PRESENT STATUS OF THE USE OF LEU IN AQUEOUS REACTORS TO PRODUCE MO-99

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Abstract: An operating aqueous homogeneous reactor, the ARGUS at Kurchatov Institute, has been used to produce fission product molybdenum-99 (Mo-99), widely used in nuclear medicine to produce technetium-99m (Tc-99m). The Mo99 has been extracted from the sulfate solution using an organic sorbent after operation at 1 kW/liter. After purification, the material has been assayed and the result is well within required specification of the US Pharmacopaeia. Operation with LEU is planned to determine if any additional purification steps are required. Calculations are presented to show the sources and quantity of alpha activity when LEU is used.

1.0 BACKGROUND

Nuclear medicine has relied on Tc-99m and its parent Mo-99 for diagnostics since its development at Brookhaven National Laboratory (USA) in 1964[1]. Over 35,000 procedures per day using Tc-99ni created by Mo-99 are performed throughout the world.

The major source of the Mo-99 is from fission of U-235. The uranium is formed into targets which are irradiated by neutrons from research or test reactors. These irradiated targets are dissolved and the fission product, Mo-99, is extracted from the solution.

In 1992, Ball[2] introduced a method of "targetless" production of fission product Mo-99 using an aqueous homogeneous reactor fueled with uranyl nitrate. The design anticipated that the uranium salt could be made with low enriched uraniurn[3,4].

The advantages of the homogeneous system compared with targets are:

1. No requirement to fabricate and transport targets
2. Full utilization of fission product Mo-99. In the target irradiation, all the fissions (and the resulting production of Mo-99) in the reactor used to make neutrons which irradiate the target are wasted. The Mo-99 remains in the solid reactor fuel.
3. Waste fission products are 1/100th of the total produced in the target method for a given quantity of Mo-99
4. Uranium consumption and heat production are 1/100th of that produced in the target method
5. Extraction processing is simplified with no uranium dissolution required

2.0 RECENT DEVELOPMENTS

In 1998, the Kurchatov Institute developed an extraction method which could remove the Mo-99 from the reactor solution and leave the uranium sulfate in solution.

The solution was irradiated at specific powers of 1 kW/liter and the solution passed through a proprietary sorbent. The Mo-99 stayed on the sorbent and the remaining products, including the uranium, passed through and could be used again as part of the reactor solution.

The Mo-99 could then be eluted from the sorbent with an acid solution and concentrated for further purification to meet medical (US Pharmacopiea) standards.

3.0 ALPHA ACTIVITY REQUIREMENTS AND SOURCES

3.1 USP Requirements

Requirments for maximum alpha activity are given in the United States Pharmacopeia, USP 23 - Official Monographs, supplements 1997. In the statement for "injection prepared from technetium 99m derived from parent molybdenum 99 formed as a result of uranium fission" under the section ALL OTHER NUCLIDIC IMPURITIES .. the USP says: "Not more than 0.001 Bq of gross alpha impurity per MBq (or 0.001 nCi of gross alpha impurity per 1 mCi) of technetium 99m is present at the time of administration." Thus, the ratio of Mo-99 activity to alpha activity must be 1 E+09 or greater.

3.2 Sources of alpha activity

3.2.1 URANIUM -

The reactor solution contains uranium salts. The requirements for the concentration are set by the need to reach a critical (self-sustaining chain reaction) concentration of the fissionable isotope. For the ARGUS reactor, the principal fissioning isotope is U-235. For a reactor made up of 20 liters of uranyl sulfate solution, the quantity of U-235 is 1600 grams for a 90% enriched uranium, HEU, and 1900 grams for a 20% enriched uranium, LEU. The inventory of U-238 is 177 grams for 90% enriched and 7600 grams for 20% enriched.

Other alpha emitters depend on the concentration of other isotopes of uranium. The quantity of these isotopes depends in part on the history of the uranium; such as: the method of enrichment, the chemical treatment, possible blending of various enrichments, and the growth of daughter alpha emitting isotopes during the lifetime of the uranium.

