RESULTS FOR THE RECENT ACTIVITIES OF REDUCED ENRICHMENT PROGRAM FOR RESEARCH REACTORS IN CHILE

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ABSTRACT

The "Reduced Enrichment for Research and Test Reactors" RERTR Program, applied to RECH-1 Chilean Research Reactor has been completed by May 11, 2006. Since this date, a core of 32 LEU fuel elements, fully fabricated in Chile, has been loaded and operated according to a project that included feasibility studies, conversion analysis, in-pile fuel qualification and licensing activities.
The Chilean Fuel Qualification Program in the HFR-Petten, the Netherlands ended in April 2006. After successful irradiation during 16 cycles (388,3 FPD), the ^{235}U depletion amounted to 64.5% ± 8.4%. A dismantled plate was submitted for PIE and the results were as expected. The final conclusion was that the irradiation qualification project was successful, and CCHEN is qualified as LEU fuel manufacturer.
Related to UMo fuel development, the effects of adding Ti, Zr, Si and Ti+Zr to U-7wt%Mo alloy are being studied, and interdiffusional reaction layers between U-7wt%Mo-Me fuel particles and Al matrix have been analyzed in dispersion miniplates. Besides, the results of comparative studies of fuel meat swelling using U-7wt%Mo, U_3Si_2 and dummy miniplates exposed to high temperatures are presented.

1. Introduction

The RERTR program, applied in CCHEN since 1978 to RECH-1 Chilean Research Reactor, has been completed by May 2006. Since this date, a complete reference core has been loaded and operated to conclude a conversion process where feasibility analysis, fuel qualification and licensing activities were included. The current reactor core loads 32 LEU fuel assemblies, fully fabricated by the Chilean Fuel Fabrication Facility (PEC). The meat of the fuel assemblies is U_3Si_2 dispersed in Al matrix. The first four LEU fuel assemblies were loaded in the reactor core in 1998-1999 and have been supporting a local fuel irradiation qualification program [1].

Since the time required for local qualification would be too long and in order to assure the safety and quality without substantial delay in the manufacturing and conversion programs, an international qualification under irradiation was required [2]. In 2001 a contractual agreement between IAEA, CCHEN and NRG (Nuclear Research and consultancy Group) as operator of HFR – Petten, was signed in order to meet this goal.

The support project for qualifying – IAEA CHI/4/021 TCP - based on contract stated above, concerns the irradiation of one test fuel element - LCC-01, containing U_3Si_2 – Al as fuel matrix with 229,1 g ^{235}U in the HFR - Petten. The execution of the irradiation and subsequent Post Irradiation Examination – PIE, is dedicated to NRG, Petten. The aim of the irradiation and subsequent PIE was to qualify the manufacturer of these LEU fuel elements [3]. In previous versions of RERTR meeting, the authors have been reporting previous stages of this TCP. In this paper, the final burnup computations, PIE of a dismantled fuel plate and final conclusions are described.

In relation to high-density fuels based on UMo alloys, last year a developing program, based on exploratory tests, was presented and approved at CCHEN in order to be up to date in a new alternative of fuel with an available back-end solution and to participate in RERTR initiatives and goals.

The U-Mo alloys are candidate materials for use as fissile phase in LEU dispersion fuels in the range of 5 to 10%wt of Mo. According to U-Mo phase diagram, at environment temperature, is possible to have a phase transformation from χ to $\alpha + \varepsilon$. Fortunately, this phase transformation occurs very slowly, then is not difficult to retain gamma phase [4]. Nevertheless, these ductile alloys cannot be converted efficiently to powder by conventional milling process used for oxides and intermetallics compounds [5]. Four methods of powder production from U-7%Mo alloys have been tested in CCHEN. The main objective was to evaluate affordable methods to produce U-Mo powders and according to the results, select a way to produce a sufficiently large amount of powder for further stages of the U-Mo program.

