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D4.1 MARINE gas puncturing and swelling behaviour of (U, Am)₂O₃

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DOCUMENT SUMMARY

Results from non-destructive examinations (NDE) and gas release analysis on irradiated fuel pins of the MARINE experiment. NDE include dimensional analysis by neutron radiography, clad strain measurements and axial scans with gamma spectrometry. Gas release analysis includes determination of pressure and elemental and isotopic composition of the plenum gas.

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Summary

The goal of the MARINE irradiation is to determine helium release behaviour and swelling of $^{241}\text{Am}_{0.15}\text{U}_{0.85}\text{O}_{2-x}$ fuel containing natural uranium, which is representative of the Minor Actinide Bearing Blanket (MABB) material to be used for transmutation in Sodium-cooled Fast Reactors (SFRs). In addition, a direct comparison under identical conditions will be made between the pellet (pin 1) and sphere-pac (pin 2) fuel forms in order to assess their performance.

MARINE was successfully irradiated for 359 Effective Full Power Days (EFPD) in 12 cycles in the High Flux Reactor in Petten, between January 5th, 2016 and May 7th, 2017.

The fluence monitor sets, retrieved during dismantling, have been measured and reporting of the results has been completed. The post irradiation nuclear analysis (or burn-up analysis) has been completed. Burnups of 1.57% and 1.98% FIMA were calculated for pin 1 and pin 2, respectively. Comparing Am-241 contents before and after irradiation, these calculations yield a reduction of 55% and 64% in pin 1 and pin 2, respectively.

An axial scan with gamma spectrometry was carried out after which the experiment was further dismantled and pins were separated from each other, after which clad strain (or profilometry) measurements were carried out. No irregular strains were observed. Maximal diameter increases of 7µm and 4µm were found at the height of the fuel columns of pin 1 (pellets) and pin 2 (Sphere-Pac), respectively.

Combining measurements on clad strains and fuel stack heights, a careful estimation of the maximum volumetric swelling was made: $\pm 10\%$ / % FIMA burn-up. More conclusive results on volumetric swelling can be made during destructive analysis.

Finally, pin 1 was punctured and the internal plenum pressure was measured to be 14.7 bars, converting to a ~86% release of the total gas produced at end of irradiation according to the post irradiation nuclear calculations. Mass spectrometry of the gas confirms dominant He-release (97%) and only a small fraction release of fission gases (2-5%).

Table of contents

Summary	3	
1	Introduction	5
2	Description of the experiment	6
2.1	Fuel pin specifications	6
3	Post irradiation nuclear analysis	9
3.1	Spectrum adjustments	9
3.2	Fuel power history	9
3.3	Gas production	12
3.4	Estimation of maximum fuel temperatures	13
4	Fuel stack elongation	15
5	Profilometry – clad strain	17
5.1	MARINE Pin 1	18
5.2	MARINE Pin 2	19
5.3	Volumetric swelling	20
6	Gamma spectrometry	21
6.1	Instrumentation	21
6.2	Pin 1 - Pellets	22
6.3	Pin 2 – Sphere-Pac	24
6.4	Stack length elongation based on gamma spectrometry	26
7	Helium and fission gas analysis	27
7.1	Instrumentation	27
7.2	MARINE Pin 1	28
7.3	MARINE Pin 2	31
8	Conclusions	32
List of tables		35
List of figures		35

1 Introduction

Americium is one of few radioactive elements, together with Pu and Np, that contribute strongly to the long-term radiotoxicity of spent nuclear fuels. Transmutation of long-lived nuclides as ^{241}Am by irradiation in fast-spectrum reactors is the only option for significantly reducing the volume and radiotoxicity of nuclear waste [1], [2].

The goal of the MARINE irradiation is to determine helium release behaviour and swelling of $^{241}\text{Am}_{0.15}\text{U}_{0.85}\text{O}_{2-x}$ fuel containing natural uranium, which is representative of the Minor Actinide Bearing Blanket (MABB) material to be used for transmutation in Sodium-cooled Fast Reactors (SFRs). In addition, a direct comparison under identical conditions is made between the pellet and sphere-pac fuel forms in order to assess their respective performance.

MARINE was successfully irradiated for 359 Effective Full Power Days (EFPD) in 12 cycles in the High Flux Reactor in Petten, between January 5, 2016 and May 7, 2017. More information is available in the dedicated irradiation report [3].

This report contains the summarized results of the post-irradiation nuclear analysis and the results of the non-destructive examinations (profilometry and gamma spectrometry) and of the puncturing test, where the internal plenum pressures of the fuel pins and their gas composition were analysed.

2 Description of the experiment

2.1 Fuel pin specifications

MARINE consists of two short fuel pins, with the minor actinide bearing oxide fuel (Am,U)O₂ in two geometries: pellets and a binary size pebble bed, called Sphere-Pac. The fuels were fabricated by JRC-Karlsruhe via two different routes. The pellets were produced by producing porous UO₂ beads, which were then infiltrated with americium solution and calcinated. The calcinated beads were compacted and sintered to obtain the fuel pellets.

For the binary-sized Sphere-Pac fuel, both the large and small bead batches were produced by starting with a uranium-americium nitrate solution, from which beads were produced by sol-gel method and sintered. Detailed information on the fabrication of both fuels can be found in a dedicated report [4].

Table 2.1 provides details on the two fuel pins. Figure 2.1 and Figure 2.2 shows the schematic drawings of the pins, including dimensions of the most important features.

Table 2.1 Fuel and fuel pin specifications

Sample holder	MARINE-01	
Fuel pin	Pin 1	Pin 2
Manufacturer	JRC Ka	JRC Ka
Type	(U,Am)O ₂	(U,Am)O ₂
Am/metal (mol %)	13.38	12.96
U/metal (mol %)	86.62	87.04
²³⁴ U (mol %)	0.00	0.01
²³⁵ U (mol %)	0.72	0.72
²³⁶ U (mol %)	0.00	0.00
²³⁸ U (mol %)	99.27	99.27
²⁴¹ Am (mol %)	93.40	93.40
²³⁷ Np (mol%) **	6.60	6.60
Bulk fuel density (g/cc)	10.46	7.45
Percentage T.D. (%)	94.61	55.36
Pellet O.D. (mm)	5.368	n.a.
Bead diameter (mm) *	n.a.	0.745 / 0.050
O/M	1.93	1.935
Fuel weight (g)	13.182	9.367
Am-weight (g)	1.561	0.256 / 0.819*
U-weight (g)	10.109	1.823 / 5.393*
Fuel column length (mm)	55.67	61.01
Fuel cladding	15/15 Ti	
Cladding O.D. (mm)	6.55	

Cladding I.D. (mm)	5.65	
Plenum gas composition	99% He / 1% Ne	99% He / 1% Ne
Plenum gas fill pressure (bar)	2.75	2.75

