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Letter to the Editor

Test irradiations of full-sized U_3Si_2–Al fuel plates up to very high fission densities

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ABSTRACT

In the course of the licensing procedure of the ‘Forschungsneutronenquelle Heinz Maier-Leibnitz’, i.e. the new 20 MW high-flux research reactor FRM II in Garching near Munich, extensive test irradiations have been performed to qualify the U_3Si_2–Al dispersion fuel with a relatively high density of highly enriched uranium (93 wt% of ^{235}U) up to very high fission densities. Two of the three FRM II type fuel plates used in the irradiation tests contained U_3Si_2–Al dispersion fuel with HEU densities of 3.0 gU/cm^3 or 1.5 gU/cm^3 (‘homogeneous plates’) and one plate had two adjacent zones of either density (‘mixed plate’). They were irradiated in the French MTR reactors SILOE and OSIRIS in the years before 2002. The local plate thickness was measured on certain tracks along the plates during interruptions of the irradiation. The maximum fission density obtained in the U_3Si_2 fuel particles was 1.4×10^{22} f/cm^3 and 1.1×10^{22} f/cm^3 in the 1.5 gU/cm^3 and 3.0 gU/cm^3 fuel zones, respectively. In the course of the irradiations, the plate thickness increased monotonically and approximately linearly, leading to a maximum plate thickness swelling of 14% and 21% and a corresponding volume increase of the fuel particles of 106% and 81%, respectively. Our results are discussed and compared with the data from the literature.

1. Introduction

The new 20 MW high flux research reactor FRM II, now called ‘Forschungsneutronenquelle Heinz Maier-Leibnitz (FRM II)’, at Garching near Munich became first critical in the year of 2004. In the course of its nuclear licensing procedure, an extensive test program has been performed to qualify its U_3Si_2–Al dispersion fuel with a high density of highly enriched uranium (HEU, with 93 wt% ^{235}U) up to very high fission densities. For that end, a total of 3 full-sized plates have been fabricated by AREVA-CERCA and irradiated in the French MTR reactors SILOE (1 plate) and OSIRIS (2 plates) [1].

The silicide dispersion fuels have been widely tested and their safe operation has been confirmed since then by many applications worldwide, mostly with low enriched uranium (LEU, below 20 wt% ^{235}U) and for uranium densities up to about 4.8 gU/cm^3 [2,3]. Later on, worldwide efforts have been undertaken to develop fuels for research reactors with even higher densities of uranium on the basis of UMo alloys. However, test irradiations of this new fuel type under realistic conditions have shown excessive swelling, and it became clear by the end of 2003 that the qualification of this fuel will be seriously delayed [4]. For the time being U_3Si_2–Al dispersion is the densest fuel available and remains the only alternative to reduce enrichment in the existing and the forthcoming research reactors. This in mind, we publish here in a comprehensive way our results of the swelling of U_3Si_2–Al dispersion fuel under the condition of very high fission density or burn-up in the U_3Si_2 particles.

2. FRM II fuel element design data

The FRM II uses a single fuel element which is cooled by light water (H_2O) and placed in the centre of a large moderator tank of heavy water (D_2O), where an unperturbed maximum of the thermal neutron flux of 8×10^{14} cm^{-2} s^{-1} is obtained at only 20 MW power [5]. The cylindrical fuel element consists of 2 tubes with 24.3 cm outer and 11.8 cm inner diameters with a total of 113 identical fuel plates of involute shape in between, so that the cooling channels between the plates have a constant width of 2.2 mm. The active zone of the plates is 700 mm long and 62.4 mm wide, and the plates are 1.36 mm thick with a fuel meat zone of 0.60 mm and two AlFeNi claddings of 0.38 mm each. The meat consists of U_3Si_2–Al dispersion fuel using HEU (93 wt% of ^{235}U) with its uranium density being graded in each plate to 3.0 gU/cm^3 in the larger inner part and 1.5 gU/cm^3 in the smaller outer part to reduce power peaking effects due to the surrounding D_2O moderator tank [5], see also Table 2.

The local maximum and plate averaged values of the fission densities and fuel meat swelling have been calculated by SIEMENS (KWU). The data refer to the unperturbed reactor, i.e. the reactor

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[1] Retired.

[2] The authors are indebted to Dr. M. Nuding, who contributed essentially to the evaluation of the test irradiations in the years before 2002. He now works at TÜV SÜD Industrie Service GmbH, an independent company for the assessment of the safety of technical facilities including nuclear reactors.

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without any installations in the D_2O moderator tank, and can be understood using the equation

$$MS[\%] = PS[\%] \times VR - V_P[\%], \quad (1)$$

where MS[%] is the meat swelling, which in a thin fuel plate with cladding (which does not swell) on both sides is identical with the increase of the fuel plate thickness t_F

$$MS[\%] = 100 \times (t_F - t_{F,0})/t_{F,0} \quad (2)$$

(with $t_{F,0}$ being the fuel plate thickness before irradiation). PS[%] is the fuel particles swelling and VR is the volume ratio of the U_3Si_2 particles in the U_3Si_2–Al meat before irradiation. The as-fabricated porosity V_P[%] of the fuel meat may vary from plate to plate and even locally within a plate; it is established during the fabrication process and it is difficult and only integrally possible to be identified (in a buoyancy flotation measurement of the whole plate) whence it is often estimated using an empirical relation as e.g. given in [2]. This porosity, expressed in vol.%, is typically higher for high uranium densities and disappears during the initial phase of the irradiation due to the irradiation-induced creep of the aluminium matrix atoms.

With FDM and FDP being the fission densities in the meat and in the particles, respectively (FDM = FDP × VR), at the end of a 52 full power days reactor cycle,[3] the calculated data concerning the fuel swelling are given in Table 1. The regions with the local maxima of the fission density or swelling of the plates comprise only a few square millimetres, which is about 0.1% of the plate area. Also given are, for these positions, the fission rates in the particles FRP at the beginning of the cycle and time-averaged over the cycle. The highest local fuel temperatures occur during the beginning of the cycle with their local maxima at the cladding surface and at the fuel centreline being about 95 °C and 110 °C, respectively, under normal operation conditions; the corresponding maximum of the heat flux density is 417 W/cm^2. Until the end of the year 2007, more than 12 fuel elements have been used for 52 full power days each without any problems.

Table 1
Typical calculated data of the FRM II fuel plates at the end of the cycle (52 full power days)

Zone	1.5 gU/cm^3	3.0 gU/cm^3
VR (volume ratio) [%]	13.3	26.5
V_P (as-fabricated porosity) [%]	0.95	2.4
Burn-up local maximum [%]	51.2	29.3
FDM plate average [f/cm^3]	1.1×10^{21}	0.9×10^{21}
FDM local maximum [f/cm^3]	1.6×10^{21}	2.1×10^{21}
FDP plate average [f/cm^3]	8.4×10^{21}	3.4×10^{21}
FDP local maximum [f/cm^3]	12.2×10^{21}	7.8×10^{21}
FRP at the beginning of the cycle [f/(cm^{3}s)]	3.5×10^{15}	2.2×10^{15}
FRP time-averaged [f/(cm^{3}s)]	2.7×10^{15}	1.7×10^{15}
MS plate average [%]	5.9	3.0
MS local maximum [%]	9.0	10.1
PS plate average [%]	50	20
PS local maximum [%]	74	47

The symbols are explained in the text. Most data are from Siemens but the porosity, V_P, of the 1.5 gU/cm^3 zone has been obtained from the plate manufacturer AREVA-CERCA [1] and that of the 3.0 gU/cm^3 zone is a typical value from [2].

