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DEVELOPMENT OF UMO/AL DISPERSION FUEL: AN OXIDE LAYER AS A PROTECTIVE BARRIER AROUND THE UMO PARTICLES

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ABSTRACT

This paper presents the out-of-pile studies carried out to select a promising advanced UMo/Al dispersion fuel. Investigations concerning UMo particles surrounded by an oxide layer as a protective barrier are detailed and discussed. The presentation is divided in three items. Firstly, a few words argue the choice of the oxide way. In the second part, the characterizations of the fuel plates fabricated with oxidized UMo are presented. The effect of silicon addition in the matrix has also been studied. At last, the behaviour of the samples under heavy ion irradiation is described, in comparison with samples without any oxide content. These irradiations evidence the positive effect of an oxide layer surrounding the UMo particles. These results may help to define one further French irradiation in OSIRIS, the IRIS4 experiment, which could be in-pile tested in 2007.

1. Introduction

Since past in-pile experiments have shown that UMo dispersion fuel in a pure Al matrix does not withstand high operating conditions [1], [2], [3], many studies have been engaged to solve the unacceptable pillowing and/or swelling of the fuel plate. As the interaction layer formed at the UMo/Al interface is incriminated, the development of an advanced UMo/Al dispersion fuel has two objectives:

- either decrease the interaction formation kinetics,
- or/and improve the interaction zone in-pile behaviour, namely its fission gas retention.

To reach these objectives, several approaches are worldwide proposed, consisting in either improving the knowledge of the U-Mo-Al system or in testing new solutions under irradiation. They are of three types:

1. Modification of the UMo particles. Mostly studied are ternary alloys. Ti, Zr, Nb and Si are the main elements added to UMo [4, 5, 6]. They aim at decreasing the reactivity between the UMo

♦ AREVA-CERCA, a subsidiary of AREVA NP, an AREVA and Siemens company

and the Al matrix or at stabilizing the UAl_3 phase which has a good in-pile behaviour. There are also trials to modify the microstructure of the UMo particles. Recently, the IRIS-TUM experiment, has tested in the OSIRIS reactor, the behaviour of fuel plates made of UMo ground powder in Al dispersion fuel. In most past experiments, atomized powders were used. The two kinds of powder differ from a microstructural, chemical and surface state point of view.

2. Modification of the Al matrix. Three ways are investigated :
   - Alloy addition in the matrix, such as Si and more general tetravalent ions [4, 7].
   - Dispersion of a stabilized porosity (ex porous material) in the aluminum matrix, to increase the fission gas retention.
   - Replacement of aluminium by magnesium in the matrix (cf. RERTR3 program). But the manufacturing of such a fuel appears to be more difficult because of the existence of the low temperature eutectic between Mg and Al from the cladding.
3. Modification of the UMo/Al interface by the use of coating materials or diffusion barriers around the UMo grain. Some studies are in progress concerning coating development (Si, Nb, Zr-Nb, Ni…) or thermochemical treatment of UMo particle (oxidation) [7, 8].

Note that monolithic UMo fuel is also under development, i.e. without any Al matrix, as an alternative way to dispersed fuel [9, 10, 11, 12].
This paper presents our out-of-pile studies, led in the framework of the CEA/AREVA-CERCA collaboration, concerning the development of an advanced dispersed fuel in which the UMo/Al interface has been modified. Especially the presence of an oxide layer surrounding the UMo particles has been studied. In section 2 we argue about the choice of the 'oxide route'. In section 3 the fabrication of the plates - made of oxide particles - is described. Their behaviour under ions irradiation, studied in collaboration with TUM and AREVA-CERCA, is presented in section 4. According to these results, this new fuel seems to be an interesting candidate for future in-pile irradiations.

2. Justification of the choice of an oxide layer at the interface

Several reasons have justified the choice of an oxide as a barrier to the UMo/Al interaction.
First, thermal treatments performed on dispersed fuel plates, but also on diffusion couples, showed that the thickness of the reaction layer at the UMo/Al interface is quite different for a sample with oxide coating compared to one without (Figure 1). The UMo alloy can be totally consumed, while a very thin interaction layer is formed for particles surrounded by an oxide layer (Figure 1.b). The oxide certainly hinders the interdiffusion of U, Mo and Al.

-a- -b-

Figure 1: Micrographs of UMo7/Al dispersion fuel annealed at 600°C/10h.

