these generators larger activities of ^{99m}Tc are obtained [2,3].

Table 1. Methods of separation of ^{99m}Tc from ^{99}Mo, characteristics of the generator operation and their perspectives [2]

Separation of ^{99}Mo and ^{99m}Tc	^{99}Mo	Separation efficiency of ^{99m}Tc(%)	Ease of generator operation	Prospect
Chromatography	(n,f)	80-95	Simple, portable	Yes
Chromatography	(n,γ)	80-95	Simple, portable	No
Extraction	(n,γ)	50-75	Complex, fixed	No
Sublimation				
High temp.	(n,γ)	30	Complex, fixed	No
Low temp.	(n,γ)	50	Complex, portable	Yes?
Gel	(n,γ)	80-95	Simple, portable	Yes

The common comment can be summarized as follows. The chromatographic generator based on $(n,f)^{99}Mo$ is due to its excellent performances predominant. Chromatographic generators based on $(n,\gamma)^{99}Mo$ vanished because they were restricted only to low activities and low radioactive concentrations of ^{99m}Tc. Extraction and high temperature sublimation generators also employing $(n,\gamma)^{99}Mo$ are already abandoned due to the complexity of the devices and poor results. Probably the best perspective offers the gel generator. However, without some essential innovation, its future is uncertain, too.

According to experience they could only be used as the source of so-called instant ^{99m}Tc. This means that ^{99m}Tc, obtained by one of the alternative procedures, is divided into portions and shipped to the users. However, this mode of the supply has several disadvantages (loss on activity, time, etc) [2].

The chromatographic ^{99}Mo/^{99m}Tc generator based on (n,f)^{99}Mo is produced also in the Laboratory for Radioisotopes (Vinča Institute of Nuclear Sciences). The technology is developed in the Laboratory and only (n,f)^{99}Mo is imported (Nordion, Canada). The plastic column is with stainless steel and silicone tubing connected into the closed elution system. The eluence is 0.9% NaCl which is in portions of 3, 5 and 10 ml passed through the column by the action of vacuum. The generator operates in the so-called dry mode. The generators are calibrated up to six days in advance so that they can be transported to the users distant from the production cite. The activities are 3.7, 7.4, 11.1 and 18.5 GBq at the time of calibration [4].

Intense investigations were performed during the development and production of these generators [5]. Two basic requirements are: high elution yield of ^{99m}Tc and low breakthrough of ^{99}Mo. It was observed that under certain experimental conditions the generator demonstrates decreased yield of ^{99m}Tc. It was found that the best results are obtained by modification of alumina layer with of divalent copper. Its content should be about 365 μg/g alumina [6].

2.3 Technetium-99m in diagnosis

Technetium-99m is from the generator column eluted in the form of sodium pertechnetate, $Na^{99m}TcO_4$ which is mainly used for labeling compounds for the production of radiopharmaceuticals. The first step is usually the reduction of nonreactive ^{99m}Tc(VII) to lower valence states - ^{99m}Tc(III), ^{99m}Tc(IV) or ^{99m}Tc(V). In dependance on the experimental conditions ^{99m}Tc is then bound to numerous chelate compounds. In the past decades a great number of coordinated chelate compounds have been developed and are now routinely used in the nuclear medical practice. Particularly important contribution to the broad use of ^{99m}Tc-radiopharmaceuticals was the development of the preparations in the kit form ("cold kits").

The production of ^{99m}Tc-radiopharmaceuticals in the Laboratory for Radioisotopes (Vinča Institute of Nuclear Sciences) started almost thirty years ago. A significant number of labeled preparations were developed. The comprehensive review can be found in ref. [1].

Table 2 reveals some ^{99m}Tc radiopharmaceuticals, indications for their use and the activities applied in the investigations [1,7].

