THE RERTR-2007 INTERNATIONAL MEETING ON
REDUCED ENRICHMENT FOR RESEARCH AND TEST REACTORS
September 23-27, 2007
Diplomat Hotel – Prague
Prague, Czech Republic

NUCLEAR FUEL DEVELOPMENT BASED ON UMo ALLOYS UNDER IRRADIATION EVALUATION OF LEU U_3Si_2 – 4,8 gU/cm^3 TEST FUEL

Luis Olivares, Jorge Marin, Mario Barrera, Gonzalo Torres and Jaime Lisboa.
Nuclear Materials Department
Chilean Commission for Nuclear Energy - CCHEN
95 Amunategui St., Author contact: lolivare@cchen.cl
Santiago – Chile

ABSTRACT

Latest irradiation reports on UMo dispersion fuel demand different solutions in order to stabilize an undesirable interaction layer zone produced by chemical reactions with the standard Al matrix. Among the different solutions, this paper addresses the evaluation of the additions of Si to the Al matrix and/or small amount of tetravalent elements (Si, Zr, and Ti) to the fuel. The results of out-of-pile swelling tests applied to dispersion mini plates made of these modified constituents and interaction tests of interdiffusion couples are discussed. These results shows that the volume changes are directly related with the uranium density. The unmodified UMo/Al system exhibited swelling levels almost three times higher than those achieved with U_3Si_2, however for the system UMo/AlSi mix, these undesired behavior was reduced to values equivalent or slightly lower than U_3Si_2. Finally, details and specifications of a LEU U_3Si_2 4,8 gU/cm^3 test fuel element, fabricated for under irradiation evaluation and design validation purposes at the RECH-1 research reactor, are reported in this paper. The planned follow-up program and initial irradiation results showing an appropriate performance that allows reaching the expected ^{235}U burn up level, slightly over 50%, are detailed and discussed in next pages.

1. INTRODUCTION

The development of U-Mo alloys based fuels is nowadays an important objective in CCHEN's goal to become an active participant in the international development and qualification of a potential solution for the majority of research reactors conversion. Furthermore, this new fuel could make possible to optimize and to improve the design of current reactors by providing a fuel with an available back-end solution [1]. Latest irradiation reports of UMo dispersion fuel confirm the necessary evaluation of the different solutions aiming to stabilize an interaction layer zone produced by chemical reactions with the standard Al matrix [2]. A local program for development of fuel based on U-Mo alloys was launched in CCHEN in 2003 [3]. Among the different solutions currently under study, this paper addresses the evaluation of the effects of the addition of Si to Al matrix and/or small amount additions of tetravalent elements (Si, Zr, and Ti) to the fuel, according to the work performed by Rodier et al., [4]. This paper presents the results of an out-of-pile swelling study carried out with mini plates made of U-7 wt% Mo modified by the addition of 1 wt% of a third element "Me" (Si, Ti, or Zr) dispersed in several Al matrixes: pure Al, Al-Si 5 wt % alloy and also a matrix composed by Al and Si (5 wt %) pre mixed powders. In order to know the interdiffusion behaviors and assure the stability of the gamma phase in these modified UMo/Al systems, interdiffusion tests were developed using UMo+Me/Al-Si couples.

Since the main technical objectives of CCHEN's Fuel Fabrication Facility are the development, qualification and fabrication of LEU fuel assemblies to satisfy the core loading requirements of Chilean research reactors, an optimized fuel design has been proposed. This new design must perform without affecting the technical and geometrical characteristics of the current fuel elements and without a major influence on reactor performance. Among other structural changes, a goal introduced is to test and evaluate an uranium density increase from the current 3,4 to 4,8 gU/cm^3. Previously a licensing process based upon a Safety Evaluation Report [5] issued by the Chilean Nuclear Regulatory Body and a Technical Report issued by RECH-1 Chilean research reactor Staff [6] for the under irradiation evaluation of a fuel test assembly was finished, and the irradiation test started at the reactor on July 12th, 2007. Details and specifications of the test fuel element, manufactured with one real fuel plate (LEU U_3Si_2 4,8 gU/cm^3) and 15 Al-6061 dummy plates are reported in this paper. The follow-up program include sipping tests [7], visual inspections [8] with immersed camera, burnup computations, neutronics flow measurements, verification of cooling channels (GAP) and cooling water quality records. The initially required ^{235}U burn up level is slightly over 50%, and after an evaluation of the fuel performance, the irradiation could be continued to higher levels.

