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RESULTS OF THE IRIS4 IRRADIATION IN OSIRIS REACTOR: OXIDIZED U-MO PARTICLES DISPERSED IN AL (WITH 0 OR 2.1%SI)

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ABSTRACT

The IRIS4 irradiation is the current step of the French U-Mo irradiation program to test an advanced oxidized U-Mo dispersion fuel for the Jules Horowitz research Reactor.

The irradiation of three of the four full-size plates has to be stopped because their thickness increase at MPF has reached the maximal acceptable value in the OSIRIS reactor without pillowing. The fourth plate is still in pile.

Based on the plate thickness increases measured at each reactor intercycle, the effect of the tailored oxide layer at the interface between fuel particles and aluminum matrix and the silicon added to the matrix is discussed:

- compared with the IRIS2 results, this effect is positive and seems to reduce or delay the interaction between U-Mo particles and aluminum matrix,
- however, considering the IRIS3 experiment, this effect seems not to be cumulative with the positive effect of the silicon added to the aluminum matrix.

1. Introduction

As described in our previous papers [1], [2], [3], [4], French CEA decided, within a specific collaboration with AREVA-CERCA for the plate manufacturing, to irradiate, since last October 2008, four full size plates made of oxidized atomized UMo particles dispersed in aluminium matrix with AlFeNi cladding.

The main aims of this IRIS 4 experiment performed in the French OSIRIS Material Testing Reactor can be summarized as follows:

- Test the efficiency of a thermochemically controlled oxide layer around UMo particles as a protective barrier to the inter-diffusion of UMo/Al to hinder the interaction between UMo particles and Al matrix.
- Confirm, at higher heat flux and temperature, the positive effect of silicon addition to the Al matrix as observed in a previously irradiated full size IRIS3 plate [5] and test its compatibility with the oxide layer.

In this paper, we present the parameters of the IRIS 4 experiment, such as the characteristics and irradiation conditions of the four plates. The non destructive plates thickness measurements collected at each inter-cycle are exposed. Their comparison to the ones registered in the course of the previous irradiations IRIS2 and IRIS3 makes the basis of a concluding discussion.

2. Plates characteristics

Considering manufacturing aspects and the difficulties to industrialize a grinding process, atomized particles have been selected. The thermochemically controlled oxidation of the atomized UMo powder has induced an oxide layer around UMo particles which mean thickness is 1.5±0.5 µm [6]. At this stage of fabrication, the oxide layer was quite homogeneous and well stuck to the UMo particles.

A set of four full size AlFeNi cladded fuel plates were then fabricated with this powder by AREVA-CERCA, using a classical hot rolling process. This hot rolling process has induced local deformations of the UMo particles, generating some unavoidable cracks in the oxide layer [6].

The plates have a high meat loading of about 7.8 gU.cm^{-3} and an enrichment of about 19.8% ^{235}U. The meat consists of oxidized atomized U7.3wt%Mo particles dispersed in either a pure (A5) aluminium matrix (plates 8053 and 8054) or an aluminium matrix containing 2.1 wt% Si (plates 8043 and 8044). As usual in the French IRIS program, two twin plates are made of the same fuel to increase statistics and to minimize the risk of technical problem during irradiation.

The main characteristics of the IRIS4 fuel plates are summarized in Table 1. The dimensions (length x width x thickness) of the fissile meat, respectively full size plate, are close to 596.5 x 55.0 x 0.51 mm^3, respectively 641.9 x 73.3 x 1.30 mm^3. They are identical to the ones of the previous IRIS experiments performed in the IRIS device designed for the OSIRIS MTR [7]. The as-fabricated porosity varies between 1.1 to 4.5% while the fuel volume fraction is close to 50%.

The fuel plate inspections, such as metallographic examinations, X-ray diffraction, blister tests, indicated that the plates met the usual OSIRIS specifications.

Plate number	U7MZ8043	U7MZ8044	U7MZ8053	U7MZ8054
Si in Al content (wt%)	0	0	2.1	2.1
Mo/UMo (wt%)	7.3	7.3	7.3	7.3
U density (gU.cm^{-3})	7.7	7.9	7.8	7.7
U total (g)	131.3	129.7	131.3	131.3
Plate thickness (mm)	1.31	1.31	1.30	1.30
Porosity (vol.%)	3.8	1.1	2.4	4.5

Table 1: Parameters of the IRIS4 oxidised UMo plates

3. Irradiation testing

These four plates were inserted in an IRIS device and loaded at the OSIRIS central core position 52, where a slight flux gradient exists between the two outer most irradiated objects (plates 8044 and 8053) and the two inner less irradiated ones (plates 8043 and 8054), as illustrated by Figure 1.

The IRIS4 irradiation was designed to test these plates in the OSIRIS MTR at its maximum irradiation conditions authorized by the French Nuclear Regulatory Commission. It corresponds to a maximum heat flux of about 290 W/cm² and an outer cladding temperature of about 100°C.