Although UMo alloys in gamma phase have the best irradiation performances, UMo/Al dispersion fuels have shown an unacceptably large pore formation due to unstable properties of an interaction layer – IL, formed between UMo particles and Al matrix [6]. In the UMo/Al system, temperature is an important consideration in the formation of porosity and reaction layer because it exerts a strong influence over the rate of reaction between fuel and matrix [7]. As IL grows, the fuel temperature increases because its thermal conductivity is poorer than aluminum matrix [6]. Since interface layer has a lower density than the rest of microconstitutive, its growth produce volume expansion of the fuel meat. Some methods were proposed by several authors in order to develop a modified UMo alloy capable to either forming a more stable IL or control its growing. One of these methods was add a third element for stabilizing the interdiffusion between UMo fuel and Al matrix [8]. In this topic, during 2006, interaction studies in U-Mo/Al and UMo-Me/Al systems in dispersed miniplates are being explored in CCHEN. Particularly, effects of alloy elements as Si, Ti and Zr in the interdiffusion and IL formation are reported. Finally, in this paper, quantitative results of volume expansion or swelling caused by thermal treatments in UMo and U_3Si_2 miniplates are discussed.

2. EXPERIMENTAL SET- UP

2.1. RECH-1 Fuel Fabrication

In May 2006, a complete core of 32 LEU fuel assemblies fabricated in Chile has been loaded and operated according to a conversion process based on RERTR goals. In order to complete the RECH-1 conversion, a LEU fuel fabrication program based on U_3Si_2 was launched in CCHEN in 1995. After 11 years, 48 fuel elements have been assembled according to international standard and Chileans technical specifications. Table 1 summarizes the RECH-1 reactor and fuel elements main features.

PARAMETER	HEU	HEU (Ex MEU)	LEU
Reactor type		Pool Type MTR	
Steady-State Power Level, MW		5	
Uranium Enrichment, % ^{235}U	80	45	19,75
Number of Reference Core Configuration		32	
Control Blade material		Cadmium	
Number of Control Blades		6	
Grid Plate		10 x 8	
Coolant, Moderator		Water	
Reflector		Beryllium	
Meat Thickness, mm		0,61±0.02	
Meat Width, mm	60,147	60,833	60,15(+0.55-0.45)
Plate Width, mm		69,8	
Active length, mm	581,05	581,05	586,0
Plate length, mm			625,5
Cladding Thickness (Al), mm		0,46 ± 0,03	
Water Channel Thickness, mm		3,17 – 0,25	
Fuel Plate Thickness, mm		1,53	
Meat Volume, cm^3		21,52	
Plates, Fuel Element		16 (14 internal + 2 external)	
Fuel Meat Composition	UAlx	UAlx	U_3Si_2
^{235}U Density in External Plate, g/cm3	0,258	0,283	1,7
^{235}U Density in Internal Plate, g/cm3	0,516	0,566	3,4
^{235}U mass in external Fuel Plate, g	5,50	6,11	7,16 ± 2%
^{235}U mass in internal Fuel Plate, g	11,0	12,21	14,32 ± 2%
^{235}U mass in Fuel Element, g	165,0	183,16	214,80 ± 2%

During five years, the parts involved in the IAEA-TCP satisfied each step of this tri-partite contract and finally it was satisfactory finished in 2006. Each stage of this project can be briefly described as follows:

Design Stage: Detailed discussions took place between CCHEN and NRG in order to arrange details of design, fabrication, characterization and quality assurance of the fuel element. Technical description, specifications, thermohydraulics computations and measurements, neutronics design computations and licensing issues were covered.

Irradiation: According to the contract, during irradiation only off-line measurements like visual inspections and coolant gap measurements were done and reported, besides the calculated burnup at the end of each HFR cycle.

Burnup computations: Neutronics computations were carried out prior, during and after irradiation in order to assess the burnup of the fuel element.

Post Irradiation Examination PIE: Post Irradiation Examinations were carried out at the Petten hot cell facility. Inspections of entire fuel element and dismantled fuel plate were done in order to study the behavior of the fuel element.