3. Documented silicide fuel performance and FRM II test program

The U_3Si_2–Al dispersion fuel to be used in the FRM II has been extensively tested in the 1980s by many groups of the world, mostly within the RERTR (Reduced Enrichment for Research and Test Reactors) program [2,3]. There it has been concluded that this fuel exhibits excellent properties under irradiation up to uranium densities of about 4.8 gU/cm^3 and at least for all fission densities accessible with LEU, i.e. up to about 2.3×10^{21} f/cm^3 meat (FDM). However, being not in the focus of this program only a few mini-plate samples with medium enrichment (MEU) and only two containing HEU have been irradiated at that time, but these tests also gave evidence of an excellent irradiation behaviour up to very high fission densities [2,3]. In conclusion it has been decided during the design phase of the FRM II to perform irradiation experiments on three full-sized test plates with HEU to confirm the existing data and extend the regime of a well documented irradiation behaviour of U_3Si_2–Al dispersion fuel up to very high fission densities particularly in the fuel particles (FDP).

For this end, three test plates – two 'homogeneous' and one 'mixed' – have been fabricated by AREVA-CERCA (France), i.e. by the same company which also was to fabricate the complete fuel elements of the FRM II. These plates had about the specifications of the realistic FRM II fuel element plates, with only small differences as required from the materials test reactors SILOE and OSIRIS of the Commissariat a l'Energie Atomique CEA, France. The characteristic data of the plates are presented in Table 2.

4. The SILOE test irradiation (homogeneous plate, 1.5 gU/cm^3)

This irradiation test has been performed in the materials test reactor SILOE (CEA Grenoble, France) during 10 reactor cycles over a total of 197 full power days in the year of 1997 [1,6,7]. Since this was immediately before the final shut down of this reactor, it was only possible to irradiate one test plate. The decision was made to choose a plate with 1.5 gU/cm^3, see Table 2, since for the FRM II fuel element the burn-up has been calculated to be highest in the 1.5 gU/cm^3 zone (Table 1).

The calculations of the local neutron flux in SILOE have been performed by the CEA and recalibrated after the second, fifth and 10th reactor cycles by gamma-scanning measurements of the radioactive fission products in the test plate to directly yield the local fission densities (3σ error = ±14%). Before irradiation and after each of the 10 reactor cycles, a plate thickness measurement using the IRIS device was performed under water in the SILOE pool. The IRIS device allows a continuous measurement of the plate thickness along certain tracks (plotting lines) with a differential method realized by two opposed linear variable differential transducer sensors, and the experimental error being ±6 μm [7]. For this experiment, 5 longitudinal (i.e. vertical) tracks have been defined that were spaced 14 mm apart, with measurements taken every 2.5 mm. Three-dimensional plots of the plate thickness data are given in [1,7] with the direct experimental curves looking very similar to the ones shown in Fig. 2.

Because of the cosine-like profile of the thermal neutron flux along the longitudinal direction of the plate – see Fig. 2 – the maximum fission density has been obtained in the plate centre. It is for this position that the results are presented in Table 3. Because of the quasi-statistical scatter of the single thickness measurements the data have been averaged over 30 measurements each, i.e. over 22.5 mm longitudinal (i.e. 10 data points on each track) and 28 mm transverse (i.e. tracks 2, 3 and 4). The cladding surface temperature and the mean heat flux density have been estimated by the CEA for this area to be about 100 °C and 87.4 W/cm^2, respectively. Using these values the thickness of the oxide layer which steadily builds up during irradiation on both cladding surfaces has been calculated with a formula given in [8], yielding about 2 × 5.3 μm at the end of the irradiation [1]. The values of the oxide layer thickness have been subtracted from the measured plate thickness values to

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[3] Only most recently, in December 2007, the Bavarian Nuclear Licensing Authority approved an extension of the FRM II cycle length to 60 full power days, which is about the maximum possible from the reactivity balance of the reactor.

Table 2
Design data of the FRM II fuel plates and of the 3 plates as used in the irradiation tests

	FRM II plate	SILOE homogeneous	OSIRIS homogeneous	OSIRIS mixed
Uranium density [gU/cm^3]	3.0/1.5	1.5	3.0	3.0/1.5
Enrichment [wt% ^{235}U]	93 nominal	≈90	90.2	92.8
Integral porosity V_P [vol.%]	2.4/0.95	0.95	1.0	1.0/1.0
Total mass of ^{235}U [g]	66.7	≈33.5	60.1	38.4
Length of plate [mm]	720	642	642	642
Length of fuel zone [mm]	700	609.5	609.5	609.5
Width of plate [mm]	76	73	73	73
Width of fuel zone [mm]	62.4 total	65.4	65.4	2 × 25.75
Plate thickness [mm]	1.36	1.27	1.27	1.27
Fuel meat thickness [mm]	0.60	0.60	0.60	0.60
Cladding thickness [mm]	0.38	0.335	0.335	0.335

The fuel is always U_3Si_2–Al and the cladding is AlFeNi, an Al alloy with 1% Fe, 1% Ni and 1% Mg. The values of the as-fabricated porosity of the test plates have been given by the plate manufacturer AREVA-CERCA (SILOE plate) or have been estimated by us based on more recent information from AREVA-CERCA (OSIRIS plates) [1] – see also the discussion in Section 7.

obtain the true meat swelling data given in Table 3, the errors being typically around ±6%. Plots of these swelling data [1,6] will be found in Sections 6.3 and 7 together with the results of the OSIRIS experiment. The time-averaged fission rate in the particles was 7.1×10^{14} f/(cm^3 s).

After the end of the irradiation and nearly one year of decay time, the plate has been transported to the hot cell laboratory LAMA of the CEA in Grenoble for post-irradiation examinations PIE's [7]. There a more precise gamma scan determination of the final fission density FD was possible due to the decay of all short-lived fission products and the reduced background in the hot cell. The result was a final burn-up of 54.6% and a final value of FDP = 12.1×10^{21} f/cm^3 with a reduced 3σ error of ±9% [1,7], this is in good agreement with the previous results given in Table 3.

The destructive PIE's started with a bending test of the irradiated plate. A device was built to fix the plate firmly in its upper and its lower part leaving a free space of 4 mm in between; in the bending test this gap was positioned 161 mm below the upper end of the fuel zone (Table 2). As a result, three bendings of 0°/+23°/0°/−23°/0° each were possible until the fracture of the plate, demonstrating a very good cohesion between fuel meat and cladding, and this good cohesion was observed even after this failure of the plate [7].

After that, two specimens of 16 × 16 mm^2 each were punched out of the plate, one of them in the region of maximum FD. They were glued into a holder, sawed, sanded, polished and inspected by optical microscopy [7]. As an example, Fig. 1 shows a micrograph of the specimen with FDP = 12×10^{21} f/cm^3. Since in every fission an uranium atom is converted into two fission fragments the volume of the fuel particles increases, in this case by about 70% (Table 3). The as-fabricated porosity (V_P = 0.95%, Table 2) has disappeared, but many small pores (bubbles) have been produced during irradiation to accommodate the gaseous fission fragments. In Fig. 1 most of these 'fission gas bubbles' have a diameter of below 2 μm with only a few larger ones. The very uniform distribution of small gas bubbles that show no tendency to interlink is the reason for the stable swelling behaviour of U_3Si_2 [2,3,9]. The interdiffusion layer which builds up at the surface of the particles is about 6–10 μm wide, which is consistent with correlations published in the literature [1]. The subject is further discussed in Section 7.

5. The OSIRIS test irradiation of the homogeneous plate (3.0 gU/cm^3)

This irradiation test has been performed at the materials test reactor OSIRIS (CEA Saclay, France) during 9 reactor cycles (F158–F168) over a total of 185 full power days in the years of 1999 and 2000 [7]. For details of the 'OSIRIS homogeneous test plate' see Table 2.

