RADIONUCLIDE PURITY OF FISSION MO-99 PRODUCED FROM LEU AND HEU. A COMPARATIVE STUDY

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Abstract

There is a huge concern around the world about the use of highly enriched uranium to operate research reactors and produce radioisotopes including Mo-99. Therefore, the current policies seek to reduce uranium enrichment.
Argentina is one of the first countries all over the world that has developed the technology to produce fission Mo-99 from LEU targets, which has been commercially available since 2002.
The aim of this work is to compare the radionuclide purity from several batches of fission Mo-99 produced routinely in Argentina from both LEU and HEU. This work characterizes some of the most commonly found contaminants in Mo-99 batches. Results form 28 HEU and 46 LEU batches of fission Mo-99 produced between 2002 (HEU) and 2003 (LEU) are presented. A general improvement in the radionuclide purity after the change from HEU to LEU targets has been observed.
Hence, the conclusion is that the radionuclide quality of Mo-99 produced from LEU targets is as good as with HEU ones and even better. It complies with all commercial standards and has excellent acceptance in the Argentine and regional market.

Introduction

There is a huge concern around the world about the use of highly enriched uranium to operate research reactors and produce radioisotopes including Mo-99. Therefore, the current policies seek to reduce uranium enrichment.
Since 1985 the Argentine National Atomic Energy Commission has been producing Mo-99 by fission of highly enriched uranium contained in targets that were irradiated in its RA-3 reactor. Due to non-proliferation concerns this highly enriched uranium had to be replaced by uranium of low enrichment. After several years of development, fission Mo-99 using LEU targets became a reality and since year 2002 is commercially available.
Regarding the production process, in both cases the targets were dissolved in basic media followed by several ion-exchange chromatographic separation steps, although, according with previously published works, some changes were made.
Prior release of the first batch of LEU produced Mo-99, studies on quality aspects were made, and results were compared with international standards.
The Argentinean domestic market uses about 75% of locally produced CNEA Mo-99, importing the rest from different origins. Some of the locally produced Mo-99 is exported, through two radiopharmaceuticals companies, to the near region via Mo-99/Tc-99m generators.
The scope of this work is to demonstrate the feasibility of LEU fission Mo-99 production and commercialization in accordance with international quality standards.

Materials and methods

Radionuclide purity was assessed between 8 and 15 weeks after production (delayed purity). Gamma spectra were acquired using high performance gamma spectrometry (Canberra) in the following conditions: Counting time 50,000 s.; Starting activity (at calibration time) 300-600 mCi . Sr-90 determinations were performed using a Liquid Scintillation Analyzer (Packard Tri-carb 2900TR). In order to separate Sr-90 from other beta emitter contaminants, Eichrom- Sr resin columns have been used. Gross alpha determinations were performed using a Ludlum 43-10 Alpha Sample Counter (Ludlum Inc.). Counting conditions were: Counting time 300,000 s.; Starting activity (at calibration time) 50-300 mCi.

Results

In order of avoid excessive data, two minor contaminants (Sb-125, Cs-137) have been chosen, representatives of other radio nuclides present in the solution, such Ru-106, Nb-95, Zr-95, etc. Nevertheless, the $\Sigma\gamma$ is represented mostly by I-131 and Ru-103 contributions. All results are expressed as contaminant to Mo-99 ratios at calibration date (usually 3 days post production). The results obtained from 28 batches of Mo-99 from HEU (Table 1) show that all of them fulfill specifications.