2. EXPERIMENTAL SET-UP

2.1 Swelling tests in dispersed UMo fuel miniplates

Thermally induced swelling tests were developed in order to compare quantitatively these volume-increasing phenomena in dispersion miniplates made of either UMo or UMo-Me dispersed in Al matrixes modified by Si additions. For volume increase evaluation, the miniplates were annealed at 500° C up to 35 hours. These studies also compared the swelling behaviors of UMo and U_3Si_2 miniplates. Test miniplates were manufactured using a traditional process for dispersion fuel plates, i.e., blending of powders, pressing, assembling with Al 6061 frame and covers, welding, hot rolling and cold rolling. By other hand, UMo and UMo-Me alloys were prepared with natural uranium and pure Mo by induction melting in an Ar atmosphere, where "Me" was 1 wt% nominal additions of Si, Ti or Zr.

2.2 Interdiffusion tests between AlSi matrix and modified UMo alloys

To compare swelling and interdiffusion behaviors, diffusion couples were prepared with pieces of Al 5 wt% Si alloy joined to pieces of UMo, UMoSi and UMoTi alloys (obtained from UMo-Me alloys produced for swelling tests). These pieces, in polish and clean condition, were maintained in intimate contact by a bolted small clamp device. The UMo/AlSi, diffusion couples were annealed at 500°C for 3 hours in a vacuum atmosphere (10^{-2} Pa). After diffusion annealing, microstructures of the interaction layers (IL) formed in the specimens were observed with optical and scanning electron microscopy (SEM). Concentration profiles and compositions were measured using a JEOL 5410 SEM with an Energy Dispersive Spectrometer–EDS by Noran Instruments

2.3 Evaluation under irradiation of U_3Si_2 Test Fuel

CCHEN's Fuel Fabrication Facility – PEC was responsible for the design and fabrication of a Test Fuel Element - TFE identified as LR-EX-01 with only one real plate, containing LEU U_3Si_2 dispersed in Al matrix. The nominal uranium density in the active fuel plate was 4,8 gU/cm^3. This TFE has the same geometry of the Standard Fuel Element (SFE) previously fabricated by PEC for the

RECH-1 reactor and the other 15 dummy plates were Al-6061. TFE was loaded at F10 core position and according to current test license, will be in that position until the test ends.
The main properties of the TFE as compared to a SFE are listed below:

- 1 fuel plate + 15 Al- 6061 plates instead of 16 fuel plates;
- Modified design using a hot stamped bottom piece with two welding cords instead three;
- Nominal ^{235}U mass per element is 20,19 g instead of 214,8 g;
- Upper piece with open inlet (square area) geometry instead of a closed filter box with filter plate;
- Extra length external dummy plates, longer than fuel and dummy plates located inside;
- Uranium density of 4,8 g/cm^3 instead of 3,4 g/cm^3.

2.3.1 Follow-up program

The main purpose of this follow-up is to obtain at programmed irradiation cycles (22 hrs.) an early detection of any deviations from expected performance including blisters, unacceptable swelling, active plate damage and its consequent fission product release. This program includes the following activities:

Water Sipping test

a) After first irradiation cycle
b) After 10th irradiation cycle
c) One cycle after core re - arrangements

Visual Inspection Using RCS-3100 – 72:1 Zoom S-Video TV Camera underwater System
This inspection will be done with the same intervals schedule for sipping test

Burn up measurements: Will be done by means a gamma spectroscopy after first core re arrangement followed by a burn up measurement after three core re-arrangement in a system installed in the auxiliary reactor pool.

Neutronics flow measurements: Axial distribution of thermal neutronics flux will be measured in TFE using gold probes, bare and cadmium coated, placed in an acrylic sword. The results, histograms, will allow correlations and comparisons between neutronics computations and gamma spectroscopy measurements. The flow measurements will be done with the same intervals schedule for burn-up measurements.

Cooling gap measurements: This measurement will be done with a SAE 316 stainless steel caliper (2,15 mm thickness) using go-no-go evaluation. The gap measurements will be done using the same intervals schedule for visual inspection.

Water quality records: Due to the relevance of the cooling water quality, during irradiation of the TFE, special records of pH, conductivity and impurities content (Fe, Cu, Cl) will be taken. Some minor deviations could be accepted in limited ranges.

3. RESULTS AND DISCUSSIONS

3.1 Swelling tests

Features of test miniplates and maximum volume increase for 30 hours at 500°C annealing in air, are summarized in Table 2. Annealing of miniplates UMo-19 and UMo-22 was stopped after first hour due to pillowing and de-bonding of cladding caused by extreme swelling. UMo 13 plate was rejected by blister test (1 hr./500°C).