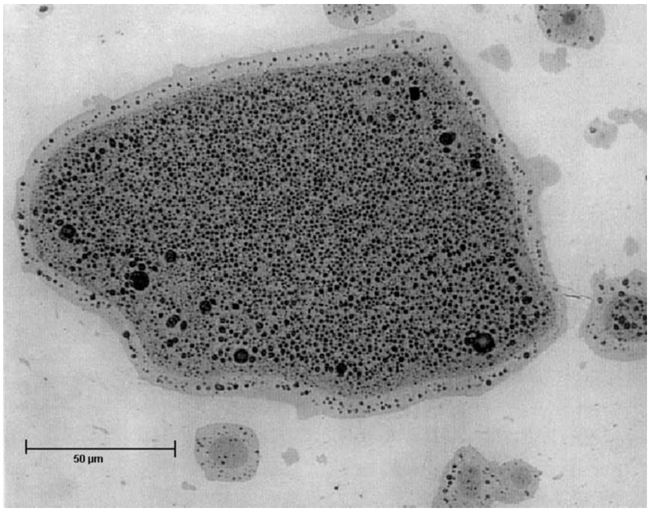

Fig. 1. Micrograph of the specimen with an uranium density of 1.5 gU/cm^3 and a fission density in the U_3Si_2 particles of 12×10^{21} f/cm^3 [1,7].

The calculations of the local neutron flux in OSIRIS have been done by the CEA and have been recalibrated after the last-but-one reactor cycle (F166) by gamma-scanning measurements of the radioactive fission products to directly yield the local fission densities (error ±4%), see Table 4. Before irradiation and after each of the 9 reactor cycles, a plate thickness measurement was done under water in the OSIRIS pool; the IRIS system used for that was identical to the one used in the SILOE reactor, but was newly built. Since a steep gradient of the thermal neutron flux existed across the plate the 5 longitudinal (i.e. vertical) tracks have been chosen in an irregular pattern, i.e. at −28 mm/−23 mm/−14 mm/0 mm/ + 28 mm as related to the transverse plate centre, with measurements taken every 2.5 mm [1,6,7]. A plot of the raw experimental data, all averaged over the two neighbouring tracks at −28 mm and −23 mm, is shown in Fig. 2.

In the longitudinal direction, the swelling was roughly cosine-like, but in the transverse direction the maximum swelling occurred at track −28 mm. Again, the single thickness measurements have then been averaged in order to reduce the statistical scatter, this time over 10 mm longitudinal around plate midplane (5 data points) and 5 mm transverse (tracks −28 mm and −23 mm). It is for this position that the results have been presented in Table 4 (an analogous evaluation for only track +28 mm, i.e. for smaller

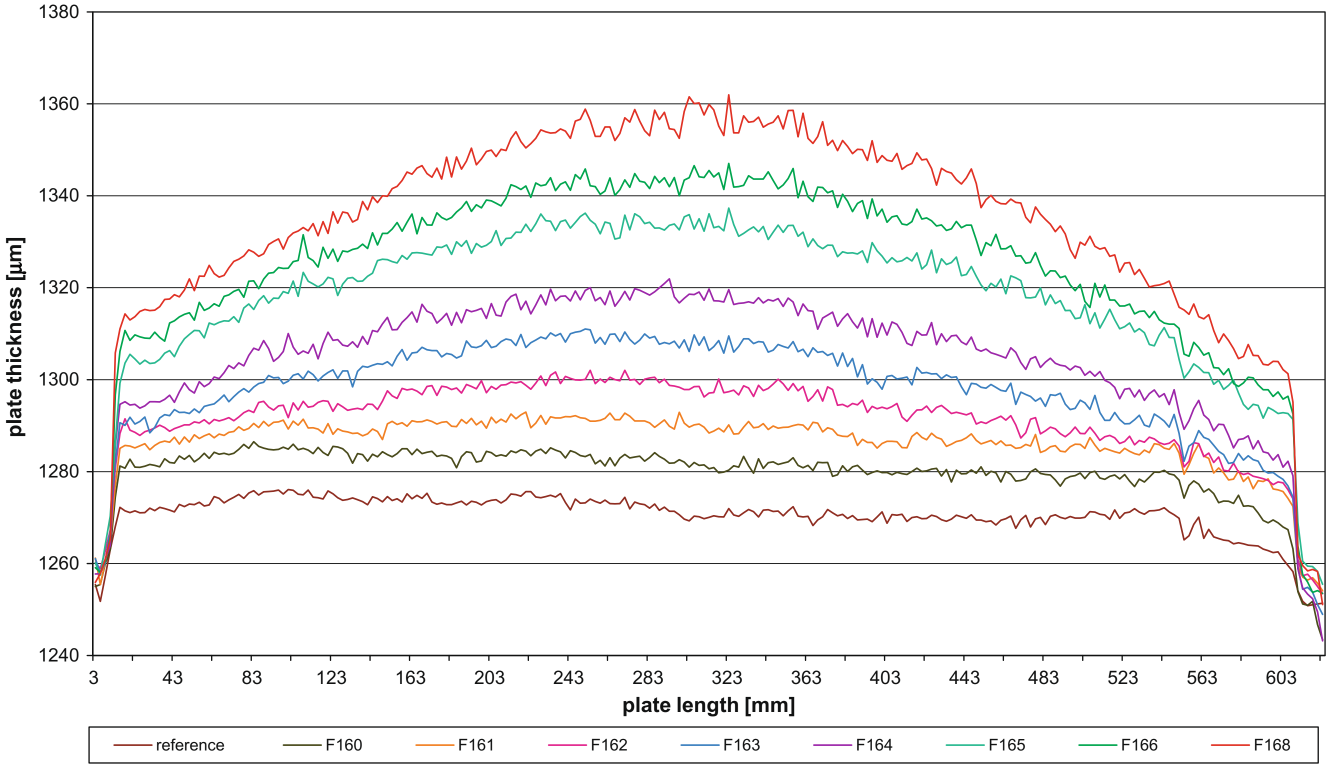

Fig. 2. Original measurements of the thickness of the OSIRIS homogeneous plate (3.0 gU/cm^3) in the longitudinal direction. Shown are the data before irradiation ('reference') and after the reactor cycles F160–F168, see Table 4. All data have been averaged over the tracks at −28 mm and −23 mm, but not yet corrected for the thickness of the claddings and oxide layers [1,7].

Table 3

Results of the irradiation experiment of the test plate with 1.5 gU/cm^3 (homogeneous) in the SILOE reactor

SILOE cycle	Burn-up [%]	FDM [10^{21}f/cm^3]	FDP [10^{21} f/cm^3]	t_F–$t_{F,0}$ [µm]	MS [%]	PS [%]
01/97	4.7	0.13	1.01	−1.96	−0.33	–
02/79	10.4	0.30	2.2	7.0	1.2	16.0
03/97	16.0	0.46	3.4	23.6	4.0	37.1
04/97	21.9	0.63	4.7	–	–	–
05/97	27.2	0.78	5.9	20.3	3.4	33.0
06/97	31.2	0.89	6.7	23.7	4.0	37.2
07/97	38.0	1.09	8.2	28.0	4.7	42.6
08/97	44.2	1.27	9.5	35.9	6.1	52.8
09/97	50.3	1.44	10.8	43.4	7.3	62.2
10/97	55.8	1.60	12.0	49.8	8.4	70.4

FDM and FDP are the fission densities in the fuel meat and in the fuel particles, respectively, and t_F–$t_{F,0}$ is the as-measured increase in fuel plate thickness after correcting for the oxide layer, which steadily increased from 0 to 2 × 5.3 µm. MS and PS are the swelling of the fuel meat and fuel particles, respectively. The data have been obtained for the position of the plate centre, averaged over 22.5 mm in the longitudinal direction (i.e. 10 data points on each track) and 28 mm in the transverse direction (i.e. tracks 2,3 and 4). The measurements after cycles 3 and 4 were perturbed by experimental problems [1].

