HEU AND LEU COMPARISON IN THE PRODUCTION OF MOLYBDENUM-99

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ABSTRACT

Fission Molybdenum-99 from LEU targets is being produced in Argentina, at the Ezeiza Atomic Centre, since 2002.
Before 2002, Argentina produced for more than 15 years fission molybdenum-99 from HEU targets.
Both production procedures involve the irradiation of the targets composed by an Uranium-Aluminium compound "meat" cladded with aluminum and a chemical processing of the targets.
A statistic relative efficiency analysis of the production results, a brief description of the LEU method and quality control data of both procedures will be presented.

Introduction

Molybdenum-99 is a radionuclide widely employed in nuclear medicine practices. For this reason, the production of fission Mo-99 always was an important concern for CNEA.
Because current international policies seek to reduce uranium enrichment in civil uses, Argentina changed in 2002 its Mo-99 production from HEU to LEU.
Before 2002, Argentina produced for more than 15 years fission Mo-99 from HEU targets, getting most of the experience that later on allowed the change from HEU to LEU.
The aim of this work is to compare the production efficiency from several batches of fission Mo-99 produced routinely in Argentina from both LEU and HEU targets.

Fission Isotopes Production Facility

The facility for the production of Mo-99 from LEU targets is the same that was used for the Mo-99 production from HEU targets, and is located at Ezeiza Atomic Center, neighbor to RA-3 reactor [1]. At the beginning, only a few changes had to be done to ensure that there were no chance of mixtures between the HEU and the LEU. Later on, all the HEU was transferred to another facility waiting for final disposition.
Production facility has two groups of four hot cells each. The first group, where digestion of targets is performed, has a shielding of 30 cm of lead. The other group, employed in the purification stages, has 20 cm of lead walls.

All the cells have α β γ tight stainless steel boxes with epoxi cover.

Low Enriched Uranium/Aluminium targets

Targets were described in previous papers, as well as the tests that led to final dissolving conditions [2].
Targets are irradiated in the RA-3 reactor core during 108 hours with a neutron flux of about $2x10^{14}$ n/cm2.seg and ten hours of cooling in reactor pool.
Transportation from RA-3 is accomplished through an internal corridor, with a motorized shielding of 23 cm of lead with a capacity for carrying up to four targets.

Digestion of LEU targets

Processing of the targets is based on the method developed by Dr. Sameh Ali at KfK in Germany for Molibdenum-99 production [3][4].
Irradiated plates are dissolved in hot alkaline solution [5] [6] [7].
Hydrogen and fission gases produced are kept in evacuated charcoal filled tanks and, after four weeks of decayment, they are released to the atmosphere of the hot cells.
When dissolution is completed, solution is filtered through a sintered stainless steel plate, leaving non-fissioned uranium in the precipitate.
The resulting solution is composed by Molybdenum-99, Iodine-131 and the other fission products soluble in alkaline medium.
The precipitate is composed by non-fissioned uranium and fission products (actinides, etc) that are insoluble in alkaline medium.
Mo-99 is present in the filtrate as molybdate anion.

Purification of Molybdenum

As first stage of purification, alkaline solution is fed through a column of ion exchange resin where some fission products, soluble in alkaline medium, pass through.
Mo-99 is retained in this column, together with some other fission products.
Then, the Mo-99 is stripped and transferred to a different hot cell where new stages of the molybdenum purification are carried on.
Two more columns of ion exchange resin and a last column of an inorganic exchanger are employed to accomplish the correct quality of the product.
Finally, the solution is delivered to another facility for quality control and dispatch.