2.3.- Uranium Molybdenum Fuel Development

Four methods of powder production from U-7%Mo alloys have been tested in CCHEN. These processes were hydride-dehydride, rotating knifes milling, cryogenic milling and high energy

milling. UMo alloys in as cast condition, homogenized by thermal treatment and cold rolled and crushed (R&C) were the feeding materials for powder production tests, with feeding sizes less than 5 mm in average.
Interaction studies were done with specimens extracted by punching from dispersion miniplates fabricated with U-7%wt Mo with addition of a third element: Si, Ti, Zr and Ti+Zr in nominal compositions of 1% wt. XRD analyses were applied to identify the influence of third elements addition in stability of gamma phase in samples of UMo and UMo-Me alloys, previously annealed to 1000°C for homogenization by two hours. The matrix of miniplates was pure aluminum powder and cladding of Al-6061. In order to obtain fuel particles fully surrounded by Al, the density of miniplates was just 1,7 gU/cm^3. The specimens were vacuum-sealed quartz tubes, annealed at 550°C for 24 and 48 hours and then quenched in water. After that, samples were examined metallographically. The microstructures of interaction tests specimens were characterized using optical microscopy and SEM with EDS.

Finally, to examine the volume expansion behavior, a set of UMo miniplates with uranium densities of 3,4 - 4,8 and 6,8 gU/cm^3 were annealed to 350°C and 550°C for accumulative times up to 30 hours. The miniplates were quenched in water after each treatment. A U_3Si_2 fuel and Al-6061 dummy miniplates were used as reference and for comparisons of swelling results. The volume expansion was controlled by means Archimedes method applied after each thermal treatment.

3. RESULTS AND DISCUSSIONS

3.1. RECH-1 fuel fabrication

The main results in relation to fuel fabrication for RECH-1 were a total of 805 fuel plates fabricated and 48 fuel elements assembled: One fuel element submitted for international qualification (Petten) and 47 transferred to RECH-1 facility. 31 of the 805 fuel plates fabricated were rejected and stored separately (3,9% of total). Some of them were used in destructive analyses. The rejections were mainly by remote island inspection

3.2. Qualification under Irradiation

For burnup computations, the most reliable neutronics computations could be done after completion of the irradiation and the PIE. These computations were done using the detailed irradiation history of the fuel element. In the depletion analyses validated versions of the software (i.e. of MCNP, FISPACT and OCTOPUS) were used. The depletion of ^{235}U as a function of time with respect to the start of the irradiation is shown in Figure 1. At the end of the irradiation, the ^{235}U depletion amounts to 64.5% ± 8.4% (81.4 gr. ^{235}U EOI vs. 229.1 gr. at BOI), higher than the required 55%. This figure also contains the results of a depletion calculation using the code HFR-TEDDI. The curves are similar within the uncertainty range of both codes, but in the HFR-TEDDI simulation the fuel burnup is slightly higher than in the MCNP/OCTOPUS values. The 3D MCNP/OCTOPUS code is considered to be more reliable and detailed than the 2D HFR-TEDDI code and therefore the MCNP/OCTOPUS results are used as the final burnup results. The dotted line indicates the level required for 55% depletion.

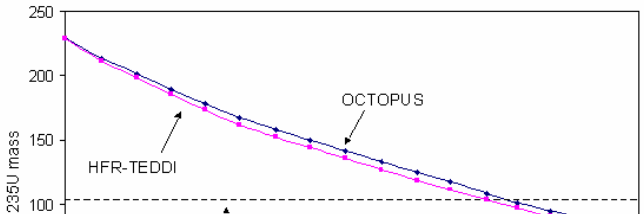


Figure 1. ^{235}U mass as function of irradiation time.