Table 1. Test miniplates and volume increase induced by 500°C – 35 hours annealing

Id Miniplate	Fuel alloy	Matrix	Compact weight (g)	Uranium Density gU/cm^3	Max Volume increase (%)
UMo-4	UMo	Pure Al	15,47	3,4	4,1
UMo-5	UMo	Pure Al	17,85	4,5	9,4
UMo-12	UMo	Pure Al	23,04	6,7	54,1 (3)
UMo-13	UMo	AlSi Alloy	11,85	3,0	Rejected (1)
UMo-14	UMo	AlSi Mix	14,23	3,6	0,5
UMo-15	UMo-1%Si	Pure Al	13,91	3,5	2,5
UMo-16	UMo-1%Si	AlSi Alloy	11,9	3,0	1,5
UMo-17	UMo-1%Si	AlSi Mix	14,49	3,7	2,2
UMo-18	UMo-1%Ti	Pure Al	16,5	4,2	5,6
UMo-19	UMo-1%Ti	AlSi Alloy	12,1	3,1	Opened (2)
UMo-20	UMo-1%Ti	AlSi Mix	14,17	3,6	2,5
UMo-21	UMo-1%Zr	Pure Al	15,23	3,9	2,7
UMo-22	UMo-1%Zr	AlSi Alloy	11,1	2,8	Opened (2)
UMo-23	UMo-1%Zr	AlSi Mix	14,7	3,7	2,2
PUS-30	U_3Si_2	Pure Al	17,37	4,8	4,8
PUS-31	U_3Si_2	Pure Al	14,62	3,4	1,9

(1) Miniplate rejected in blister test
(2) Miniplates opened at beginning of annealing
(3) Test canceled in the first hour by excessive swelling and distortion

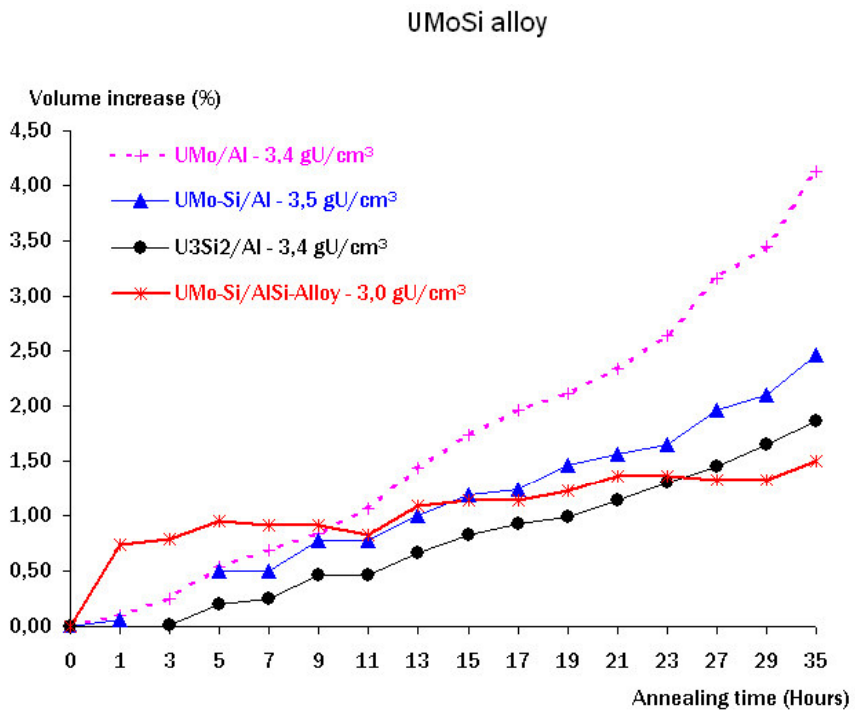

Figure 1. Volume increase v/s annealing time for UMoSi fuel alloy dispersed in pure Al or AlSi alloy matrixes.