Relative Efficiency's Productions Results

For the production relative efficiency analysis, we have considered the period between 1998 to October of 2002, for the HEU process, and between August of 2002 to 2006, for

the LEU one. These are nine straights years of CNEA's experience in the production of molybdenum.
In order to avoid non comparison data, all the batches obtained with an uncompleted cycle of irradiation or a different configuration of the irradiation period, were not considered, trying to compare, in the best possible way, both targets and processes.
Results from 103 HEU and 135 LEU batches of fission Mo-99 produced between 1998 and 2006 are considered in this work.
As a first stage, we calculated, with the total production activity and the mass of U-235 used in this batch, the specific activity for each batch, and then we obtained the mean of the specific activity for each year.

$$\overline{E}_j = \frac{\sum_{i=1}^{n} \frac{\text{Activity Produced in Batch i}}{\text{total grams of } U^{235} \text{ used in Bach i}}}{n} \qquad \left[\overline{E}\right] = \frac{Ci}{g\,U^{235}}$$

n = number of batches considered in the year

j = number of years considered for each technology

For a better comparison, a "factor" was created in order to relate the different average specific activities in each year with the best production result obtained with HEU. In this way, it is possible to read directly the increases or decreases in the relative production efficiency in each year compared with the best HEU result.
The factor "f" has the following form:

$$f_k = \frac{\overline{E}_k}{\overline{E}_{MAX(HEU)}}$$

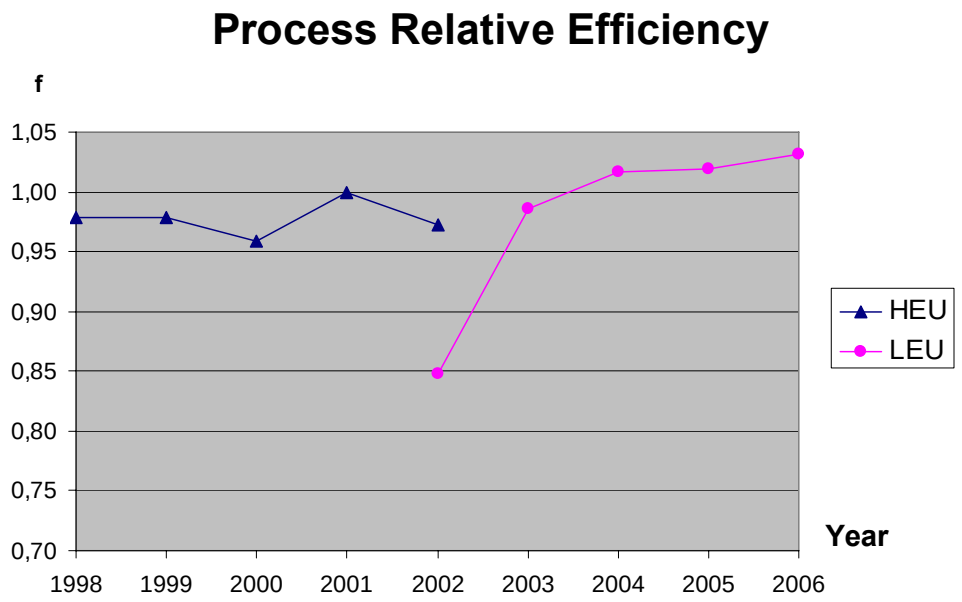
k = number of years considered for each technology

The $\overline{E}_{MAX(HEU)}$ was in 2001, so this is the value against we are going to compare the rest of the periods considered.

Calculating f_k for each year:

Year	f_{HEU}	nº batches considered
1998	0.98	27
1999	0.98	31
2000	0.96	34
2001 (base)	**1.00**	32
2002	0.97	21