The irradiation started in October 2008 and ended after 6 cycles for plates 8053 and 8054, 7 cycles for plate 8044. The irradiation of the last plate 8043 will restart this autumn to reach, at least, a mean burn up of 50% ^{235}U. As it will be explained in section 4, the plate 8053 and 8054 have been removed from the irradiation device at the end of the 6th cycle because they reached the maximal swelling of 250 µm admitted in the OSIRIS reactor. The same reason conducted to remove the plate 8044 after the 7th irradiation cycle.

To illustrate this irradiation history, the following characteristics, Peak Heat Flux (PHF) (displayed in figure 2), plate average Fission Density (FD) and Burn Up (BU) of the four plates, are given in

Table 2 for each cycle. They are determined by specific preliminary neutronic calculations. A gamma-ray spectrometry of the plate 8053 is foreseen to validate them.

At the current stage of the irradiation, 8 cycles have been performed, for a total duration of 121.5 EFPD. The mean fission density, respectively BU, are 3.8 10^{21} f.cm^{-3}$_{UMo}$, respectively 55.1 ^{235}U at. % at the Maximal Flux Plane (MFP) of the plate 8044. For this plate, the Peak Heat Flux was of 269 W.cm^{-2}.

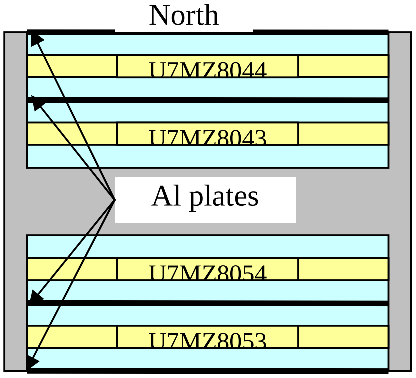

Figure 1: Loading of the IRIS device

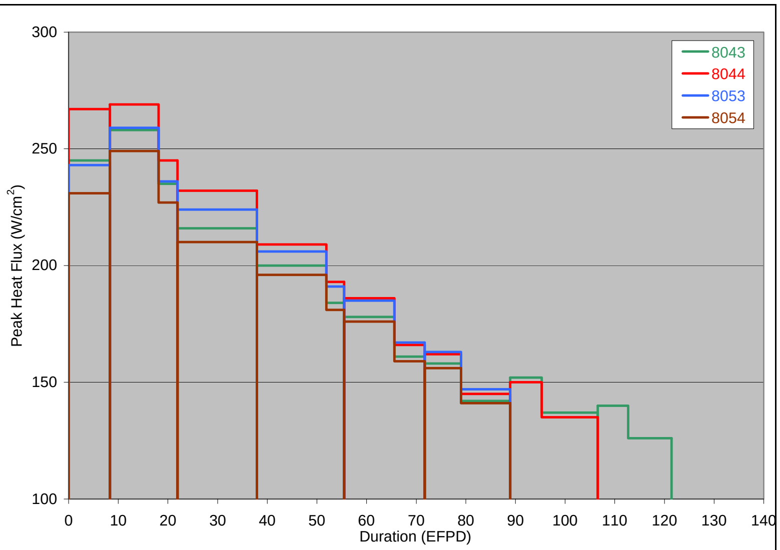

Figure 2: Evolution of the PHF during the IRIS4 testing

Plate		U7MZ8043			U7MZ8044			U7MZ8053			U7MZ8054		
Cycle	EFPD	FD	BU	PHF	FD	BU	PHF	FD	BU	PHF	FD	BU	PHF
F229	8.3	0.36	5.2	245	0.40	5.8	267	0.36	5.2	243	0.34	5.0	231
F230	21.9	0.92	13.3	258	1.01	14.6	269	0.93	13.5	259	0.89	12.9	249
F231	37.9	1.51	21.8	216	1.65	23.8	232	1.53	22.3	224	1.46	21.1	210
F232	55.5	2.06	29.9	200	2.24	32.4	209	2.11	30.6	206	2.01	29.1	196
F233	71.7	2.49	36.1	178	2.69	38.9	186	2.56	37.1	185	2.43	35.3	176
F234	88.9	2.86	41.4	158	3.07	44.4	162	2.94	42.6	163	2.80	40.6	156
F235	106.6	3.13	45.4	152	3.34	48.3	150	Swelling>250µm			Swelling>250µm		
F236	121.5	3.32	48.1	140	Swelling>250µm								
End	MFP	3.76	54.6	258	3.82	55.1	269	3.41	49.5	259	3.23	47.0	249

Table 2: IRIS 4 irradiation conditions

Fission density (FD) in 10^{21} f.cm^{-3}$_{UMo}$ and burn-up (BU) in ^{235}U at. % are plate average values. PHF stands for Peak Heat Flux (in W.cm^{-2}) and EFPD for Equivalent Full Power Day

4. Plates thickness measurements

At each intercycle, the plates were removed from the IRIS device and inserted in a specific measuring bench to collect for each of them their thickness profiles. The measurements were performed along five axial traces (at -26, -13, 0, +13 and +26 mm) and one transverse at 307 mm from the bottom of plates.

