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## 2009 Progress Report on RERTR Activities in Argentina

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### ABSTRACT

Since last RERTR meeting, CNEA has deployed several related tasks. The RA-6 MTR type reactor, converted from HEU to a new LEU silicide core is scaling up the power, according to a protocol requested by ARN.

CNEA is deploying an intense R&D activity to fabricate both dispersed U-Mo (Al-Si matrix and Al cladding) and monolithic (Zry-4 cladding) miniplates to develop possible solutions to VHD dispersed and monolithic fuels technical problems. Some monolithic 58% enrichment U8%Mo and U10%Mo are being delivered to INL-DoE to be irradiated in ATR reactor core. A thoroughful study on compound interphase formation in both cases is being carried out. 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:

- 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 U-Mo fuel plates with Zry-4 cladding as a part of the RERTR program.
- Plan for future irradiation of full scale VHD fuel plates as part of standard assemblies in RA-3 reactor core.
- Optimization of LEU target and radiochemical techniques for radioisotope production.

### 1. RA-6 reactor with its new LEU core: from restart to its regime power

The RA-6 reactor is a pool-type one sited at San Carlos de Bariloche city, Province of Río Negro, Argentina, and its core was recently converted into a new LEU-based one<sup>1</sup>. This successful conversion process started in October 30<sup>th</sup>, 2005 with the signature of two contracts between CNEA and NNSA-DoE and comprised swapping of HEU-LEU inventories, exportation of HEU SNF US-origin to USA, and fabrication of the

conversion core and new graphite reflectors and improvements on primary and secondary loops. During 2009 the following steps took place:

- Criticality start-up operations, started on January 19<sup>th</sup>, 2009.
- Formal re-inauguration took place on March 16<sup>th</sup>, 2009.
- Progress on first step to 1 MW in power scaling-up towards 3 MW, according to a plan authorized by ARN, the national nuclear regulatory agency.

Once completed the first step, activities as research on BNCT applications for melanoma treatment and Si doping by neutron capture will be full operational.

## **2. Applied R&D on dispersed and monolithic U-Mo fuels.**

- The final analysis on the interaction U(Mo,1%Zr)/Al (7%Si) developed in out-of-pile tests using high intensity synchrotron X-rays diffraction techniques performed in the LNL Campinas, Brazil, was carried out. It showed the precipitation of Zr as Zr<sub>3</sub>Al<sub>5</sub> in the interaction zone.
- Other research activities already performed are the irradiation of a set of microplates in the RA3 reactor and computational methods applied to thermodynamic and kinetic calculations. Concerning calculations, a thermodynamic database was built to calculate phase equilibria. The diffusion problem was simulated using the DICTRA package which articulates thermodynamic data with a mobility database. The equilibria obtained in the Al-U system and a first model of UAl<sub>3</sub>/Al diffusion couple that successfully simulates the growth of the Al<sub>4</sub>U phase are presented in the poster session.

## **3. Development and irradiation of promissory solutions to VHD monolithic and dispersed fuels technical problems**

- Improvements in the development of dispersed and monolithic fuels made with Uranium-molybdenum alloy were done. The purpose is to have additional alternatives to cover HEU-LEU conversion possibilities.
- CNEA is working in the fabrication of depleted U-7Mo based dispersed miniplates. Once this process is stable and repetitive, the fabrication of a LEU prototypic fuel assembly will follow. To enhance the expertise of human resources in rolling techniques, the technical visit of Dr. Wiencek to CNEA laboratories, an US-ANL expert was produced. This visit was done through the IAEA'S Technical Cooperation MANPOWER program.
- The project ARG/4/092, in the frame of IAEA'S Technical Cooperation, looks for the irradiation in a high flux reactor and PIE a full scale LEU-Mo/Al-Si based dispersed fuel assembly prototype. Procurement activities are finished and the contract with the irradiation and PIE services provider will take place soon.
- In order to avoid an undesirable porosity in the aluminium side of the interaction zone with U-Mo due to the migration of gas fission products, and according to studies done on the convenience to add a proper component to matrix powder, Al-Si alloys are being tested. After results, fabrication conditions of dispersed U-Mo plate full size will be obtained.
- Concerning very high density monolithic MEU-Mo miniplates with Zry-4 cladding to be delivered to INL-DoE for irradiation in the ATR Reactor core, are being fabricated.