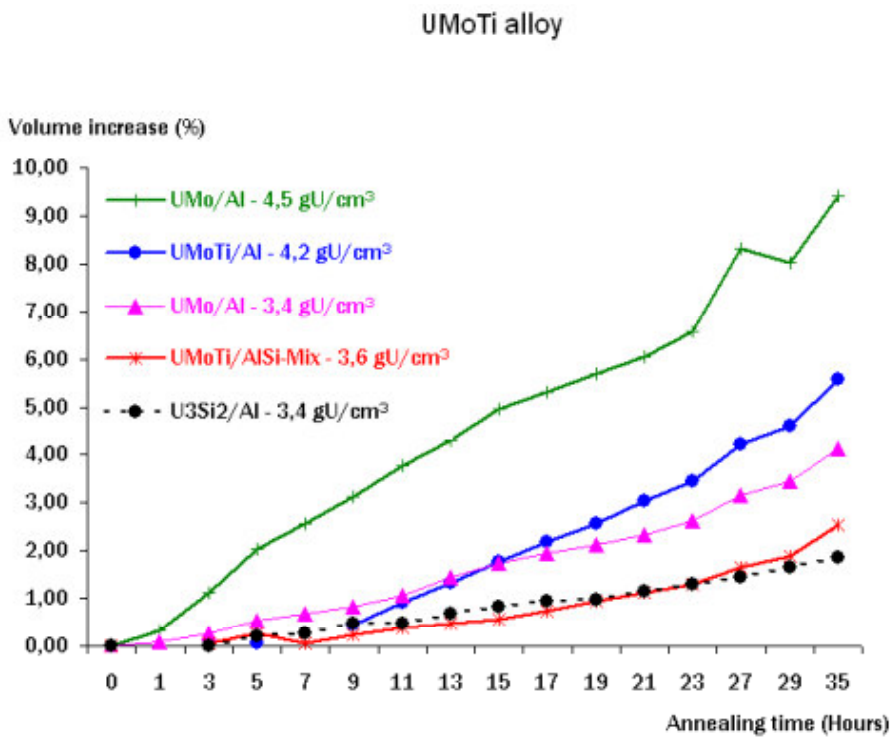

Figure 2. Volume increase v/s annealing time for UMoTi fuel alloy dispersed in pure Al or AlSi mixed matrixes.

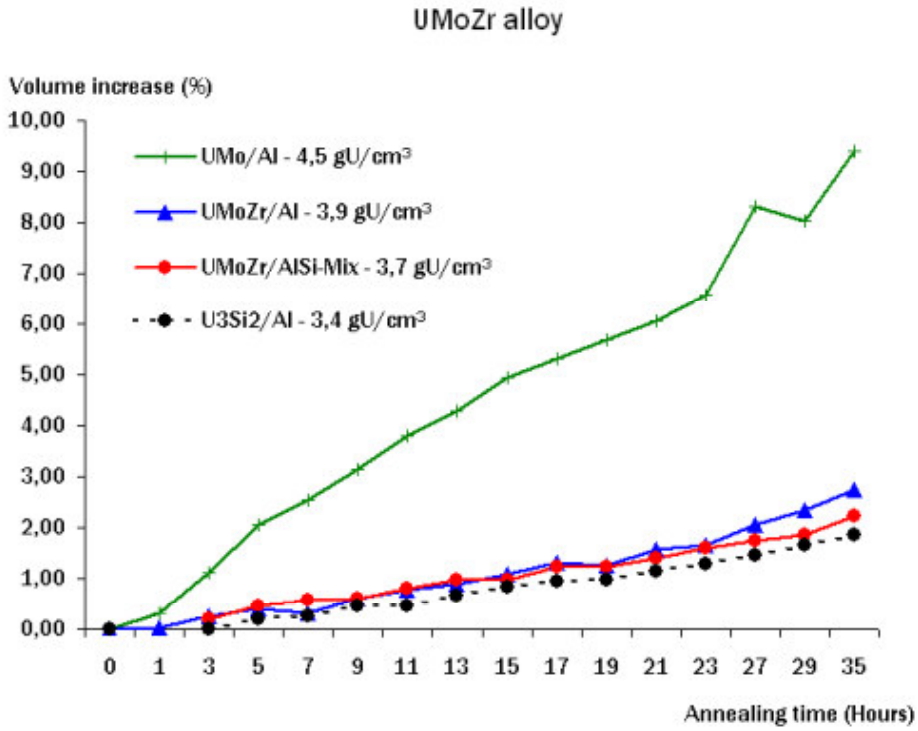

Figure 3. Volume increase v/s annealing time for UMoZr fuel alloy dispersed in pure Al or AlSi mixed matrixes.