The thickness increase is obtained by subtracting an intercycle measurement to the one performed before irradiation. The measurements after the 2nd cycle F230 are not available due to technical problems. The precision of these measurements is about 15 µm.

The figures 3a to 6a display the thickness profiles of the four IRIS 4 plates along one vertical direction chosen to cross the maximal thickness value. The figures 3b to 6b display a pseudo-3D zoom of the thickness profiles of the four IRIS 4 plates collected between the horizontal levels 210 mm and 450 mm from the bottom of plates.

4.1. Plates 8053 and 8054: pure Al matrix

The plates 8053 and 8054 made of oxidized atomized UMo particles dispersed in pure aluminium matrix have been irradiated during six cycles up to respective mean fission densities of 3.41 and 3.23 10^{21} $f.cm^{-3}{}_{UMo}$ at MFP.
As can be seen in the figures 3 and 4, a smooth swelling has occurred during the first three cycles. Then an abnormal thickness increase has begun during the fourth irradiation cycle. The testing of these two plates has to be stopped at the end of the sixth cycle, because they reached the maximal swelling value of 250 µm admitted in the OSIRIS reactor.
The maximal local thickness increase of the plate 8053, respectively 8054, measured by local scanning is 251 µm, respectively 272 µm, at the end of the sixth cycle F234.
No failure of the cladding has occurred as far as no fission products have been detected.

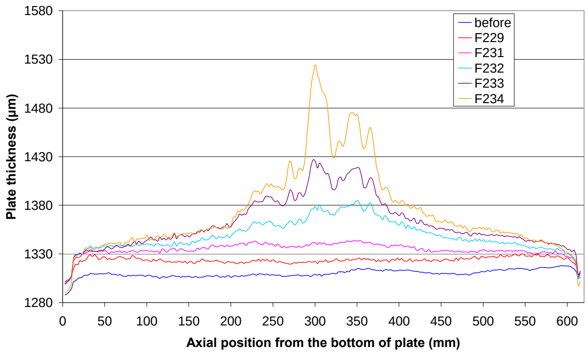

Figure 3a: In-pile evolution of the longitudinal thickness profiles of the plate 8053 measured along the axial direction 0 mm

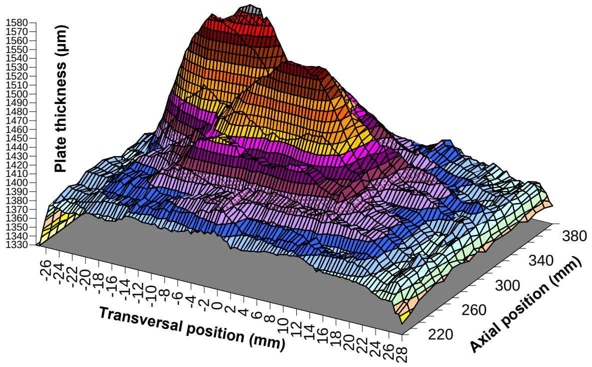

Figure 3b: Pseudo-3D zoom of the thickness profiles of the plate 8053 around MFP at the end of irradiation

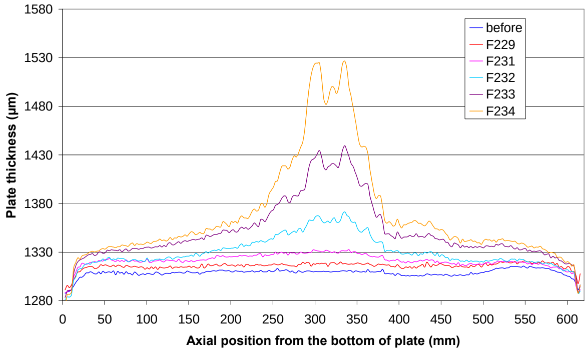

Figure 4a: In-pile evolution of the longitudinal thickness profiles of the plate 8054 measured along the axial direction +13 mm

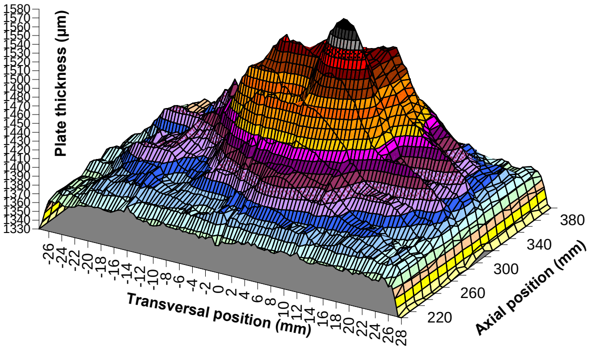

Figure 4b: Pseudo-3D zoom of the thickness profiles of the plate 8054 around MFP at the end of irradiation

4.2. Plates 8043 and 8044: Si addition to the Al matrix

The plate 8044 made of oxidized atomized UMo particles dispersed in an aluminum matrix containing 2.1 wt% Si has been irradiated during seven cycles up to a mean fission density of 3.82 10^{21} fissions.cm$^{-3}{}_{\mathrm{UMo}}$ at MFP.