Batch	I-131	Ru-103	Sb-125	Cs-137	Sr-90	Gross Alpha
1	7,57E-06	1,25E-08	2,24E-09	3,52E-10	1,76E-09	3,80E-12
2	2,01E-06	5,53E-09	1,50E-09	1,95E-10	6,02E-09	4,40E-11
3	4,89E-06	4,46E-09	2,13E-09	3,27E-10	3,47E-10	3,60E-11
4	2,48E-06	1,83E-08	4,99E-09	3,00E-10	5,02E-10	1,20E-11
5	1,22E-05	5,09E-09	1,44E-09	2,80E-10	2,93E-10	9,80E-12
6	1,49E-05	9,06E-07	1,32E-07	2,75E-10	2,14E-09	3,60E-11
7	1,11E-05	1,17E-07	6,71E-09	2,42E-10	2,76E-09	2,10E-11
8	1,81E-06	3,46E-07	1,32E-10	2,73E-10	1,13E-08	4,40E-12
9	9,50E-07	3,37E-06	2,17E-08	4,96E-10	3,88E-08	7,20E-12
10	2,79E-07	2,39E-07	4,78E-09	2,10E-10	4,06E-09	6,50E-12
11	7,44E-07	2,28E-07	4,80E-09	2,86E-10	5,44E-09	7,80E-11
12	1,69E-07	1,49E-07	3,88E-09	2,72E-10	3,90E-09	2,30E-11
13	1,37E-07	2,01E-07	3,11E-09	2,40E-10	6,92E-09	1,10E-11
14	9,27E-08	1,83E-07	2,85E-09	2,00E-10	6,04E-09	7,20E-11
15	2,20E-08	9,96E-08	1,62E-09	3,55E-10	6,05E-09	9,90E-12
16	4,73E-08	6,58E-08	2,62E-09	2,63E-10	8,76E-09	2,10E-11
17	3,31E-07	2,74E-07	2,79E-09	3,82E-10	3,59E-09	9,10E-12
18	8,35E-08	1,69E-07	2,01E-09	8,48E-10	1,70E-10	3,30E-12
19	4,35E-08	2,44E-07	1,40E-09	2,97E-10	1,01E-09	6,30E-12
20	1,98E-07	8,11E-08	1,22E-09	3,03E-10	1,78E-09	2,30E-12
21	5,58E-08	1,33E-07	1,37E-09	2,53E-10	7,21E-09	5,60E-12
22	8,93E-08	4,39E-08	7,37E-10	2,78E-10	1,93E-08	1,20E-11
23	4,71E-06	2,02E-08	7,44E-10	2,81E-10	2,49E-09	8,80E-12
24	4,12E-08	2,27E-08	9,04E-10	2,09E-10	2,42E-09	5,50E-12
25	1,34E-06	1,14E-08	3,67E-10	2,63E-10	2,05E-09	2,30E-11
26	9,08E-07	2,36E-07	9,32E-10	1,39E-10	2,75E-09	3,40E-11
27	1,20E-07	1,93E-07	1,23E-09	2,26E-10	2,84E-09	9,80E-12
28	1,78E-07	3,88E-07	1,36E-10	2,12E-10	1,95E-09	3,10E-11

Table1: Contaminants found in Fission Mo-99 produced from HEU targets.