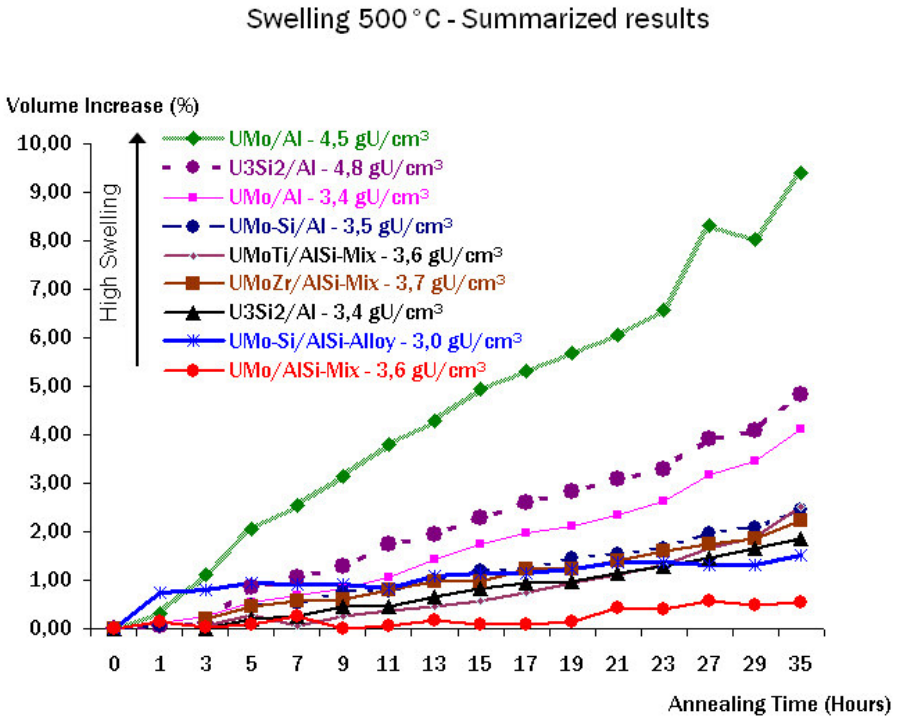

Figure 4. Volume increase v/s annealing time for UMoSi fuel alloy dispersed in Al or AlSi alloy matrixes.

According to the swelling test results, the volume changes are directly related with the uranium density, and in general, third element additions resulted in improvements in swelling behaviors. Comparatively, the best result was obtained for dispersed miniplates made of UMo/AlSi-Mix followed by UMoSi/AlSi alloy, both slightly better than U_3Si_2 for the same range of uranium density (3,4 gU/cm^3). The volume increase for unmodified UMo/Al system achieves levels almost three times higher than those achieved with U_3Si_2, however for the system UMo/AlSi mix, these undesired behavior was reduced to values equivalent or slightly lower than U_3Si_2. The result indicates that the modification by silicon addition is most effective in the matrix than in the fuel alloy. UMo alloy without a third element in a standard Al matrix, exhibited swelling levels five times higher than those obtained with U_3Si_2. The expected effect for the modified UMo/Al dispersed fuel was to reduce this undesired behavior down to an accepted level and the latest results showed promissory and decreasing swelling values from five to less than two times.

3.2 Interdiffusion tests

3.2.1 UMo/AlSi interdiffusion couple: The interface layer (IL), exhibited a discontinuous morphology and thickness up to 5 µm with a strong Si atoms migration from AlSi alloy to IL. It was also possible to detect a decrease in Mo content towards IL and U and Mo atoms migration into AlSi alloy. In AlSi alloy, EDS analyses of silicon detect a remarkable increasing towards IL zone up to a content higher than 20 wt% at 1 mm from IL. In UMo alloy area, the Mo content decrease to 3,9 wt% at 3,3 mm from IL and in the center of IL 5,5 wt% of Mo was detected. Metallographic examination of AlSi alloy shown a microstructure of Al grains surrounded by rich in silicon phase with lamellar morphology typical of eutectic micro constituent. The presence of high amounts of U (79,8 wt% at 1 µm and 6,1 wt% at 1mm from IL) and Mo, (0,78 and 0,42 wt% at 1µm y 1 mm respectively) showed a pronounced diffusion and consequential chemical interaction between couples constituents.

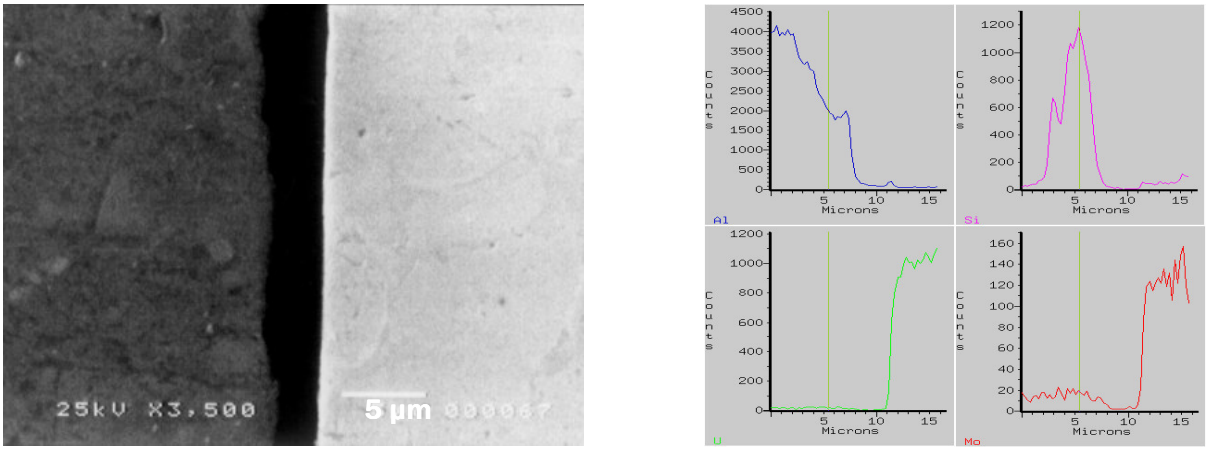

Figure 5. MEB image and line scanning of IL in UMo/AlSi alloys couple after annealing.

