# A secular technetium–molybdenum generator

Wagner L. Araujo, Tarcisio P.R. Campos

*Departamento de Engenharia Nuclear, Universidade Federal de Minas Gerais, Belo Horizonte, 31.270-901, Minas Gerais, Brazil*

## ARTICLE INFO

## ABSTRACT

A compact secular molybdenium generator is subject of this paper. This generator represents a nuclear system that comprises a hydrogen-isotopes fusor, moderator, reflector and shield. Deuterium fusion reactions in a tritiated or deuterated target provide the neutron source. A moderation fluid slowdown the neutron energy which increases $^{98}Mo(n,\gamma)^{99}Mo$ capture reaction rates. Neutron reflection minimizes the neutron escape and the radiation shield encloses the device. The neutron yield calculation along with electromagnetic and nuclear simulations were addressed. Results revealed the accelerator equipotential surfaces ranging from $-30$ to 150 kV, the ion trajectories and the energy beam profile define a deuteron current of 1 A with energy of 180 keV at the target, the spatial distribution of the neutron flux, and the $^{99}Mo$ and $^{99m}Tc$ activities in function of transmuter operation time. The kinetics of the $^{99m}Tc$ correlated to its precursor activity demonstrates a secular equilibrium providing 2 Ci in a operational time of 150 h. As conclusion, the investigated nuclear and electromagnetic features have demonstrated that such generator shall have a notable potential for feeding the $^{99m}Tc$ clinical application.

© 2015 Elsevier B.V. All rights reserved.

## 1. Introduction

Radiopharmaceuticals based on $^{99m}Tc$ become, over the last 50 years, important tools in the diagnosis of various diseases and organ disorders. The main reasons of such success is based on its prompt availability by a radionuclide generator associated with its plurality of chemical valences and relative low cost [1]. Several complexes labeled with technetium turn over to diagnosis routine in nuclear medicine, yielding 80% of the clinical protocols [2–5]. The $^{99m}Tc$ radionuclide is generally produced by $^{99}Mo$ decayment enclosed in a column inside a radionuclide generator. The column is filled with adsorbed material made of alumina or zirconia, in which the $^{99}Mo$ irradiated isotope was adsorbed. The $^{99m}Tc$ daughter radionuclide grows as the result of the decayment of an initial mass of $^{99}Mo$ radionuclide until a transient equilibrium is reached. The activity of the daughter is eluted by a saline solvent, leaving the residual $^{99}Mo$ radionuclide and the $^{98}Mo$–Al or Zr columns. Ideal chemical and physical $^{99m}Tc$ properties, like physical half-life of 6.01 h, decay by isomeric transition, emission of a 140 keV gamma ray, quickly elution from a $^{99}Mo/^{99m}Tc$ generator system and the diversity of oxidation states with possible large number of coordination links held by the $^{99m}Tc$ ion giving rise to different radiopharmaceuticals based on a simple reconstitution of lyophilized reagent sets, justify its high rate of application. Modern portable gel-column generators have been produced [6–8]. In these devices, the radiochemical purity

of $^{99m}Tc$ is higher than 99%, and the final product contains less than 0.02%) of $^{99}Mo$. Generally the technology for preparing radioactive columns for radionuclide generators, such as $^{99}Mo/^{99m}Tc$, involves large and complex installations.

The $^{99}Mo$ can be generated by neutron activation of $^{98}Mo$ in a fission reactor, based on the $^{98}Mo$ neutron capture reaction. However, the fission route in nuclear reactor is the common choice to the commercial production of $^{99}Mo$. Two types of fission target are often used: high-enriched uranium (HEU) and low-enriched uranium (LEU), based on aluminate dispersion targets, are used. $^{99}Mo$, $^{133}Xe$ and $^{131}I$ are all produced together. Another possibility is $^{99}Mo$ production based on nuclear reactor, operating in low power, whose fuel is a uranium solution. In this case, the $^{235}U$ target are not solid as HEU or LEU-targets, but in a homogeneous fluid fuel. Thus, during the operation, $^{99}Mo$ is produced and removed continually [9–12]. Fig. 1 provides the microscopic cross-sections of the neutron nuclear reactions addressed to produced $^{99}Mo$.