As shown by the figure 5, the same abnormal thickness increase, observed on the plates without silicon addition to the aluminum matrix, as described in the previous section 4.1, has begun at the end the fifth irradiation cycle. The testing of this plate 8044, the hottest, has to be stopped at the end of the seventh cycle. The maximal thickness increase of this plate was then 269 µm.

The addition of Si to the aluminium matrix has somehow delayed the plate thickness increase, improving the behaviour of the plate under irradiation (see following discussion in section 5).

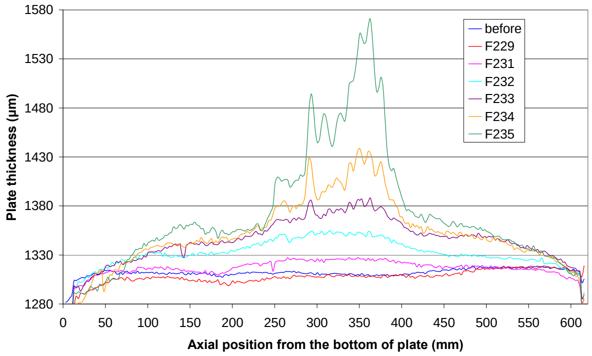

Figure 5a: In-pile evolution of the longitudinal thickness profiles of the plate 8044 measured along the axial direction 0 mm

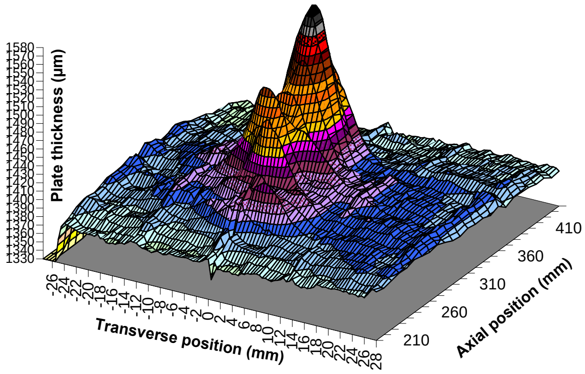

Figure 5b: Pseudo-3D zoom of the thickness profiles of the plate 8044 around MFP at the end of irradiation

The plate 8043 also made of oxidized atomized UMo particles dispersed in an aluminum matrix containing 2.1 wt% Si has been irradiated during 8 cycles up to a mean fission density of 3.76 10^{21} fissions.cm$^{-3}{}_{\mathrm{UMo}}$ at MFP.

On the opposite of the previous statements, almost no abnormal thickness increase has been observed for this plate, as can be seen in the figure 6. The maximal thickness increase is currently of 132 µm. Just a beginning of swelling seems to occur after the eighth irradiation cycle. One or two other cycles are foreseen for this plate before the end of 2009 to reach the objective of a mean plate burn up of at least 50% ^{235}U.

In this case, the addition of Si to the aluminium matrix has clearly delayed the plate thickness increase, improving the behaviour of the plate under irradiation (see following discussion in section 5).

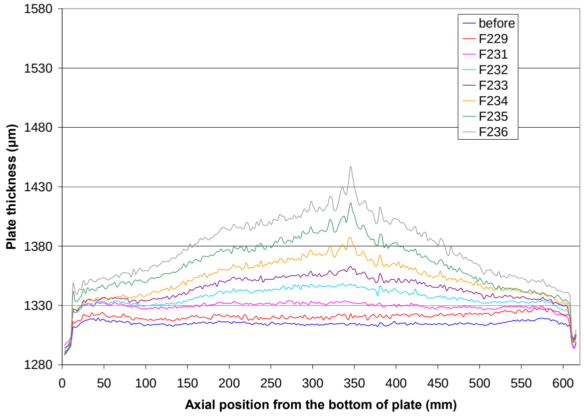

Figure 6a: In-pile evolution of the longitudinal thickness profiles of the plate 8043 measured along the axial direction -13 mm

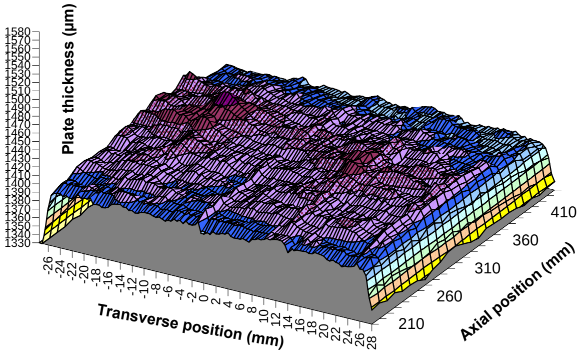

Figure 6b: Pseudo-3D zoom of the thickness profiles of the plate 8043 around MFP at the end of irradiation

5. Discussion

5.1. IRIS4 without Si versus IRIS2: Effect of the oxide layer

The IRIS2 fuel plates were made of atomized UMo particles dispersed in pure aluminium matrix. The irradiation has been performed in the OSIRIS core position 52 (the same as IRIS4) and lasted three or four cycles before the plates pillowed [8].