Table 2. Indications for the application of some ^{99m}Tc-radiopharmaceuticals, their chemical forms and maximum activity per test, Adapted from refs [1,7]

Indication	Chemical form of the radiopharmaceutical	Max. act. per test (MBq)
Brain imaging (static)	Pertechnetate (TcO_4^-)	500
Brain imaging (SPECT)	TcO_4^-, DTPA, gluconate, glucoheptonate, HM-PAO	500
Cerebral brain flow	HM-PAO	800
Thyroid imaging	TcO_4^-	200
Radionuclide ventriculography	Labeled red blood cells	1110
Myocardial imaging (SPECT)	Isonitriles	300
First pass blood flow studies	DTPA, TcO_4	800
Blood pool imaging	Human albumin complex	40

Indication	Chemical form of the radiopharmaceutical	Max. act. per test (MBq)
Lung ventilation imaging	DTPA-aerosol	80
Lung perfusion imaging (with venography)	Human albumine (macroaggregates or microspheres)	160
Lung imaging (SPECT)	Macroaggregated albumin (MAA)	200
Liver and spleen imaging	Labeled colloid	80
Functional biliary system imaging	Iminodiacetates and equivalent agents	150
Spleen imaging	Labeled denaturated red blood cells	100
Liver imaging (SPECT)	Labeled colloid	200
Renal imaging (static)	DMSA	160
Renal imaging/renography	DTPA, gluconate, glucoheptonate, PAH, MAG_3	350
Bone imaging (planar)	PyP, MDP, DPD	600
Bone imaging (SPECT)	PyP, MDP, DPD	800
Detection of inflammatory bowel diseases	Sucralphate	200
Salivary gland imaging	TcO_4^-	40
Gastrointestinal bleeding	Labeled colloid	400
Oesophagal transit and reflux	Labeled colloid	40
Bacterial infection	Ciprofloxacin	555
Tumor imaging	DMS (V)	400

SPECT- single photon emission computerized tomography; DTPA- diethylenetriamine pentaacetic acid, HM-PAO - hexamethyl propyleneamine oxime, MAA - macroaggregated albumin, DMSA - dimercaptosuccinic acid, PAH - p-aminohippurate, MAG_3 - mercaptoacetyl triglycine, PyP - pyrophosphate; MDP - methylene diphosphonate; DPD - dicarboxypropane diphosphonate

Together with the production of generators and radiopharmaceuticals the modern quality control procedures, radiation protection measures, etc, were included and developed [8].

3. RADIONUCLIDES IN THERAPY

In contemporary nuclear medicine alpha-, beta- or beta-/gamma- emitters can be used for targeted therapy. The achievements in oncology, endocrinology, rheumatology and, a short while ago, in interventional cardiology, has refined the role of radionuclidic therapy as an important alternative to more common therapeutic regimens. Two radionuclides of rhenium - ^{186}Re and ^{188}Re, are of particular interest. Their physical characteristics are shown in Table 3 [9].

Table 3. Physical characteristics of ^{186}Re and ^{188}Re

Radionuclide	$T_{1/2}$	E_β(max), MeV	E_γ ,MeV,Yield (%)
Re-186	90 h	1,07	0,137 (12)
Re-188	16,9 h	1,95; 2,11	0,155 (25)

3.1 Production of rhenium-186 and rhenium-188

Rhenium-186 is routinely produced in nuclear reactor by using ^{185}Re(n,γ)^{186}Re reaction. The target is usually metallic rhenium with ^{185}Re in natural abundance (37.07%) or enriched in ^{185}Re up to 96%. Rhenium-186 decays to stable ^{186}Os.

For the production of ^{188}Re two reactions in nuclear reactor can be employed. In ^{187}Re(n,γ)^{188}Re reaction the target is metallic rhenium or oxide in natural abundance or enriched in ^{187}Re. Due to the high costs of the enriched target material this reaction has no importance for the routine production.

Rhenium-188 has a significant advantage as it can be obtained by using the generator system ^{188}W/^{188}Re. Thus the known benefits of the generators can be used. The parent radionuclide ^{188}W, formed by the double neutron capture on ^{186}W, by β-decay produces ^{188}Re:

$$^{186}\mathrm{W(n,\gamma)}^{187}\mathrm{W(n,\gamma)} \rightarrow {}^{188}\mathrm{Re}$$

3.2 Tungsten-188/rhenium-188 generator

The first generators contained ion exchange resins with adsorbed ^{188}W. The generated ^{188}Re was eluted by HCl or $HClO_4$ [10]. The mayor drawback was low resistance of the resin against radiation.