Proton accelerators can also be used to produce $^{99}Mo$ and $^{99m}Tc$. It is produced by $^{100}Mo(p,2n)^{99m}Tc$ reaction. A proton beam of 20 MeV accelerated in a isochronal cyclotron can bombard a molybdenum metallic target, which can be highly enriched in $^{100}Mo$ ($> 99\%$)). Indeed, this type of device has been applied to produce $^{18}F$, $^{15}O$, $^{11}C$, $^{13}N$ and now $^{99m}Tc$ through the $^{100}Mo(p,2n)^{99m}Tc$ reaction. An estimated activity is about 16 Ci mA$^{-1}$h$^{-1}$(Ci of $^{99m}Tc$ EOB) at 24 MeV [9]. The corresponding saturated $^{99m}Tc$ activity is 141 Ci mA$^{-1}$(Ci of $^{99m}Tc$ EOB) for a beam of protons (6 mm diameter) with 25 MeV and metallic Mo targets (density of 10.3 g cm$^{-3}$), and a total stopping power of about 13 MeV mm$^{-1}$ [9].
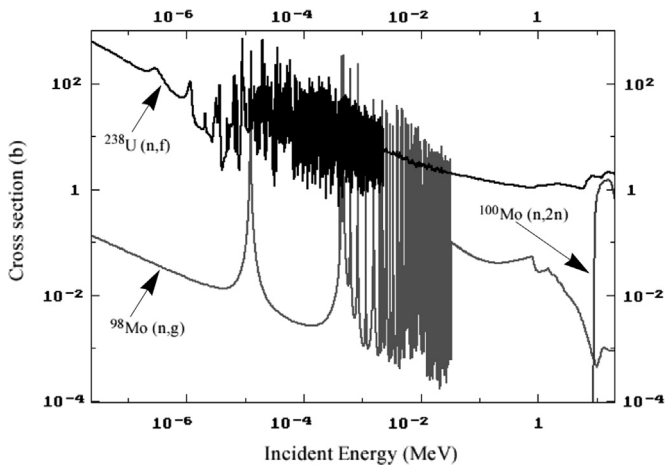
*E-mail addresses:* wagnerleite@ufmg.br (W.L. Araujo),
tprcampos@pq.cnpq.br (T.P.R. Campos).

Fig. 1. Microscopic cross-sections of $^{100}$Mo(p,2n) $^{99m}$Tc, $^{100}$Mo(n, 2n),$^{99}$Mo, $^{98}$Mo(n,g) $^{99}$Mo and $^{235}$U fission [13].

The $^{99}$Mo can also be production by photo fission route. Two reactions can be used based on U or Mo target, as $^{100}$Mo($\gamma$,n) $^{99}$Mo or $^{238}$U($\gamma$,f) $^{99}$Mo. In this case, intense photon flux can be produced by linear electron accelerators [9].

Spallation neutron sources provide high intense neutron flux, whose neutron fission can be used to produce $^{99}$Mo. Particularly , $^{100}$Mo(n, 2n) $^{99}$Mo reaction are of interest. The cross-section of $^{100}$Mo(n,2n) $^{99}$Mo reaction is close to 1.5 b at the 10–17 MeV energy interval. A 40 MeV deuteron beam bombarding a natural carbon converter can generate intense 14 MeV neutrons, which can be interact via $^{100}$Mo(n,2n) $^{99}$Mo reaction [9]. The spallation neutron sources can also support the $^{235}$U(n,f) $^{99}$Mo reactions [14].

In summary, uranium fission in research reactors (HEU and LEU-alumine dispersion targets), liquid-fuel reactor technology and neutron activation of $^{98}$Mo in nuclear reactor are based on various technological scenarios together with direct $^{99m}$Tc production with isochronal cyclotrons are the methods of $^{99}$Mo/ $^{99m}$Tc production at the present time. Spallation neutron source by $^{100}$Mo(n,2n) have been investigated to produced $^{99}$Mo, but it has not been applied commercially yet. Nevertheless, $^{99}$Mo from fission fragments of $^{235}$U generated into nuclear reactor is the most common method for assembling $^{99}$Mo/$^{99m}$Tc alumina-based column [15].

Nowadays, the world demand for $^{99}$Mo is about 450,000 GBq per week and the annual demand for $^{99}$Mo is considered to have an 8–12% growth over this decade [16]. Currently, five main nuclear research reactors located in Canada, Belgium, France, Holland and South Africa [17] produce $^{99}$Mo at commercial scale. Factors associated mainly to the cost, complexity and security involving nuclear reactor technology can make difficult the application of the $^{99m}$Tc, considering that all other nonproducer countries import the $^{99}$Mo radioactive columns from countries that holds technology of production. Such radioactive columns have been used to assemble the radioisotope generator which shall be weekly transferred to the nuclear medical clinics.