-a- some particles react (strong reaction),

-b- just a slight reaction for some particles having an initial surface oxidation. The UO_2 layer became thicker after thermal treatment and spread inside the UMo.

Moreover, in past French IRIS1 experiment (ground powder instead of atomized UMo) [13] UMo plates were successfully irradiated to a high burn-up. The PIE of IRIS1 fuel showed that the interaction layer was thicker around oxidized UMo. The oxidation was induced by the grinding

process. However a maximum cladding temperature around 75°C and a peak heat flux of 140 W/cm^2 is considered as too low for being representative for future applications in high flux neutron sources.
Ground powders (UMo8) have also been used in a recent experiment, IRIS-TUM, ended in September 2006. Partial results have been presented at the last RRFM meeting [14]. For a heat flux up to a 300 W/cm^2 and a fission density of 3.8 10^{21} f/cm$^3_{UMo}$ no abnormal swelling has been observed, independently whether a pure Al matrix or an Al matrix with Si addition has been chosen [15]. Actually, atomized and ground powders differ in the induced plate microstructure like porosity, fissile particle morphology.... It is expected, that future PIE on the plates of the IRIS-TUM irradiation will reveal details of the microscopic storage and swelling behaviour.
At last, oxidation effects have been intently tested in a current Russian experiment [7]. It consists in a dispersed fuel elaborated with a 1-2 µm UO_2 layer around UMo atomized particles. The latest results will be probably presented soon.

3. Fuel samples (miniplates) manufacturing

The oxide layer should have the following features, as a compromise between a sensitive barrier effect, a decrease of the uranium density and an alteration of the thermomechanical properties:

- a thickness of a few microns,
- a cubic symmetry phase, like UO_2
- a good adherence.

In the following tests, the depleted UMo alloy is made of atomized powders, with 7 wt % Mo content. The particle diameter is in the range of 0-40 µm with an average size of about 15 µm. The UMo powders were oxidized [16].
The miniplates (typically a few cm^2) are manufactured at the CERCA by conventional rolling. They are characterized by a uranium density of 7 gU/cm^3. The matrix is either pure Al or AlSi matrix (2 wt%Si). Indeed, recent in-pile experiments (RERTR6, IRIS3, IRIS-TUM) state the benefit of silicon addition to aluminium matrix [4, 14, 17].
The two oxidized samples (samples 3 and 4 in table 1) are compared to those previously studied without any oxidation layer surrounding UMo, and considered as reference (samples 1 and 2 in table 1) [18].
Thus, four samples, noted sample 1 to 4 will be discussed.

	Sample 1	Sample 2	Sample 3	Sample 4
Notation	UMo/Al	UMo/Al,Si	UMoO/Al	UMoO/Al,Si
Matrix type	Al A5	Al98Si2	Al A5	Al98Si2

Table 1: Sample composition.

The oxide layer is characterized after roll-bonding (Figure 2). The average thickness of the oxide layer has been measured in the range of 1.4-1.7 µm. The behaviour of the oxide layer is satisfactory during the fabrication process. It is altered in only a few areas (Figure 2.b). On XRD pattern three phases are detected (Figure 2.c), Al (matrix), UMo (in γ-phase) and UO_2. The UO_2/UMo ratio thus determined is quite consistent with the oxide thickness and the UMo particle size. It's also interesting to notice that the weight fractions of the three phases are close to those of the IRIS 1 type sample, the ground powder based fuel (Figure 2.d). This means that the global oxide content is similar in a thermal controlled oxidized sample and in a mechanically oxidized sample, as opposed to the sample 1 (UMo/Al fuel with atomized powder), in which no UO_2 content is measured.

