CURRENT STATUS AND DEVELOPMENT OF FUEL FOR RESEARCH REACTORS IN CHILE

Jorge Marín, Jaime Lisboa, Mario Barrera, Luis Olivares, Gonzalo Torres

Department of Nuclear Materials
Chilean Commission for Nuclear Energy - CCHEN
Amonategui 95, Santiago 6500687, Chile
Author contact: jmarin@cchen.cl

Abstract

CCHEN has developed, fabricated, and qualified MTR type fuel since 20 years, all of them have been loaded in both Chilean research reactors. Recently, more than 48 LEU uranium silicide fuel assemblies have been delivered to the Chilean research reactor La Reina- RECH-1. New local development deals with U-Mo fuel where, several activities has been completed such as casting of U-Mo alloys, phase stabilization studies, techniques for powder production, interaction, interdiffusion and out of pile swelling studies of standard and modified UMo/Al system. In parallel, for fission Mo, UMo foil targets are under development in the framework of an IAEA’s Coordinated Research Project, and some of the achievements are included in this paper.

1. Introduction

CCHEN has been involved in development of fuel for research reactors since 1980's. Actually 48 LEU high density dispersion fuel assemblies have been fabricated of \( U_{3}Si_{2} \) LEU with a uranium density of 3.4 g/cm\(^3\) for La Reina research reactor - RECH-1 (over 800 LEU fuel plates). The work was launched in 1987 when was necessary to disassemble and re-assemble 31 fuel elements for the other Chilean research reactor, RECH-2 at Lo Aguirre.

These task included inspection, X-ray examination of meat distribution, plates cold examination, redesign of some fuel parts, and re-assemble of fuel elements.

In 1998 new LEU fuel was designed for conversion of RECH-1, starting with loading in the reactor core four test fuel elements for irradiation behaviour surveillance. No fuel defects were observed and no performance problems were observed. Complementary, a Chilean test fuel element was fabricated for and irradiated in HFR, Petten, The Netherlands [1], achieving high burn up performance and an excellent PIE results.

CCHEN continues on the development of new fuel designs and new fuel technologies. In 2003 has started a programme for developing U-Mo compound. As a result of it, several activities have been carried out, [3], [4] such as casting of U-Mo alloys with Mo contents from 7 to 10wt%, phase transformations, gamma phase stabilization studies and several techniques for powder production, including cryogenic milling, high energy milling and grinding milling of machined chips. Particularly, interesting results from efficiency point of view, were obtained through hydration – milling – dehydration or HMD process applied to an UMo with special condition, deformed by cold rolling and crushed by impact. Also, they were carried out interaction, interdiffusion and out-of-pile swelling studies. Last year, UMo foil manufacturing, by means rolling, is under development. The final stage on this programme considers under irradiation evaluation of dispersed and monolithic miniplates.

Based on the irradiation results, is necessary to evaluate the different solutions aimed to stabilize an interaction layer zone produced by reactions of UMo fuel with standard Al matrix [5], as it is generally accepted. Among the different options studied, in this paper is included an experimental evaluation of the effects of Si addition to Al matrix and/or addition of a third element (Si, Zr, and Ti) to the UMo fuel.
In the other hand, suppressing the Al matrix employing just monolithic UMo as fuel meat seems to be an promising alternative solution based on the hypothesis that this fuel type can solve the U-Al interaction problem. In monolithic fuel the entirety of the fuel meat is comprised of a single foil of the fuel alloy. This fuel configuration represents the optimum in fuel meat density. The highly reduced fuel surface/volume relation and the fact that fuel-aluminium interfaces are in the cooler region of the plates should minimize the fuel-aluminium reaction [6]. Following this trend, CCHEN has started the development of technologies to obtain UMo foil as the first stage of the final aim; to have their own methodology for UMo monolithic fuel plates.

2. Experimental activities

UMo ingots were produced using an induction furnace placed inside a multipurpose chamber with controlled atmosphere. Ingots were obtained by melting natural uranium and Mo metal inside a high density alumina crucible and poured into a graphite mould. After casting, the ingots were annealed at 950°C for 24 hours in vacuum atmosphere (10–5 Torr) and cooled in argon in order to induce micro structural homogenisation and residual alpha phase transformation for gamma phase stabilization.

For dispersion fuel and interaction studies, fuel grade fine powder was necessary. To produce these powders, four techniques have been evaluated: Hydration - Milling – Dehydration (HMD), cryogenic grinding and mechanical grinding using high speed rotating blades made of several materials.

Fuel/Matrix interaction tests and out of pile swelling studies required more than twenty test miniplates. These dispersion miniplates, of pure UMo or modified by third element addition dispersed in Al matrixes, pure or alloyed with silicon, were manufactured employing the powder metallurgy conventional method.