3.3.2 UMoSi/AlSi interdiffusion couple: A continuous IL, thicker than 3 µm was observed. Interdiffusion of silicon from both sides of couple was revealed by EDS, but the silicon content in the center of IL is lower than UMo/AlSi couple. The presence of silicon in both sides of IL produced a decrease in concentration gradient that allowed its behavior as a moderator agent for Si interdiffusion. EDS at the center of IL showed 70 wt% of U, 15 wt% of Si, 9 wt% of Mo and 6 wt% of Al. In the UMoSi alloy near to IL, U and a slightly higher Mo contents were detected (74 wt% and 9.4 wt% respectively), here the Si content decreased to a few 0,27 wt%. Inside the AlSi alloy, as detected by EDS, among Al and Si, some contents of U and Mo (13,1 and 3 wt% respectively), much lower than UMo/AlSi.

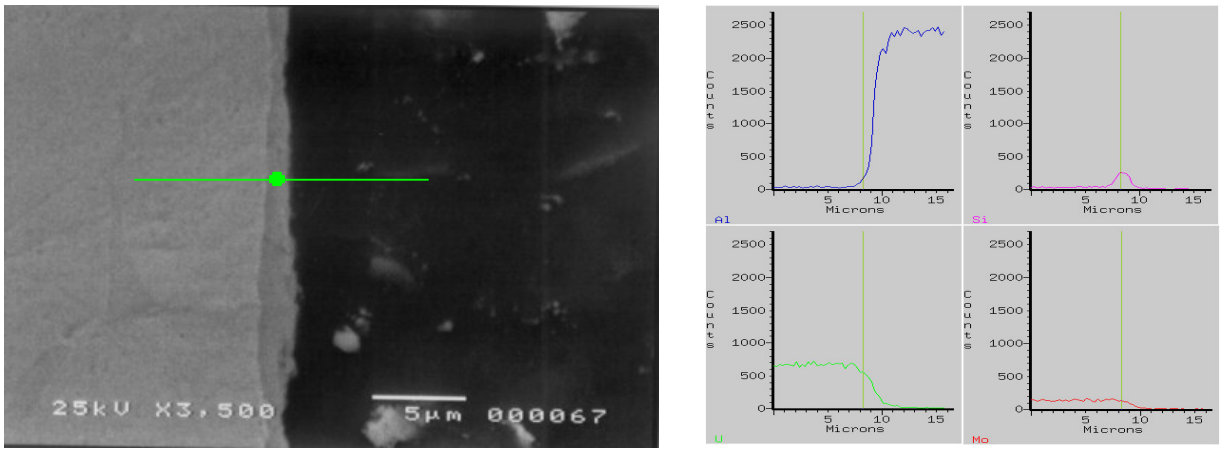

Figure 6. MEB image and line scanning of IL in UMoSi/AlSi couple after annealing.

3.2.3 UMoTi/AlSi interdiffusion couple: This couple showed a continuous IL thicker than 1 µm. Silicon concentration profile shown migration towards IL, where it was found amounts three times higher than nominal AlSi alloy content. In UMoTi alloy, at 6 µm of IL, presence of Mo (4,5 wt%) and small amounts of Al were detected. In the center of the IL, EDS results shown majority U and Al (9,7wt%), Si (4,6 wt%), Mo (2,4 wt%) and Ti (0,5 wt%). In AlSi, near to IL, among Al, 3,4 wt% of U and a few 0,2 wt% of Si were detected.

