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Progress on RERTR activities in ARGENTINA

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ABSTRACT

During 2006 several activities related to RERTR topics are taking place: the progress on the conversion project of the RA-6 core reactor from HEU to LEU, where CNEA has minimized the HEU inventory plates by swapping them for LEU material to fabricate the new LEU core; LEU U-Mo and UZrNb monolithic miniplates using Zry-4 cladding are being produced for its irradiation in ATR and RA-3 reactors, in the first case as a part of the RERTR 9 irradiation experiment. U-Mo/Al-Si and U-Mo/ Zry-4 compounds using atomistic modeling prediction (BFS techniques and first principles) were studied, finding some trends on the interaction phases, which were found on diffusion couples tests and were characterized.
Cold process to treat and recover actinides from Zry-4 cladding U-Mo monolithic miniplates was started and has promissory first results.
Progress on LEU technology for radioisotope production: since September 2005, CNEA began the regular production of high quality fission I^{131}, a by-product of Mo-99 production, meeting international quality standards.

1. Introduction

During 2006 RERTR R&D activities in CNEA were focused on

- The progress on the RA-6 reactor core conversion project. from HEU to LEU one,
- Continuing on a deeper and comprehensive understanding of U-Mo/Al and U-Mo/Zry-4 alloys interaction zone formation in dispersed and monolithic fuels,
- Developing promissory solutions for VHD monolithic and dispersed fuels,
- Developing cold test on actinides separation for final conditioning of Zry-4 coated U-Mo monolithic plates by recovering U from the chopped cladding.
- The progress on LEU target and radiochemical technology for radioisotope production: since September 2005, CNEA began the regular production of high quality fission I^{131}, a by-product of Mo-99 production, meeting international quality standards. A special work will be presented in technical sessions

2. RA-6 conversion

RA-6 reactor is a pool-type one with a thermal power of 0.5MW, sited in Bariloche Atomic Center, Provincia de Rio Negro. At present and since its inauguration in1983 it is working with a HEU core. On October 30th, 2005 two contracts between CNEA and NNSA-DoE were signed to give place to its conversion and to take part of the FRR SNF program. Several tasks took and are taking place:

- As part of the minimization of the use of HEU in Argentina, a swapping of HEU (Argentina)-LEU (USA) materials was agreed and done on July 17th, 2006
- Fuel design and fabrication of new silicide based LEU core for RA-6 reactor and neutronic reflectors are in progress.
- Construction of interim storage facilities to store LL and MLW was rescheduled for austral summer 2006-7
- Removal and conditioning of HEU core were rescheduled for austral spring 2007
- Transportation of HEU fuels were rescheduled for austral spring 2007

3. Very High Density fuel development

An intense activity both on dispersed and monolithic VHD fuels are taking place

- The development of UNbZr monolithic fuels with Zry-4 cladding continued. The phase diagram zone where the ternary UZrNb retains the metastable gamma phase was characterized. The work is focused on testing such specimens[1].
- 2 CNEA´s Zry-4 cladding UMo monolithic miniplates were irradiated at ATR reactor (INL), as a part of RERTR 7 irradiation experiment. One of them achieved 80% burnup.[2]
- Plans to irradiate a full scale Zry-4 cladding UMo monolithic FA in the RA-3 reactor are in progress
- The IAEA´s TC irradiation project named ARG2005002 of a dispersed UMo in Al-Si matrix FA in a high flux reactor was approved. It will run during 2007-8 in parallel to an irradiation alike in RA-3 reactor.
- A contract between BEA(LLC)(INL)-CNEA for the provision of several U-Mo and UZrNb monolithic miniplates for irradiation during RERTR 9 experiment was signed and is operative.
- The codes PLACA/DPLACA designed to describe the irradiation behavior of plate-type fuels under normal operation conditions were developed. They contain about thirty interconnected and mutually dependent models. The growth of an interaction layer around fuel particles in dispersed fuels was modeled assuming that the kinetics of both layer boundaries is determined by diffusion of U and Al through the layer. The associated Stefan problems are numerically solved. Both codes simulated several irradiation histories. Comparison of the calculation results to the experimental data showed a correct performance of the models involved and a good coupling of the ensemble.[3]

4. Characterization of phases in U-Mo/Al – U-Mo/Zry-4 alloys interaction zone

- o Theoretical calculation using BFS (Bozzolo, Ferrante Smith, 1992) method for determining the atomic system energy as a function of it geometrical configuration was performed. This provides a virtual model of an alloy formation process. This methodology was applied to compare UMo/Al – UMo/Zr interaction zones behavior. Simulations showed that instead of the vast interdiffusion zone presented in the first case, in UMo/Zr case predict that the presence of Mo inhibits but does not avoid the Zr diffusion although the trend exhibited at high temperature suggests the formation of an stable small interaction zone[4].