The $^{99}$Mo radionuclide has a half-life of 65.94 h and decays by beta minus emission; however, 87%) goes to the metastable state of technetium, generating $^{99m}$Tc; while 13%) to the ground state of technetium which has a half-life of $2.1 \times 10^5$ year. The technetium decay to $^{99}$Ru isotope by beta emission. On this nuclear process, photons of 740 and 780 keV are emitted. The daughter nuclide $^{99m}$Tc decays by isomeric transition; however, 10% decays by internal conversion. A typical decay-growth kinetics of $^{99m}$Tc and $^{99}$Mo nuclide shows activities in transient equilibrium due to the fact that half-live of $^{99}$Mo is about 10 times greater than that of $^{99m}$Tc, which is 6.01 h, where the yield of $^{99m}$Tc is maximum close to 24 h after elution.

In order of transmutation of $^{98}$Mo to $^{99}$Mo, the following three major processing can be addressed: nuclear reactors, radioisotope sources and particle accelerators. Nuclear reactors can provide high level of neutron flux; however, reactors are complex, expensive and has large dimensions. Sealed radioisotope source emits radiation whose strengths decay with time. Hence, these sources are associated to continuum radiation protection requirements. Another method of neutron generation is represented by particle accelerators. This device may provide advantages over the other two available neutron sources, since it is able to turn off.

The essence of a modern and compact neutron generator based on deuteron fusion should comprise the design of a gas-control reservoir, a plasma and ion source to generate and gather the ions in a beam shape, a set of electrodes, including a metal target electrode loaded with deuterium ($^2$H) or tritium ($^3$H) hydrides. The plasma source produces the ions generally through magnet and electrode configurations or radio frequency antenna. Subsequently, the deuterium or tritium ions, deuterons (d) or tritons (t), are accelerated by an electrode system toward a hydride target loaded with deuterium, tritium, or a mixture of both; in which the fusion reaction occur generating neutrons, that are emitted with energy of 2.45 MeV from d-d and 14.1 MeV from d–t reactions [18]. Although the d–t reaction is more prolific in terms of neutron generation, tritium is a radioisotope while deuterium is stable. In this case, a generator can be assembled in compact dimensions due to the appreciable isotope hydrogen fusion cross-section at relative low-energy acceleration, depicted on Fig. 2.

In an attempt to improve the commercial technetium generation method, a generator based on neutron activation of the precursor is presented, whose technology includes a potential route to $^{99m}$Tc and other radioisotopes production. The main issues related to a fusor generator for radioisotope production can be summarized as:

(i) supplement of radioisotopes rich in neutrons of short physical half-life;
(ii) independence of the high flux reactor technology in the activation of $^{99}$Mo, available only in few industrialized countries;
(iii) replacement of complex technologies represented by reactors or particle accelerators;
(iv) supporting the philosophy of compact radioisotope generators, without dependence on preexistent radioactive parent nuclide;
(v) improvement of radiological safety, allowing full facility shutdown and consequently interruption of the radiation produced in the device, similar to a x-ray machine with minimum radioactive internal contamination.

Herein, a device named Hemispherical Fusor Technetium Generator (HFTG) [20] is presented based on a particle accelerator for
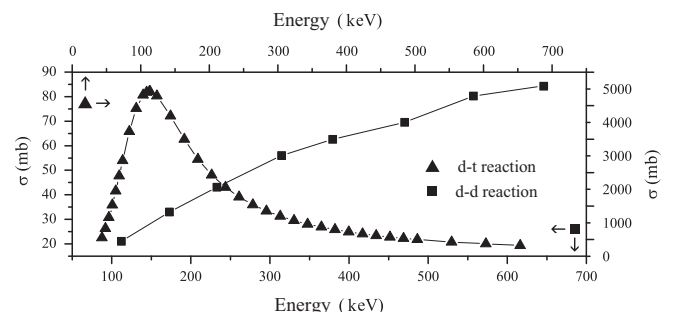


Fig. 2. Cross-sections of neutron production for $^2$H and $^3$H target bombarded by deuterons [19].