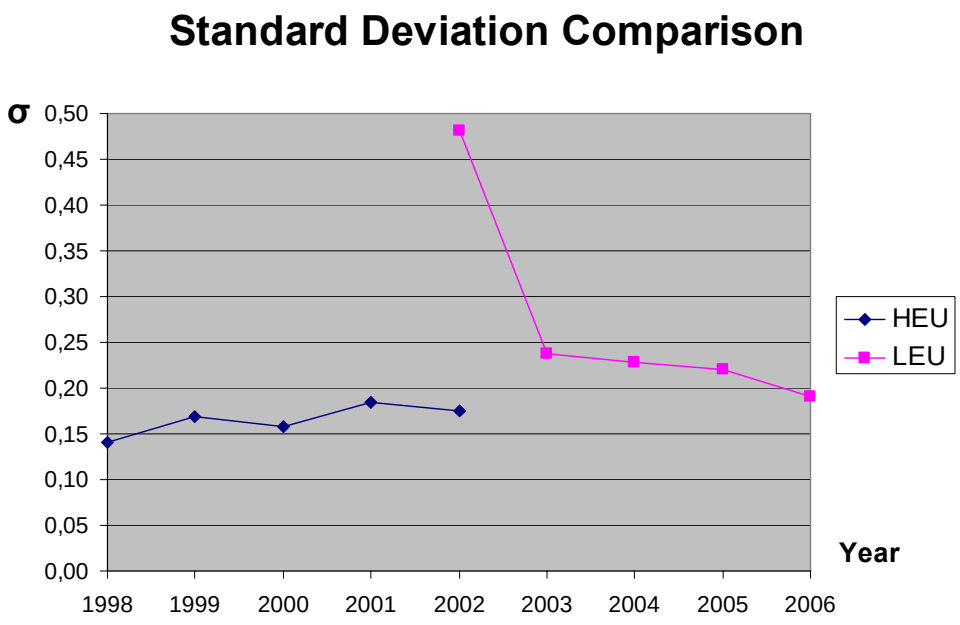
Year	f_{LEU}	nº batches considered
2002	0.85	15
2003	0.99	44
2004	1.02	33
2005	1.02	42
2006	1.03	43



To have an idea of the variability of the whole production process, which involves the irradiation of the targets and the chemical processing, the standard deviation of each period was calculated.

Year	σ_{HEU}	nº batches considered
1998	0.14	27
1999	0.17	31
2000	0.16	34
2001	0.19	32
2002	0.17	21

Year	σ_{LEU}	nº batches considered
2002	0.48	15
2003	0.24	44
2004	0.23	33
2005	0.22	42
2006	0.19	43



Quality Results:

In order to have an idea not only of the efficiency but also the quality of both technologies, a table of a previous CNEA's work is showed [8]. Results form 28 HEU and 46 LEU batches of fission Mo-99 produced between 2002 (HEU) and 2003 (LEU) were used in that work.

	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

It is clear that in the LEU case, the concentration of most of the contaminants has been diminished in about an order of magnitude.
With the intention of comparing results, the international specifications are showed.

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	$< 5 \times 10^{-7}$

Conclusions:

There is a huge concern around the world about if it is possible to achieve the same efficiency and quality standards using LEU and HEU targets to produce radioisotopes.
In a previous work presented in 2004's RERTR, the main conclusion was that the radionuclide quality of Mo-99 from LEU targets is as good as with HEU ones and even better.
The new and most important conclusion in this work is that it is feasible to produce fission Mo-99 from LEU targets at least with the same production relative efficiency, in terms of produced activity related with the use of U-235, than with HEU targets. In our case, the results show that a little improvement was achieved after a period of LEU's technology adaptation to production scale.
Another important conclusion related with the statistic analysis of the sample, is concerning the standard deviation. For the correct analysis, first you have to find out if the variability is caused by special or common reasons, because the way to deal with them are quite different. By common causes or reasons are understood those variability's sources that are under statistic control. That means that all the values are between the limits of the normal

variability, and these limits generally are calculated adding and subtracting three standard deviation to the mean.

When there are only random variability's causes in the process, the results show a stable and predictable distribution upon the time. On the contrary, the presence of special variability's causes is evidenced by unpredictable results.

Analyzing the batches obtained during nine years, the results show, with the 2002`s LEU exception, that all the values are between two standard deviation, indicating that the process is under statistic control. The 2002`s LEU exception matches with the LEU's technology adaptation to production scale.

The solution of these random variability's causes involves, first of all, identifying them, and then, take the corrective actions, but only those ones that consider the process or the system as a whole, as these common causes are peculiar and inherent of each technology.

The analysis of which are the contributors to the standard deviation of each technology is not the purpose of this work, and could be analyzed in future papers.

The main conclusion of this paper is that the "behavior", in terms of efficiency and stability of the CNEA's LEU process, is quite similar to the HEU one, and we hope that these conclusions can help the other Mo-99 producers, to have a mark of reference for a correct transition in reducing the use of high enriched uranium.

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