The comparison of the results of this IRIS2 test plates with the IRIS4 8053 and 8054 plates, whose matrix is pure aluminium, should bring direct information on the effect of the oxide layer around atomized UMo particles.

The main manufacturing data of the IRIS2 plates and their irradiation characteristics are compared with those of the two IRIS4 8053 and 8054 plates in the table 3. The evolution of the PHF during these two experiments is illustrated by the figure 7(left). It appears that the IRIS2 experiment has been a little “colder” than the IRIS4 one. The figure 7 (right) represents the plate thickness increase as a function of the mean fission density at the MFP of these five plates.

	Experiment	IRIS2			IRIS4	
Manufacturing data	Plate	U7MT2002	U7MT2003	U7MT2007	U7MZ8053	U7MZ8054
	Mo in UMo (wt%)	7.6	7.6	7.6	7.3	7.3
	Enrichment (^{5}U wt%)	19.8	19.8	19.8	19.8	19.8
	Si in Al matrix (wt%)	0	0	0	0	0
	Matrix type	A5	A5	A5	A5	A5
	Fuel loading (gU/cc)	8.3	8.3	8.3	7.8	7.7
	As fab meat porosity (%)	1.5	1.6	1.4	2.4	4.5
	Cladding material	AG3NE	AG3NE	AG3NE	AlFeNi	AlFeNi
Irradiation data	Max heat flux at BOL (W/cm^2)	238	217	231	259	249
	Max clad surface temp. (°C)	93	90	92	~ 100	~ 100
	Number of cycles	4	4	3	6	6
	Duration (EFPD)	58	58	40	87	87
	Plate mean BU (^{235}U %)	32.5	30.5	23.4	42.6	40.6
	Mean BU at MFP (^{235}U %)	39.1	36.6	28.4	49.5	47.0
	Plate mean FD (f/cm$^3_{UMo}$)	2.2 10^{21}	2.1 10^{21}	1.6 10^{21}	2.2 10^{21}	2.2 10^{21}
	Mean FD at MFP (f/cm$^3_{UMo}$)	2.7 10^{21}	2.5 10^{21}	2.0 10^{21}	3.4 10^{21}	3.2 10^{21}
	Max thickness increase (µm)	1237	257	371	251	272

Table 3: Main features of the testing of the IRIS2 and IRIS4 plates without Si

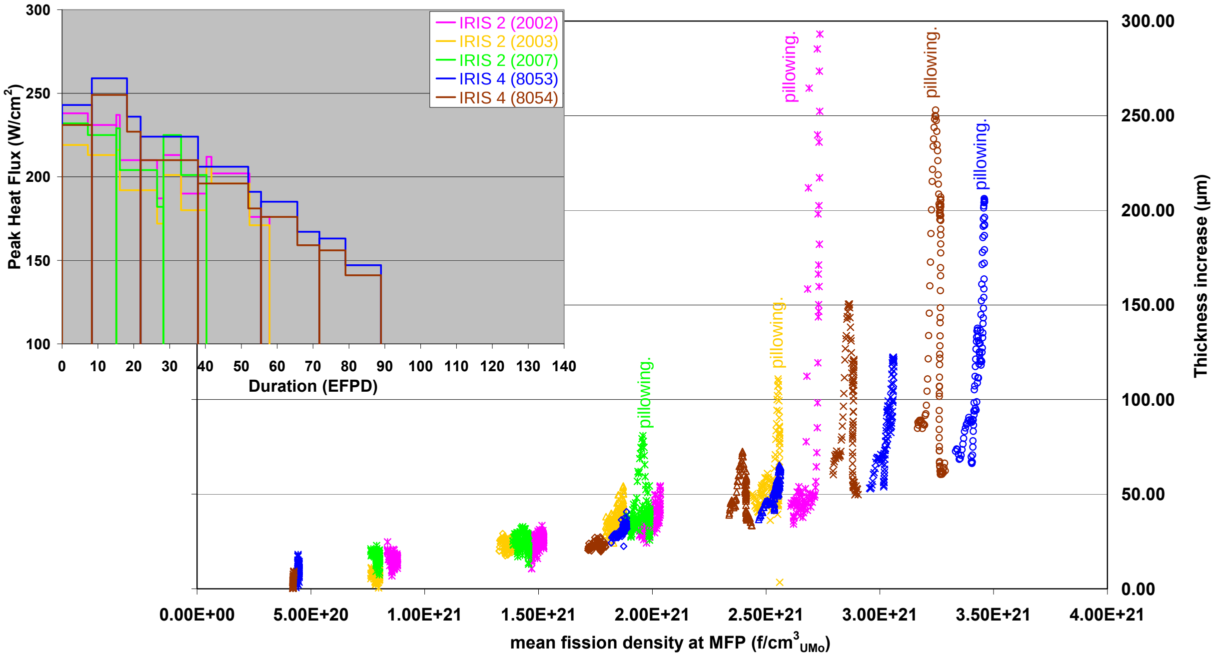

Figure 7: Left: History of the PHF of the IRIS2 and IRIS4 plates. Right: Plate thickness increase as a function of the mean fission density at the MFP