The results obtained from 46 batches of Mo99 from LEU are shown in Table 2

Batch	I-131	Ru-103	Sb-125	Cs-137	Sr-90	Gross Alpha
3	7,71E-08	2,30E-09	1,30E-10	7,10E-11	1,70E-09	6,10E-12
4	6,13E-08	1,75E-09	2,36E-10	6,14E-11	1,76E-10	3,20E-11
5	3,84E-08	1,25E-09	2,81E-10	3,41E-10	2,25E-10	5,50E-11
6	1,64E-07	9,04E-10	6,28E-10	2,31E-10	2,16E-10	3,50E-11
7	6,87E-08	1,11E-09	4,24E-10	2,97E-10	2,55E-10	7,40E-12
8	1,67E-07	1,37E-09	7,23E-10	2,80E-10	8,33E-10	1,40E-11
9	2,01E-07	4,39E-09	3,56E-10	8,15E-10	3,47E-09	2,20E-11
10	5,04E-07	1,15E-09	5,31E-10	2,50E-10	1,49E-10	3,50E-12
11	2,35E-07	2,10E-09	1,44E-10	9,57E-11	1,73E-10	6,30E-12
12	1,96E-07	8,20E-10	1,40E-10	9,50E-11	2,21E-10	8,40E-12
13	8,21E-08	7,28E-09	7,78E-10	1,48E-09	3,03E-10	1,10E-11
14	5,52E-08	7,40E-09	6,42E-10	4,66E-10	2,34E-10	9,30E-12
15	1,14E-07	5,60E-09	4,99E-10	2,46E-10	3,73E-10	8,50E-12
16	1,71E-07	9,85E-08	1,07E-09	7,82E-10	2,00E-09	2,00E-11
17	6,84E-08	2,08E-08	9,33E-10	1,79E-10	8,39E-10	9,80E-12
18	2,26E-07	3,15E-08	9,49E-10	7,50E-10	1,78E-09	8,60E-12
19	1,12E-07	2,46E-08	1,38E-09	2,78E-10	7,55E-10	9,10E-12
20	1,60E-07	2,75E-08	7,00E-10	7,46E-10	6,83E-10	8,30E-12
21	2,31E-07	1,50E-08	1,02E-10	1,26E-10	1,52E-09	5,50E-12
22	9,15E-08	9,91E-09	3,40E-10	9,81E-11	7,63E-10	2,10E-12
23	1,42E-07	3,13E-08	9,80E-10	4,50E-10	8,73E-10	7,50E-12
24	1,16E-07	4,51E-08	2,30E-10	9,15E-10	1,21E-09	8,30E-12
25	3,21E-07	7,80E-09	3,40E-10	3,33E-10	5,56E-10	9,20E-12
26	6,57E-08	2,17E-08	1,44E-10	5,10E-11	8,32E-10	5,50E-12
27	8,34E-08	4,13E-08	5,30E-10	4,50E-10	8,45E-10	2,50E-11
28	1,05E-07	8,32E-09	1,02E-09	7,50E-10	9,66E-10	1,50E-11
29	3,17E-07	1,38E-08	3,15E-10	2,10E-10	9,56E-10	9,80E-12
30	7,34E-08	2,44E-09	4,23E-10	7,52E-10	9,05E-11	1,00E-11
31	9,15E-08	7,39E-09	2,44E-10	8,93E-11	5,90E-10	8,60E-12
32	1,43E-07	4,11E-08	6,31E-10	8,98E-11	8,93E-10	7,10E-12
33	2,17E-07	7,30E-09	1,99E-09	1,23E-10	5,72E-10	2,00E-11
34	8,49E-08	5,90E-08	3,15E-10	4,44E-10	1,06E-09	6,30E-12
35	1,12E-07	1,22E-08	2,10E-10	2,19E-10	6,01E-10	7,20E-12
36	7,60E-08	1,43E-08	6,20E-10	3,23E-10	7,68E-10	5,50E-12
37	2,14E-07	5,57E-09	1,25E-09	6,45E-10	5,62E-10	9,80E-12
38	1,31E-07	1,23E-08	3,55E-10	5,33E-10	5,54E-10	9,60E-12
39	4,32E-07	3,14E-08	5,28E-10	1,02E-10	1,15E-09	3,40E-11
40	9,17E-08	9,81E-09	5,44E-10	3,48E-10	6,74E-10	6,50E-12
41	1,05E-07	6,35E-09	3,30E-10	7,40E-11	5,31E-10	7,30E-12
42	7,13E-08	2,32E-08	8,90E-10	3,25E-10	1,02E-09	6,00E-12
43	1,42E-07	1,65E-08	2,25E-10	5,55E-10	1,15E-09	9,00E-12
44	8,18E-08	3,13E-08	9,33E-10	8,16E-11	4,18E-10	1,00E-11
45	2,40E-07	8,38E-09	6,25E-10	2,65E-10	6,36E-10	7,30E-12
46	1,00E-07	1,32E-08	3,34E-10	1,03E-10	5,34E-10	7,50E-12

Table2: Fission Mo-99 produced from LEU targets contaminants

With the intention to compare results, international specifications are showed in Table 3.

