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Semi-homogeneous Reactor for ^{99}Mo Production: Conceptual Design

Enrique E. Pasqualini
Nuclear Nanotechnology Laboratory
Constituyentes Atomic Center
National Atomic Energy Commission
Buenos Aires. Argentina
pascua@cnea.gov.ar

ABSTRACT

This work identifies and discusses the more relevant characteristics of the engineering concepts of semi-homogeneous reactors for the production of nuclear medicine radioisotopes coming from fission products, such as molybdenum-99 (^{99}Mo).

Similarities and differences in design concepts between homogeneous reactors (uranium salts dissolved in an acid media) and semi-homogeneous reactors (UO_2 particles suspended in water) are emphasized for radioisotopes production.

Some of the design parameters considered are: power density of 1 kilowatt per liter of suspension, volumetric concentration of solids around 3% using low enriched uranium (20% ^{235}U) in a 50 liters vessel at low working temperature and atmospheric pressure with heat extraction by means of an independent refrigeration circuit and control bars. Stainless steel (AISI 316L) is the preferred material to be used. Forced fluid circulation is needed to maintain homogeneity, avoid decantation and control gas bubbles evolution. Batch extraction of fission products will be performed with convenient by-pass circuits.

Introduction

Technetium-99 metastable (^{99m}Tc), the daughter isotope of ^{99}Mo, is used in more than 20 million radiotracer and diagnostic nuclear medical procedures (10-30 mCi each) per year, half of which are bone scans, and the other half are roughly divided between kidney, heart and lung scans. The world demand estimated of ^{99}Mo, used in 85 percent of diagnostic imaging procedures in nuclear medicine, is approximately 12.000 six-day curies per week [1,2].

The major production of ^{99}Mo around the world is concentrated in ten heterogeneous nuclear reactors using high (HEU) or low (LEU) enriched uranium fuel and targets (*Table 1*). Most of these reactors have been working for more than 40 years.

The large-scale producers capacity of ^{99}Mo is indicated in *Table 2*. As shown in this table the total processing capacity of 21.425 six-day curies per week is greater than the world demand. These resources are not fully exploited since the ^{99}Mo distribution has to be done from the processing plants that have to be near the reactors location.

Table 1. *Major current ^{99}Mo production reactors [2,3].*

Reactor	***Country***	***Annual operating days***	***Production 6-day curies/week***	***Weekly % of world demand***	***Fuel/Targets***	***Commis-sioning year***
BR-2	Belgium	140	5200	25-65	HEU/HEU	1961
HFR	Netherlands	300	4680	35-70	LEU/HEU	1961
LVR-15	Czech Republic	Just started	> 600	No data	HEU/HEU	1957
MARIA	Poland	Just started	700-1500	No data	HEU/HEU	1974
NRU	Canada	300	4680	35-70	LEU/HEU	1957
OPAL	Australia	290	1000-1500	No data	LEU/LEU	2007
OSIRIS	France	180	1200	10-20	LEU/HEU	1966
SAFARI-1	South Africa	305	2500	10-30	LEU/HEU	1965
RA-3	Argentina	230	200	< 2	LEU/LEU	1967
RSG-GAS	Indonesia	147	150	< 2	HEU/LEU	1987

Table 2. *Processing capacity of ^{99}Mo [2].*

Facility	***Country***	***Processing capacity six-day curies EOP/wk***
ANSTO	Australia	> 1000
Covidien	Netherlands	> 3500
CNEA: Ezeiza Atomic Centre	Argentina	> 600
IRE	Belgium	> 3000
MDS Nordion	Canada	> 7200
NTP	South Africa	> 3000
NTP – in development	South Africa	2625
Total		> 21425

These mentioned two aspects of old reactors and distribution capacity are the basic realities in which statements of shortage of medical isotopes production in the world [4] can be based, requiring upgrades of installed capacities [5] and improvements in the distribution chains.
Since it is expected that the clinical demand for ^{99m}Tc will remain strong for decades to come [1], the future of a significant part of nuclear medicine requires new reactors being built, incorporation of new technologies that can be developed and better distribution capacity.

1. Production alternatives

Almost all ^{99}Mo production today comes from research reactors. Targets containing uranium are placed inside the reactor where neutrons cause fission of approximately 3% of the ^{235}U atoms present inside the target. After about 1 week of irradiation, the targets are taken from the reactor for chemical processing. The targets are dissolved in either an acid or alkaline solution and the ^{99}Mo is extracted chemically. The production rate of this technology is determined by the combination of three factors: the number of neutrons available that cause fission (flux) -coming from the ^{235}U fissions inside the nuclear fuel-, the mass of ^{235}U available in the target, and the probability of fission when a collision occurs (cross section). Since a large proportion of the molybdenum produced is the desired ^{99}Mo, the end product has a high level of radioactivity per gram of molybdenum (high specific activity).