* Large/small bead fraction

** impurity in Am-batch

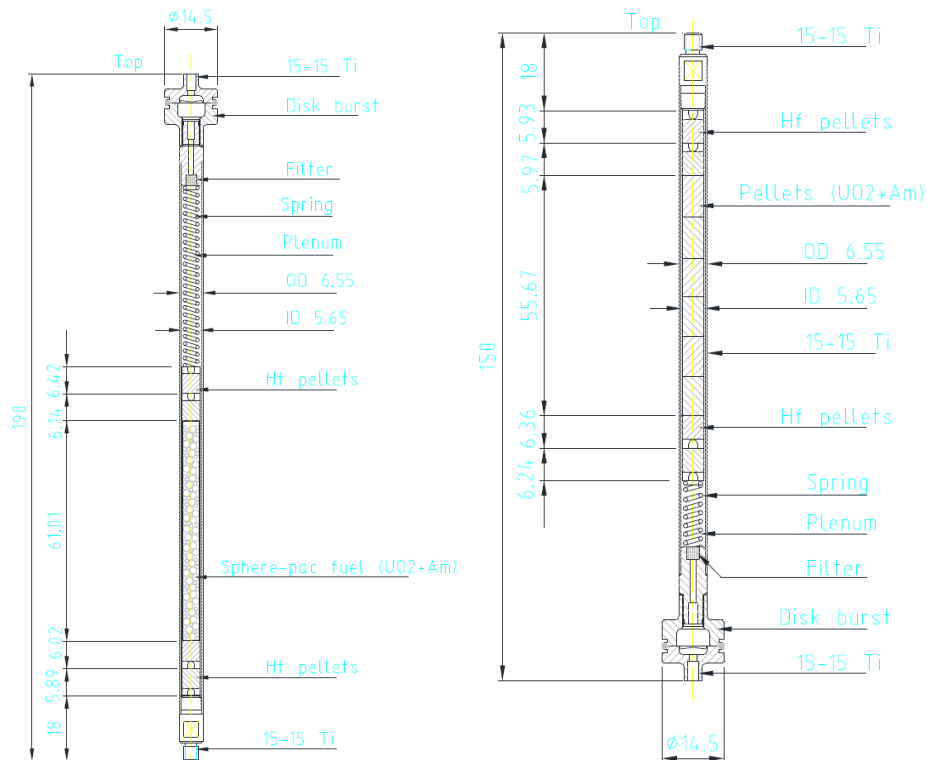


Figure 2.1 Schematic drawings of the two fuel pins

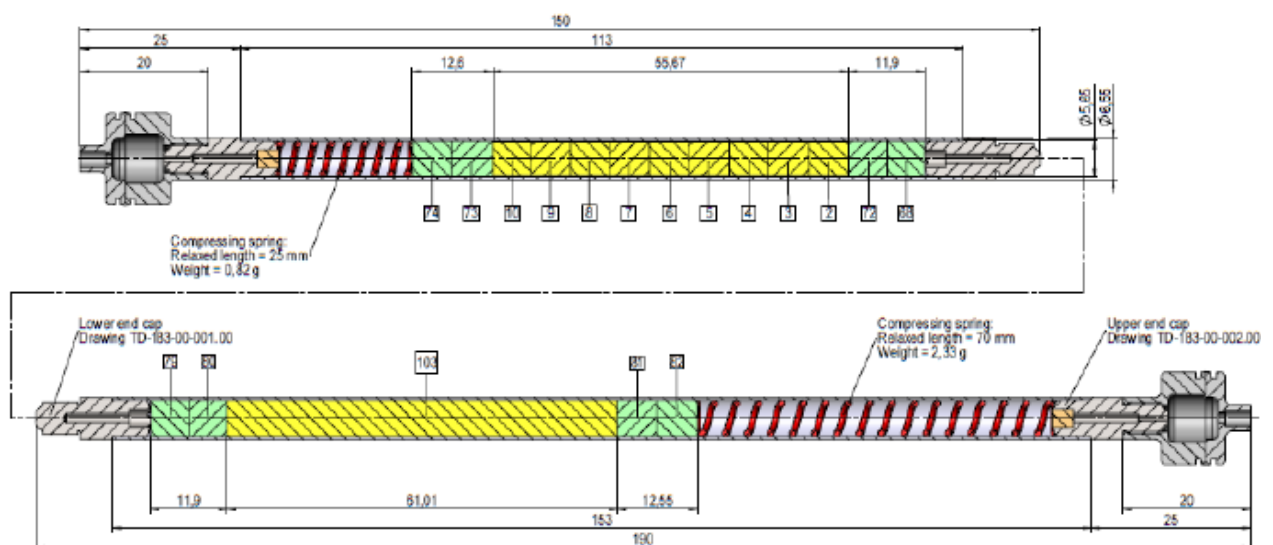


Figure 2.2 Schematic drawings of the two fuel pins

Figure 2.3 shows an X-ray tomography image, produced by JRC Karlsruhe after pin fabrication, that reveals a difference in packing / smear density along the fuel pin, due to a shortage of small bead fraction material.

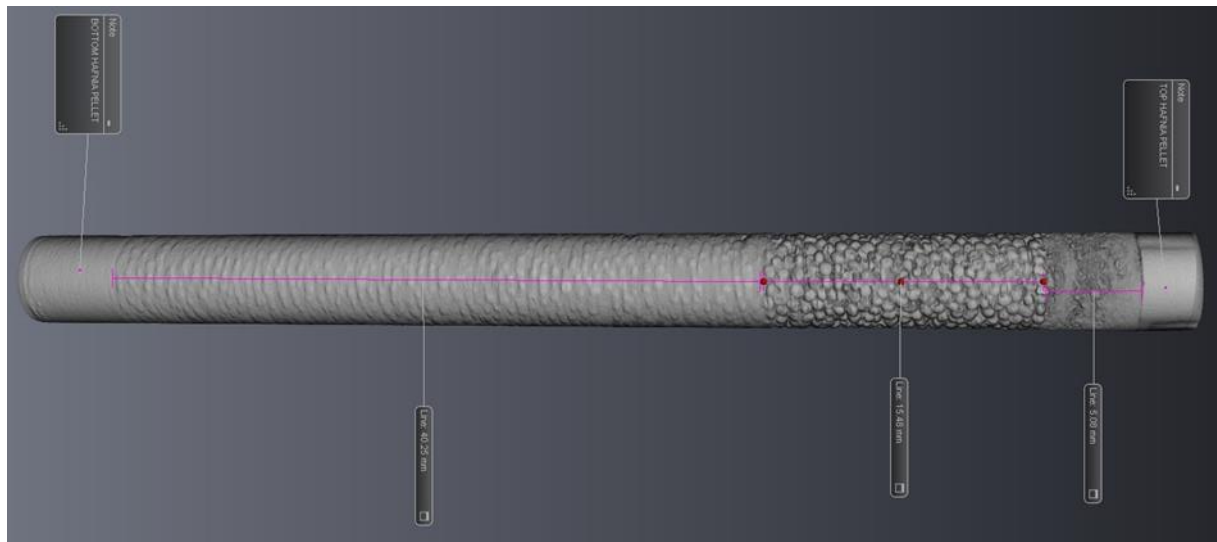


Figure 2.3 X-ray tomography image, made by JRC to show the difference in fuel smear density along the fuel column

3 Post irradiation nuclear analysis

3.1 Spectrum adjustments

MARINE was equipped with six activation monitor sets, located at two axial positions near the centre of the two fuel columns. These monitor sets consist of small samples of Co, Nb, Ti and Fe, which have dominant nuclear reactions with neutrons of a certain energy resulting in their activation. After the activation monitor sets were retrieved during dismantling and their activities were measured, the calculated reaction rates of the various elements were reconstructed and compared to a coupled MCNP simulation and FISPACT activation analysis [5]. Table 3.1 shows the E/C of the dominant reactions for the thermal part and fast part of the neutron spectrum, i.e. ratio of experimentally deduced reaction rates (E) and reaction rates calculated (C) through MCNP/FISPACT, which are used for renormalization of the neutron spectrum at the height of both pins.

Table 3.1 E/C for reaction rates in MARINE

	$^{59}\text{Co} (n,\gamma)^{60}\text{Co}$ (thermal)	$^{54}\text{Fe} (n,p)^{54}\text{Mn}$ (fast)
FD4: Top, North	1.11	1.17
FD5: Top, South East	1.03	0.95
FD6: Top, South West	1.07	0.97
FD1: Bottom, North	1.05	1.15
FD2: Bottom, South East	1.02	0.93
FD3: Bottom, South West	1.05	0.94
Average value for FD4, FD5 and FD6 (Top pin)	1.07	1.03
Average value for FD1, FD2 and FD3 (Bottom pin)	1.04	1.01

3.2 Fuel power history

The total sample-average nuclear power (photons and neutrons contributions, fission included) are calculated by MCNP and FISPACT, keeping track of the depletion and activation of the nuclear fuels during the course of 12 irradiation cycles in the HFR (359 full power days in a run-time of 469 days). The resulting (fuel pin averaged) power histories of MARINE pin 1 (bearing pellet fuel) and pin 2 (Sphere-Pac fuel) are presented in Figure 3.1 and Figure 3.2, respectively. In these figures, also the expected power history, calculated prior to irradiation is shown for comparison. These pre-irradiation calculations are carried out with more generic parameters, i.e. with a single core configuration for each cycle and 90 day cycles (representing 3 consecutive 30-day cycles), followed by a 30-day stop.

The fuel stacks of both pins were divided into three equal parts to see if an axial gradient in the heating would be visible across the fuel stacks. This resulted in variations that statistically were not significant enough. For further analysis, it is therefore assumed that the linear heat rate across the fuel stacks is roughly uniform. The increase of linear heating during irradiation is shown in Figure 3.3.