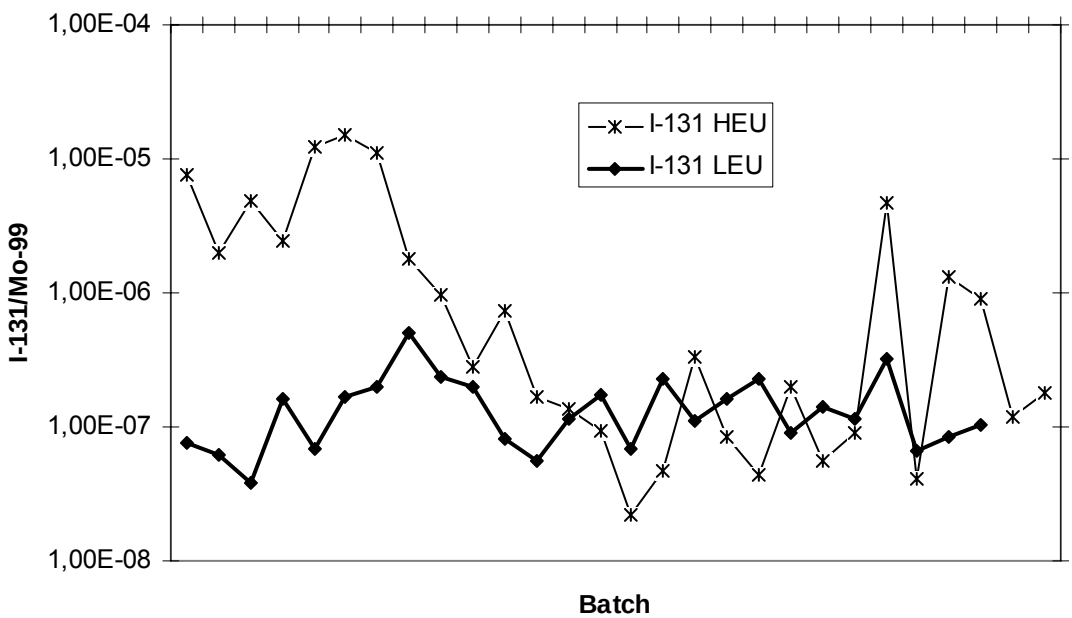
Nucleide	Specification
I-131 : Mo-99	$< 5 \times 10^{-5}$
Ru-103 : Mo-99	$< 5 \times 10^{-5}$
Sr-90 : Mo-99	$< 6 \times 10^{-8}$
$\Sigma\alpha$: Mo-99	$< 6 \times 10^{-8}$
$\Sigma\gamma$: Mo-99	$< 5 \times 10^{-7}$

Table 3: International specifications

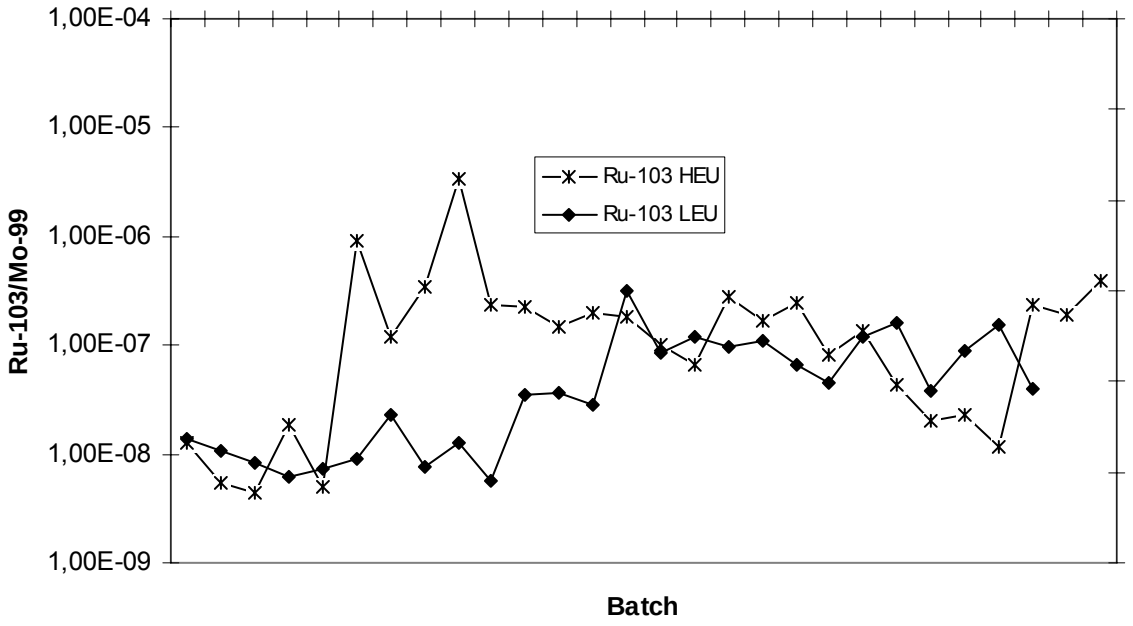
The average values calculated from both targets are shown in Table 4. It is clear that in the LEU case, the concentration of most of the contaminants has been diminished in about an order of magnitude.