Another way of producing ^{99}Mo is with the irradiation of molybdenum targets in a reactor: neutrons can be captured by ^{98}Mo atoms to form ^{99}Mo [4,6]. Separation of ^{99}Mo from the rest of the molybdenum to increase its specific activity can be performed using Szilard Chalmers reactions, improving the low yield of this process [7]. Also, protons and photons can induce nuclear reactions obtaining ^{99}Mo as product [4] in accelerators but with low production rates. A brief review of production methods with several references can be found [8].

Liquid reactors, in which uranium is either in solution or as suspended particles, avoids the use of targets and circumstantially, fission products have been extracted from them [9,10]. In liquid reactors the whole inventory of ^{235}U is the target and, as all fission products in the reactor can be collected, the power needed to produce the same quantity of Curies of ^{99}Mo than the most efficient conventional target method is dramatically reduced. Also follows that nuclear waste is extensively diminished. Homogeneous reactors for the production of ^{99}Mo do not need to have powers much greater than 200 kWth [11,12] and are being developed to work with low enriched uranium.

Once a week, in an homogeneous salt solution reactor, the acidic solution is transferred to a hot cell for processing and the molybdenum chemically and physically extracted. Afterwards, the uranium containing solution has to be fed back into the reactor to be used in a next cycle. In the case of a suspended particles or semi-homogeneous reactor, the particles can be filtered and only the liquid with fission products and no uranium can be processed; fresh liquid is used in the next cycle.

2. Semi-homogeneous reactors

Semi-homogeneous reactors with suspended particles have been designed many years ago to overcome low solubility of thorium salts (used in breeder reactors) and solubility misbehavior dependence with temperature of uranium sulphate salts in the development of homogeneous nuclear reactors for electric power production [13]. Nowadays, it is interesting to study the feasibility of this type of reactors to extract ^{235}U fission products, in particular ^{99}Mo.

3.1. Suspended particles

The size of the insoluble uranium containing particles in semi-homogeneous reactors should be smaller than the penetration depth of fission products, such that they will end their trajectory in the surrounding light water moderator -principle of fission recoil separation- [14]. Uranium dioxide (UO_2) spherical particles of approximately 5 microns diameter [15,16,17] are chemically stable in water and if the distance between them is twice their diameter the probability of fission product penetrating other particle is greatly reduced. This characteristic defines an approximately volume concentration of the particles -theoretical density of 10,96 g/cm^3 and 20% ^{235}U- smaller than 3% V/V with light water as moderator. The sedimentation limit velocity of 5 microns diameter UO_2 particles in water at room temperature is 0.14 mm/sec, at 50 ºC is 0,25 mm/sec and at

100 ºC is 0.5 mm/sec. Obviously the liquid should be in movement to maintain the particles in suspension.

The size of particles allow to use sintered metal fiber filters available in a variety of alloys with absolute ratings from 1 μm that can control a previously specified volume where the particles can be with an overhead volume of clean water; this configuration makes irrelevant any fluctuations ought to surface waves because the reactive volume is not involved. An important consideration of the particles sedimentation and the filtering metal sintered fibers sieves is the liquid with the radioisotopes can be extracted without the suspended particles, avoiding in this way any possibility of having a critical volume outside the reactor. Another benefit of this extraction procedure is that the processing facility has to be necessarily aside the reactor, since no critical mass is involved outside the reactor vessel.

One of the preferred synthesis method for producing spherical particles with size control and spherical shape is by means of nucleation and growth by precipitation of uranium containing nanoparticles and subsequent controlled agglomeration; heat treatments are performed for reduction of the uranium compound to UO_2 and sintering purposes [18]. Fission products will produce interior damage and as they exit the surface of the particles, some matrix atoms can be dragged out by being knocked out or even the thermal burst produced can eject matrix atoms [19]. These situations can be estimated by means of transport of ions in matter (TRIM) calculations and experimentally checked. If needed, coverage of the particles with an inert material can produce some desired benefit.