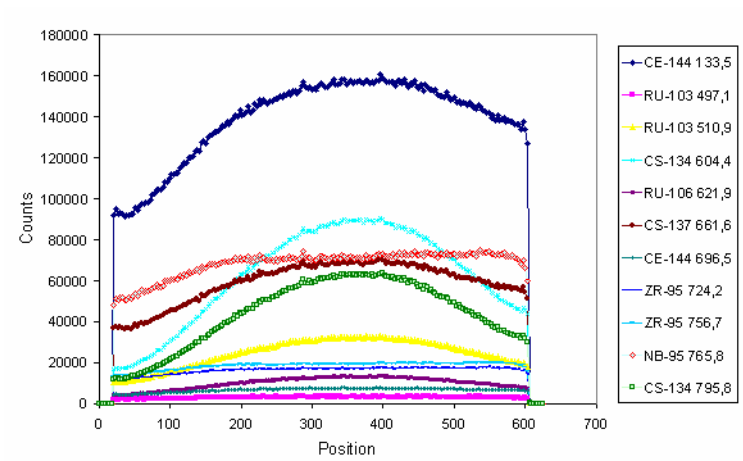


Figure 2. *PIE results*: Gamma-scan spectrum on the dismantled plate. The position 600 mm is the lower side of the element.

The difference in shape of the curves between the various isotopes in figure 2, can be understood from their decay time or from the number of neutrons required to form the isotopes. Oxide layer thickness measurements were done on the dismantled plate using the eddy-current technique. The oxide layer thickness is 15.00 micron with an uncertainty of ± n

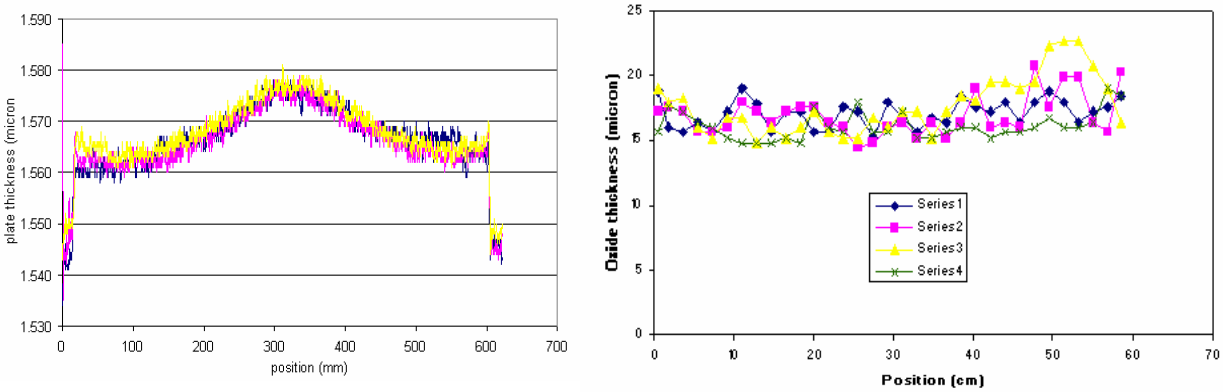


Figure 3. *PIE Results*: Plate thickness measurements (left) and the measured oxide layer thickness (right) [9].

Reference data [10] on the volume change (swelling) as a function of the fission density is shown in Figure 4. Note that the volume change is shown as the volume change in the fissile particles and that the volume change in the plate should be derived from this. This conversion is made with data shown in Table 3. Using this data it can be computed that approximately 0.433 gU/cm^3 (0.645*0.1975*3.4 gU/cm3) has been fissioned. This amounts to approximately (0.433*6*1023/237.4) = 1.1*10^{27} fissions/m^3. This corresponds (Figure 4) to a volume change of the particles of about 6.6 %vol This volume change of fuel is generally only a change in thickness of the plate (not of the height and width). The thickness variation can then be computed as 0,32*0,61mm*0,066 = 13 micron. Given the uncertainty in various parameters and the fact that the oxide thickness may be slightly different between the fuelled zone and the non-fuelled zone it can be concluded that good agreement exists between the measured thickness change and the literature data [10]. Note that the curves shown in Figure 4 [10] only show general trends, assuming that the as-fabricated porosity has been completely filled. The initial porosity distribution has not been determined for the present fuel.

