FRM II AND AREVA-CERCA COMMON EFFORT ON MONOLITHIC UMO PLATE PRODUCTION

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ABSTRACT
Since 2003 the Technische Universität München (TUM) is strongly engaged on the UMo fuel development program. Dispersive UMo fuel type was firstly investigated through collaborative efforts involving CEA, TUM and AREVA-CERCA. IRIS TUM irradiation and associated PIEs were presented during international conferences. The European consortium also studied the fabrication of UMo monolithic fuel plates during 2005-2007 and tentatively manufactured full size plates for the irradiation program IRIS V. As the program unfolded technical information were obtained and gathered with the international community. UMo foils were produced at laboratory scale and different methods to clad the UMo foils within aluminium were investigated. Based on these first results TUM and AREVA-CERCA have decided to pursue a common effort on the development of monolithic fuel plates with the purpose of minimizing the enrichment in use. Emphasis will be more dedicated to the foil and cladding package preparation and further studies will be performed in order to investigate various processing techniques to join the fuel foil with the cladding. As a complete new approach for the manufacturing of monolithic foils and for foil cladding the DC magnetron sputtering technique is investigated. A full R&D program was defined between TUM and AREVA-CERCA. This paper aims at presenting the program, discusses the selected options and first results will be presented.

1. Introduction
The monolithic fuel concept presented by INL early in 2002 allows the possible LEU conversion of high performance reactors which can’t be converted using dispersed UMo fuel. The basic principle is to replace a dispersive fuel form with a density of 8 gU/cc by a solid form of UMo where a density close to 17 gU/cc can be reached.

The technical challenges for this kind of fuel are multiplex: Instead of having numerous interfaces between the matrix and each grain the interface is now reduced to two large interfaces on top and the bottom of the monolithic foil. Which kind of interlayer is best suited for an excellent adhesion of the cladding and for suppressing the interdiffusion layer? How to produce large foils with a typical thickness of 300 – 500 µm on an industrial scale to affordable costs? How to introduce gradients in thickness for the foils? Can the assembled fuel plate be shaped to geometries others than flat? And last but not least, how do large tests plates behave under in-pile irradiations which simulate the heat load and burn-up of high performance reactors?

• AREVA-CERCA, a subsidiary of AREVA NP, an AREVA and SIEMENS company
2. State of the art

For fuel plate design, new processes technologies as friction welding (FW), AREVA-CERCAs welding and hot isostatic pressure (HIP) were proposed and evaluated during the last few years. Miniplates manufactured by INL through FW and HIP processing were irradiated in ATR according to the RERTR irradiation program. These irradiation results have brought a basic set of information regarding monolithic fuel concept behavior under irradiation. Mainly the weak interaction layer (IL) which is formed during irradiation at the UMo foil and Al cladding front edge interface should be eliminated to prevent any detrimental debonding in the course of the fuel burn-up during operation.

Anyhow at the same time a diffusion barrier in between the different materials to be bounded must be observed in order to withstand the mechanical stress in use as well as guaranty the heat dissipation required by the performance of the fuel during irradiation. A dedicated material which can simultaneously bring a good mechanical bound between the cladding and UMo foil and also reduces the formation of the IL should be defined.

A full R&D program is then required to down select such materials and to study a processing method to form this barrier. The beginning easy principle becomes more complex and a monolithic plate appears as a challenge to assemble a multilayer material: cover, barrier and UMo foil. AlSi or Zr layer have been proposed as a remedy for the bonding and diffusion problem and are still being investigated by INL [1].

More challenging, the fuel meat which is a solid UMo foil has to be produced at a large scale and throughout a cost effective processing way. UMo foils elaboration at a laboratory scale was described previously [2] and developments are ongoing at Y12 complex to scale up the UMo foils production [3].

The UMo foil production must include a step where the needed barrier that should wrap up the UMo foil surface has to be processed. Specifically shaped fuel plates including gradients in the thickness of the UMo foil have also to be considered as it may be necessary for research reactors with highest power density. This aspect needs to be further studied in order to integrate this demand early in the manufacturing foil development stage. Dedicated program was recently launched by INL to evaluate the production of shaped UMo foils [4].