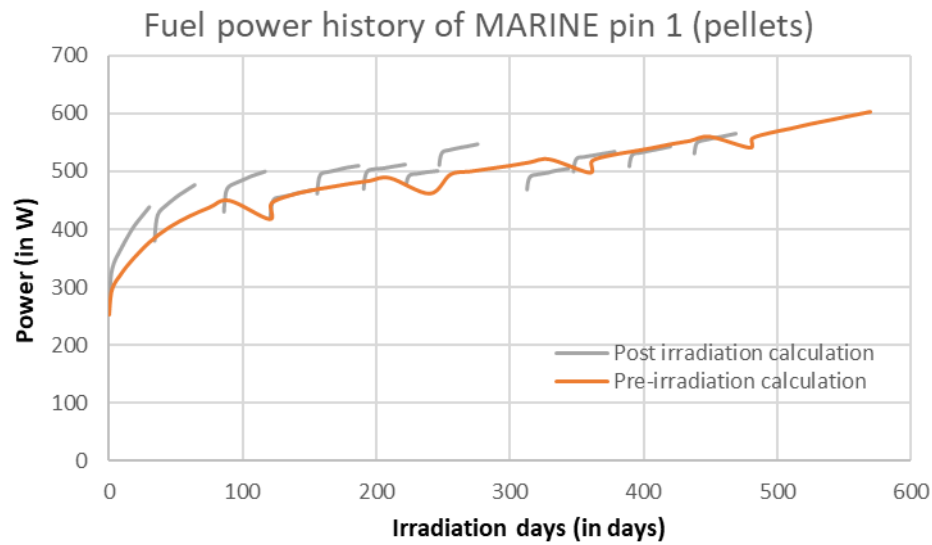


Figure 3.1 The fuel power history calculated prior to irradiation (pre) and after irradiation (post) with spectrum corrections applied for MARINE pin 1.

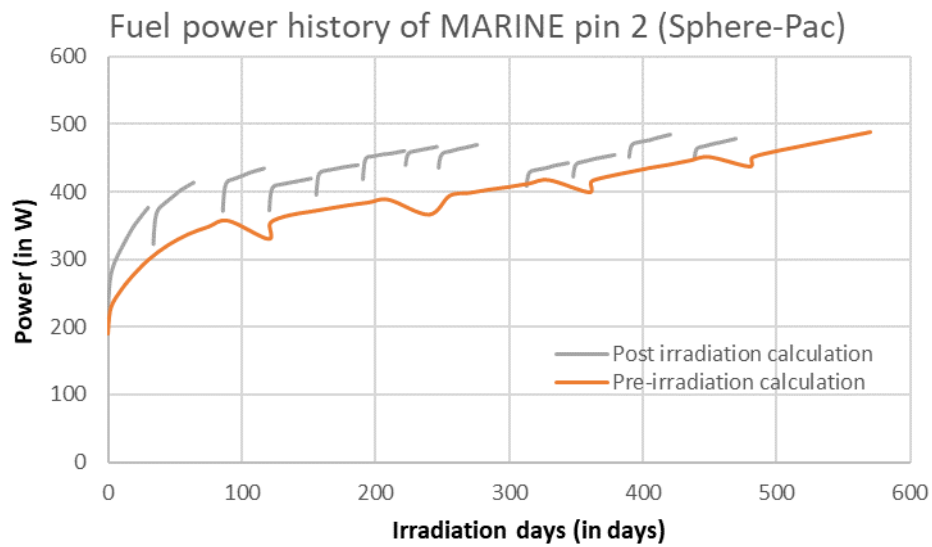


Figure 3.2 The fuel power history calculated prior to irradiation (pre) and after irradiation (post) with spectrum corrections applied for MARINE pin 2.

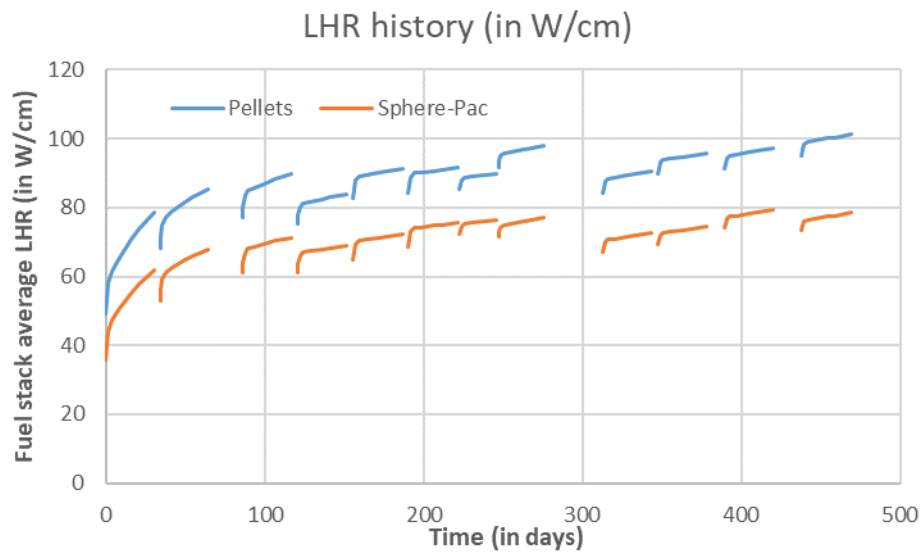


Figure 3.3 The linear heat rate of both fuel stacks during irradiation. The nuclear analysis did not yield statistically significant axial gradients across the fuel stacks

The pin average burn-up is calculated to be 1.98% and 1.57% FIMA (fissions per initial metal atom) for pin 2 and pin 1, respectively. Table 3.2 lists the main actinides before and after irradiation to assess the efficiency of Am-burning, e.g. directly comparing the Am-241 contents at BOI and EOI shows an Americium-241 reduction of 55% and 64% in pin1 and pin 2, respectively. The overall effect of the 'burning' still needs to be assessed, e.g. the Pu-242 contents significantly increased, when looking at the numbers in the same table.

Table 3.2 Selection of actinide contents in fuel at BOI and EOI in atoms/cc as used in / obtained from the nuclear analyses

	Pin1 (pellet)		Pin 2 (SpherePac)	
	atoms/cc		atoms/cc	
	BOI	EOI	BOI	EOI
U234	8.2E+17	3.8E+18	6.6E+17	2.5E+18
U235	1.4E+20	7.7E+19	8.3E+19	4.0E+19
U238	2.0E+22	2.0E+22	1.2E+22	1.1E+22
Pu238	1.5E+18	7.1E+20	8.5E+17	4.6E+20
Pu239	3.9E+19	2.3E+20	2.2E+19	1.6E+20
Pu240	1.8E+19	3.7E+19	1.0E+19	2.8E+19
Pu241	1.1E+19	2.3E+19	6.4E+18	1.6E+19
Pu242	4.5E+18	2.4E+20	2.5E+18	1.6E+20
Np237	2.2E+20	1.7E+20	1.3E+20	8.9E+19
Am241	3.1E+21	1.4E+21	1.8E+21	6.4E+20

3.3 Gas production

Besides the evolution of the generated fuel power, also the production of activation and fission products is calculated in the same MCNP-FISPACT routine.

Both pins were filled with 100% He at a pressure of 2.75 bars during assembly of the experiment. Based on the gas production numbers, a.o. presented in Figure 3.4 and Figure 3.5, an upper limit can be put on the internal pressure in the pins at the end of irradiation (EOI) and at the moment of fuel pin puncturing. It is assumed, however, that gas in the fuel at EOI and gas produced after EOI is retained and does not contribute to the internal plenum pressure. In Section 7, the assumptions and the relations on He and fission gas release after EOI are discussed in more detail.

Table 3.3 presents the maximum amount of gas in the plenum of each pin, taking into account the fill pressure and assuming total release of the produced gas, at 3 years after the end of irradiation (EOI+3yrs). From this conservative amount of released gas, an upper limit on the internal pin pressure is calculated.

Table 3.4 gives the same information as the previous table, but at the end of irradiation, which is considered the more realistic estimation because He produced after the end of irradiation is trapped in the cold fuel.

