RERTR 2010 — 32nd INTERNATIONAL MEETING ON
REDUCED ENRICHMENT FOR RESEARCH AND TEST REACTORS

October 10-14, 2010
SANA Lisboa Hotel
Lisbon, Portugal

2010 NATIONAL PROGRESS REPORT ON R&D ON LEU FUEL AND TARGET TECHNOLOGY IN ARGENTINA

S. Aricó, S. Balart, A. Bonini, P. Cristini, L. de Lio, L. Dell'Occhio, D. Gil, A.G. Gonzalez, R. González, C. Komar Varela, M. López, M. Mirandou, H. Taboada
COMISIÓN NACIONAL DE ENERGÍA ATÓMICA
Av. Del Libertador 8250 (1429) Buenos Aires, Argentina

ABSTRACT

After the core conversion of the RA-6 reactor in 2008, an extension of the original CNEA-NNSA DoE contract was signed to enhance the final national HEU inventories minimization. This contract extension, done in the frame of the GTRI, mainly implies the recovery, purification and down-blend to LEU of almost all the fresh and irradiated HEU remnant inventories coming from fuels and Mo99 irradiation targets fabrication and production filters. A small inventory of HEU for R&D uses in fission chambers, neutronic probes and standards was reserved.

CNEA is also developing the fabrication of both dispersed U-Mo (Al-Si matrix and Al cladding) and monolithic (Zry-4 cladding) miniplates to support the qualification activities of the RERTR program. Some monolithic 58% enrichment and LEU 8%Mo and U10%Mo miniplates and plates were and are being delivered to INL-DoE to be irradiated in the ATR reactor core.

CNEA, a worldwide leader on LEU technology for fission radioisotope production is providing Brazil with 1/3 of the national requirements on Mo99 by weekly deliveries. Australia has started the fission radioisotope production through several batches by week, based on CNEA's LEU technology provided by INVAP SE. CNEA is also committed to improve the diffusion of LEU target and radiochemical technology for radioisotope production and target and process optimization. Future plans include:

- Plans to recover and purify the LEU based inventories in Mo99 production filters, once the HEU to LEU campaign is over.
- Fabrication of a LEU dispersed U-Mo fuel prototype following the recommendations of the IAEA's Good Practices document, to be irradiated in a high flux reactor in the frame of the ARG/4/092 IAEA's Technical Cooperation project.
- Development of LEU very high density monolithic and dispersed U-Mo fuel plates with Zry-4 or Al cladding as a part of the RERTR program.
- Optimization of LEU target and radiochemical techniques for radioisotope production.

1. **HEU minimization in Argentina.**
   In March, 2010 a supplementary agreement to the contract DE-SC55 05NA25735 between CNEA and NNSA-DoE was signed by both parties. It includes the completion of several tasks related to the recovery, purification and blending down of fresh and irradiated inventories contained in scraps from fuel and target fabrication and in fission Mo99 production filters. These tasks are taking place and the expected deadline is December 2012. The materials are classified into 6 groups, as it is shown in Table 1

Table 1: HEU remnants classification

Description	Group Number	Form	U Mass (kg)	Enrichment	235 U Mass (kg)
Irradiated Mo-99 Targets And Solutions	1	Solid and Liquid	1.928	89.73%	1.73
UF6 Cylinder	2	Gas / Solid (UO2F2)	0.65	90.14%	0.59
Miscellaneous Solids (alloys, metal)	3	Solid	0.397	87.15%	0.346
Miscellaneous Solutions	4	Liquid	0.228	89.91%	0.205
Materials declared waste to dispose	5	Solid	0.505	89.97%	0.453
Ingot for MEU-Mo/Zr Miniplates Fabrication	6	Solid	0.344	88.66%	0.305
TOTAL			4.05		3.63

Group 1 comprises the refurbishment of a radiochemical laboratory (LFR lab), licensing of two transport casks, for irradiated solutions and solids contained in cartridge filters, among of the proper recovery, purification and downblending of the HEU inventory.

Group 2 comprised the opening of a 5A type cylinder containing a partial hydrolized UF6 inventory, which valves were stucked and the recovery, purification and downblending of the HEU inventory into LEU. This task is already finished[I]

Group 3 implied the downblending of an HEU-Al inventory through cast melting. This task was finished during September, 2010.