3.2. Power density

No problems have been reported for homogeneous reactors with power densities lower than 1.8 kW per liter of solution. A design parameter of 1 kW/lt of suspension for a semi-homogeneous reactor is chosen to perform initial calculations with prototype capabilities -such as heat extraction- to be extended five times to increase and test extended performance parameters. With a power density of 1 kW/lt a volume of a 50 liters vessel is the minimum to be considered to have the possibility of accommodating the required pumps, filters, circuits, and other accessories needed for appropriate working of a prototype device. This makes a nominal total power of 50 kW, generated from the fissions of ^{235}U nucleus. Knowing that each ^{235}U thermal neutron fission produces approximately 200 MeV [20], a fission rate of 3.1×10^{10} fission/sec will generate 1 Watt, and hitherto the 50 kW power will be sustained by 1.55×10^{15} fissions/second. Since 6.1 % of the produced fission products is ^{99}Mo a total of 9.5×10^{13} atoms will be produced per second. ^{99}Mo is produced at a constant rate (α), depending of the power sustained by the reactor, and decays with a mean life of 66 hours ($\tau = 1/\lambda$). The net quantity *N* of ^{99}Mo atoms produced can be derived resolving the following differential equation:

$$\frac{dN}{dt} = \alpha - \lambda N$$

Rearranging this equation and multiplying all terms by $e^{\lambda t}$[21] the rate equation can be expressed as:

$$\frac{d\left(N e^{\lambda t}\right)}{dt} = \alpha e^{\lambda t}$$

After Integrating it is obtained the following expression

$$N e^{\lambda t} = \frac{\alpha}{\lambda} e^{\lambda t} + C$$

,

And resolving for the constant, knowing that $N\ (t = 0) = 0$, the total quantity of ^{99}Mo present at each irradiation time will be:

$$N = \frac{\alpha}{\lambda}\left(1 - e^{-\lambda t}\right)$$

The bracket expression is represented in Fig.1 (left). For the case of a 50 kW reactor the asymptotic value of ^{99}Mo production will be the value of α/λ that is 2.26 x 10^{19} atoms; for a 7 days production the quantity of ^{99}Mo atoms will be near 90 % of this value. The asymptotic production value corresponds to 0.0037 grs of ^{99}Mo with a specific activity of 5 x 10^4 Ci/gr [22,23]. With an extraction time of 5 days irradiation (80% of maximum) will generate 150 Ci. Considering that extraction efficiency is around 90% and after 6 days of irradiation a 17% portion is retained, the calibrated quantity of this production scheme will be around 25 six-day Curie in one week with a 50 kW reactor. In figure 1 (right) it is shown the decay of the ^{99}Mo radioisotope in the corresponding exponential decreasing form:

$$N = N_o\, e^{-\lambda t}$$

N_o will be some percentage of the produced ^{99}Mo at the end of irradiation that will depend on the extraction yield of the radioisotope.

A continuous process of fission products extraction during power generation is not recommended since retarded neutrons will not be any more in the critical zone and the reactivity control will loose versatility. Radioisotopes should be extracted at the end of pre-designed working cycles, in batch conditions. A first approximation of the 50 litres reactor is a 40 cm diameter closed cylinder of 40 cms height, with a maximum temperature of 50 ºC and working near atmospheric pressure. The construction material can be AISI 316 stainless steel. It is not expected that corrosion can be an issue but it has to be observed in detail the action of direct fission products bombardment at the vessel walls. Considering a 3% v/v concentration of suspended particles, the total quantity of 20% enriched uranium in UO_2 (ρ = 10.96 grs/cm^3) is of 15 kgs; the quantity of ^{235}U is 2.6 kgs. This fuel can surely be used up to more than 50 % burn up.

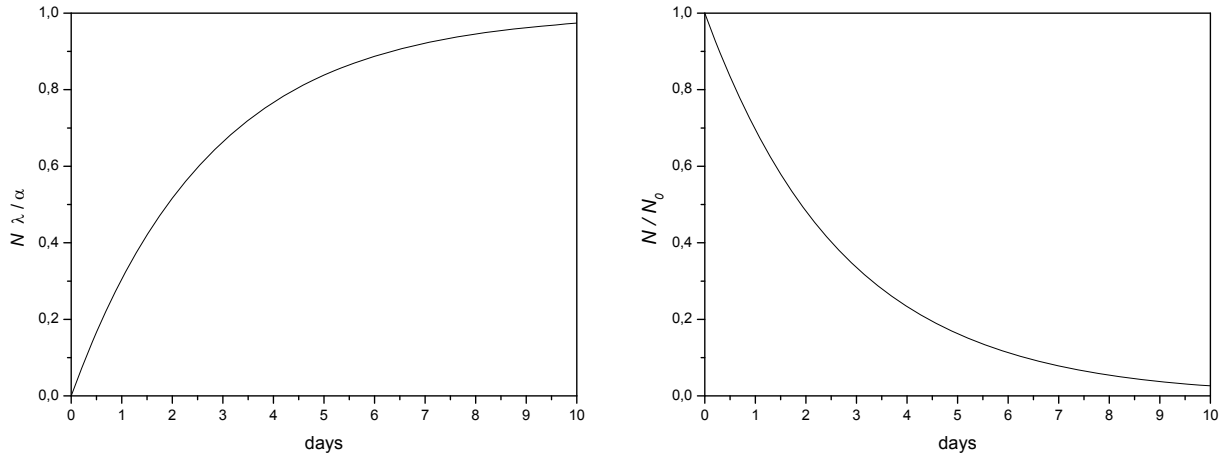

***Figure1**. ^{99}Mo production dependence as a function of target irradiation time (left) and decade mode of ^{99}Mo after irradiation (right).*