It appears from the figure 7 (right), that the initial evolution of the plate thicknesses is quite similar for the five different plates. Indeed, during the first two cycles, the swelling of the UMo particles consumes the as-fabricated initial porosity. Then, an abnormal swelling, the so-called pillowing, appeared around a plate mean fission density of 2.0 10^{21} f.cm$^{-3}_{\mathrm{UMo}}$ for the IRIS2 2007 plate, 2.5 10^{21} f.cm$^{-3}_{\mathrm{UMo}}$ for the IRIS2 2003 plate and 2.7 10^{21} f.cm$^{-3}_{\mathrm{UMo}}$ for the IRIS2 2002 plate. This swelling is weaker and delayed of one or two cycles for the IRIS4 8053 and 8054 plates. Their plate mean fission densities are then 3.4 and 3.2 10^{21} f.cm$^{-3}_{\mathrm{UMo}}$, respectively.

The oxide layer around UMo particles of the IRIS4 plates has played a clear role in reducing and/or delaying the interaction between UMo particles and aluminium matrix. This interaction is responsible for the fission gas release associated to the large pores formation described after the post-irradiation examinations performed on some samples of the IRIS2 [8] and FUTURE [9] plates, among others.

The PIEs foreseen on IRIS4 samples in 2010 should give more information whether this oxide layer act as a physical barrier to prevent this interaction and/or modify the properties of the UMoAl interaction layer formed during irradiation. If any, the influence of the cladding materials will be also checked.

5.2. IRIS4 with Si versus IRIS4 without Si: Effect of the silicon added to the Al matrix

The table 4, as well as the figure 8, recapitulate the main features and measurements performed on the four IRIS4 plates.

	Experiment	IRIS4			
Manufacturing data	Plate	U7MZ8053	U7MZ8054	U7MZ8043	U7MZ8044
	Mo in UMo (wt%)	7.3	7.3	7.3	7.3
	Enrichment (^{5}U wt%)	19.8	19.8	19.8	19.8
	Si in Al matrix (wt%)	0	0	2.1	2.1
	Matrix type	A5	A5	A5	A5
	Fuel loading (gU/cc)	7.8	7.7	7.7	7.9
	As fab meat porosity (%)	2.4	4.5	3.8	1.1
	Cladding material	AlFeNi	AlFeNi	AlFeNi	AlFeNi
Irradiation data	Max heat flux at BOL (W/cm^{2})	259	249	258	269
	Max clad surface temp. (°C)	~ 100	~ 100	~ 100	~ 100
	Number of cycles	6	6	7	8
	Duration (EFPD)	87	87	121	107
	Plate mean BU (^{235}U %)	42.6	40.6	48.1	48.3
	Mean BU at MFP (^{235}U %)	49.5	47.0	54.6	55.1
	Plate mean FD (f/cm$^{3}_{UMo}$)	2.2 10^{21}	2.2 10^{21}	3.3 10^{21}	3.3 10^{21}
	Mean FD at MFP (f/cm$^{3}_{UMo}$)	3.4 10^{21}	3.2 10^{21}	3.7 10^{21}	3.8 10^{21}
	Max thickness increase (µm)	251	272	132	269

Table 4: Main features of the testing of the IRIS4 plates

The figure 8 (right) clearly shows that the addition of 2.1 wt% silicon to the aluminium matrix has reduced the plate thickness increase and delayed of at least one cycle the appearance of unacceptable swelling. Indeed, while this swelling has occurred at 3.4 and 3.2 10^{21} f.cm$^{-3}_{UMo}$ respectively for the plates 8053 and 8054, it has appeared at 3.8 10^{21} f.cm$^{-3}_{UMo}$ for the plate 8044 and it is still not evidenced at 3.7 10^{21} f.cm$^{-3}_{UMo}$ for the plate 8043.

The positive effect of silicon addition is confirmed by this IRIS4 full size plate experiment at higher irradiation condition than the previous IRIS3 test [5], but this improvement is not sufficient to reach the Jules Horowitz Reactor (JHR) objectives.