Using the ARGUS[5'6] inventory for uranium enrichments of 90% and 20%, Table 1 was prepared to show alpha activity in the solution. Supporting calculations were made by Kurchatov specialist V. Palvanov.

Parameter	Units	Value 90 % Enr HEU	Value 20 % Enr LEU
Inventory of U-235	grams	1600	1900
Inventory of U-238	grams	177	7600
Alpha activity from U-235	Bq	1.3 E8	1.5 E8
Alpha activity from U-238	Bq	2.2 E6	9.4 E7
Total alpha activity from uranium	Bq	1.3 E8	2.4 E8
Total alpha activity from uranium	mCi	3.5	6.6

Table 1 - Alpha Activity in ARGUS Solutions

Since the burnup of uranium is insignificant at the low power levels of the system, the inventory of both uraniums remains nearly constant for the life of the solution (expected to be 10 years).

3.2.2 PLUTONIUM

As the reactor operates, the U-238 absorbs neutrons. The reaction rate is proportional to the number of U-238 atoms and is, therefore, higher with the LEU. The absorption cross section of U-238 for thermal neutrons is 2.68 barns (2.68E-24 cm^2). The product of the absorption is U-239 which decays with a 23.5 minute half life to Np-239 which further decays with a 2.35 day half life to Pu-239. Pu-239 has a long half life, 24,000 years, and decays with alpha emission.

Thus, the buildup of Pu-239 is proportional to the power of the reactor which, in turn, is proportional to the flux and U-235 concentration. At 20 kW, the average flux of the ARGUS reactor with HEU is 2.5 El1 n/cm^2-sec. The average flux with LEU is 2.1 El1 n/ cm^2-sec.

Table 2 compares the production rate of Pu-239 for a reactor fueled with HEU and LEU.

Parameter		Value 90 % Enr HEU	Value 20 % Enr LEU
Thermal flux - 20 kW	n/cm^2-s	2.5 Ell	2.1 Ell
Production rate of Pu-239 - 20 kW	grams/day	1.04 E-05	3.75 E-04
Alpha activity from Pu-239 3 days op	mCi	1.9 E-03	7.0 E-02
Alpha activity from Pu-239 3 years op	mCi	6.9 E-01	2.5 E+01
Ratio LEU/HEU alpha activity from Pu-239		36	

Table 2 - Comparison of HEU and LEU Pu-239 Alpha Activity

As the Pu-239 builds up in the solution, there is increased absorption of neutrons. The cross section for thermal neutrons, both fission and absorption, in Pu-239 is approximately 1000 barns. The determination of the net balance of Pu-239 is from the solution of the differential equation:

Change in Pu-239 = Production by U-238 abs. - Loss by Pu-239 abs. 1)

The solution has a time constant proportional to the flux. For the solution reactor operating at 20 W, the time constant is 144 years. Thus, in the operating life of a single solution, the loss of Pu-239 by absorption is negligible.

4.0 CONCLUSIONS

4.1 HEU Operations

The ARGUS reactor has been used to produce Mo-99 using the HEU uranyl sulfate solution. The Mo-99 has been extracted using a sorbent which holds up the Mo-99 and passes through the uranium solution. The Mo-99 is then washed from the sorbent and further purified.

The alpha activity for an HEU solution has been found to be within USP limits**.** Thus, further work on elimination of alpha emitting material does not appear necessary.

4.2 LEU Operations

Further investigation is required to determine that the alpha activity of the final Mo-99 product made from an LEU solution is within USP specifications. As shown in Table 1, the initial uranium alpha activity of the LEU is only slightly higher than the HEU (6.6 mCi vs 3.5 mCi). It may be possible that present sorption and purification techniques will suffice for the uranium alphas in LEU.

The growth of the alpha emitting Pu-239 is small and for an HEU reactor is only 20% of the uranium alpha activity after three full power years at 20 M

The increase in Pu-239 alpha activity is a factor of 36 greater in LEU than in HEU. (Note that this is also true in the target system of Mo-99 production.)
Thus, it must be demonstrated that the product, Mo-99, will meet USP specifications for LEU. This may be done by modification of the purification process or by the addition of another purification step.

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