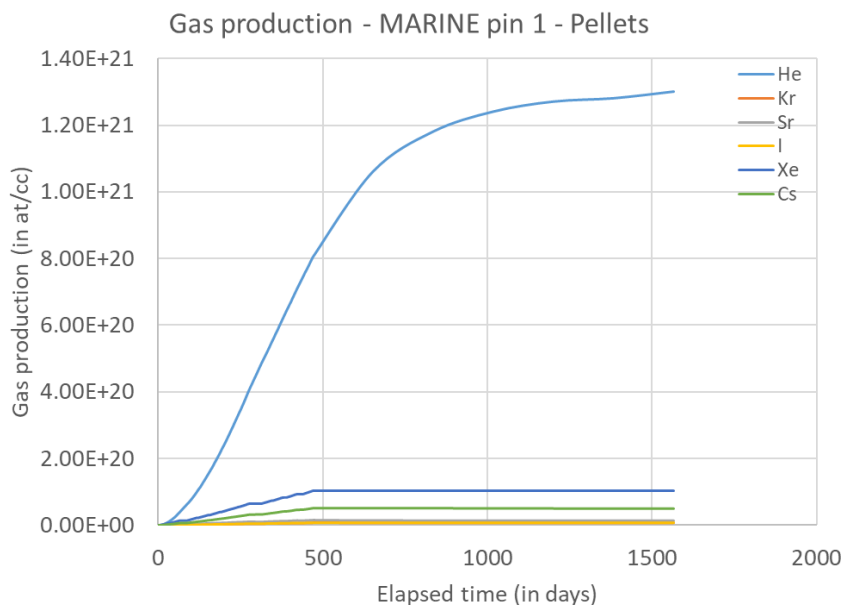


Figure 3.4 The production of gaseous and volatile fission products in MARINE pin 1 during irradiation and the following period of cooling

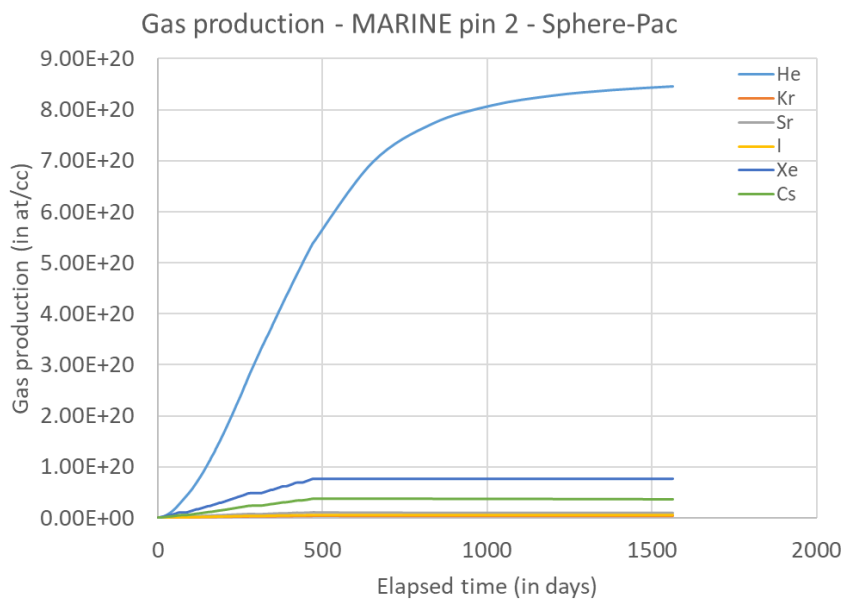


Figure 3.5 The production of gaseous and volatile fission products in MARINE pin 2 during irradiation and the following period of cooling

Table 3.3 Calculated gas production amount from filling and production (at EOI + 3 years), with a maximum estimated pressure build-up in case of total release of gaseous products to the pins' plenums

	total gas			He		Kr		Xe	
	# atoms	Free volume (cc)	Maximum internal pressure (bara)	# atoms	relative	# atoms	relative	# atoms	relative
Pin 1	2.00E+21	3.28	24.64	1.86E+21	93.2%	8.08E+18	0.4%	1.3E+20	6.4%
Pin 2	1.67E+21	3.77	17.97	1.55E+21	92.6%	7.16E+18	0.4%	1.2E+20	7.0%

Table 3.4 Calculated gas production amount from filling and production (at EOI), with a maximum estimated pressure build-up in case of total release of gaseous products to the pins' plenums

	total gas			He		Kr		Xe	
	# atoms	Free volume (cc)	Maximum internal pressure (bara)	# atoms	relative	# atoms	relative	# atoms	relative
Pin 1	1.37E+21	3.28	16.92	1.23E+21	90.0%	8.19E+18	0.6%	1.3E+20	9.4%
Pin 2	1.20E+21	3.77	12.89	1.08E+21	89.7%	7.25E+18	0.6%	1.2E+20	9.7%

3.4 Estimation of maximum fuel temperatures

In order to interpret the PIE data, it is important to know the fuel conditions at peak power, which occurs at the end of irradiation. While a detailed nuclear analysis was performed after completion of irradiation, this was not done for the thermal-mechanical (TM) analysis. Moreover, physical and

chemical processes in the fuel determine the fuel's thermal conductivity and in particular that of the fuel-clad gap (i.e. gap conductance), which is not taken into account in the TM-model. Also, the thermal conductivity of the sphere-pac fuel column has a relatively large uncertainty.

At a later stage, detailed calculations preferably should be done with dedicated fuel performance codes (FPC).

In order to have a rough indication, however, this section summarizes the main results from the TM analysis performed before irradiation [6] and in particular the resulting temperature profiles as a function of the fuel power. In this case average fission heatings of 108.1 W/cm and 80.0 W/cm are used for pin 1 and 2, respectively, which approximate the heating values from the post irradiation nuclear analysis of 101.3 and 79.5 W/cm for pin 1 and 2 respectively, see Figure 3.3. In the earlier TM-analysis an axial profile of radiation heating was used, which had a maximum of 3.06 W/g, see also e.g. [7].

From this model, the maximum temperatures of the fuel and at cladding outer diameter (OD) are derived, but note that uncertainty exists because of the limited definition of a.o. gap conductance and fuel thermal conductivity during irradiation. The results are shown in Table 3.5.

Table 3.5 Estimations of maximum temperatures at cladding OD and fuel centre, derived from pre-irradiation calculations

	Maximum temperature in last cycle			LHR @EOI [W/cm]
	Thermocouple max/ avg	Cladding OD max	Fuel Centre max	
Pin 1	391 (376)	428	1019	108
Pin 2	379 (367)	413	1066	80.1

4 Fuel stack elongation

Neutron radiograms were recorded in the Pool-Side Facility of the High Flux Reactor (HFR) on CN85 nitrocellulose film sandwiched between BN1 natural boron plates, providing an image resolution of 170 μm . After etching of the CN85 film in a NaOH bath, digital images were obtained by scanning the film at a pixel size of 50 μm using a microdensitometer. The pellet height analysis was carried out using the ImageJ software[8].

Neutron radiography images were produced before irradiation, i.e. at beginning of irradiation (BOI), and at the end of irradiation (EOI). Figure 4.1 and Figure 4.2 show the full images and a detail of pin 1 with analysis markings at BOI, respectively.

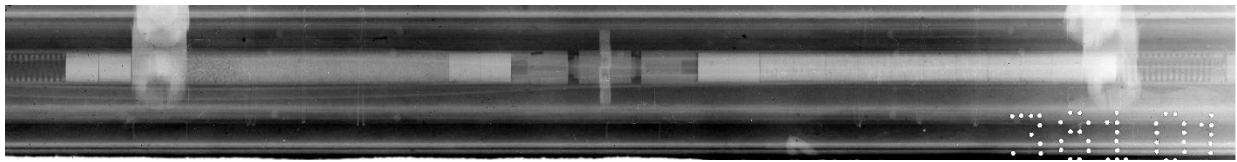


Figure 4.1 The full neutron radiogram of the MARINE pins at begin of irradiation

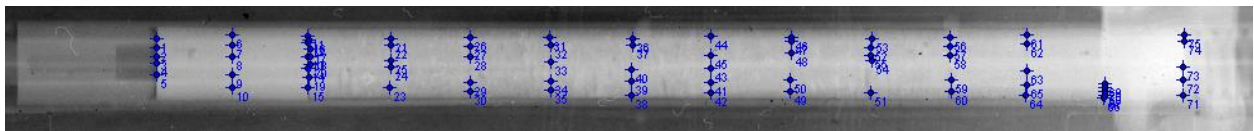


Figure 4.2 Close-up of the fuel column at BOI with the pellet edges marked during analysis in ImageJ

Table 4.1 and Figure 4.3 show the hafnia and fuel pellet heights as provided in the fabrication report and the heights from the analysis of the neutron radiographs. The pellet edges in the BOI- and EOI- neutron radiographs were marked 5 times to get a sense of accuracy on the measurement. This standard deviation of these five markers is also shown in the table (column *SD*). The standard deviation on the EOI-measurements are substantially higher because of the lower quality of the neutron radiography image.