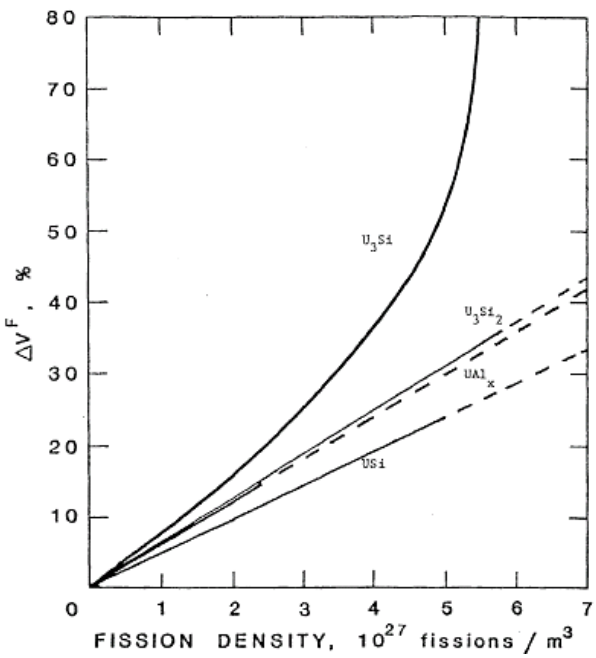


Figure 4. Swelling of silicides and UAlx fuel particles vs fission density in the particle.

Table 3. Data used to thickness discussion

Thickness fuel meat	0,61 mm
Vol fraction fuel in meat	32%
Burnup (EOI)	64,5% ±8,4% of initial ^{235}U
U235 enrichment	19,75%
U density	3,4 gU/cm^3

3.3. U-Mo fuel development

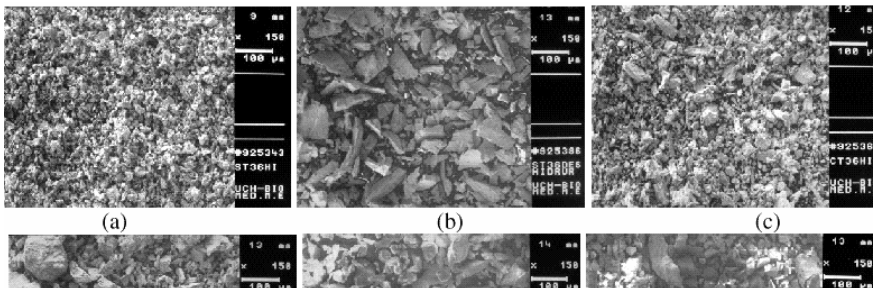
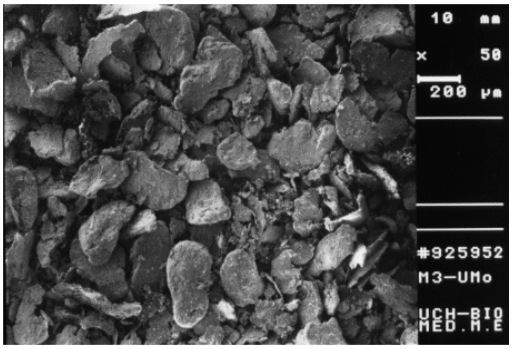


Figure 5 Powders produced by means HMD method: (a) UMo as cast Hydrided (b) UMo as cast dehydrided (c) UMo homogenized Hydrided, (d) UMo homogenized Dehydrided, (e) UMo – R&C Hydrided and (f) UMo R&C Dehydrided (SEM images)

In general, dehydride powders were coarser than hydrided due probably to sintering of small particles, except in UMo R&C powders (e), (f) pictures.