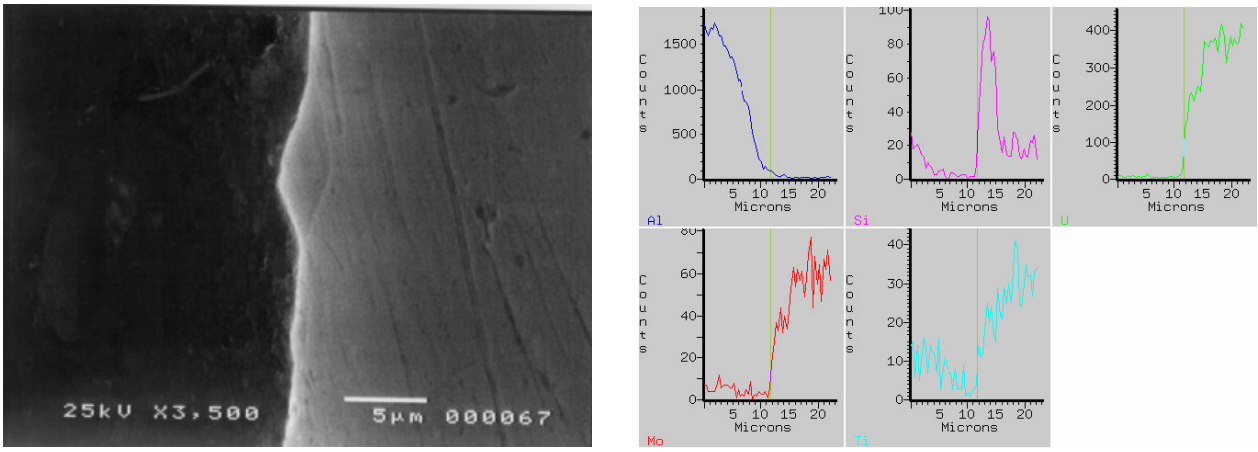

Figure 7. MEB image and scanning of IL in UMoTi/AlSi alloys couple after annealing.

From these interdiffusion studies it was possible to confirm that the interaction layer formation could be controlled by means of the addition of a third element without detectable effects in gamma phase stabilization. The additions of silicon to both couples constituents seems to be a way to control the IL, but the silicon effect in fuel may produce alterations in UMo alloy due to its affinity with U and its tendency to form U-Si compounds.

3.3 Under irradiation evaluation of U_3Si_2 Test Fuel

The cross sections diagrams for the original and fully modified designs are included in Figure 8 and the features of the Test Fuel Element manufactured at CCHEN are summarized in table 2.

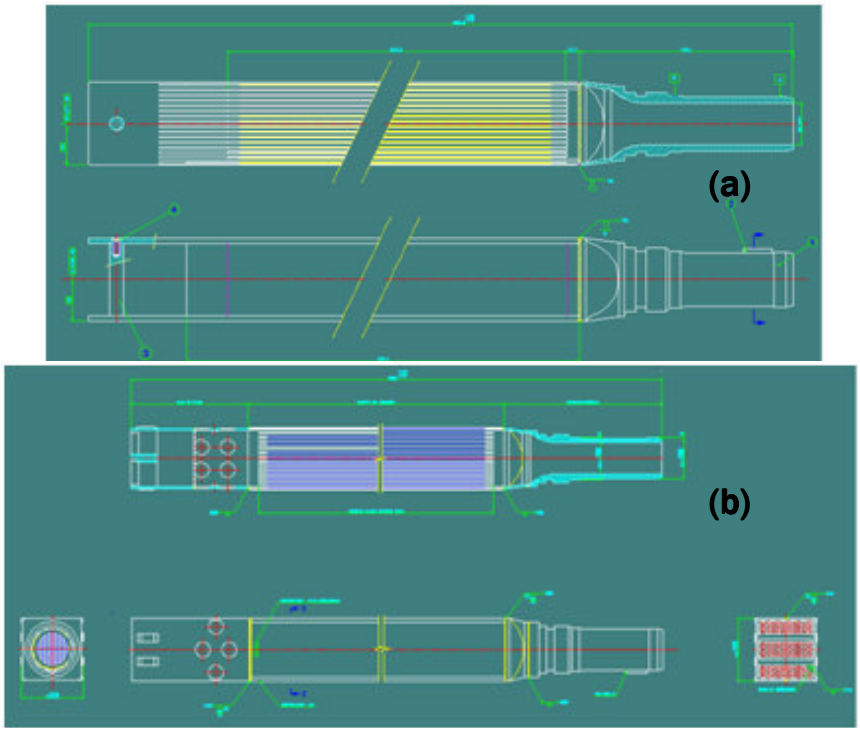

Figure 8. Longitudinal cross sections of SFE (a) and TFE (b) with modified design.