average fission densities, has also been performed [1] but the results will be presented here only in the summarizing Fig. 6). The time-averaged fission rate in the particles was FRP = 4.9 × 10^{14} f/(cm^3 s) in the position of the maximum. The cladding surface temperature and the heat flux density have been estimated by the CEA for this area to vary in the ranges of 63–75 °C and 100–148 W/cm^2, respectively [7]. Once again, the thickness of the oxide layer has been calculated using a formula given in [8], resulting in a maximum value of 2 × 1.8 µm at the end of the irradiation (which is lower than that given in Section 4 because of the lower temperatures). These values of the oxide layer thickness have been subtracted from the measured plate thickness values (with $t_{F,0}$ = 600 µm and assuming a constant cladding thickness of 2 × 335 µm) to obtain the true meat swelling data given in Table 4, the statistical error being around ±10% [1]. Plots of these swelling data will be found in Sections 6.2 and 7 together with the results of the 3.0 gU/cm^3 zone of the mixed plate. PIE's have not been performed.

6. The OSIRIS test irradiation of the mixed plate (3.0 and 1.5 gU/cm^3)

6.1. General

This irradiation test has also been performed at the materials test reactor OSIRIS (CEA Saclay, France) during 13 reactor cycles (F158–F172) over a total of 284 full power days in the years of 1999 and 2000 [7]. Many of the OSIRIS cycles were identical to those referred to in Section 5. For details of the 'OSIRIS mixed test plate' see Table 2. There were no PIE's.

Table 4
Results of the irradiation experiment of the homogeneous test plate with 3.0 gU/cm^3 in the OSIRIS reactor

OSIRIS cycle	Burn-up [%]	FDM [10^{21} f/cm^3]	FDP [10^{21} f/cm^3]	t_F–$t_{F,0}$ [μm]	MS [%]	PS [%]
F158	0.9	0.05	0.19			
F160	6.1	0.35	1.3	11.2	1.9	10.8
F161	10.6	0.61	2.3	19.2	3.2	15.8
F162	15.3	0.88	3.3	26.4	4.4	20.4
F163	19.4	1.11	4.2	36.5	6.1	26.7
F164	23.3	1.39	5.3	45.8	7.6	32.6
F165	28.7	1.64	6.2	61.5	10.3	39.6
F166	32.7	1.87	7.1	71.3	11.9	48.6
F168	35.9	2.06	7.8	86.1	14.4	57.9

For the meaning of the abbreviations see the caption of Table 3. The swelling t_F–$t_{F,0}$ refers to the as-measured increase in the fuel plate thickness, corrected for the oxide layer steadily increasing from 0 to 2 × 1.8 μm [1,7]. The data have been obtained by averaging over 10 mm longitudinal around plate midplane and over 5 mm transverse (i.e. tracks −28 mm and −23 mm).

Table 5
Results for the 3.0 gU/cm^3 zone of the mixed test plate of the irradiation experiment in the OSIRIS reactor

OSIRIS cycle	Burn-up [%]	FDM [10^{21} f/cm^3]	FDP [10^{21} f/cm^3]	t_F–$t_{F,0}$ [μm]	MS [%]	PS [%]
F158	0.8	0.05	0.18			
F160	5.6	0.33	1.2	9.6	1.5	9.5
F161	9.7	0.57	2.2	15.9	2.5	13.4
F162	14.0	0.83	3.1	19.7	3.1	15.6
F163	17.8	1.05	4.0	31.2	5.0	22.5
F165	22.0	1.30	4.9	42.5	6.8	29.3
F166	25.9	1.53	5.8	52.1	8.3	35.1
F167	30.6	1.80	6.8	62.0	9.9	41.0
F168	33.5	1.97	7.4	74.2	11.8	48.4
F169	36.4	2.15	8.1	78.5	12.5	50.9
F170	41.0	2.42	9.1	90.9	14.5	58.4
F171	44.6	2.63	9.9	110.2	17.5	70.0
F172	48.1	2.84	10.7	128.9	20.5	81.2

The data have been averaged over 10 mm length of the +23 mm track (Fig. 3). For the meaning of the abbreviations see the caption of Table 3. The swelling t_F–$t_{F,0}$ includes the correction for the oxide layer, which steadily increased from 0 to 2 × 3.0 μm [1,7].

The calculations of the local neutron flux in OSIRIS have again been done by the CEA and have been recalibrated after the reactor cycles F166 and F172 (i.e. the last one) by gamma-scanning measurements of the radioactive fission products to directly yield the local fission densities (error ±4%), see Tables 5 and 6. The gamma-scanning and plate thickness measurements have been performed in the same way as already mentioned in Section 5. For the longitudinal plate thickness measurements 4 tracks have been chosen at −23 mm/−5 mm (1.5 gU/cm^3 zone) and at +5 mm/+23 mm (3.0 gU/cm^3 zone) as related to the transverse plate centre, see vertical lines in Fig. 3. In this transverse direction, the thermal neutron flux was characterized by a strong gradient with a maximum beyond track −23 mm and a minimum between tracks +5 mm and +23 mm. In the longitudinal direction the flux profile was again cosine-like with its maximum in the longitudinal plate centre, whence a single track was chosen at this position for the transverse (i.e. horizontal) plate thickness measurements with readings taken every 0.46 mm [1,7].

The original data of the transverse measurements – not yet corrected for the oxide layer and cladding thickness – are presented in Fig. 3. The lowest curve represents the reference curve before irradiation, but the data after the first, very short irradiation (cycle F158) have been omitted for clarity. One sees that the plate thickness increases monotonically during irradiation and that there is no anomaly across the step in the uranium density. At this position – and even stronger beyond track +23 mm – the local maximum in the 3.0 gU/cm^3 zone is a consequence of the well-known ‘reflector maximum’ of the thermal neutron flux, and the local minimum in the 1.5 gU/cm^3 zone just corresponds to the inverse effect. These local maxima, however, have not been further evaluated since not being positioned on the longitudinal tracks whence the experimental uncertainties would have been particularly high.

6.2. Mixed plate results for the 3.0 gU/cm^3 zone

The results for the 3.0 gU/cm^3 zone of the mixed plate are presented in Table 5. The data have been averaged over 10 mm length of track +23 mm (see Fig. 3) around the longitudinal plate centre [1]. The time-averaged fission rate in the particles was FRP = 4.4 × 10^{14} f/(cm^3 s) at this position. The cladding surface temperature and the heat flux density have been estimated by the CEA for this area to vary during irradiation from 73 °C to 63 °C and from 142 W/cm^2 to 98 W/cm^2, respectively [7]. With these input values the thickness of the oxide layer has been calculated using a formula given in [8], resulting in a maximum value of 2 × 3.0 μm at the end of the irradiation. These values have been subtracted from the measured plate thickness data. By assuming a constant cladding thickness of 2 × 335 μm and $t_{F,0}$ = 628.2 μm as derived from the measurement before irradiation, the true meat swelling data have been obtained as listed in Table 5. The statistical error of PS is around ±2% [1].