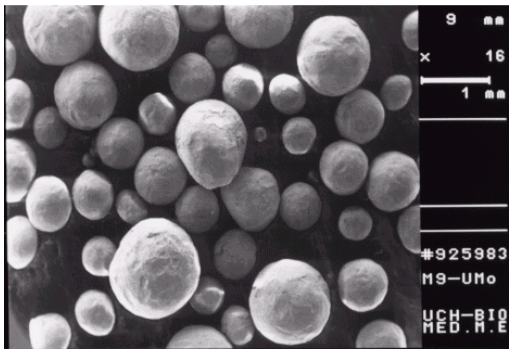
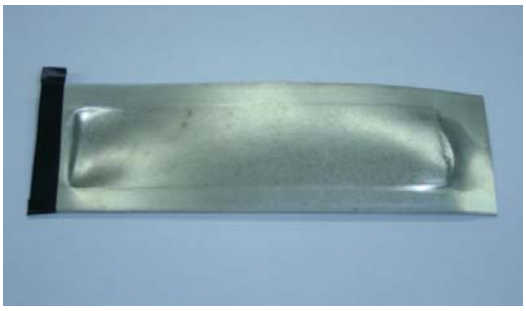


Figure 6. At left UMo, As cast condition. At right, UMo, homogenized, R&C condition both milled by means of titanium rotating knifes mill.

Besides HMD method, other techniques for powder production were tested [11]. In general terms, all processes of grinding and milling were found inefficient and in rotating knifes mill, high levels of contamination were detected (0,83 wt% of Ti and 0.03 wt% Co + 0.07wt% W for Ti and tungsten carbide knifes, respectively). The major efficiency was found for tungsten carbide knifes.

The Figure 7 shows miniplates after swelling annealing. The distortion due to swelling in UMo-6.8gU/cm^3 miniplate (right) was so extreme that debonding and opening effects were observed after only one hour at 550 °C. This effect was less dramatic at 350 °C, by which was possible to conclude the tests.



(a) (b)

Figure 7. Miniplates after swelling annealing at 550 °C. (a) Left Al-6061 (dummy) - UMo 3,4 –UMo 4,8 - UMo 6,8 gU/cm^3 and U_3Si_2 by 30 hours and (b) One end opened miniplate UMo-6,8 gU/cm^3. - after1 hour.

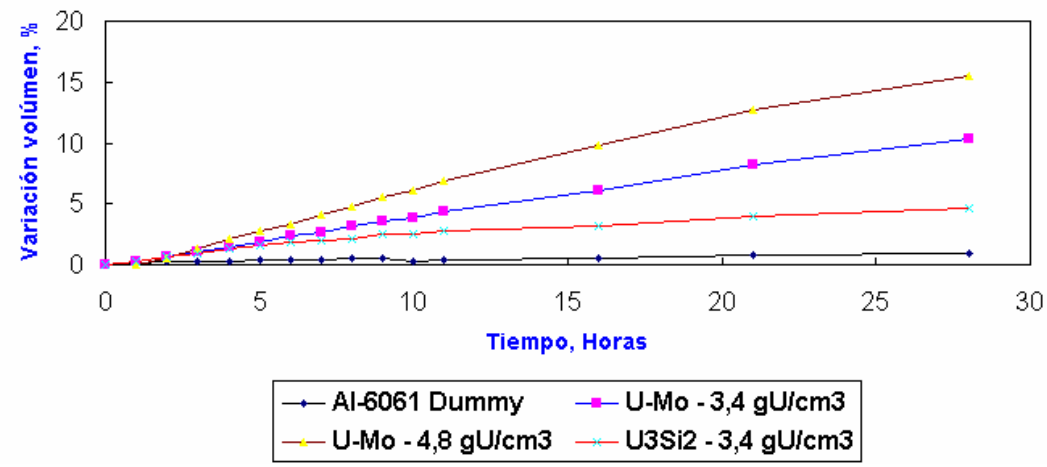


Figure 8. Volume variations of plates as a function of the annealing time at 550 °C.

In figure 8 is possible to verify that swelling effect are increased with U density, the swelling due to UMo/Al interaction produces volume expansion, always growing in UMo miniplates but with a trend to stabilization in U_3Si_2 miniplate (red curve). The slight volume expansion in dummy plate is due just to surface oxide layer formation. To same U density, UMo exhibits more than double volume expansion than U_3Si_2 miniplate.