Table 4.1 Pellet heights as indicated in the fabrication report [4], and as measured by analysis of the neutron radiographs at beginning and at end of irradiation

Fabrication report		BOI n-graphy		EOI n-graphy	
Pellet	Height (mm)	Height (mm)	SD (mm)	Height (mm)	SD (mm)
Hf4	5.93	5.906	0.039	5.966	0.033
Hf3	5.97	5.926	0.039	5.970	0.038
Pellet 2	6.288	6.299	0.033	6.554	0.071
Pellet 3	6.275	6.234	0.030	6.386	0.090
Pellet 4	6.193	6.247	0.045	6.448	0.109
Pellet 5	6.296	6.338	0.047	6.445	0.092
Pellet 6	6.155	6.162	0.027	6.289	0.061
Pellet 7	6.237	6.214	0.031	6.658	0.063
Pellet 8	6.128	6.237	0.033	6.272	0.033
Pellet 9	6.069	6.135	0.067	6.435	0.060
Pellet 10	6.031	5.978	0.081	6.181	0.067
Hf2	6.36	6.100	0.066	6.201	0.082
Hf1	6.24	6.227	0.067	6.213	0.078
Total Fuel Stack	55.672	55.844	0.142	57.668	0.225

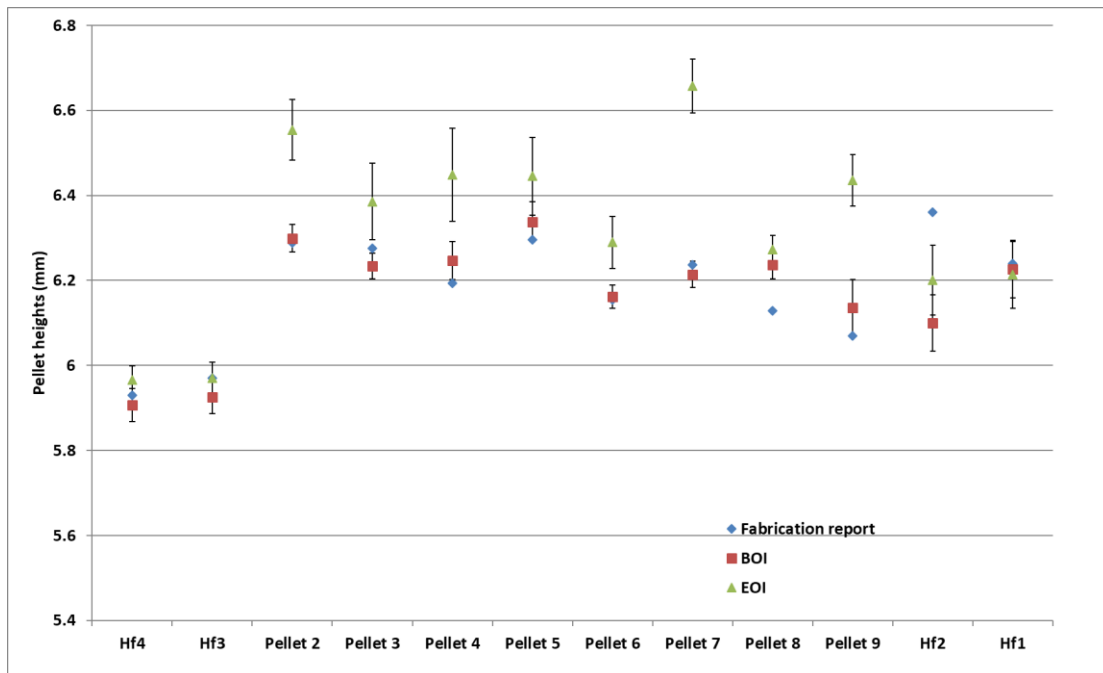


Figure 4.3 Pellet heights as indicated in the fabrication report [4], and as measured by analysis of the neutron radiographs at beginning and at end of irradiation (BOI and EOI)

The fuel stack height shows an apparent increase of approximately 3.3% by comparison of BOI and EOI neutron radiographs.

A second estimation of the fuel stack length is made, based on axial gamma spectrometry: 58.51 ± 0.05 mm. This corresponds to a stack height increase of 5.1% compared to the fabrication report [4] and 4.8% compared to the BOI neutron radiography data.

5 Profilometry – clad strain

The fuel pins were placed on a displacement bench, placed near the rear wall of the hot cell. The axial positioning has a reach of $\sim 1200\text{mm}$ and is controlled by means of a magnetic Vernier system. The smallest axial displacement amounts to $0.10 \pm 0.01\text{ mm}$ for each position. The smallest rotation step is 0.1° .

The setup is equipped with a pair of adjacently placed, pneumatically activated gauges that measure a sample's thickness. The two gauges have a combined systematic uncertainty of $\sim 1.5\mu\text{m}$. The left hand side of Figure 5.1 shows an example of a fuel pin loaded into the setup for profilometry. The step size, is set by setting the step size of the displacement table.

At this point, the MARINE pins were dismantled, with the (closed) gas tube and pressure transducer still attached. The right hand side of Figure 5.1 shows MARINE pin 1 clamped in the chuck, ready to be placed onto the setup.

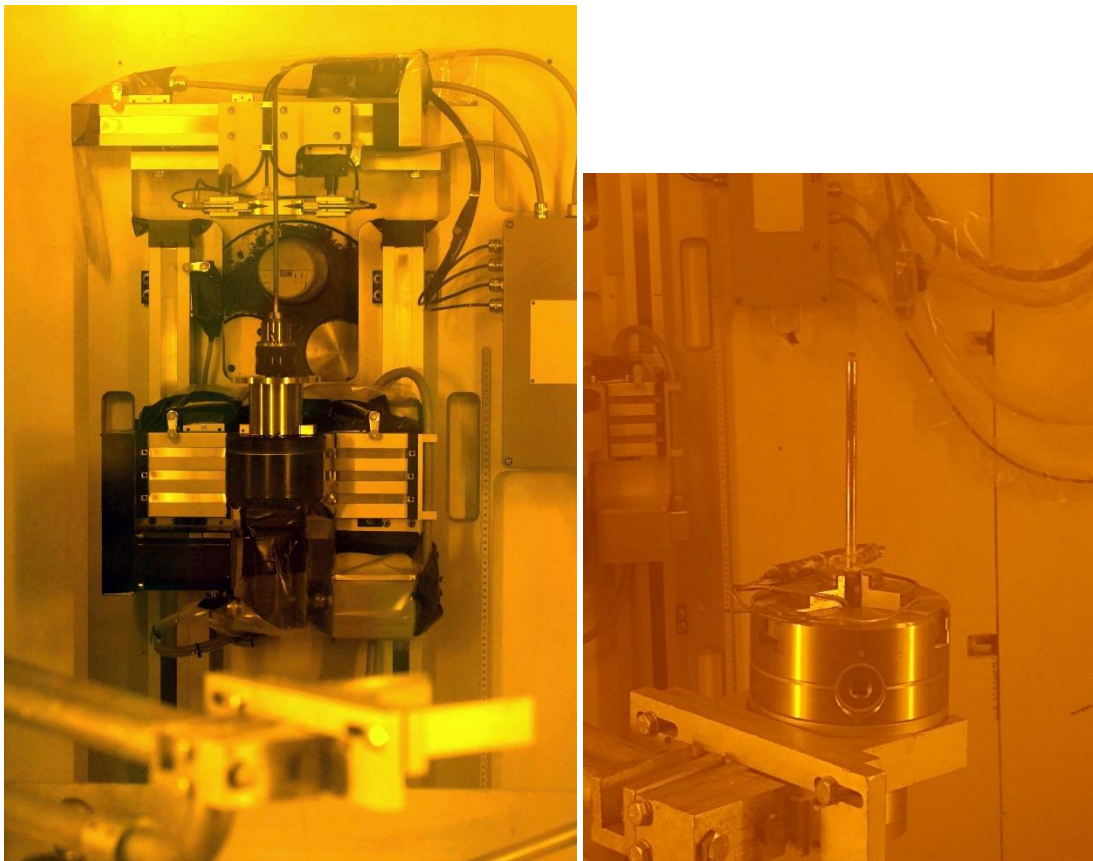


Figure 5.1 Left: the setup with a fuel pin (not MARINE) loaded for profilometry. Right: MARINE pin1 clamped in the chuck, ready to be loaded onto the setup.

The results of the profilometry are described in more detail in [9].

5.1 MARINE Pin 1

Figure 5.2 shows results of axial diameter scans along fuel pin 1, at three independent angles. The plot also shows a schematic of the fuel pin to scale and markers to indicate the location on the pin. The plenum region at the height of the cladding shows a relatively flat profile with diameter between 6.549mm and 6.555mm, depending on rotational orientation of the pin. At the height of the fuel column, the diameter gradually increases towards the lower end cap to a maximum value of 7-8 μ m compared to the value at the height of the plenum. The direction of increase is consistent with the orientation in the neutron flux field, i.e. direction of increasing linear heat rate (LHR).