All our results of the fuel plate thickness measurements (t_F–$t_{F,0}$) for the U_3Si_2–Al dispersion fuel with an uranium density of 3.0 gU/cm^3 are shown in Fig. 4 [1]. The data are displayed as a function of the fission density in the fuel particles (FDP) and comprehend not only those of the +23 mm track of the mixed plate (as in Table 5) but also those of the +5 mm track (see Fig. 3) which have not been tabulated in this paper (tables are given in [1]). Additionally, the results for the homogeneous plate described in Section 5 (Table 4) have been included. As one can see, all data nicely fit to one another and confirm a practically linear, stable and well predictable swelling behaviour of the plates. Even more, this excellent irradiation behaviour of U_3Si_2–Al dispersion fuel with 3.0 gU/cm^3 has been verified up to very high irradiation doses with a maximum value of FDP = 10.7 × 10^{21} f/cm^3, which corresponds to a maximum

Table 6
Results for the 1.5 gU/cm^3 zone of the mixed test plate of the irradiation experiment in the OSIRIS reactor

OSIRIS cycle	Burn-up [%]	FDM [10^{21} f/cm^3]	FDP [10^{21} f/cm^3]	t_F–$t_{F,0}$ [μm]	MS [%]	PS [%]
F158	1.2	0.03	0.26			
F160	7.8	0.23	1.7	9.9	1.6	19.3
F161	13.5	0.40	3.0	20.4	3.2	31.8
F162	19.4	0.57	4.3	25.4	4.0	37.7
F163	24.5	0.72	5.4	32.8	5.2	46.4
F165	29.9	0.89	6.7	39.8	6.3	54.8
F166	34.9	1.03	7.8	44.7	7.1	60.6
F167	40.7	1.20	9.1	51.1	8.1	68.2
F168	44.3	1.31	9.9	58.7	9.3	77.3
F169	47.9	1.42	10.7	65.9	10.4	85.8
F170	53.5	1.58	11.9	68.8	10.9	89.3
F171	57.9	1.72	12.9	84.1	13.3	107.4
F172	61.9	1.84	13.8	83.0	13.1	106.1

The data have been averaged over 10 mm of length around the longitudinal plate centre of the −23 mm track as indicated in Fig. 3. For the meaning of the abbreviations see the caption of Table 3. The swelling t_F–$t_{F,0}$ is the as-measured increase in fuel plate thickness after correcting for the oxide layer, which steadily increased from 0 to 2 × 1.5 μm [1,7].

FDM = 2.8 × 10^{21} f/cm^3, see Table 5. This is far more than the design values of the FRM II as listed in Table 1.

6.3. Mixed plate results for the 1.5 gU/cm^3 zone

The results for the 1.5 gU/cm^3 zone of the mixed plate are presented in Table 6. The data have been averaged over 10 mm length of track −23 mm (see Fig. 3) around the longitudinal plate centre [1]. The time-averaged fission rate in the particles was FRP = 5.6 × 10^{14} f/(cm^{3}s) at this position. The cladding surface temperature and the heat flux density have been estimated by the CEA for this area to vary during irradiation from 63 °C to 52 °C and from 97 W/cm^2 to 55 W/cm^2, respectively [7]. With these input values the thickness of the oxide layer on the plate has been calculated to be 2 × 1.5 μm at the end of the irradiation. These values have been subtracted from the measured plate thickness data, and by assuming a constant cladding thickness of 2 × 335 μm and $t_{F,0}$ = 632.9 μm as derived from the measurement before irradiation, the true meat swelling data have been obtained as listed in Table 6. The statistical error of PS is around ±2% [1].

All our results of the fuel plate thickness measurements (t_F–$t_{F,0}$) for the U_3Si_2–Al dispersion fuel with an uranium density of 1.5 gU/cm^3 are shown in Fig. 5 [1]. The data are displayed as a function of FDP and comprehend not only those of the −23 mm track of the mixed plate (as in Table 6) but also those of the −5 mm track (see Fig. 3; tables are given in [1]); here, the fission densities

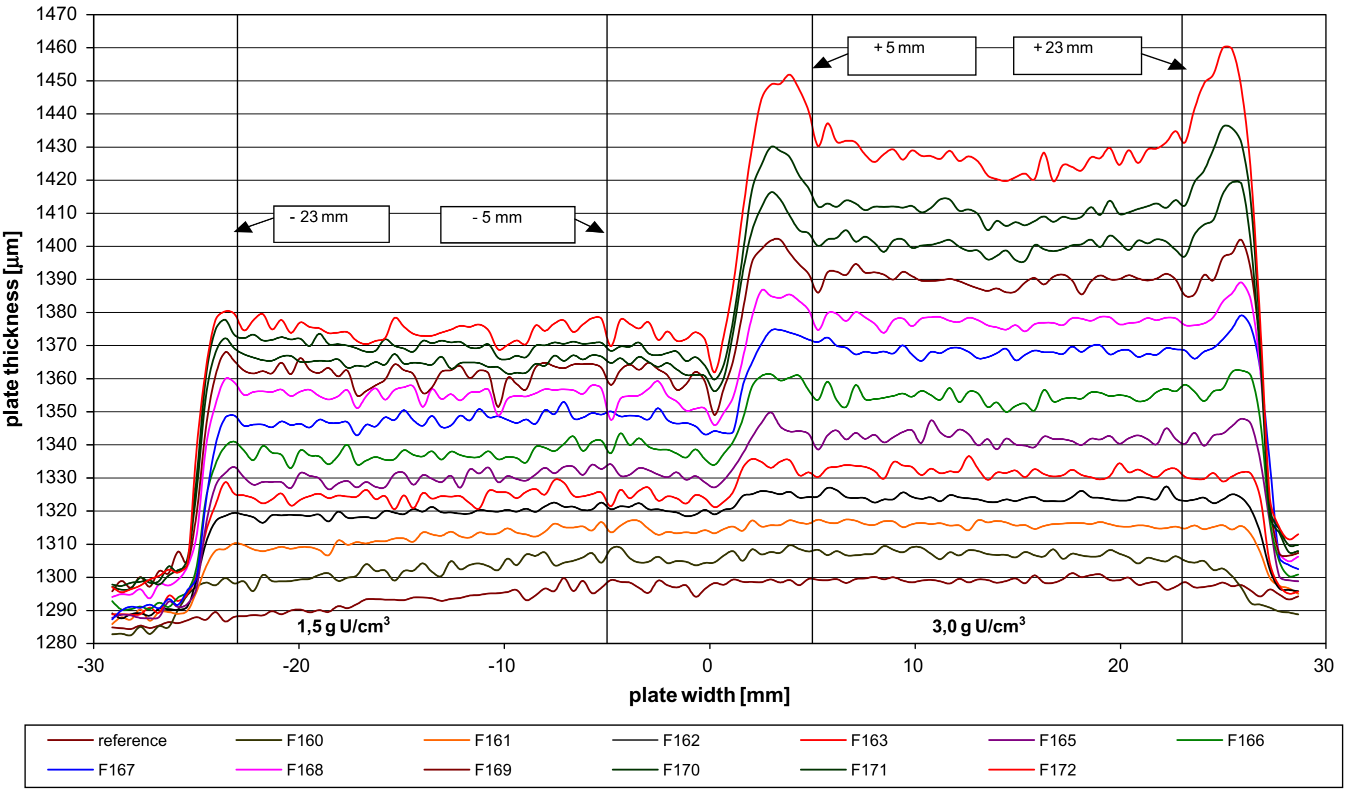

Fig. 3. Original measurements of the thickness of the OSIRIS mixed plate (in the longitudinal midplane) in the transverse direction, i.e. across the step in the uranium density from 1.5 gU/cm^3 (left) to 3.0 gU/cm^3 (right). Shown are the data before irradiation ('reference') and after the reactor cycles F160–F172, see Tables 5 and 6, without correcting for the claddings and oxide layers. The vertical lines indicate the position of the four tracks of the longitudinal measurements [1,7].