As previously described a stainless material barrier can also be advantageously used to protect the UMo foils from oxidation prior or during monolithic plate processing [5].

Recently 4 full size monolithic plates composed of AlSi and Zr barrier processed by FW were irradiated through AFIP 2 experimental test with a peak power up 350 W/cm² (US/DOE program carried-out by INL). PIEs results are awaited for this year. The preliminary ultrasonic test inspection results performed by INL seem to indicate a relative good plate behavior under irradiation. The prototypes manufacturing results presented by INL state the difficulties to master routinely the monolithic plate fabrication.

Following a common effort started 5 years ago through a collaborative program carried out between CEA, AREVA-CERCA and TUM, TUM and AREVA-CERCA have commonly defined a new R&D program aiming at evaluating and testing the main challenges on the road of monolithic plate processing. In order to save time and taking into account to the ongoing development dedicated to UMo foil production our program will be conducted using depleted UMo foils coming from dedicated outside entities.

The monolithic program has been split down into sub items as shown in Table 1.
The program is scheduled to start this year and final results are forecasted at the latest beginning of 2011. As soon as successful results will be available an irradiation program encompassing the best down selection options will follow. In the following section each item is further detailed.

<table>
<thead>
<tr>
<th>Items</th>
<th>Objective</th>
<th>Dedicated tools</th>
</tr>
</thead>
<tbody>
<tr>
<td>Depleted UMo Foil</td>
<td>To have foils in order to carry-out the program (uncoated and coated foils)</td>
<td></td>
</tr>
<tr>
<td>Material down selection &amp; processing</td>
<td>To down select an appropriate material barrier between UMo foil and cladding</td>
<td>Heavy ion irradiation of a set of UMo samples by FRM II</td>
</tr>
<tr>
<td></td>
<td>To deposite a selected material on UMo foils</td>
<td>Sputtering at FRM II</td>
</tr>
<tr>
<td>Monolithic plate processing</td>
<td>To investigate the material compatibility through the selected monolithic plate processing</td>
<td>AREVA-CERCAs machineries</td>
</tr>
<tr>
<td></td>
<td>To further evaluate monolithic plate processing through AREVA-CERCAs processing options i.e. C2TWP and/or HIP</td>
<td>FRM II characterization tool box</td>
</tr>
</tbody>
</table>

Table 1: FRM II & AREVA-CERCA monolithic program summary

3 Material down selection & processing

3.1 Options to reduce the formation of an unwanted IL at the UMo-Al interface

Regarding the UMo/Al system there are in principal three possibilities reported to avoid the formation of an undesired IL during pile irradiation. A literature inquiry in current and older publications revealed the most promising candidates:

a) Addition of a diffusion-limiting element to the Al-matrix which contains the UMo particles.

It has been found that the addition of Silicon to the Aluminium matrix limits the formation of an interdiffusion layer at the interface UMo-Al. However, uncertainties remain for the optimal Silicon addition that should be added to the Aluminium matrix [6, 7, 8]. Moreover, diffusion coefficients given in older literature show, that the addition of Titan, Bismuth, Beryllium or Antimony to the Aluminium matrix is even more powerful in suppressing the formation of the interdiffusion layer than Silicon. Especially Bi has been found to be most promising due to its low neutron cross section and its high density [9].

b) Usage of ternary UMo alloys.

Creation of ternary U-Mo-X alloys seems to have in some cases a suppressing effect on the formation of the interdiffusion layer. The main reason for this effect is the stabilization of the UMo \(\gamma\)-phase by the third element [10,11]. Consequently, the addition of some wt\% of Ti, Nb, Pt, Si or Pd to the U-Mo has been considered [12,13].

c) Insertion of a diffusion barrier at the interface UMo-Al.
It is known since the early days of metallic fuel element development that an oxide layer at the interface Uranium-Aluminum prevents the formation of a diffusion layer very effectively [9]. Also in UMo-Al test fuel plates irradiated in-pile an oxide layer around the UMo particles has proven its effectiveness [14]. Furthermore, Nb, Ta, Ti or Zr has been proposed as a diffusion barrier [14, 15, 16].

d) Any combination of a), b) and c)

e) Usage of a completely different matrix material, such as Magnesium.