Group 4 is made of several HEU solutions in acqueous and organic media. It is being characterized and during October 2010 will be downblended.

Group 5: a consultation of the Argentinean Regulatory Authority (ARN) to IAEA on a specific proposal to dispose this inventory was made. After the answer this inventory will be disposed.

Group 6 includes the blending down of a HEU ingot to 58% enr. U to make ME U-10%Mo and U-8%Mo based miniplates with Zr cladding. The agreement includes the exportation to INL of miniplates for irradiation testing under the RERTR program. This task is being accomplished with a first delivery in April, 2010 and a final second one scheduled in October 2010[II]. A special work about is presented during this meeting (see Lopez et al.). Finally, the blending down of the remaining material to LE U-10%Mo and U-8%Mo for full scale plate fabrication.

2. R&D on VHD fuels

a. Investigations of the interaction between U(Mo) and Al(Si) alloys*.*

Investigations of the interaction between U(Mo) and Al(Si) alloys done during the last years gave rise to new lines of work. To increase the knowledge of the interaction layer between U(Mo) and Al(Si) alloys, systematization is considered very important at this stage. So, diffusion couples with one U(Mo) alloy and a series of binary Al(Si) alloys will be studied to obtain information about the presence or absence of different phases in the interaction layer, in relation with the Si concentration. Up to now, six binary Al-Si alloys with Si concentration of 0.6, 2.0, 4.0, 5.2, 6.0 y 7.1 wt% and a U-9Mo alloy are already melted, and stabilized at the temperature to be used. A new device is being tested to fabricate the diffusion couples instead of the stainless steel clamps used before. With this new device, external variables (as pressure or heat removing during cooling) will be the same for all the diffusion couples making more reliable comparisons among them.

b. Fall-back option: stability of the γU-phase in U-Zr-Nb alloys

The aim of this work is to optimize the parameters of the fabrication process of a high-density monolithic-type nuclear fuel with U-Zr-Nb alloys as meat and different possible claddings such as Zry-4 or Al 6061. The first step is to define a range of concentrations in which the γU-phase doesn't decompose during this fabrication process.

Ten U-Zr-Nb alloys with concentrations between 13.9-43.7 wt.% Zr and 0-6.4 wt.% Nb were melted. The $\gamma U \rightarrow \delta UZr_2$ and $\gamma U \rightarrow \alpha U$ transformations are being evaluated by measurements of thermo-electrical resistivity with cooling rates between 4-120 ºC/min.

First results indicate that, with a cooling rate of 4ºC/min, a Nb concentration of at least 17.8 wt. % would inhibit the transformation $\gamma U \rightarrow \delta UZr_2$ while 23.3 wt % Nb would inhibit the γU → αU transformation. It's expected that a higher cooling rate would lead to a lower Nb concentration to retain the γU-phase.

3. **Largest contribution of CNEA to HEU minimization: LEU target and radiochemical technology for Mo99 and other radioisotopes production:** to assess the supply to the Argentinean fission radioisotope domestic market under the final cutoff to HEU supply, CNEA turned in 2001 into LEU material for target fabrication, maintaining other characteristics of the production like the alkaline chemical digestion process. CNEA began producing Mo-99 using HEU targets in 1985[III] and developed and converted to LEU-based production in 2002. CNEA manufactures its own uranium aluminide alloy plate LEU targets[IV].
   - CNEA has developed and is using high-density LEU-aluminum dispersion targets. The target meat has a density of 2.9 gU/cm3 obtained by increasing the ratio of uranium aluminide to aluminum in the target meat. The mass of U-235 in the target meat is about twice that of conventional uranium-aluminum alloy targets.
   - CNEA was able to convert to LEU-based production in the same set of hot cells that were being used for HEU-based production, without interrupting Mo-99 production
   - Targets are irradiated in the RA-3 reactor at CNEA's Ezeiza Atomic Center near Buenos Aires. Target processing is carried out in a hot cell facility at the Ezeiza site. Process wastes are also managed at the site.
   - CNEA's development showed that there are no technical barriers to conversion of Mo-99 production from HEU targets to LEU targets. Production using LEU targets is technically feasible and is being carried out by CNEA in Argentina and will be shortly by the Australian National Nuclear Science and Technology Organisation (ANSTO) using CNEA technology using CNEA-developed LEU targets and target dissolution process to produce Mo-99.
   - This new LEU technology satisfies the most stringent requirements of quality for its use in nuclear medicine applications. Mo-99 purity has been consistently higher than that produced using HEU targets[V]
   - Also in September 2005, CNEA began the regular production of high quality fission I-131, a by-product of Mo-99 production, meeting also international quality standards.
   - New results are that HEU-LEU production process comparison costs reveals that this new technology has no significant over cost [VI]. Overall costs for LEU-based production compared to HEU-based production increased by about 5 percent.
   - Since CNEA has duplicated the LEU-based radioisotope weekly production rate to provide Mo99 to Brazil.
   - Conclusions: no technical, quality or financial reasons make disadvantageous to change from HEU to LEU radiochemical technology for Mo99 and other radioisotopes production. CNEA becomes a leader in LEU based isotope production technology: the production plant built up with CNEA's technology in Australia by INVAP is producing several batches of RI per week, and a similar one in Egypt is scheduled to start production during 2010. Future