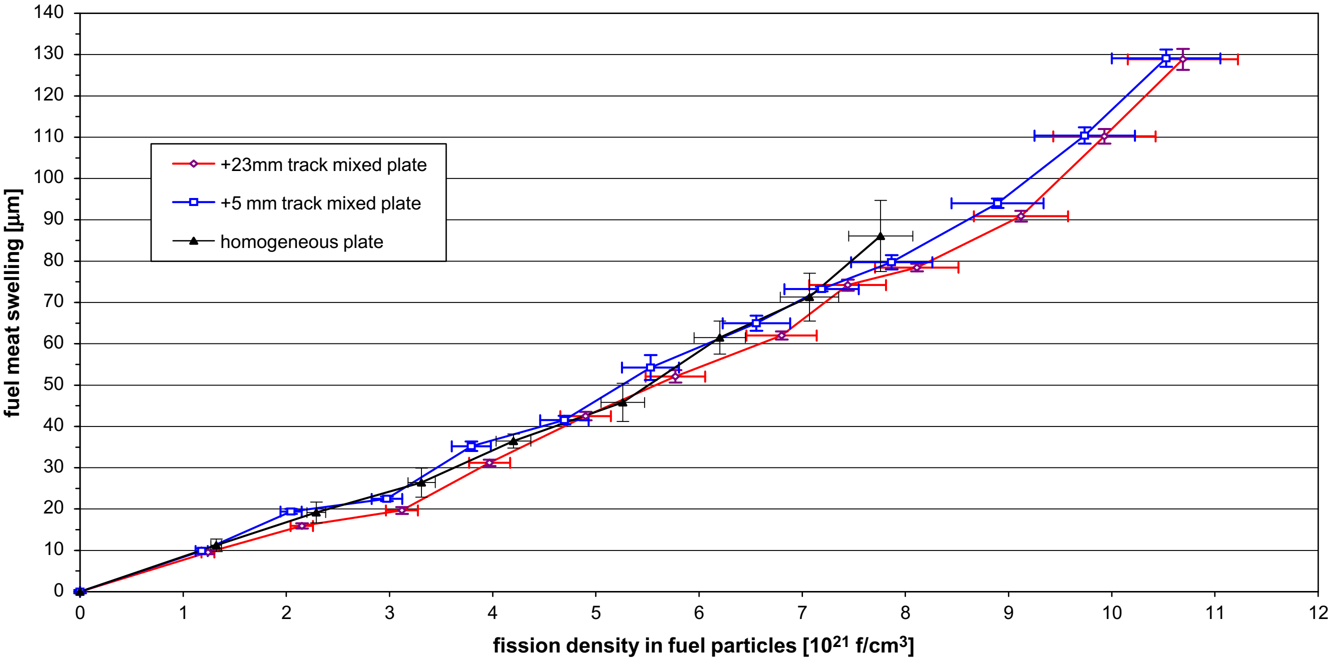

Fig. 4. U_3Si_2–Al fuel with 3.0 gU/cm^3 – plot of the increase of the fuel meat thickness (t_F–$t_{F,0}$) as a function of the fission density in the U_3Si_2 fuel particles (FDP). Shown are the data for the +23 mm track of the OSIRIS mixed plate together with those of the +5 mm track and also those of the −28 mm/−23 mm tracks of the OSIRIS homogeneous plate described in Section 5 (Table 4). The data have always been averaged over 10 mm of track length. The thickness of the oxide layer has been subtracted in all cases [1].

achieved are significantly lower because of the strong gradient of the thermal neutron flux (Section 6.1). The last-but-one data point of the −23 mm track is very probably not quite realistic, but is an outlier originating from the measuring system, as can be concluded since the very last point again fits very well to the other data. Additionally, the results of the SILOE experiment described in Section 4 (Table 3) have been included, although the measurements after reactor cycles 3 (outlier data point) and 4 (missing data) have been perturbed by experimental problems. As discussed in Section 2, the initial decrease of the plate thickness must be a consequence of a locally high value of the as-fabricated porosity, which disappears during the initial phase of the irradiation due to the radiation-induced diffusion. In the further progress of the irradiation, the slope of the SILOE swelling curve is practically the same as that of the OSIRIS curves.

Keeping all this in mind one sees that all data nicely fit to one another and confirm a practically linear, stable and well-predictable swelling behaviour of the plates. Even more, this excellent

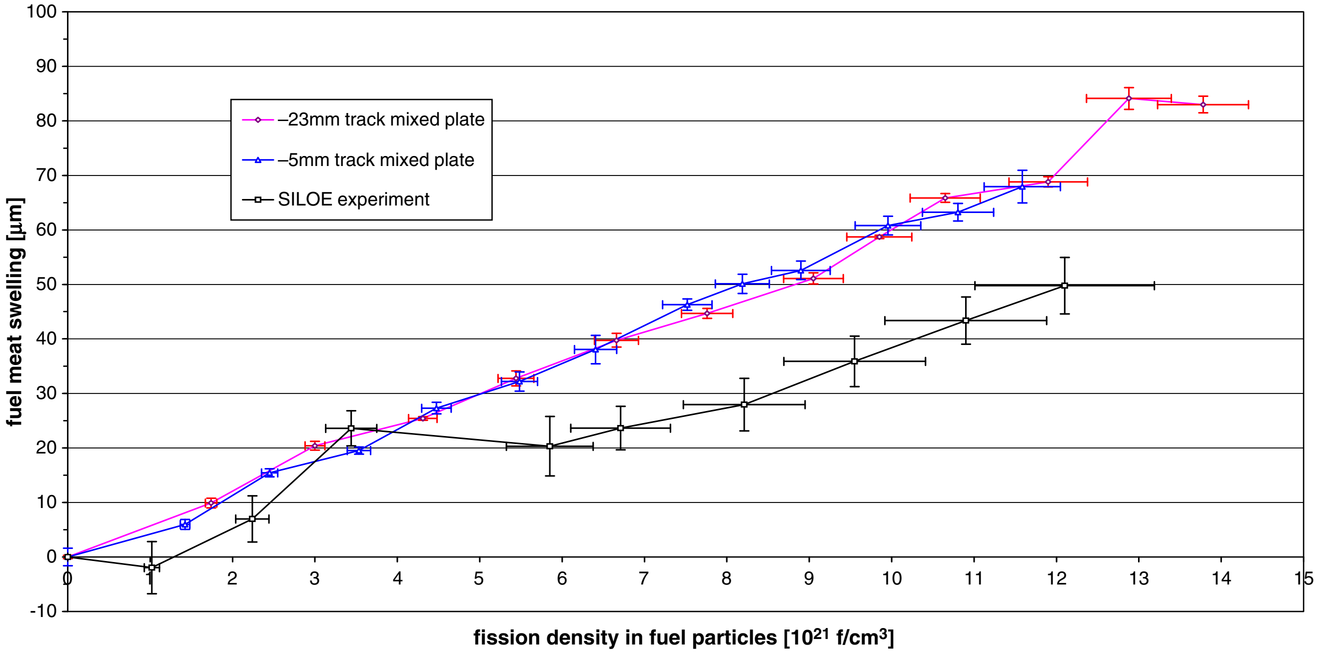

Fig. 5. U_3Si_2–Al fuel with 1.5 gU/cm^3 – plot of the increase of the fuel meat thickness (t_F–$t_{F,0}$) as a function of the fission density in the U_3Si_2 fuel particles (FDP). Shown are the data for the −23 mm track of the OSIRIS mixed plate (Table 6) together with those of the −5 mm track (not contained in Table 6), always averaged over 10 mm of track length. Also shown are the results of the SILOE experiment described in Section 4 (Table 3). The thickness of the oxide layer has been subtracted in all cases [1].