The biggest diameter at the weld on the lower end cap is clearly visible, with a value of ~6.60mm, matching the diameter stated in the fabrication report [4]. The end cap weld near the burst disc house, was not within reach of the setup's probes.

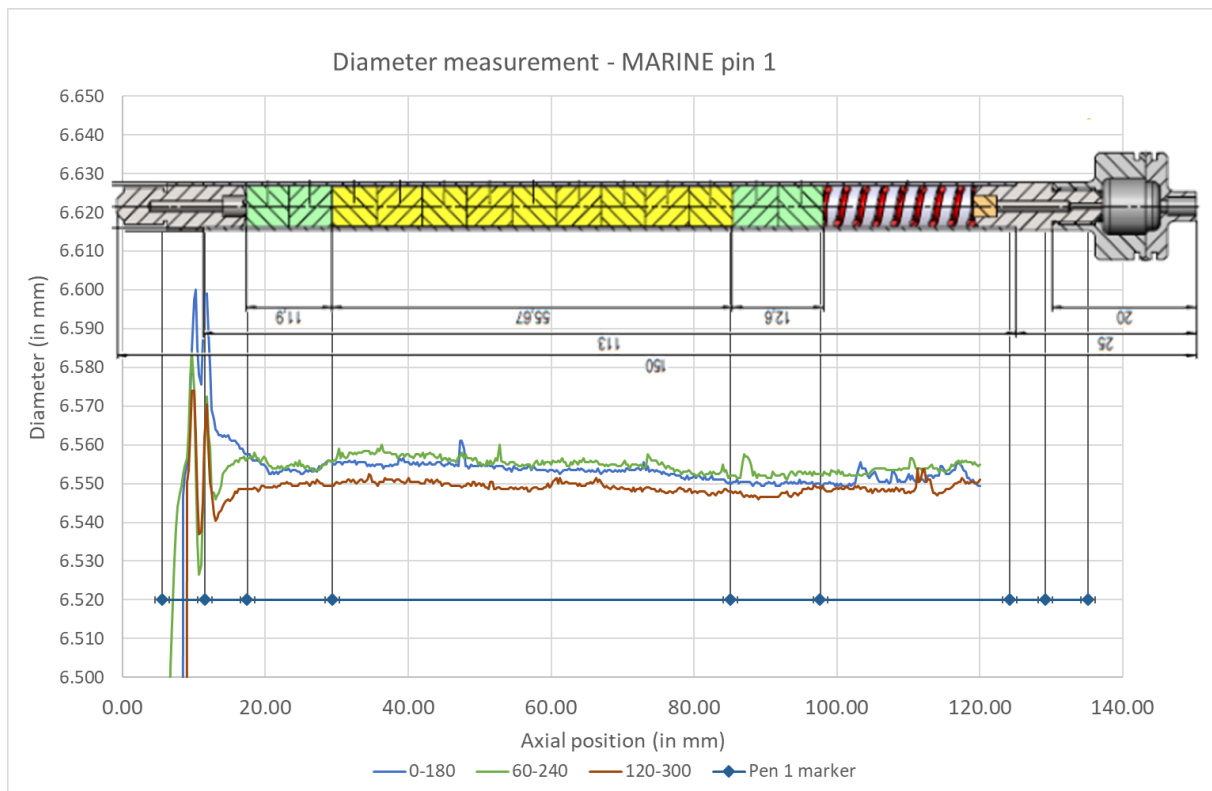


Figure 5.2 Axial scans along pin 1 (pellets) of MARINE at various angles.

With an initial average pellet diameter of 5.368mm, taken from [4] and an initial cladding inner diameter of 5.65mm, the increase of the fuel diameter can be estimated by the closure of the initial fuel-clad gap and the observed increase in clad diameter from profilometry, being $\Delta d = 5.650 - 5.368 + 0.008 = 0.290$ mm. Without known tolerances for the cladding's thickness and inner diameter and using the nominal values, this indicates a maximum relative radial increase of 5.4% .

5.2 MARINE Pin 2

Figure 5.3 shows results of axial diameter scans along fuel pin 2, at several angles. The plot also shows a schematic of the fuel pin to scale and markers to relate the measurement to the location on the pin.

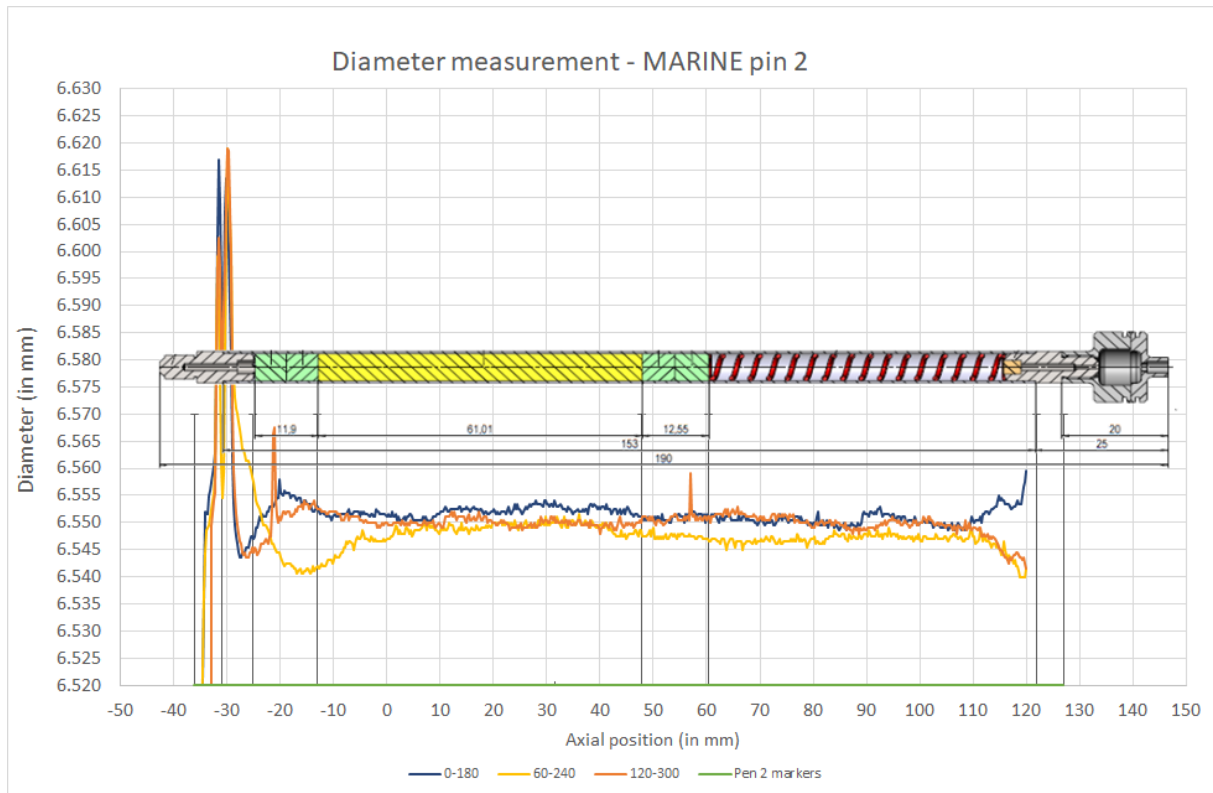


Figure 5.3 Axial scans along pin 2 (Sphere-Pac) of MARINE at various angles.

Beyond the HfO_2 pellets, a slightly increasing trend of the clad diameter is visible and the maximum measured diameter is incidental at 6.554mm, i.e an increase of 4 μm , compared to the clad diameter at the plenum. With Sphere-Pac fuel, less clad strain is expected, compared to pellet fuel, due to its increased porosity (due to stacking of spheres), which accommodate swelling of the fuel.

The bigger diameter at the bottom end cap weld is in line with the value reported in the fabrication report, i.e. 6.62 mm [4].

Near either end of the pin, near the end caps, a slight clad deformation/ ovality is observed, varying between 6.540mm and 6.560mm. The origin of this deformation is assumed to be an effect from the welding, i.e. clamping or stress relaxation during welding.

5.3 Volumetric swelling

Combining the maximum increase in fuel diameter and height, reported in sections 4 and 5.1, an estimation can be made for volumetric swelling. Axial swelling observations vary between 3.3% and 5.1%, while the diametric swelling is 5.4% at maximum, which combine to an upper limit on the volumetric swelling of 16%.