Interaction tests, which results have been reported previously [2]-[4], involved metallographic preparation and inspection of samples extracted through punching of miniplates and annealed in quartz capsules vacuum sealed. After annealing for diffusion tests, the samples were analysed with SEM and EDS micro analyses of interaction layer (IL) regions formed by UMo particles surrounded by aluminium matrixes. Following the kinetics considerations given by the TTT curves of the U7Mo alloy, thermal annealing were performed to 550 °C for times up to 48 hours.

Based on interaction tests results and according to our experimental UMo program [7] the following step was to develop the swelling tests were performed. Taken into account that the swelling phenomenon produces thickness increasing, volume changes in miniplates can be assumed as thickness changes. Then, for these studies, out of pile tests were applied to dispersion miniplates. Air annealing carried out at 500°C followed by immersion density measurements were applied after each annealing treatment to all miniplates. This methodology permits leads to obtain global increasing trends for each fuel/matrix combination and it’s comparisons with reference U₃Si₂ miniplates.

In relation to monolithic UMo fuel, foil manufacture starts with casting of U-7Mo ingot. This casting was performed employing an induction melting furnace using a gravity pour into a graphite closed mould. Prior to hot rolling, the ingot is removed from the mould and annealed in a vacuum atmosphere to homogenize the microstructure. Because the ingot has enough thickness, the surfaces were machined under water to remove casting defects and to improve the surface ruggedness. Finally, the ingot was divided in four sections to produce equal a certain number of coupons. UMo foils for the monolithic test plates were produced by hot rolling of these coupons, which were sealed in a plain low carbon steel can (A37-24ES) to isolate the UMo alloy from the atmosphere during processing. The coupon/steel assemblies were repeatedly heated to 680°C and rolled at this temperature to reduce the thickness of the fuel meat from 5,7 mm until an average value of 0,49 mm. In a previous rolling test the thickness of a UMo fuel alloy was reduced from 2,5 mm to 0,32 mm also using only hot rolling.
3. Results and Discussion

3.1 Casting and microstructure homogenisation of U-7% wt Mo alloy

![Images of microstructure](a) (b) (c) (d)

Figure 1. Optical microscopy and SEM fracture surfaces (at room temperature) of U - 7% wt Mo alloy (a), (b) As cast, (c), (d) homogenised by vacuum annealing.

In a cast alloy, Fig. 1(a), the presence of two phases, a light matrix of gamma phase and a second phase, darker, precipitated in the gamma grain boundaries is observed and accordingly to X-RD analyses, it corresponds to alpha phase. In 1(c) image, the presence of the second phase is very few, product of its dissolution an homogenisation during the thermal treatment. Related to fractography analyses of images (b) and (d) of as cast and annealed samples respectively, the predominant fracture mechanisms corresponding to transgranular ductile fracture via micro void coalescence combined with minor evidences of cleavage along crystallographic planes (brittle fracture). According to Charpy tests carried out from −120 and +20°C, U-7% Mo alloy shows a brittle-ductile transition temperature in the range of 10 to 15 °C. Any mechanic method for powder production could overcome easily this temperature, promoting ductile fracture conditions.

2.2. Powder production

![Images of powder production](a) (b) (c) (d) (e)

Figure 3. SEM images of powder produced by several methodologies from U7Mo previously homogenised by annealing, cold rolled and crushed (R&C): (a) U7Mo-CTT-R&C cryogenic grinding, (b) U7Mo-CTT-R&C grinded by Ti blades, (c) U7Mo-CTT-R&C grinded by WC blades, (d) U7Mo-CTT-R&C Hydrated and (e) U7Mo-CTT-R&C Hydrated and Dehydrated.

In general terms, all grinding methods for powder production results with very low efficiency and in grinding with WC blades, small amounts of Co contamination was detected in powders. In the other hand, HMD process shown be efficient, specialty applied to cold rolled and crushed UMo alloy. Anyway, in order to produce UMo powder for subsequent dispersion test miniplates, enough amounts of UMo alloy were produced by means mechanical grinding using Ti blades. The next stage will be powder production and characterization of UMo-Ti and UMo-Zr alloys in R&C condition using HMD method.
3.3. Interaction tests in dispersion fuel miniplates

Figure 4. Morphology of Interaction Layers after 48 hours/550°C annealing. Comparison between UMo/Al (a) and modified UMo+Si/Al (b).