plans: at present, CNEA is expanding Mo-99 production within its current facilities by increasing target throughputs.

.

4. **Conclusions:**

FINAL HEU MINIMIZATION: CNEA is minimizing the remnants HEU inventories, both fresh and irradiated from fuel and target fabrication scraps and fission RI production solutions and filters. All these tasks are scheduled to finish during December 2012.

R&D ON LEU VHD FUELS: CNEA is actively supporting both R&D activities to achieve solutions for core conversions.

LEU TECHNOLOGY FOR FISSION RI PRODUCTION: No technical, quality or financial reasons make disadvantageous changing from HEU to LEU for fission Mo99 and other RI production. CNEA leads LEU based isotope production technology, and with INVAP built all LEU-based production systems in Australia and Egypt. This is by far the largest contribution of CNEA to the HEU minimization for civilian uses.

[I] L. N. Aldave, H. Blanco Bello, A. A. Bonini, L. I. De Lio, L. A. Dell'Occhio, M. Falcón, T. Feijoo, A. Gauna, D. A. Gil, A. Rodriguez y J. Valdez. 2010 RERTR International Meeting, Lisbon, Portugal, 10-14 October 2010.

II M.López, A. González, R. González, F. Rice, H. Taboada, D. Wachs. 2010 RERTR International Meeting, Lisbon, Portugal, 10-14 October 2010.

III Cols, H. J., P. R. Cristini, and A. C. Manzini. 2000. Mo-99 from low-enriched uranium. 2000 International RERTR Meeting, Las Vegas, Nevada, USA, 1-6 October, 2000. Available at http://www.rertr.anl.gov/Web2000/PDF/Cristi00.pdf

IV Kohut, C., M. de la Fuente, P. Echenique, D. Podesta, and P. Adelfang. 2000. Target development of low enrichment for production of 99Mo for fission. 2000 International RERTR Meeting, Las Vegas, Nevada, USA, 1-6 October, 2000. Available at http://www.rertr.anl.gov/Web2000/PDF/Fuente00.pdf

V Durán,A. 2005. Radionuclide Purity of Fission Mo-99 Produced from LEU And HEU. A Comparative Study. 2005 International RERTR Meeting, Boston, Massachusetts, USA, November 6-10, 2005. Available at http://www.rertr.anl.gov/RERTR27/PDF/S8-3_Duran.pdf.

VI Cestau D., A. Novello, P. Cristini, M. Bronca, R. Centurión, R. Bavaro, J. Cestau, E. Carranza. HEU and LEU cost comparison in the production of molybdenum-99. 2008 International RERTR Meeting, Washington, DC, USA, 5-9 October 2008, and Cestau D., A. Novello, P. Cristini, M. Bronca, R. Centurión, R. Bavaro, J. Cestau, E.Carranza. 2007. HEU and LEU comparison in the production of molybdenum-99. 2007 International RERTR Meeting, Prague, Czech Republic, Sep. 23-27, 2007. Available at http://www.rertr.anl.gov/RERTR29/PDF/6-4_Cestau.pdf