Considering the calculated fuel-stack average burn-up of 1.57%, this gives 10vol% swelling / %FIMA

6 Gamma spectrometry

Before puncturing for the gas analysis, the gamma scanning was performed on the two pins. Results of the gamma spectrometry have been reported in [10].

6.1 Instrumentation

The MARINE fuel pins were analysed using a high purity germanium detector that is placed just behind a collimator system outside the rear wall of the hot cell. A schematic representation of the setup is shown in Figure 6.1. The collimator has a total length of 1.4 m. The slit height is adjustable from 0 to 8 mm and the width is adjustable from 0 to 15 mm.

The fuel pins were placed on a displacement bench near the rear wall of the hot cell. The axial position of the gamma scan transport unit has a reach of $\sim 1200\text{mm}$ and is controlled by means of a magnetic Vernier system. The smallest axial displacement amounts to $0.10 \pm 0.01\text{ mm}$ for each position. The smallest rotational step is 0.1° . The distance of the source (fuel pin) to detector in this setup is 1840mm .

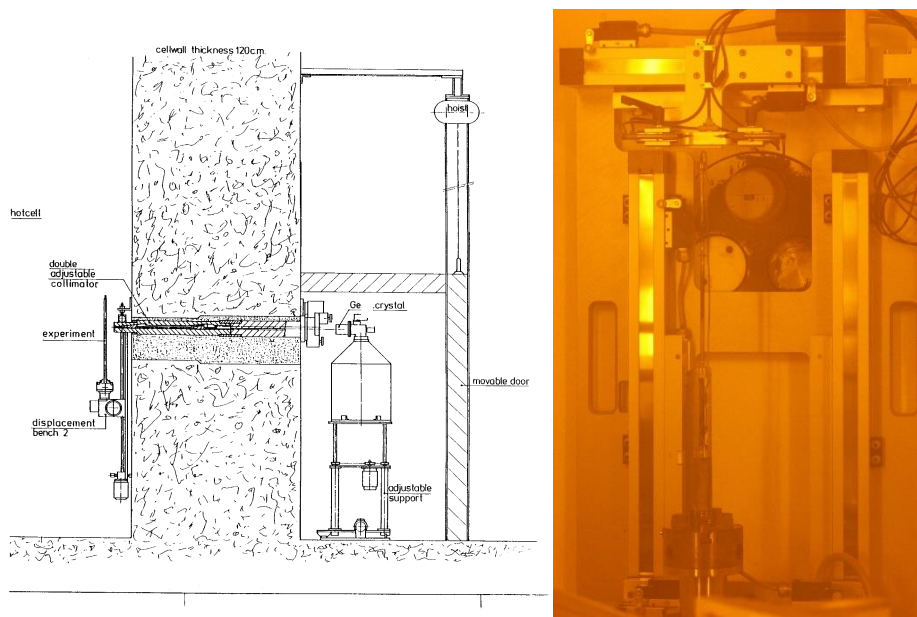


Figure 6.1 Left: Schematic cross section of the gamma spectrometry setup in the D-cell in the HCL. Right: Image of the setup in-cell; MARINE is loaded in the setup. The collimator can be seen in the rear wall.

The collimator dimensions were set at 1mm in height and 15mm in width. The step size for the axial displacement of the pin was 1mm. At each position a scan of 600s (live) was made. For both pins a scan was made over $\sim 110\text{mm}$, from the lower end cap, along the fuel column and a big part of the fuel pin plenum.

6.2 Pin 1 - Pellets

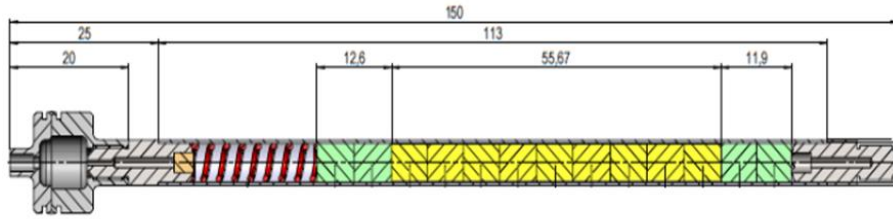


Figure 6.2 This schematic serves only as clarification of the orientation of the various gamma spectra, presented in the following figures

Figure 6.2 shows the schematic of pin 1, as indicator of the orientation of the scans, presented in the following figures. In the following plots the axial heating profile in MARINE's irradiation position is added, which is related to the axial neutron flux profile, for reference. Also the HFR-axial coordinates of the Hf-blanket pellets and fuel stack are shown, taken from the as-built drawing of the experiment. These serve as indicative and might be shifted slightly with respect to the activity profiles due to uncertainties in the coordinates and fuel stack displacement.

Figure 6.3 shows the Am and Cm activity distributions along the pellet stack in MARINE pin 1. A slight decrease in activity towards the max flux plane is observed in the Am-241 activity, though the uncertainty on the reconstructed activity is relatively high for peaks from the low gamma energy regime. At the same time, the Cm-243 distribution is more flat. A slight dispersion of Am-241 beyond the fuel column is observed, deposited on the top Hf-pellets.

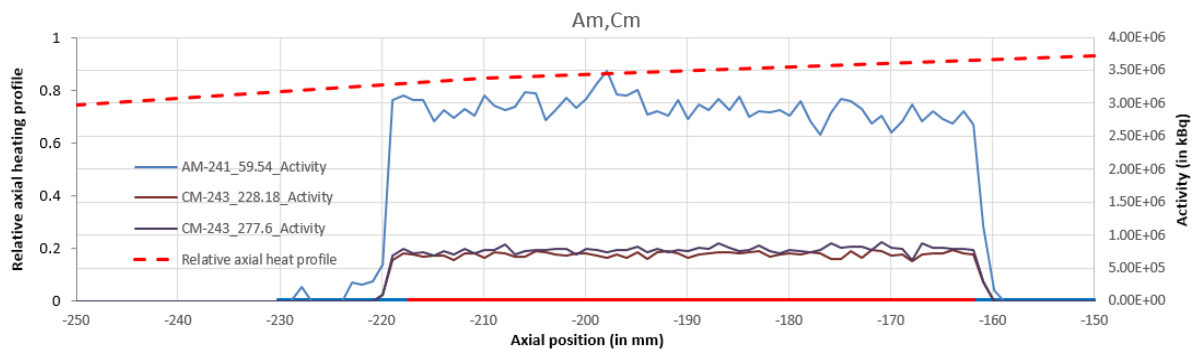


Figure 6.3 The Am and Cm activity distribution along the pellet fuel column of MARINE pin 1. The relative axial heating profile is indicated as a relation to the axial neutron flux profile.

Figure 6.4 shows the Cs-134 and -137 activity distribution in the pellet stack. The 9 individual fuel pellets (each 6mm high) can be distinguished by local dips in the Cs-137 activity. A gradual increase in activity is seen towards the lower end of the fuel column, i.e. the direction of the increasing neutron flux, which is consistent with the decreasing trend observed in the Am-profile. Very limited diffusion of Cs is observed beyond the fuel column, which is understood by the relatively low temperature the fuel experienced during irradiation. Besides this slight diffusion into/onto the Hf-pellet, also Cs-134 is not observed beyond the fuel column. As Cs-134 is dominantly produced by the transmutation of Xe-133, this is also an indicator of limited release of the Xe fission gas.

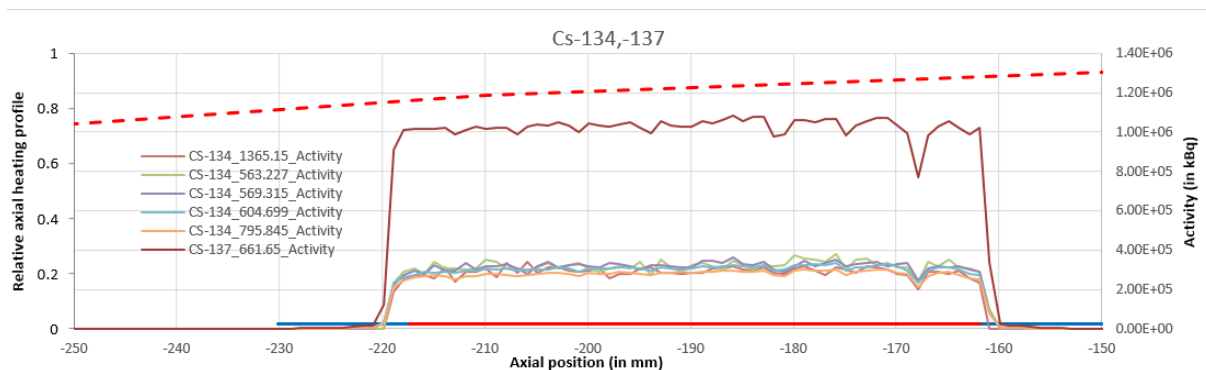


Figure 6.4 The Cs-134 and Cs-137 activity distribution along the pellet fuel column of MARINE pin 1. The relative axial heating profile is indicated as a relation to the axial neutron flux profile.