	I-131	**Ru-103**	**Sb-125**	**Cs-137**	**Sr-90**	**Gross Alpha**	
HEU	2,41E-06	2,77E-07	7,50E-09	2,95E-10	6,88E-09	1,95E-11	n=28
LEU	1,50E-07	1,67E-08	5,68E-10	3,62E-10	7,88E-10	1,21E-11	n=46

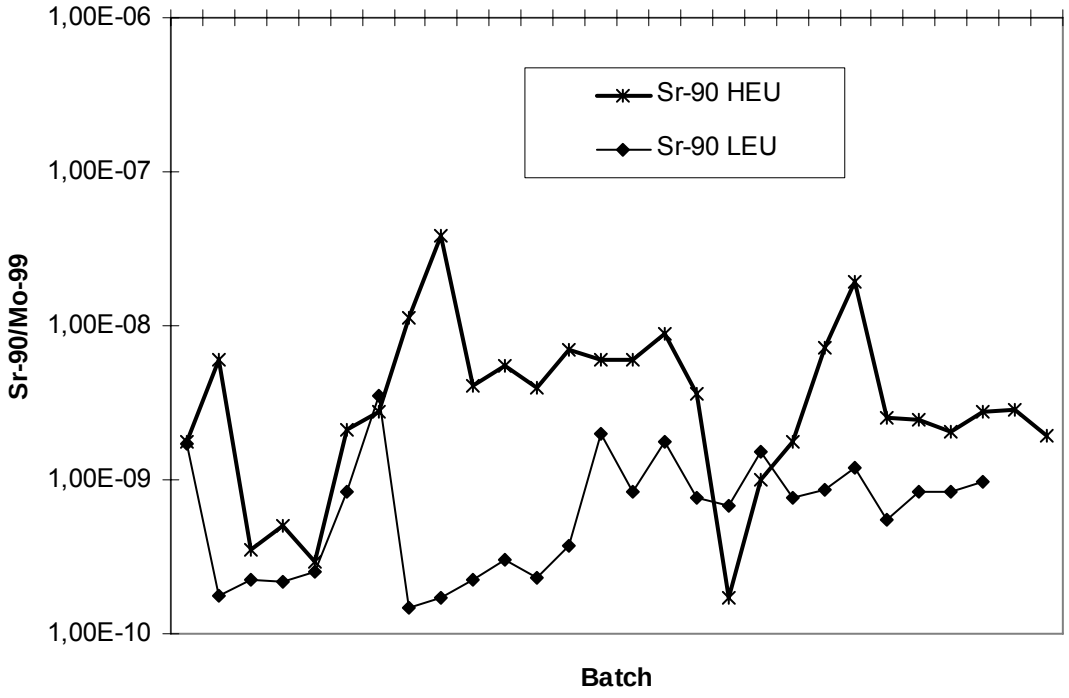
Table 4: Average values



Graph. 1: I-131 content (log scale)



Graph. 2: Ru-103 content (log scale)



Graph. 3: Sr-90 content (log scale)

It is interesting to note that, practically, there is no difference between the gross alpha content in Mo-99 produced from either target. The reason could be that almost all results are expressed as minimal detectable activity (MDA). These results refute the concern that the increase of total uranium mass in LEU targets could affect gross alpha content.
Other distinctive behavior found in LEU batches is the lower standard deviation in any purity parameter, which shows the reliability of the process.

Conclusions

There are two important conclusions. The first one is that it is feasible to produce fission Mo-99 from LEU targets and the second one is that it is possible to obtain a product that complies with the international requirements in order to prepare medical devices (generators) for producing radiopharmaceuticals (labeled with Tc-99m) in accordance with specifications established in the most important Pharmacopeias.
These results should serve to realize that the potential future positioning of LEU targets into the market, for fission Mo-99 production, should be based on economical or political factors and not in quality consideration.

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