In 2006 it has been shown that it is possible to emulate the IL growth during in-pile irradiation of UMo/Al specimens by out-of-pile irradiation with \(^{127}\)Iodine at 80 MeV. Typical burn-up fission densities are achieved within a few hours [17,18]. Because the energy of the ions usually is far below the Coulomb barrier the UMo samples are not activated during heavy ion irradiation. They are therefore easily accessible with normal laboratory equipment (SEM, EDX, and XRD).

Since then, considerable progress has been made to improve the reliability of this method. In collaboration with CEA-Cadarache, TUM has build up a complete new irradiation setup at the tandem accelerator in Garching (Maier-Leibnitz Laboratorium) which allows monitoring and controlling the irradiation conditions like flux, fluency, vacuum and sampling temperature automatically. The new setup allows the quick irradiation of different kinds of samples [19].

Consequently, we decided to start an irradiation campaign at the tandem accelerator in Garching to screen as many combinations of the materials mentioned above as reasonably possible. For the irradiations UMo powder dispersed in an Aluminum matrix has been chosen. The results are also valid for the here described monolithic fuel development program, because anyhow the interaction between Al matrix, IL and UMo will be the focus of our characterization.

We decided to divide the options mentioned into three parts (compare Tab. 2). For each option, one miniplate (in total 20) has been provided by AREVA-CERCA.

The first part consists of atomized U7wt%Mo powder dispersed in an Al matrix with and without addition of secondary elements. 2wt%Si, 5wt%Si and 7wt%Si have been chosen as addition to the Al matrix to find the best Silicon concentration. Furthermore, 2wt%Ti, 2wt%Bi and 5wt%Bi have been added to the Al matrix, respectively, to check the positive effect of these elements. It has been reported, that the addition of Magnesium to Aluminum accelerates the diffusion at the U-Al interface [8]. In consequence, 2wt%Mg has been added to the matrix to check this effect. In each case the samples have been prepared with and without an oxide layer (UO\(_2\)) of ~2µm thickness around the UMo particles, to check its effectiveness as a diffusion barrier – second part.

The third part consists of ternary U8wt%Mo-x ground powder dispersed in a pure Aluminum matrix. To study the principal effect of alloying the UMo, 1wt%Ti, 1,5wt%Nb, 3wt%Nb and 1wt%Pt have been added. One miniplate consisted of UMo dispersed in Magnesium, covered with AlFeNi. It was not possible to prepare a sample for irradiation from this miniplate. The meat was grayish and very brittle. It broke apart during cutting and polishing.

It was not possible to obtain UMo particles coated with metals like Ta or Ti to check the impact of those elements on the formation of the diffusion layer during bombardment with heavy ions. However, examinations on this issue are planned on monolithic UMo/Al layer systems that will be prepared with the newly installed sputtering device.

From the 20 miniplates provided by AREVA-CERCA 60 samples have been prepared for irradiation with \(^{127}\)Iodine at 80 MeV. First irradiations have been performed since fall 2008 with an integral of 1x10\(^{17}\) ions/cm\(^2\) for each sample. This corresponds to a full burn-up of a
high flux research reactor. The irradiation temperature was \( \sim 200^\circ C \). The irradiation campaign will be finished until summer 2009. Examinations on already irradiated samples are ongoing. The full set of data will be available until end of 2009.