Due to the chemical similarity to the ^{99}Mo/^{99m}Tc pair, most efforts have been focused on an alumina-based system. Tungsten is adsorbed on alumina as WO_4^- and ^{188}Re is, as perrhenate, preferably eluted with NaCl solution.

As in the case of ^{99}Mo/^{99m}Tc generator the choice of the process for the separation of the daughter radionuclide, i.e., which type of the generator will be produced, depends on the specific activity of the parent ^{188}W. It depends on the characteristics of the reactor i.e. on its neutron flux. The dependence of the specific activity of ^{188}W on the neutron flux is given in Table 4 [11].

Table 4. Dependence of the specific activity of ^{188}W on the neutron flux [11]

Nuclear reaction	Target; Enrich. (%)	Flux (n cm^2 s)	Irradiat.time (h)	Spec.act. (GBq/mg)
^{186}W(2n,γ)	WO_3 (96%)	10^{14}	500	22.2
Generator		10^{15}	5000	222

Due to the double neutron capture process and the rather long ^{188}W half-life, relatively long irradiation periods are required. After irradiation the metallic target is usually processed by oxidation with hydrogen peroxide and/or hypochlorite in the presence of base. The oxide targets are dissolved in base with concomitant oxidation [12].

For the chromatographic generator ^{188}W of high specific activity is essential. The neutron flux should be about 10^{15}n m^2 s. A typical generator of 18.5 GBq consists of the column with 5-6 g of alumina. Elution is performed by saline. The bolus volume is about 10-12 ml. The principal radioactive concentration is 1110-1480 MBq/ml [12,13].

The radioactive concentration of ^{188}Re in the eluate decreases with time. To prolong the shelf-life of the generator several attempts were made for the concentration of ^{188}Re in the generator eluate. The procedure developed recently is based on the incorporation of the disposable tandem cation-exchange/anion-exchange columns. As the result ^{188}Re activity is obtained in a very low volume of 0.9% NaCl (<0.1 ml) [11,12].

In principle, the use of rhenium radionuclides depends on their specific volume. If it is low rhenium-186,188 can be used only for labeling of particles or diphosphonates. However, in the case of peptides or antibodies ^{188}Re of high specific volume is needed.

The separation of ^{188}Re from ^{188}W of low specific activity is performed, similar as in the case of (n,γ)^{99}Mo, by the alternative procedures like, e.g. gel technology. These procedures are also in this case more convenient for the production of "instant" ^{188}Re.

Preliminary experiments in the Laboratory for Radioisotopes show that the adsorption coefficients of alumina for tungstate and perrhenate anions differ, in dependence on the experimental conditions. The investigations in the pH region of interest (pH = 4-6) and concentrations of the eluence (0.9-1.2% NaCl) are in progress.

3.3 Rhenium-186 and rhenium-188 in therapy

There are several fields of applications of the radiopharmaceuticals labeled with ^{186,188}Re. For the bone pain palliation the most often used are ^{186}Re-HEDP and ^{188}Re-DMSA. For the synovectomy, ^{186}Re-sulphide in the kit form is already commercially available. The endovacular radiation therapy is performed by using ^{188}Re-perrhenate or ^{188}Re-MAG_3. The labeling of peptides and antibodies with ^{188}Re is also reported [14-16].

The main indications for the application of radiopharmaceuticals labeled with radionuclides of rhenium are given in Table 5.