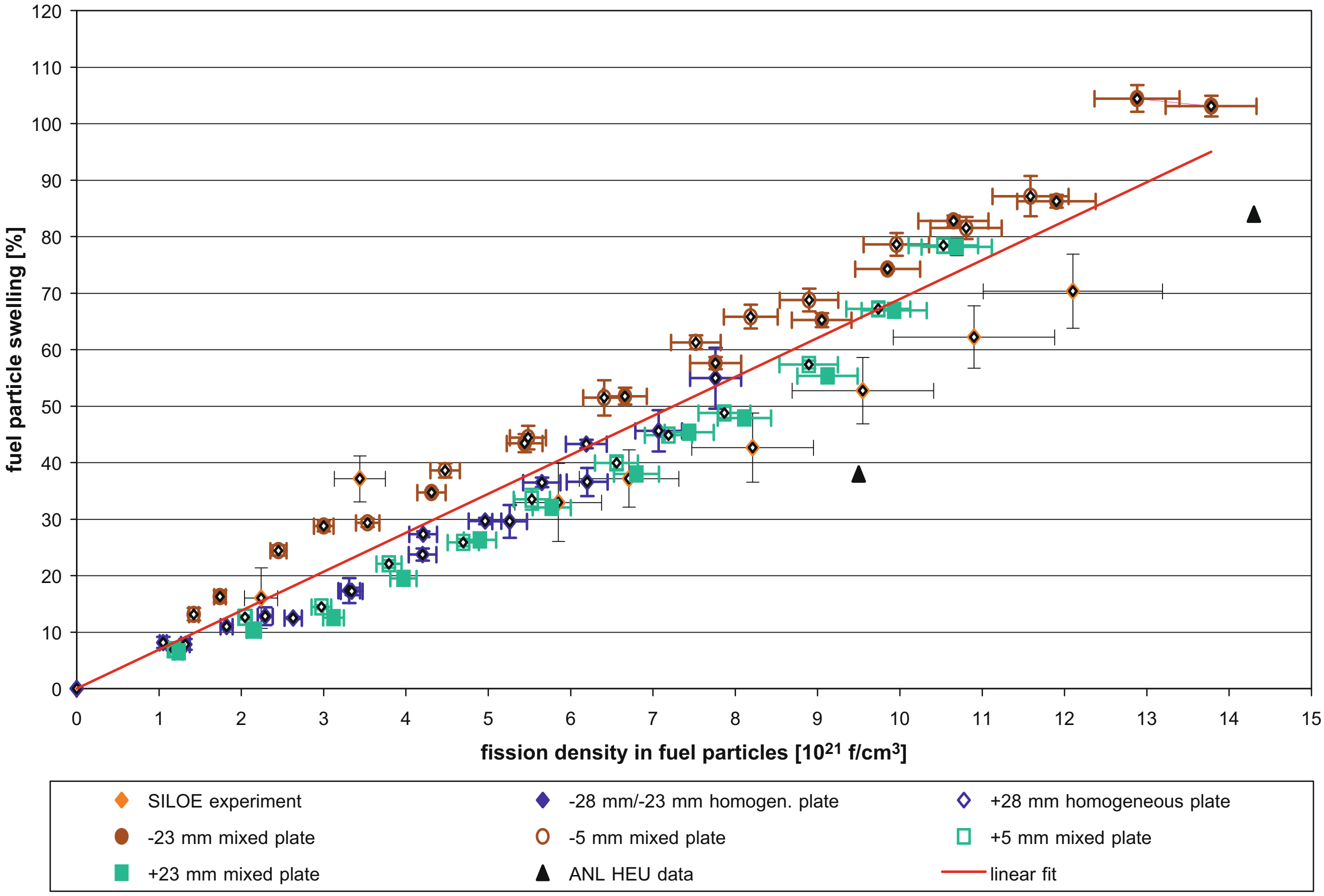

Fig. 6. Increase of the volume of the U_3Si_2 fuel particles (PS) as a function of the fission density in the particles (FDP). This is a comprehensive plot of all results [1], but in contrast to the tabulated PS values as given above the unknown as-fabricated porosity V_P has been adjusted for each curve of the OSIRIS experiments to yield an initially smooth relationship. Shown are the data for the OSIRIS homogeneous plate (tracks −28/−23 mm and +28 mm) and the OSIRIS mixed plate (tracks +23 and +5 mm), all with 3.0 gU/cm^3, as well as tracks −23 mm and −5 mm of the OSIRIS mixed plate and the SILOE results, all with 1.5 gU/cm^3. The straight line with a slope of 6.86×10^{-21}%cm^3/f represents a fit of all our data. Also shown are the ANL data of Refs. [2,10] as obtained from a HEU sample with 1.7 gU/cm^3.

irradiation behaviour of U_3Si_2–Al dispersion fuel with 1.5 gU/cm^3 has been verified up to very high irradiation doses with a maximum fission density in the particles of FDP = 13.8×10^{21} f/cm^3 corresponding to a maximum fission density in the meat of FDM = 1.84×10^{21} f/cm^3 (see Table 6). This is far more than the design values of the FRM II as listed in Table 1.

7. Discussion

The summarizing Fig. 6 shows the increase of the volume of the U_3Si_2 fuel particles (particle swelling PS) as a function of the fission density in the particles (FDP). This is a comprehensive plot of all data we have obtained – however, including an adjustment of the as-fabricated porosity V_P for each curve of the OSIRIS experiments [1]. It has been pointed out in Section 2 that V_P is difficult to be determined and that the radiation-induced disappearance of V_P in the initial phase of an irradiation may give rise to even negative values of the fuel thickness swelling (as in Fig. 5, SILOE experiment). However, it is physically plausible to expect that PS should depend more or less linearly on FDP in the initial phase of an irradiation, where any production of larger gas bubbles in the particles can be excluded (Fig. 1). So for all the curves in Fig. 6, originating from the OSIRIS experiments the local values of V_P have been adjusted to yield an initially smooth and monotonic increase of PS with FDP. The results of the corrected V_P are given in the footnote.[4] This procedure also reduces the scatter of the various data points, as can be seen from the correlation coefficient of the linear fit to all our data, which increases from 0.918 without porosity adjustment to 0.925 (the ideal case would correspond to 1.000). The consequences of this modification of V_P for the resulting values of the particle swelling PS are small, as can be derived from differentiating Eq. (1). So the maximum values of PS as given in Tables 4–6 are only reduced by not more than 3% (absolute), as e.g. in Table 4 the maximum value of PS ≈ 58% reduces to PS ≈ 55%. All our conclusions in this paper remain unaffected by this [1].

Fig. 6 demonstrates once again that the swelling PS of the fuel particles, which is independent of the uranium density in the U_3Si_2–Al fuel meat, is a monotonic and well-predictable function of FDP over this very large range. The various data do show some scatter which, however, was anticipated considering the experimental difficulties of measuring very small changes of different radioactive samples under tough conditions in the reactor pools of different reactors after different reactor irradiation histories.

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[4] As a result the values of the as-fabricated porosity V_P of the OSIRIS plates have been modified from their previous estimates given in Table 2 to 0.2% and 0.1% (homogeneous plate, tracks −28/−23 mm and +28 mm, respectively), to 0.2% and 0.25% (mixed plate, tracks +23 mm and +5 mm, respectively), and to 0.6% and 0.8% (mixed plate, tracks −23 mm and −5 mm, respectively).

It is clear that the swelling is a uniform function of the fission density – without any indication of a rapid increase which would have been related to a build up and interlinkage of very large gas bubbles in the fuel particles ultimately leading to excessive swelling (pillowing) and failure of the fuel plate. This uniform behaviour of PS also excludes any build up of larger pores in the Al matrix of the fuel since such an effect would be misinterpreted as a particle swelling according to Eq. (1).