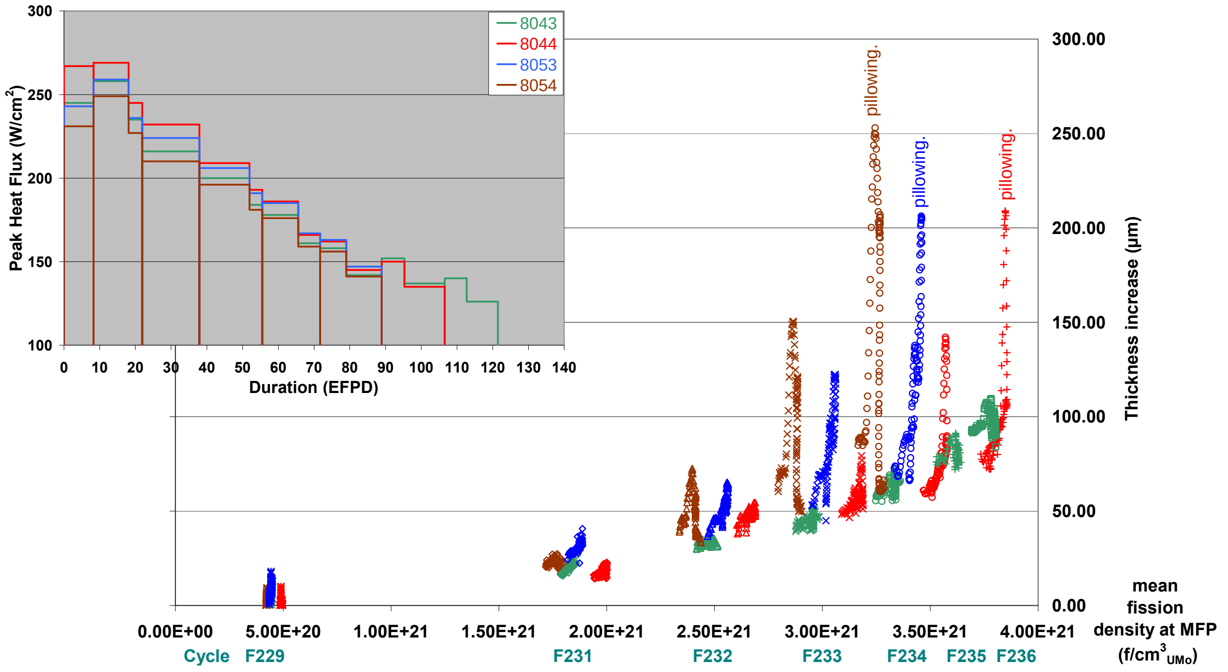

Figure 8: Left: History of the PHF of the IRIS4 plates. Right: IRIS4 plate thickness increase as a function of the mean fission density at the MFP

5.3. IRIS4 with Si versus IRIS3: Effect of the silicon and compatibility with the oxide layer

In this part, the comparison will be made between the IRIS3 and IRIS4 fuel plates whose aluminium matrix contained 2.1% Si, to underline the impact of this peculiar element on the behaviour of the fuel plates under irradiation. It concerns the IRIS3 8021 plate and the IRIS4 8043 and 8044 ones. In other words, the comparison of the thickness increase of these three plates should confirm, at higher heat flux and temperature, the positive effect of silicon addition to the Al matrix as observed in the IRIS3 8021 plate and its compatibility with the oxide layer.
The main manufacturing data of the IRIS3 8021 plate and its irradiation characteristics are compared with those of the two IRIS4 8043 and 8044 plates in the table 4.

	Experiment	IRIS3	IRIS4	
Manufacturing data	Plate	U7M8021	U7MZ8043	U7MZ8044
	Mo in UMo (wt%)	7.3	7.3	7.3
	Enrichment (^{5}U wt%)	19.8	19.8	19.8
	Si in Al matrix (wt%)	2.1	2.1	2.1
	Matrix type	A5	A5	A5
	Fuel loading (gU/cc)	8.0	7.7	7.9
	As fab meat porosity (%)	2.2	3.8	1.1
	Cladding material	AG3NE	AlFeNi	AlFeNi
Irradiation data	Max heat flux at BOL (W/cm^2)	201	258	269
	Max clad surface temp. (°C)	93	~ 100	~ 100
	Number of cycles	7	7	8
	Duration (EFPD)	130	121	107
	Plate mean BU (^{235}U %)	48.8	48.1	48.3
	Mean BU at MFP (^{235}U %)	56.5	54.6	55.1
	Plate mean FD (f/cm$^3_{UMo}$)	3.4 10^{21}	3.3 10^{21}	3.3 10^{21}
	Mean FD at MFP (f/cm$^3_{UMo}$)	3.9 10^{21}	3.7 10^{21}	3.8 10^{21}
	Max thickness increase (µm)	91	132	269

Table 4: Main features of the testing of the IRIS3 and IRIS4 plates with Si

The evolution of the PHF of these three plates is illustrated by the figure 9 (left). It appears that the IRIS3 8021 plate has been much colder than the IRIS4 8043 and 8044 ones. The figure 9 (right) represents the plate thickness increase as a function of the mean fission density at the MFP of these three plates.