Table 5. Application of the radiopharmaceuticals labeled with ^{186}Re and ^{188}Re; adapted from refs. [9,14-16]

Field of application	Radiopharmaceutical
Bone pain palliation	^{186,188}Re-HEDP, ^{188}Re-DMSA
Synovectomy	^{186}Re-sulphide
Endovascular radiation therapy	^{188}Re-perrhenate, ^{188}Re-MAG3
Tumor therapy	^{188}Re-peptides
Endoradiotherapy of tumors - catheter administration	^{188}Re - particles
Marrow ablation prior to stem cell rescue	^{188}Re- antigranulocyte antibodies

Hydroxyethane diphosphonate, DMSA-dimercaptosuccinic acid, MAG_3- mercaptoacetyl triglycine

4. CONCLUSION

Technetium-99m remains the dominant radionuclide in diagnosis in nuclear medicine. The main source is ^{99}Mo/^{99m}Tc generator based on ^{99}Mo obtained by uranium fission. These generators are due to their performances some kind of the golden standard that any other technology of ^{99m}Tc production must match in order to be recognized as a viable alternative. This type of generators based on the technology developed in the Laboratory for Radioisotopes and imported (n,f)^{99}Mo is regularly produced also in the Vinča Institute of Nuclear Sciences. The problem of low elution yield was solved by introduction of Cu(II) into the alumina adsorbent.

Due to its favorable nuclear properties ^{188}Re was proposed as a suitable candidate for therapeutic applications. A further advantage comes from the fact that scintigraphy can also be performed by using the associate gamma-emission. It is expected that wide use of ^{188}Re could be ensured with the development of the chromatographic ^{188}W/^{188}Re generator and the import of ^{188}W of high specific activity. From the chemical point of view, an important feature lies also in the similarity between the chemical behavior of rhenium and technetium. So experience gained with ^{99}Mo/^{99m}Tc generator and ^{99m}Tc-radiopharmaceuticals can be ex-

ploited also in the development of $^{188}W/^{188}Re$ generator and ^{188}Re-radiopharmaceuticals. The preliminary experiments in the investigations of the adsorption of tungsten and rhenium on alumina show that an efficient separation of ^{188}Re from ^{188}W could be achieved. The eluence is 1-2% NaCl solution.

Alternative technologies based on extraction or gel technology are also considered. This form of supply is in the case of therapy more appropriate than in diagnosis. Rhenium-188 is relatively long-lived and therapy is more easily to plan so the user can order activity in advance. Skilled personnel and appropriate equipment necessary for the manipulation with beta-emitters are available in the central laboratories. However, these procedures are, as in the case of ^{99m}Tc, more suitable for the production of "instant" rhenium.

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RADIONUKLIDNI GENERATORI ZA DOBIJANJE TEHNECIJUMA-99M I RENIJUMA-188

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Prikazani su neki od najvažnijih aspekata proizvodnje i primene otvorenih izvora radioaktivnog zračenja u dijagnostičkoj i terapijskoj nuklearnoj medicini. Pored fizičkih, bitne su i hemijske i biohemijske osobine datog radionuklida odnosno radiofarmaceutika. Uslov za redovnu primenu su i dostupnost, kvalitet i cena. Radionuklidni generatori predstavljaju vrlo pogodan izvor dobijanja radionuklida kraćih vremena poluraspada. Najvažniji dijagnostički radionuklid je ^{99m}Tc. Odlične fizičke osobine, visok kvalitet i laka dostupnost po prihvatljivim cenama korišćenjem molibden-99/tehnecijum-99m generatora i razvijena hemija doveli su do toga da ovaj radionuklid postane praktično nezamenjiv u ispitivanju skoro svih organa i tkiva u ljudskom organizmu. Dati su savremeni trendovi u razvoju proizvodnje radionuklida i radiofarmaceutika za primenu u terapijske svrhe. Prikazani su načini dobijanja radionuklida renijuma (^{186}Re i ^{188}Re) sa posebnim naglaskom na ^{188}W/^{188}Re generatoru. Dati su hemijski oblici i indikacije za primenu nekih radioaktivnih lekova obeleženih ovim radionuklidima. Prikazana su istraživanja sprovedena u Laboratoriji za radioizotope (Institut za nuklearne nauke «Vinča»).

Ključne reči: *tehnecijum-99m, renijum-186, renijum-188, nuklearna medicina, dijagnostika, terapija*