The groups at Argonne (ANL) and Oak Ridge (ORNL) National Laboratories developed a metallurgical model to understand the irradiation behaviour of U_3Si_2–Al fuel in a two-stage process [3,9]. At low values of FDP, the swelling of the particles (PS) first increases linearly with a certain slope. In this phase the fission gas forms very small bubbles in the original U_3Si_2 matrix, so small that they cannot be seen even with the scanning electron microscope. At a certain value of FDP the stress in the particles becomes so large that they undergo complete recrystallization into a large number of submicron-size grains ('grains refinement'). Fission gas precipitates at the boundaries of these small new grains resulting in a rather uniform distribution of small noninteracting gas bubbles that grow in size with increasing fission density – this situation is shown in Fig. 1. In this second stage, the slope of the PS versus FDP curve should be about three times of that of the first stage, and the onset of the second stage should shift to a higher fission density with increasing fission rate [3,9]. Concerning the results of our test irradiations, see Fig. 6, we did not perform such a two-slope fit of our data since the one-slope fit was very satisfying. It is, however, important to realize that our tests were also conservative with respect to the fission rate since this is higher at the FRM II than in the tests whence the fuel swelling should even be lower in the FRM II than reported in this paper.

We have also included in Fig. 6 the only swelling data known to us of other HEU U_3Si_2–Al samples irradiated to very high fission densities. These are the results of the ANL [2,3,10], see Table 7, where the meat swelling MS has been obtained from immersion density measurements (buoyancy flotation measurements) of the complete miniature test plates. The fission rate in the particles was FRP ≈ 6.4×10^{14} f/(cm^3 s), which is of the same order as in our tests but lower than at the FRM II. Allowing for the general observation [11] that integral measurements of the whole specimen lead to somewhat smaller swelling values than local measurements, the agreement with our data is quite good.

The fuel temperatures of the FRM II irradiation tests, as listed above, were somewhat lower than the maximum centreline fuel temperatures of 110 °C in the FRM II fuel element at nominal conditions (Section 2). Since the heat conduction in the U_3Si_2–Al fuel is dominated by the very good heat conductivity of the matrix Al, the difference between fuel centreline temperature and outer cladding surface temperature in the FRM II fuel element is only about 14 °C at the beginning of life. During irradiation some of the matrix Al is consumed due to the build up of U(Al,Si)$_3$ interdiffusion layers around the fuel particles [3,12]. In the PIEs after the SILOE experiment (1.5 gU/cm^3, see Fig. 1) the remaining volume ratio of the Al matrix in the fuel has been experimentally determined to be $VR_{Al} \approx (77 \pm 7)\%$ in the zone with FDP ≈ 12×10^{21} f/cm^3 [1]. A theoretical treatment, assuming spherical fuel particles with a mean diameter of 50 μm, resulted in $VR_{Al} \approx 68\%$ for a fuel temperature of 110 °C; larger particle diameters give rise to larger values of VR_{Al} [1]. The same calculation, applied to the case of the OSIRIS mixed plate irradiation where PIE's have not been performed, yielded $VR_{Al} \approx 63\%$ for the zone with 1.5 gU/cm^3 and FDP = 13.8×10^{21} f/cm^3 (Section 6.3), and $VR_{Al} \approx 41\%$ for the zone with 3.0 gU/cm^3 and FDP = 10.7×10^{21} f/cm^3 (Section 6.2). All these values are significantly higher than the conservative lower limit of $VR_{Al} \approx 15\%$ as established by the ANL for a stable behaviour of U_3Si_2–Al fuel under irradiation [12]. In our calculations the fuel temperature has again been assumed to be 110 °C – which is conservative since the actual temperature in the tests was lower than that.

Table 7
Swelling data of two ANL miniature plate specimen with U_3Si_2–Al HEU fuel with 1.7 gU/cm^3 [2,3] – shown in the table are the revised data after a correction which has been performed later by the ANL [10]

Burn-up [%]	FDM [10^{21} f/cm^3]	FDP [10^{21} f/cm^3]	MS [%]	PS [%]
42	1.4	9.5	4.9	38
63	2.1	14.3	11.6	84

The volume ratio was reported to be VR = 14.7% and the as-fabricated porosity V_P = 0.8%. As always in this paper, FDM and FDP are the fission densities in the fuel meat and particles, respectively, and MS and PS are the fuel meat swelling and particles swelling, respectively.

Quite recently higher temperatures have been realized during the test irradiation of full-sized U_3Si_2–Al fuel plates over 69 days in the BR2 reactor in Mol, Belgium [13].[5] These were LEU plates (19.9 wt% ^{235}U) with an uranium density as high as 4.8 gU/cm^3. The local maximum of the heat flux was calculated to be over 400 W/cm^2 and the maximum temperature at the outer cladding surface to be in the range of 120–140 °C so that the fuel centreline temperature had to be even higher than that. As was demonstrated in a PIE, the fuel behaved as expected 'with ample matrix Al still left within the meat' even after a peak burn-up of 86% yielding a value of FDP = 4.8×10^{21} f/cm^3 corresponding to FDM = 2.1×10^{21} f/cm^3 [13].

A significant dependence of the fuel swelling on the temperature could only be expected if the diffusion would no longer be activated by radiation only but also by temperature. Unirradiated U_3Si_2–Al fuel had to be annealed for several hundreds of hours at temperatures around 400 °C to show a measurable formation of an interdiffusion layer; however, at 300 °C no interaction was observed after more than 1000 h [12].

The HANS test irradiations were performed in the USA to qualify the U_3Si_2–Al fuel with HEU (93% ^{235}U), but with low uranium density (about 1.7 gU/cm^3) for the Advanced Neutron Source (ANS, a 330 MW research reactor project of the US abandoned in 1995) under unprecedented experimental conditions. In the HANS-1 and HANS-2 tests, cold-compacted mixtures of fuel particles and Al powder were irradiated [9] and in the HANS-3 experiment small samples punched from realistic fuel plates as produced by the standard hot-rolling process [14]. The aim was to study the fission gas bubbles morphology and chemical interactions after irradiation with very high fission rates (initial values FRP up to 2.8×10^{16} f/(cm^3 s)) at elevated temperatures (230–425 °C) to extremely high burn-ups. The fuel swelling has not been measured. The authors concluded that the interdiffusion of matrix Al into the fuel particles at temperatures below about 250 °C is athermal and fission enhanced. At the highest burn-up of about 90% (FDP ≈ 20×10^{21} f/cm^3), the change in the chemical composition of the HEU U_3Si_2 fuel particles is substantial, since about 84% of the original uranium has been transmuted into fission products (two each). Depending on temperature and burn-up, a coarsening of the fission gas bubbles into relatively large ones has been observed in the centres of practically all fuel particles. The authors point out, however, that the dispersion fuel would still perform satisfactorily under irradiation, if the individual fuel particles all remain surrounded by Al matrix material to prevent them from interlinking with other particles, which would lead to the formation of large interparticle gas bubbles and excessive swelling

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[5] In that paper some values of FDM and FDP have been confused as can be verified by comparing these values and the burn-ups given in Section 2.1 of [13] with each other or with data from the literature, e.g. [2,3].

(pillowing). And U_3Si_2–Al fuel with a volume ratio VR (Table 1) up to at least 20% would meet these requirements in the ANS reactor even at temperatures up to about 425 °C [9,14].

More generally the maximum permissible value of FDP is higher, the lower is the uranium density in the fuel. The maximum possible FDP values can be accomplished only with HEU fuel since, e.g., in LEU fuel with 20% ^{235}U a burn-up of even 100% only yields a FDP value of about 5.5×10^{21} f/cm^3.

In conclusion, it is evident that U_3Si_2–Al dispersion fuel – under realistic operating conditions as always being realized in practice, and for not too high uranium densities so that there is always sufficient matrix aluminium available in the fuel meat – represents an excellent fuel for being used in high performance research reactors up to very high fission densities, fission rates and fuel temperatures.

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