CNEA is also refurbishing the laboratory and mounting new equipment to develop full size plates.

- In order to improve material performance and plate dimensioning fabrication conditions were studied and modified, like hot co-lamination of U-Mo and Zry-4 sheaths. Several depleted uranium prototypes were elaborated, characterized and tested to set up process variables and fabrication conditions.

#### **4. Improvement of the LEU target and radiochemical technology for Mo99 and other radioisotopes production.**

- It was already presented that CNEA has decided on 2001 to turn into LEU material for target fabrication, due to the final cutoff in HEU provision. It was done maintaining other characteristics of the production, i.e. the alkaline chemical digestion process. CNEA has been producing Mo-99 using LEU since 2002. CNEA produces Mo-99 primarily for its domestic market and also for export to other South American countries. It began producing Mo-99 using HEU targets in 1985<sup>[ii]</sup> and developed and converted to LEU-based production in 2002. CNEA manufactures its own U-Al alloy plate LEU targets<sup>[iii]</sup>.
- CNEA has developed and is using high-density LEU-aluminum dispersion targets (density = 2.9 gU/cm<sup>3</sup>) to produce Mo-99 for its domestic market. The mass of U-235 in the target meat is about twice that of conventional U-Al alloy targets.
- 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 converted to LEU-based production using the same set of hot cells that were being used for HEU-based production. Moreover, no interruption in Mo-99 production was made.
- 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 by the Australian National Nuclear Science and Technology Organisation (ANSTO) using CNEA's technology: 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<sup>[iv]</sup>
- 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.
- HEU-LEU production cost comparison reveals that this new technology has no significant over cost. CNEA recently presented a comparison of its variable costs for producing Mo-99 using LEU and HEU targets<sup>[v]</sup>, where variable costs for Mo-99 production ( HEU: 1998-2001; LEU: 2003-2007) were compared. Costs were presented in three categories: (1) labor; (2) materials; and (3) services, maintenance, taxes, and miscellaneous, normalized on a per curie basis for the number of curies produced in 2007. Overall costs for LEU-based production compared to HEU-based production increased by about 5 percent.

- This year CNEA has duplicated the LEU-based radioisotope weekly production rate to provide Mo99 and other radioisotopes 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, and with INVAP built all-LEU production systems in Australia and Egypt
- Future plans: at present, CNEA expanded Mo-99 production within its current facilities by increasing target throughputs. Such an expansion would put CNEA in the ranks of large-scale producers

Conclusions: CNEA continues deploying development activities on LEU technology for core reactor conversion and Mo99 and related radioisotope production. Future plans include prototypic fuel irradiation and optimization of LEU targets and alkaline digestion process.

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<sup>i</sup> Balart, S; Cristini, P; Fernández, C; González, AG; López, M; Taboada, H. 2009 CNEA Progress Report on the Development of LEU Fuels and Targets in Argentina. 2009 RRFM International Meeting, Vienna, Austria, 22-25 March, 2009

<sup>ii</sup> 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>

<sup>iii</sup> Kohut, C., M. de la Fuente, P. Echenique, D. Podesta, and P. Adelfang. 2000. Target development of low enrichment for production of <sup>99</sup>Mo 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>

<sup>iv</sup> 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](http://www.rertr.anl.gov/RERTR27/PDF/S8-3_Duran.pdf).

<sup>v</sup> 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](http://www.rertr.anl.gov/RERTR29/PDF/6-4_Cestau.pdf)