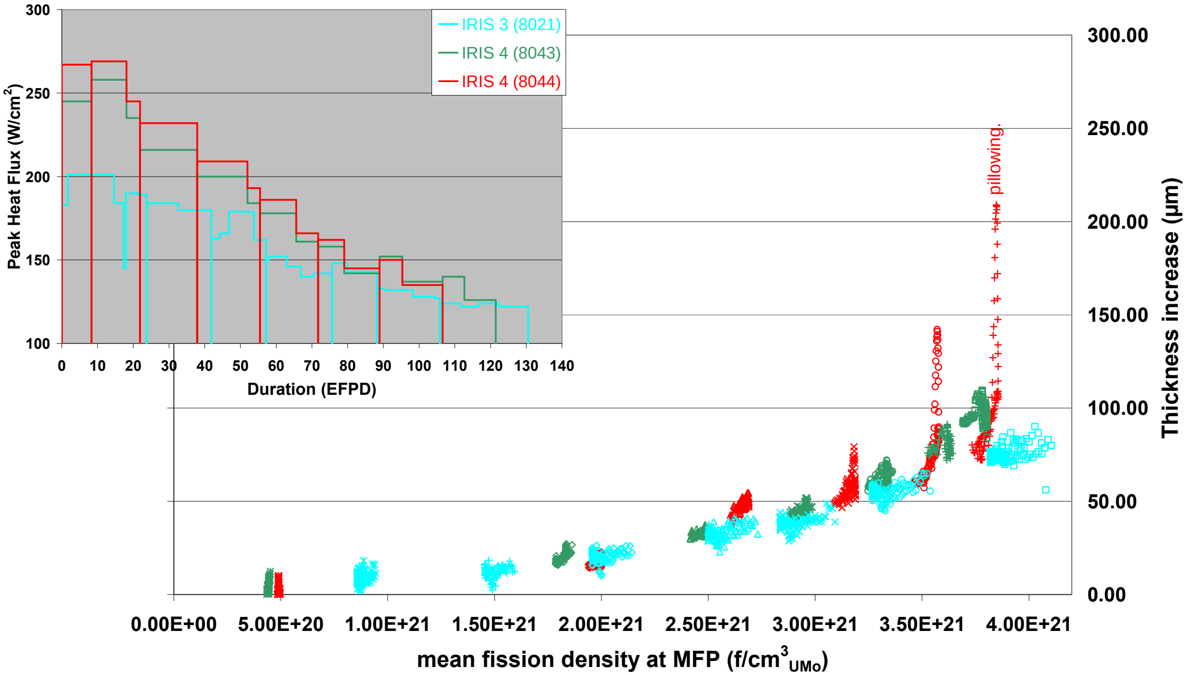

Figure 9: Left: History of the PHF during IRIS4 tests. Right: Plate thickness increase as a function of the mean fission density at the MFP

As described in the section 5.2, the addition of silicon in the Al matrix improves the behaviour of oxidized atomized UMo fuel plates. But this effect seems to be weaker than what was observed in the case of the IRIS3 8021 plate (cf. figure 9 right).

The PIEs performed on this plate have shown that the Si effect is particularly pronounced if a rich Si layer is close to the UMo particles before the beginning of irradiation [10]. It thus acts as a physical barrier to avoid or minimize the interaction between UMo and Al.
In the IRIS4 case, we can imagine that the oxide layer might have obstruct or impede the close contact between UMo particles and Al matrix. The efficiency of silicon is then certainly reduced but not completely annihilated, because, as described in section 2, some cracks in the oxide layer have been caused by the hot rolling process. Some pathways still exist for the silicon to be efficient.
Another point to bear in mind lies in the temperatures of the plates during irradiation. Indeed, the PHF of the IRIS3 8021 plate is 201 W/cm^2 instead of 258 and 269 W/cm^2 for the IRIS 4 8043 and 8044 plates (cf. figure 10 left). The amount of interaction layer, driven by temperature as explained in [10], should be higher in the IRIS4 plates than in the IRIS3 one. The behaviour of the latter is expected to be better because of the lower temperatures reached during irradiation.
Those two explanations have to be completely checked in the PIEs foreseen on some IRIS4 samples in 2010.

6. Conclusion

The IRIS4 experiment has been designed to test four full size plates made of oxidized atomized UMo particles dispersed in pure or added with silicon aluminium matrix. The IRIS4 plates were irradiated at the maximal heat conditions authorized in the OSIRIS reactor (about 290 W.cm^{-2}.and 100°C). The irradiation of three of the four full-size plates has to be stopped because their thickness increase at MPF has reached the maximal acceptable value in the OSIRIS reactor. The fourth plate is still in pile.

Based on the plate thickness increases measured at each reactor intercycle, the effect of the tailored oxide layer at the interface between fuel particles and aluminum matrix and the silicon added to the matrix can be summarize as follows.:

- In comparison with the IRIS2 plates, the oxide layer has delayed the pillowing of the IRIS4 plates, but has not been able to prevent it. The oxide layer has a positive effect on the fuel behavior, but is not, alone, a solution to reduce sufficiently the UMo/Al interaction responsible for this abnormal swelling.
- The positive effect of silicon addition to the Al matrix, as observed in the IRIS3 test, has been confirmed at higher heat flux and temperature. But considering this IRIS3 experiment, this effect seems not to be cumulative with the benefit of oxide layer around UMo particles.

The difference in the shape of the swellings, observed in IRIS4 compared to IRIS2 and 3, might be due to a modification of the fission gas release. The oxide layer and its association with silicon could have modified the characteristics of the interaction layer and the fission gas release.

The real influence of this oxide layer and its association with silicon has to be specified through the future PIEs foreseen in 2010.

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