<table>
<thead>
<tr>
<th>Sample Nr.</th>
<th>Alloy</th>
<th>Matrix</th>
<th>Comment</th>
</tr>
</thead>
<tbody>
<tr>
<td>MAFIA-I-1*</td>
<td>U-7Mo</td>
<td>Al-atomized</td>
<td>Reference specimen</td>
</tr>
<tr>
<td>MAFIA-I-2*</td>
<td>U-7Mo-ox</td>
<td>Al-atomized</td>
<td></td>
</tr>
<tr>
<td>MAFIA-I-3*</td>
<td>U-7Mo</td>
<td>Al98-Si2</td>
<td></td>
</tr>
<tr>
<td>MAFIA-I-4*</td>
<td>U-7Mo-ox</td>
<td>Al98-Si2</td>
<td></td>
</tr>
<tr>
<td>MAFIA-I-5*</td>
<td>U-7Mo</td>
<td>Al95-Si5</td>
<td>Different Si concentrations to find the best concentration</td>
</tr>
<tr>
<td>MAFIA-I-6*</td>
<td>U-7Mo-ox</td>
<td>Al95-Si5</td>
<td></td>
</tr>
<tr>
<td>MAFIA-I-7*</td>
<td>U-7Mo</td>
<td>Al93-Si7</td>
<td></td>
</tr>
<tr>
<td>MAFIA-I-8</td>
<td>U-7Mo-ox</td>
<td>Al93-Si7</td>
<td></td>
</tr>
<tr>
<td>MAFIA-I-9</td>
<td>U-7Mo</td>
<td>Al98-Mg2</td>
<td>Mg accelerates formation of IDL.</td>
</tr>
<tr>
<td>MAFIA-I-10</td>
<td>U-7Mo-ox</td>
<td>Al98-Mg2</td>
<td>Reproduction of this effect.</td>
</tr>
<tr>
<td>MAFIA-I-11</td>
<td>U-7Mo</td>
<td>Al98-Ti2</td>
<td>Study the effect of Ti on IDL formation</td>
</tr>
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<td>U-7Mo-ox</td>
<td>Al98-Ti2</td>
<td></td>
</tr>
<tr>
<td>MAFIA-I-13</td>
<td>U-7Mo</td>
<td>Al98-Bi2</td>
<td>Different Bi concentrations to find the best concentration</td>
</tr>
<tr>
<td>MAFIA-I-14</td>
<td>U-7Mo-ox</td>
<td>Al98-Bi2</td>
<td></td>
</tr>
<tr>
<td>MAFIA-I-15</td>
<td>U-7Mo</td>
<td>Al95-Bi5</td>
<td></td>
</tr>
<tr>
<td>MAFIA-I-16</td>
<td>U-7Mo-ox</td>
<td>Al95-Bi5</td>
<td></td>
</tr>
<tr>
<td>MAFIA-I-17</td>
<td>U-7Mo</td>
<td>Mg</td>
<td>Mg matrix, did not work, brittle matrix, matrix with no adhesion to cladding</td>
</tr>
<tr>
<td>MAFIA-I-18</td>
<td>U-7Mo-1Ti</td>
<td>Al-ground</td>
<td>To study principal effect of alloying the UMo on formation of IDL.</td>
</tr>
<tr>
<td>MAFIA-I-19</td>
<td>U-7Mo-1.5Nb</td>
<td>Al-ground</td>
<td></td>
</tr>
<tr>
<td>MAFIA-I-20</td>
<td>U-7Mo-3Nb</td>
<td>Al-ground</td>
<td></td>
</tr>
<tr>
<td>MAFIA-I-21</td>
<td>U-7Mo-1Pt</td>
<td>Al-ground</td>
<td></td>
</tr>
</tbody>
</table>

Tab 2: List of mini-plates provided by AREVA-CERCA for heavy-ion irradiation. The irradiation has been completed for miniplates earmarked with (*).

### 3.2 Sputtering techniques

The sputtering process offers the advantage, that perfect layers from any material can be grown on any substrate in any size. This means to the first, that monolithic full size foils and blank sheets from any given UMo alloy can successfully be produced. It means to the second, that a given UMo foil or blank sheet can be surrounded with a layer of any desired material, be it as a diffusion barrier or as a cladding. It means to the third, that bonding between the different layers is not a problem at all, because the layers will have the maximum physically possible adhesion to each other.

On laboratory scale the sputtering technique provides the opportunity to quickly produce small numbers of high quality full size fuel plates for irradiation tests and further examinations, which is our primary aim. The applicability of sputtering to industrial scale seems promising but has to be further examined.