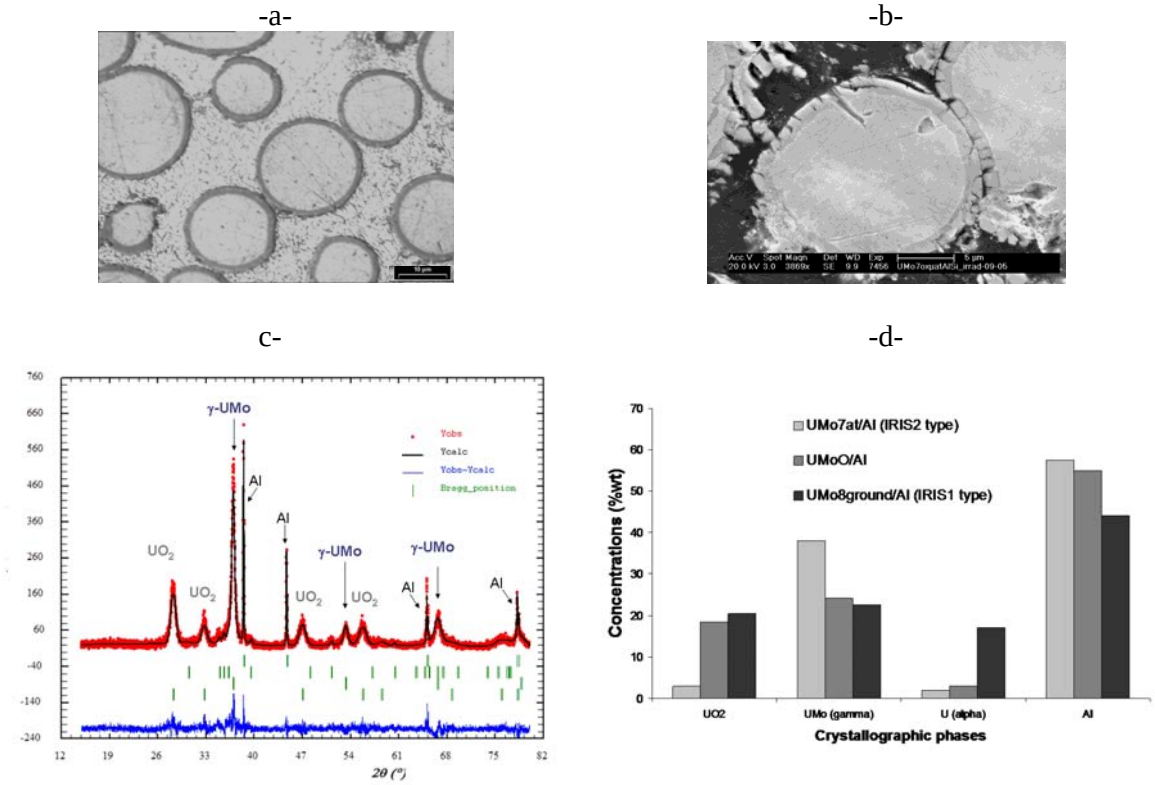


Figure 2: Characterization of sample 3 (UMoO/Al). Optical micrograph (a), SEM micrograph (b) XRD pattern (c), weight % of each crystalline phase (d).

The samples were cut from plates and processed for the heavy ion irradiation (cf. Figure 3 [18]).

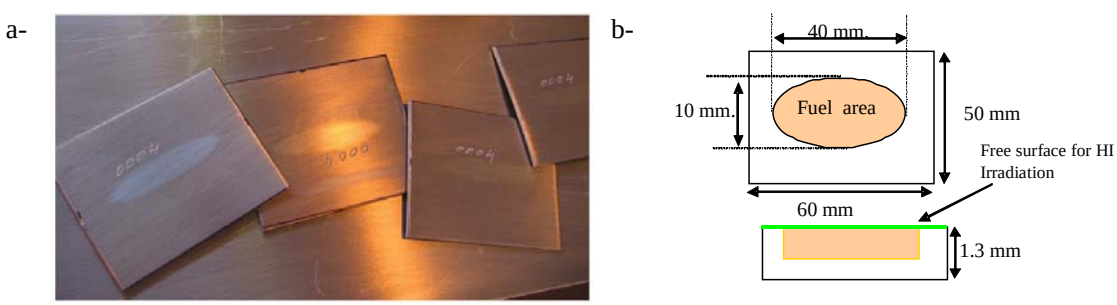


Figure 3: Mini-plates description (photos (a-) and schemes (b-)).

4. Heavy ion irradiation experiment

4.1. Experimental procedure

As previously reported [18], the experiment has been carried out at the Munich 14MV tandem accelerator in Garching, Germany, under the conditions stated in Table 2). Each sample was irradiated with ^{127}I at 80 MeV under secondary vacuum. A maximum heavy ion flux of $1.4*10^{12}$ sec^{-1} (i.e.18 Watt) was chosen in order to limit the specimen temperature to a maximum of 170°C, which was measured in-situ by a thermocouple on the rear side of the specimen. This temperature conditions should avoid a thermally activated diffusion or decomposition of the metastable γ-UMo phase. It is low compared to annealing test, but slightly higher or similar to in-pile conditions. The samples were screwed on a water-cooled aluminium bloc. After almost four hours of irradiation, the final fluence was achieved in an area of 4*4mm². The final ion density in the UMo particles (8.10^{20}

ion/cm^3) can be compared to the meat fission fragment density obtained in past in pile experiments, 2-4.10^{21}FF/ cm^3 for IRIS1 and FUTURE [2].