- o Characterization of phases in UMo/ Al alloys interaction zone: during 2006, work was related to study the effect of the consumption of Si in the Al alloy while interdiffusion is in progress. U-7w%Mo / Al-7w%Si diffusion couples, specially prepared for this objective (fig.1) were made by FSW. In addition to the results previously reported at 550ºC, prolonged treatments at 340 ºC were performed and analysed. Results do not show clearly that the condition of no Si in the Al alloy was reached during this last treatment. The irradiation plan continues as scheduled. PIE are intended to be started in 2007.[5]

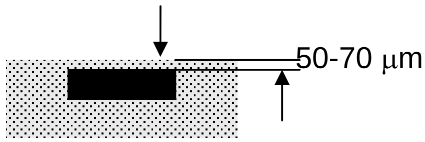

Fig.1

- The pseudo-binary phase diagram USi_3-UAl_3: The formation of $(U,Mo)(Al,Si)_3$ has been shown in diffusion couples of U-7%wt Mo and Al-5.2wt%-7.1 wt% Si (out-of-pile experiments, temperature 340º-580ºC. The Si/Al ratio within this phase was found to be 1/3 and no $(U,Mo)Al_4$ was found. Recently, the Russian researchers have shown that the intermetallic $U_4Al_9Si_3$ (Si/Al ratio = 1/3, cubic lattice, elementary cell of 256 atoms) coexist in equilibrium with the $U(Al,Si)_3$ phase below 1340°C. It was evaluated the thermodynamic functions of the pseudo-binary and the UAl_4 compound by using first principles calculated total energies in a cluster expansion method. The minimal cluster expansion exhibits a good convergence and predicts the experimental ground state with the stability of the $U_4Al_9Si_3$ compound. It is also shown that the $U_4Al_9Si_3$ compound containing more than 8at.% Si interacts only with Al without UAl_4 phase formation[6]
- $U(Al,Si)_3$ stabilization by Zr addition: promising results have already been obtained showing that Si addition to Al matrix is able to inhibit UAl_4 formation. It was also noticed that minor Si quantities should be required in the presence of fourth element collaboration: Zr is an already suggested candidate. Stabilization experiments in 50U49.9Al0.1Si alloys with 0, 1, 3 and 6% Zr addition (weight percentages) were performed. Heat treatments at 600°C (100h and 1000h) were undertaken and results were analyzed by x-Ray diffraction, metallographic and composition measurement techniques. Slight evidence of UAl_4 presence was still found in heat treated samples with 6%Zr content, although UAl_4 related x-Ray peaks intensities diminish with higher Zr contents. It suggests that this way enables the retard of the peritectic reaction that leads to UAl_4 formation.[5]

5. Cold test for silicide and U-Mo Zry-4 coated SNF final conditioning

- Silicide production scrap recovery: the development presented in former RERTR meeting was finished. It is an important step to have available a process to separate actinides and some radionuclides for nuclear medicine applications for final conditioning of spent fuels. U-Mo monolithic Zry-4 coated scrap recovery: first results using a chopping machine to cut, process and U recovering were satisfactory[7]

6. Improvement of the LEU target and radiochemical technology for Mo99 and other radioisotopes production.

- Due to the final restriction on the supply of HEU material for the production of fission Mo99, CNEA has decided on 2001 to turn into LEU material for target fabrication.
- It was done maintaining other characteristics of the production, i.e. the alkaline chemical process. CNEA achieved successfully an adequate replacement meat.
- Our LEU technology satisfies the most stringent requirements of quality for its use in nuclear medicine applications.
- This LEU technology was sold to ANSTO (Australia) and Egypt’s EAEA.

- Since September 2005, CNEA began the regular production of high quality fission I-131, a by-product of Mo-99 production, meeting international quality standards. A special work will be presented in technical sessions
- CNEA has a project in progress to optimize the Mo99 production (CiMo99/grU).
- CNEA-US DoE collaboration on LEU technologies: during Dec. 2006, USDoE monolithic targets and a process digestor will be tested at the RA-3 reactor and production plant.

o A project to recover irradiated HEU, blend it down to LEU and separate Cs137 and Sr90 is ongoing. The separation of Sr-90 will be used in Y-90 generators. Both radioisotopes are of interest for nuclear medicine applications.

o CNEA is participating in the IAEA Coordinated Research Project on developing techniques for small scale indigenous LEU Mo^{99} production as an agreement holder

7. Conclusions

CNEA continued deploying an intensive activity on R&D on RERTR technologies. Concerning VHD fuels, we focused our work some promissory lines for technological solutions both on dispersed and monolithic fuels in the range of 8-16 gU/cc. Concerning LEU technologies for radioisotope production we are deeply involved on its development and diffusion of the technology. Future plans includes:

o Completion during 2007 the RA-6 reactor conversion to LEU

o Improvement on fuel development and production facilities to implement new technologies, including NDT techniques to assess bonding quality.

o Irradiation of miniplates and full scale fuel assemblies in RA-3 and in high flux reactor, both dispersed and monolithic fuels.

o Optimization of LEU target and radiochemical techniques for radioisotope production

8. References

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