and Mo atoms from the fuel particles toward UMo/Al interlayer zone. Evidences of Al atoms migration from the matrix toward the outlying areas of UMo particles, where combines with U to form binary aluminides (UAlx) or ternary compound U-Mo-Alx were detected. Towards the centre of the fuel particles, also the presence of Al was detected in UMo+Si sample (b), which confirms the occurrence of the interdiffusion phenomenon in the interlayer zone. The addition of a third element allows to delay the interdiffusion phenomenon or at least to have some influence on the kinetics of growth of the interface region. These effects are evident when observing the thickness and morphology of the interface regions. (Figure 4). These results confirm the hypothesis outlined in previous works [2] in the sense that the second phase formed by the addition of the third element, and it’s preferable location in grain boundaries of UMo, it could constitute barriers to diffusion or atomic migration of the UMo/Al system. Compositions analyses verify the spontaneous migration of atoms of Si present in the Al-6061 cladding (0.6% wt%) toward the particles of UMo where, probably it form compounds with U and/or Mo. For the UMo with Si addition, this diffusive phenomenon is also verified but the presence of Si in the fuel particles makes less strong the concentration gradient and the interdiffusion of this element, appears in some grade, controlled. In consequence, if silicon atoms are presents in the UMo particles and Al matrix, it’s mobility appears slowed, thus they can constitute diffusion barriers by themselves. Other authors suggest that the addition of Si just has effect in the Al matrix [8]-[9], while the outlined hypothesis is coherent with other authors [10] in the sense that the addition of Si to the fuel phase (UMo) can help to avoid or limit the interdiffusion due to the action of precipitates (second phase) and also as effect of decreasing of the silicon concentration gradients. On the other hand, the Zr addition produces a very fine and homogeneous dispersion of this element in the entire UMo microstructure, without preferential location or segregations. By means of this mechanism the Zr could be causing restrictions to movement of dislocations and vacancies and/or formation of precipitated in the grain boundaries, all mechanisms that constitute barriers to the diffusion. Titanium act in very similar form inside the UMo particles, with the difference that Ti experienced preferential location in the interface, probably, for their

Figure 5. SEM images of (a) UMo, (b) UMo+Si, (c) UMo+Ti and (d) UMo+Zr particles dispersed in Al matrixes after 48 hours/500°C (vacuum) annealing.

SEM combined with EDS concentration profiles analyses applied to UMo samples shown in figure 5 reveal the occurrence of mechanisms of interdiffusion of U
affinity with the Al. The mechanisms for which the third element is capable to control the thickness and the composition of the reaction layer are relatively clear and keep certain relationship with disincentive, for some mechanism, the atomic mobility.

### 3.4. Out-of-Pile swelling tests applied to dispersion fuel miniplates

![Graph](image)

**Figure 6.** Volume increase v/s annealing time for UMoMe alloy dispersed in Al matrixes Summarized result for 500°C

According to the swelling test results, the volume changes are directly related with the uranium density, and in general, third element additions result in improvements in swelling behaviours. Comparatively, the best result was obtained for dispersed miniplates made of UMo/AlSi-Mix followed by UMoSi/AlSi alloy, both slightly better than U₃Si₂ for similar range of uranium density.

The volume increase for unmodified UMo/Al system achieves levels almost three fold higher than those achieved with U₃Si₂; however for system UMo/AlSi mix, these undesired behaviour was reduced to values equivalent or slightly lower than for U₃Si₂.

### 2.5. Development of monolithic U-Mo fuel

![Images](image)

**Figure 7.** Sequence of UMo foil manufacture in CCHEN. (a) Ingot casting, (b) underwater machining of ingot surfaces, (c) UMo ingot, (d) ingot divided in four coupons, (e) UMo coupon sealed in steel assembly for hot rolling, (f) foils manufactured by hot rolling (0.49 mm thickness) and (g) exploratory test of cold rolling (0.32 mm thickness).

In figure 7, the sequence to obtain UMo foils includes ingot casting, machined and cutting in four sections or coupons, which were encapsulated in steel assemblies. The coupons are hot rolled and, after several rolling steps, the thickness was reduced from 5.7 to 0.49 mm (91.4%). In a previous test, an UMo foil with thickness of 320 µm was achieved by hot rolling, with total reduction of 86.9%. For the next step, new steel cans will be required to continues hot rolling until reduce foil thickness to about 180-220 µm. Finally, limited cold rolling (5% or less) would be applied to UMo foils just to improve the surface finish and stiffness increasing. The following step would be UMo-10wt% alloy foil manufacture and, finally with the
U7Mo and U10Mo foils, to select a suitable UMo/Al6061 bonding methods in order to begin the manufacture of monolithic fuel plate.

4. Conclusions

Based on results of characterization and testing described above for dispersion fuel miniplates and monolithic fuel, the following conclusions can be drawn:

The volume changes are directly related with the uranium density and for similar annealing condition, the unmodified UMo/Al system exhibited swelling levels almost three times higher than those achieved with $\text{U}_3\text{Si}_2$. However for the system UMo/AlSi mix, this undesired behaviour was reduced to values equivalent or slightly lower than for $\text{U}_3\text{Si}_2$

Out of pile swelling results indicates that the modification by silicon addition is more effective in the matrix than in the fuel alloy.

Manufacture of UMo foil for monolithic fuel has been achieved successfully.

5. Acknowledgements

The authors are grateful for the support received from CCHEN through it’s Nuclear Materials Department and specially from technical staff members of Fuel Element Plant – PEC.

6. References