Fluence (Ion/cm²)	2.0*10^{17}
Energy (MeV)	80
Penetration depth into UMo (µm)	5
Penetration depth into Al (µm)	12.9
Max. surface temperature (°C)	170
Ion-density in UMo (Ion/cm^3)	8.0*10^{20}
Ion-density in Al (Ion/cm^3)	3.1*10^{20}

Table 2: Conditions for heavy ion irradiation.

4.2. PIE

The irradiated samples have been characterized using optical microscopy and scanning electron microscopy with EDX. Results concerning samples 1 and 2 have already been presented [18].

Optical microscopy

In the case of sample 1, an interaction layer surrounding each of the spherical UMo particles is visible (Figure 4.a). Its thickness appears to be homogeneous (5-10 µm).

In the case of samples 2, 3 and 4 (Figure 4.b to d), the optical technique appeared to be inefficient to distinguish topographic effects (induced by irradiation) from the presence of an interaction layer. Nevertheless, in both case, it would be thinner than in sample 1.

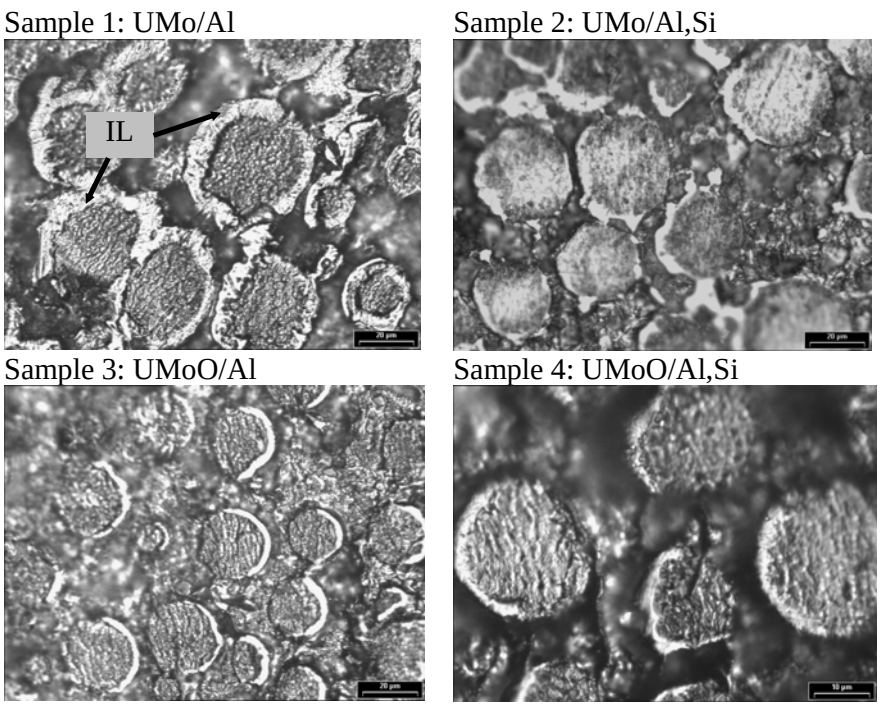


Figure 4: Optical micrographs of the irradiated zone of the four samples (IL=Interaction Layer).

SEM and EDX analyses

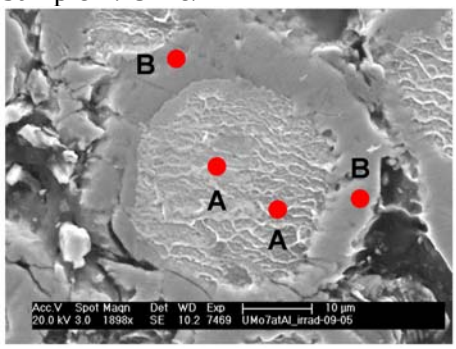
In order to specify the optical observations, SEM (scanning electron microscopy) pictures and EDX (energy dispersive X-ray spectroscopy) analyses have been carried out.