3.3. Fission products

The fission of ^{235}U produces, with more than half percent yield, the following chemical elements Br, Kr, Rb, Sr, Zr, Mo, Ru, Rh in the light side of the distribution mass spectra between 83 to 106 amu and Sn, I, Sb, Xe, Cs, Ba, Ce, Nd, Sm, Pm in the heavy mass side between 128 and 151 amu. The mean fission fragment energy of the lighter nucleus is approximately 60 MeV and of the heavier nucleus is 95 MeV [20,24]. These, as generated highly positively charged fission products, will evolve in the surrounding water as gas, elements in solution, insoluble products or compounds. The volatile species fundamentally are the noble gases Krypton and Xenon; bromine has a boiling point at 60 ºC at atmospheric pressure. Insoluble products will adsorb at the particles surface and vessel and piping walls. High specific surface materials such as activated carbon nanoparticles can be used to adsorb insoluble products. The volatile species should be gathered at the top end of the closed vessel using high adsorption materials (zeolites, activated carbon, activated alumina, etc).

The processing of the fission products can be schematically outlined. The liquid with the activated carbon is extracted from the reactor using a by pass circuit. The activated carbon can be again filtered and the soluble and adsorbed species be treated in separate ways. Known processes should follow for specific radioisotopes extractions. As soon as the liquid is extracted from the reactor a fresh charge of water and activated carbon can be used and a new irradiation cycle begins.

3.4. Fluid circuits

The reactor facility should contemplate several fluid circuits. The first that can be considered is a natural one inside the reactor where some of the fission products evolve as gaseous species that are adsorbed at high specific surface materials at a plenum space at the top of the closed reactor vessel. Another evolution of gaseous species has

its origin in the water decomposition. Hydrogen and oxygen should be catalytically recombined in a nearby circuit.

A main circuit is used to keep particles in suspension and to force the fluid threw serpentines working as heat exchangers. The serpentines are part of an independent refrigerating water circuit. A bypass circuit is connected to the main one for the liquid extraction at the end of each irradiation cycle with intercalated filters or hydrocyclones. A solid reflector is preferred with a convenient air cooler and heat exchanger.

3.5. Calculations

Several calculations can be performed. The neutron calculations for tuning the ratio between fissile material and moderator, control bars design, power evolution with particle decantation, etc. Computational fluid dynamics is important to evaluate general and local suspension characteristics. TRIM calculations can be used to compare theoretical and experimental contaminations and performance of suspended particles. Catalytic recombination can also be simulated.

3. Discussion

The performance of suspended particles reactors have already been tested in the 50s and 60s. One of the main interests in the construction of an aqueous suspended particles prototype reactor is testing the behavior at power densities higher than 2 kW/lt and the extraction yields of ^{99}Mo. The power density can be tested in an experimental prototype that can be instrumented and with simple considerations of closed vessel, low temperature and normal pressure working conditions, refrigerating serpentine, control bars and refrigerated reflector. The extraction yields can be tested in small size irradiation experiments in experimental reactors.

4. Conclusions

Although aqueous suspension reactors have not been used extensively it is important to notice that the development of this technology was practically finished after more than 20 years work in several countries. The new application proposed in this work of using them for the production of medical radioisotopes is simpler than the energy production and breeder conditions for which they were initially tested. The fundamental reasons for these differences are that smaller powers, temperatures and pressures are involved.

Aqueous suspension reactors for radioisotope production is worthwhile the most serious feasibility studies. This affirmation is based in two main aspects. One of them is the general advantages of homogeneous solution reactors for fission radioisotopes production that are also common to suspended particles reactors. The second reason is that suspended particles reactors have extra advantages compared with homogeneous solution reactors: no acids are involved, higher uranium concentrations can be

achieved, critical liquid zones can be confined using filtering surfaces, initial physical separation of fission products and uranium with direct consequences in extraction yields, no need of extraction of uranium from the reactor vessel, smaller temperatures and probably higher power densities can be used.

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