Over the last two years TUM has built up two DC magnetron sputtering assemblies: a small tabletop setup for the production of samples sized 100mm x 100mm as well as a large plant – see Fig. 1 - for the fabrication of plates sized 700mm x 65mm. Both assemblies have successfully been operated in the past with surrogate materials as copper or stainless steel and the process of full size foil production as well as the process of cladding respectively depositing a barrier layer on these foils have been shown successfully [20].
End of 2008 first DU-8wt.%Mo (depleted Uranium) foils have been sputtered successfully. DU-8wt.%Mo foils with 120mm x 50mm in size and 150 µm thickness were produced inside the tabletop assembly in 28 hours of sputtering. The surface quality of these foils is still poor due to thermal stress effects. A phase analysis of the deposited layer showed the DU-8wt.%Mo to be in the desired $\gamma$-phase after the sputtering process. These experiments are continued with the aim to reduce the thermal stresses.

In parallel the large sputtering plant was mounted inside a glove box to enable operation under inert atmosphere. The inert atmosphere guarantees, that the concentration of oxygen during the production of foils, barrier layers or cladding and even during handling of the material is always below 10ppm, which results in a nearly complete suppression of oxidation and oxide layer formation in all process steps.

The sputtering plant is currently installed in a radioisotope laboratory and will be ready for operation within the next weeks.

The main emphasis of the large sputtering device will be barrier coating and cladding deposition. Manufacturing of full size foils by sputtering DU-8wt.%Mo will be continued, too. But the perspectives to introduce this technique for an industrial production of UMo foils are still in the future.

4 Monolithic plates processing

As the main conclusion of the monolithic plate processing program carried-out in 2005-2007 we found that the difficulty encountered to clad an UMo foil with Al covers using an AREVA-CERCAs methodology was linked to the UMo foil oxidation during plate processing. This effect was especially observed during the production of UMo full size plates [2]. Replacing the UMo by a stainless steel foil of the same thickness a perfect bounding junction was always obtained. During these tests the UMo foil surface was free of any material barrier which is today strongly advised for preventing any wrong irradiation behaviour due to the IL growth formation at the Al/UMo interface. This barrier can also be advantageously used to prevent the oxidation of the UMo foil and then improved the efficiency of our processing method. Processing parameters can also be adjusted in order to reduce the temperature preventing oxidation.
The new program is structured into several phases where a material compatibility study shall be performed first. This stage should be performed to validate the compatibility of the barrier material we want to use with the Al covers and also the processing tool deployed. After this material down selection (heavy ions studies and compatibility aspects) miniplates and full size plates will be processed. In case of unexpected results recorded with this first manufacturing option a processing back-up program will be launched. The back-up option is based on HIP processing.

As previously did the quality of multilayer interfaces bounding will be checked through ultrasonic test (UT), metallographic inspection and dedicated mechanical test at TUM. The characterization tools available are presented in the following section. As proposed and already under evaluation in the US side the first evaluation will start with Zr coated depleted UMo foil.

5 Characterization

The here presented R&D program will benefit from the huge variety of methods and competences for the physical and chemical characterization, which are offered by the university campus at Garching. The nuclear operation licences of the Institute of Radiochemistry and the FRM II itself allow the handling of α-emitters like Uranium. Scanning electron microscope and EDX can be used with open α-emitters. XRD will serve for structural and phase analysis. FRM II itself is one of the leading neutron scattering facilities and offers among others non-destructive analysis of the internal strains introduced by the processing techniques (deposition, plate processing and bending). Micro focus beams of synchrotron radiation allow the spatial resolution of structures on a scale smaller than 1 µm [21]. Precise chemical element analysis serves as a gauging for locally resolved EDX analysis. Mechanical pulling tests will check the adhesion of the sputtered layers.

6. Conclusion

A new R&D monolithic program was defined in collaboration with AREVA-CERCA and TUM.

From barrier material down selection to monolithic plate processing our program encompasses the overall identified aspect which needs to be investigated to successfully produce a monolithic UMo plate.

The tools used will be adapted according to the maturity of the technology from a R&D lab scale to an industrial workshop. A part of the results will benefit to the ongoing UMo dispersive development program.

As soon as our program will answer to the main challenges, an irradiation of monolithic UMo LEU plates will be considered.
References