Figure 9 shows microstructures of UMo-Me alloys after thermal homogenization annealing (1000 °C-2 hours in vacuum). XRD revealed gamma phase UMo and second phases precipitated in grain boundaries. Surely in this second phase is concentrated the third element and probably this precipitated may became barriers to reduce the migration of U and Mo atoms. This theory requires further studies, but our idea is that the grain boundaries, more than the outer surface of particles, is the key to reduce the reaction between UMo and Al. As shown the figure 10, the morphologies of IL is clearly modified by Si addition. In UMo-Si/Al sample, more clean and thinner IL are observed, probably by adding of the third element. In UMo/Al specimen, porosity is evident in the outer surface of UMo particles and greater than UMo-Si/Al sample. Similar effects were observed in the specimens with Zr, Ti and Ti+Zr additions.

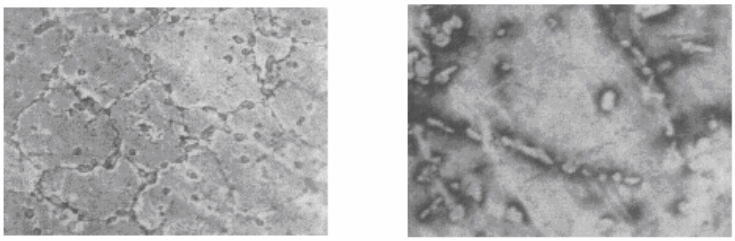


Figure 9. Optical micrographs of UMo-Me alloys after homogenization annealing

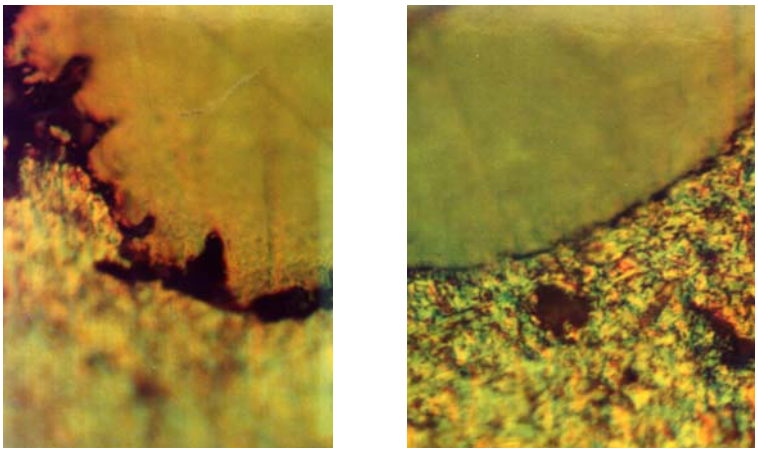


Figure 10. Details of Interaction Layers IL after 48 hours at 550°C.Left UMo/Al. At right UMo-Si/Al

4.- Conclusions

The PIE on the LCC01 fuel element has been done successfully. All measurements show the proper behavior of the fuel element, being only a small swelling of the plate, smaller than expected. The small oxide layer thickness and the fact that visual inspection through the elements shows that all plates are stable. No remarkable features are observed.

Detailed neutronics computations have been done showing that the 235U depletion amounts to 64.5% ± 8.4% (81.4 gr. 235U EOI vs. 229.1 gr. at BOI), which is more than the required 55%.

The final conclusion is that according to inspections of fuel fabrication process, QA management system and results observed during irradiation of test fuel element at the HFR

in Petten, the qualification under irradiation project was successful and qualifies CCHEN as a LEU silicide fuel manufacturer.

The possibility of simulating the effects of the irradiation by means thermal treatment applied to natural uranium plates is a validate and affordable alternative applicable for preliminary tests and studies. The results obtained in thermally induced swelling tests are in concordance with the LEU samples tested under irradiation and reported in the references.

5.- ACKNOWLEDGEMENTS

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