On different locations of each SEM picture, the chemical composition has been measured by EDX. Three kinds of local composition are revealed, spot A type with the presence of U and Mo (without

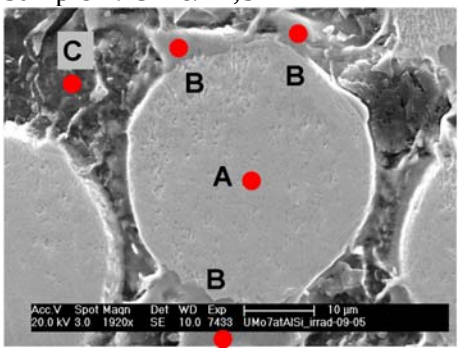
any Al i.e UMo alloy zone), spot B type with the three elements U, Mo and Al (interaction zone) and possibly spot C type in the Al matrix. Of course the projectile iodine was also detected but is not mentioned here any further.

These analyses show the presence of a quite homogeneous interaction layer in sample 1, composed of UAl_3. The average thickness of this layer is 7.6 µm (measured by image analysis on about 20 particles). These results prove the efficiency of ion irradiation to stimulate the interaction at the UMo/Al interface. However, in oxidized UMo based sample (sample 3) no evidence of any interaction layer presence around the UMo particles has been found (many particles have been analysed). In this condition, the addition of silicon in the Al matrix has no additional benefit effect (sample 4), while its presence decreases the interaction phenomenon without any interface barrier presence (sample 2). Indeed, only a local interaction is observed with an extremely inhomogeneous thickness. In order to determine the efficiency of modified matrix in combination with oxidation route, other ion irradiation condition would be essential.

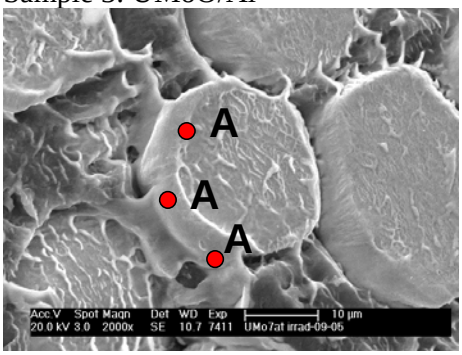
Sample 1: UMo/Al



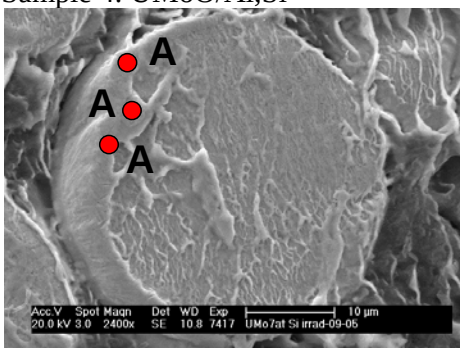
Sample 2: UMo/Al,Si



Sample 3: UMoO/Al



Sample 4: UMoO/Al,Si



Spot A	Spot B	Spot C
U, Mo	U, Mo, Al	Al

Figure 5: SEM micrographs of the four different irradiated samples (secondary electrons) and elementary composition derived from EDX analyses. Note: Probed volume for EDX is 1 μm^3.

6. Conclusion

In the framework of the high density UMo fuel development, several solutions are worldwide studied as an alternative fuel to UMo dispersed in pure Al. In this work a modification of the UMo/Al interface is considered. The paper presents the out-of-pile studies on this subject.

An oxide layer, mainly composed of UO_2, has been thermally obtained at the atomized UMo particle surface. During roll processing, the oxidation layer is maintained without any noticeable damage. The samples have been irradiated by ions. While the reaction is important in a standard UMo/Al fuel with a 7-8 µm thickness interaction, the presence of an oxide between UMo and Al totally prevents the

interaction formation (at least in the chosen irradiation conditions). The experiments should be continued to determine the actual benefit of silicon addition to Al matrix.
This latest results attest that this advanced fuel could be promising as regard to the in-pile behaviour. But, because the out-of-pile mechanisms are probably different from the in-pile ones, this benefit has to be verified for in-pile conditions.
Therefore, a future French irradiation, named IRIS4 experiment, is foreseen in OSIRIS reactor in 2007. The objective is to test and discriminate the influence of an oxide layer coating atomized UMo particles.
This irradiation would permit to have a better understanding of the oxidation role on the UMo/Al interface evolution, in comparison with the recent IRIS-TUM experiment. The latter was performed at high irradiation conditions and with ground UMo powders (and thus a high amount of surface oxide, but without any distribution control).

7. References

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