Table 2. Main features of RECH-1 standard and modified test fuel elements

	Standard RECH-1 Fuel (LEU)	HD Test Fuel (LEU)
Element identification code	LR-XY	LR-EX-01
Number of fuel plates	16	1
Enrichment [%]	19,75	19,75
Total U mass [g]	1087,2	102,21
^{235}U mass [g]	214,8	20,19
Fuel matrix	U_3Si_2-Al	U_3Si_2-Al
U-density [g/cm^3]	3,4	4,8
Fuel volume fraction [%]	32	43
Plate width [mm]	69,8	69,83
Plate length	625,5	625,36
Meat thickness [mm]	0,61	0,61
Meat width [mm]	60,15	60,5
Upper Cladding [mm]	0,46	0,47
Lower Cladding [mm]	0,46	0,46
Plate thickness [mm]	1,53	1,57
Coolant gap [mm]	2,92	2,92
Meat length [mm]	583,0	586,0
Structural Modifications		
Lower end assembly (Nozzle)	Two pieces, two welding Nozzle machined welded to a hot formed transition part	One piece, one welding Nozzle hot formed included the transition shape all in just one part
Upper inlet section Upper end assembly	Closed filter box, square area Filter box Filter plate Handling transverse parts All pieces are joined by multiple welding	Open, square area Not filter plate Filter box Handling transverse parts
Side plates	Longer than fuel plates until filter box welding	Longer than fuel plates until filter box
Welding	3 total rounded cord	2 total rounded cords: One to join lower end piece to fuel box and other to join filter box

The first follow up activities were done in July 25th, 2007. At that date the test fuel element was submitted to visual inspection and gap measurements. The results were according to expected. Until now, no abnormal performance or deviations has been observed.

4. CONCLUSIONS

- From experimental results it was possible to control the interaction layer formation by means of the addition of a third element without detectable effects in gamma phase stabilization
- The volume changes are directly related with the uranium density.
- In general, third element additions produce improvements in swelling behaviors.
- The best result was obtained for dispersed miniplates of UMo/AlSi-Mix and UMoSi/AlSi alloy, even better than U_3Si_2 for medium uranium density (3,4 gU/cm^3).
- The volume increase for unmodified UMo/Al system is five times higher than U_3Si_2, ten times higher than UMo/AlSi mix matrix and two times higher than UMoSi/pure Al matrix.
- The effect for the modified UMo/Al dispersed fuel was according to expected and the latest results showed decreasing in swelling values from five to less than two times.

5. ACKNOWLEDGEMENTS

The authors wish to express gratefully acknowledge to CCHEN by its support to this research and particularly the cooperation of the Fuel Facility staff in the miniplates manufacture and test performing.

6. REFERENCES

[1] J. L. Snelgrove, A. Languille "Qualification of uranium-molybdenum alloy fuel- Conclusions of an International Workshop". 4th International Topical Meeting of Research Reactor Fuel Management, RRFM-2000, Colmar, France, 19-21 March 2000.

[2] D. M. Wachs, R. G. Ambrosek, G. S. Chang, M. K. Meyer "Design and Status of RERTR Irradiation tests in the Advanced Test Reactor". International Meeting on Reduced Enrichment for Research and Test Reactors, RERTR, Cape Town, South Africa, 30 Oct-2 Nov, 2006.

[3] Mario Barrera, Jaime Lisboa, Jorge Marin, Luis Olivares, Klass Bakker, Fred Wijtsma, "Results for the recent activities of reduced enrichment program for research reactors in Chile" International Meeting on Reduced Enrichment for Research and Test Reactors, RERTR, Cape Town, South Africa, 30 Oct-2 Nov, 2006.

[4] Rodier, X. Iltis, F. Mazaudier, M. Cornen, S. Dubois, P. Lemoine "Effects of Ti in the UMo/Al system: Preliminary Results". 11th International Topical Meeting of Research Reactor Fuel Management, RRFM and Meeting of the International Group IGORR, Lyon, France, March, 11-15, 2007

[5] Autorización de Operación Unitaria Reactor Experimental Chileno RECH-1, AOU*RECH-1*01, CCHEN, DSNR, May 22th, 2007.

[6] Klein, J, Medel, J., Bustamante, S., "Irradiación de UN Elemento Combustible Experimental Fabricado con una Placa Activa de 4,8 g/cm3. Cálculos Neutrónicos y Termohidráulicos", CCHEN's Technical Report, Reactors Sub-Dept., March 2007.

[7] Technical Procedure PROC-RECH1- 04: Medición de Fuga de Productos de Fisión en Elementos Combustibles Mediante Ensayo Sipping, CCHEN, DAN, RECH-1 Reactor, June 21th, 1999.

[8] Novara, O., Lafuente, J., Rosa da Silva, J., Alencar, D., Klein, J., Loya, A., Salazar, A., Sindelar, R., " Recomendaciones para la Elaboración de un Programa de Inspección Visual de Combustibles Gastados Almacenados en Pileta", Project RLA/4/018, 2º Workshop of Characterisation of Spent Fuels, Buenos Aires, Argentina, 2 to 6 December 2002.