Figure 6.5 and Figure 6.6 show activity profiles of other identified nuclides, i.e. the non-mobile Ce-144, Ru-106, Sb-125 and Eu-154. The activity profiles are very similar to the Cs-profiles and confirm the slight Linear Heat Rate (LHR) increase towards the lower end of the fuel stack and the limited diffusion of Cs.

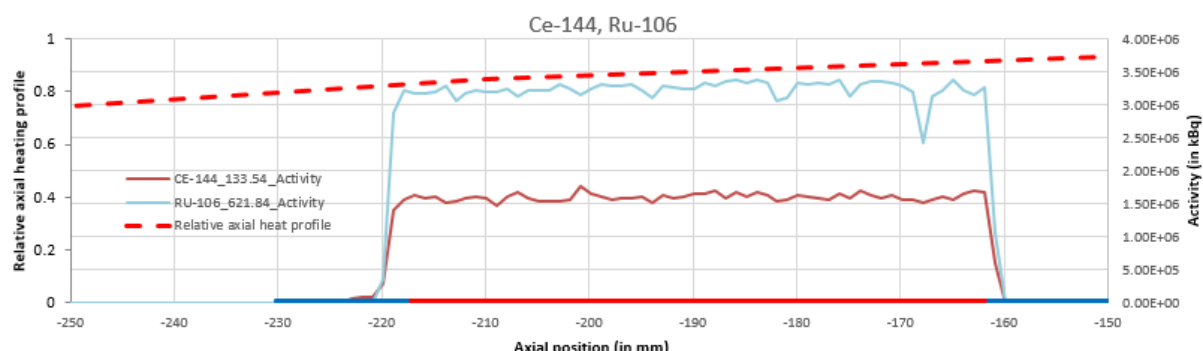


Figure 6.5 The Ce-144 and Ru-106 activity distribution along the pellet fuel column of MARINE pin 1. The relative axial heating profile is indicated as a relation to the axial neutron flux profile.

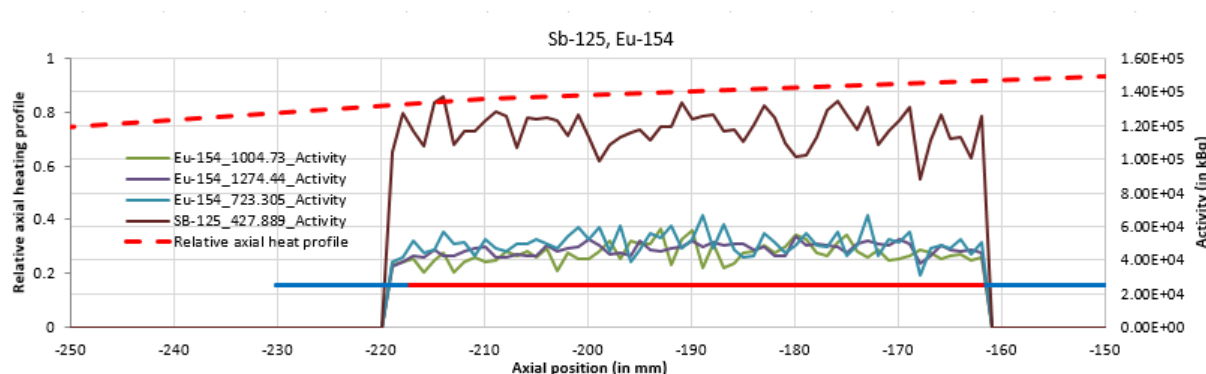


Figure 6.6 The Sb-125 and Eu-154 activity distribution along the pellet fuel column of MARINE pin 1. The relative axial heating profile is indicated as a relation to the axial neutron flux profile.

6.3 Pin 2 – Sphere-Pac

Figure 6.7 shows the schematic of pin 2, as indicator of the orientation of the scans, presented in the following figures. In the following plots, the axial heating profile in MARINE's irradiation position is added, which is related to the axial neutron flux profile, for reference. Also the HFR-axial coordinates of the Hf-blanket pellets and fuel stack are shown, taken from the as-built drawing of the experiment. These serve as indicative and might be shifted slightly with respect to the activity profiles due to uncertainties in the coordinates and fuel stack displacement. The fuel section indications refer to the variation in density that was observed by JRC in the tomography images (see Figure 2.3).

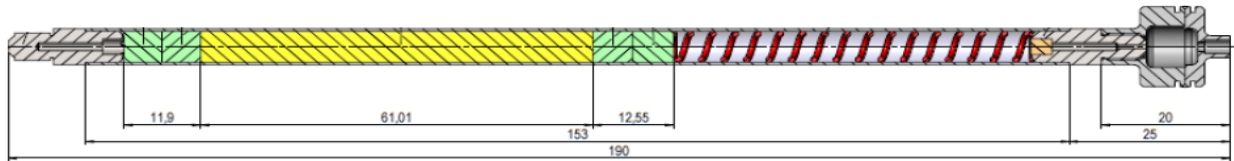


Figure 6.7 This schematic serves only as clarification of the orientation of the various gamma spectra, presented in the following figures

Figure 6.8 shows the distributions of the activities of Am-241 and Cm-243. Am-241 seems to slightly decrease towards the top section of the fuel column, which coincides with the direction towards the max flux plane (indicated by the related heating profile), though uncertainty is generally relatively high with determining activities from low energy gamma emissions. The increasing activity profile of Cm-243 (transmutation product of Am-241) towards the top of the fuel stack also indicates a higher transmutation rate towards the top of the fuel column.

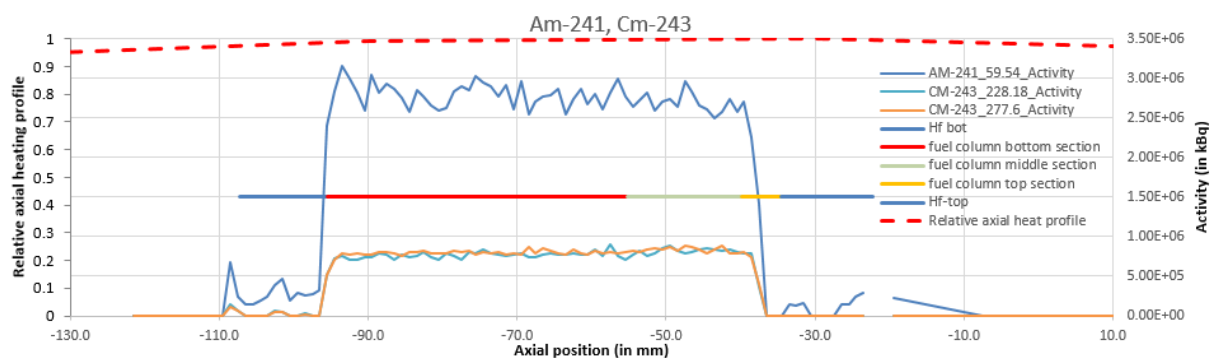


Figure 6.8 The activity distributions of Am-241 and Cm-243 along the Sphere-pac column in MARINE pin 2. The relative axial heating profile is indicated as a relation to the axial neutron flux profile and the fuel columns of various densities are indicated.

The Cs-134 and Cs-137 activity distributions, shown in Figure 6.9, show an increasing activity towards the top of the fuel stack. The Cs-137 activity clearly shows an additional activity increase that coincides with the middle section of the fuel stack which has a relatively low density (see Figure 2.3).

This increased activity is also observed in other fission product distributions (Ru -106 and Eu-154) and is thus not (fully) explained by the volatility of Cs and deposition on locally colder fuel. Also, a potential misinterpretation in the fuel density, would be visible in e.g. Am-Cm profiles.

The limited activity outside the fuel column indicates a fuel temperature which allows only very limited diffusion of Xe-133 into the plenum.

Some activity is found at the axial position of the bottom Hf-pellets. Because also Am (Figure 6.8) , Ce and Ru (Figure 6.10) activities are found there, it is likely explained by some small fuel ('dust') particles located there, rather than diffusion and deposition processes of some volatile products.

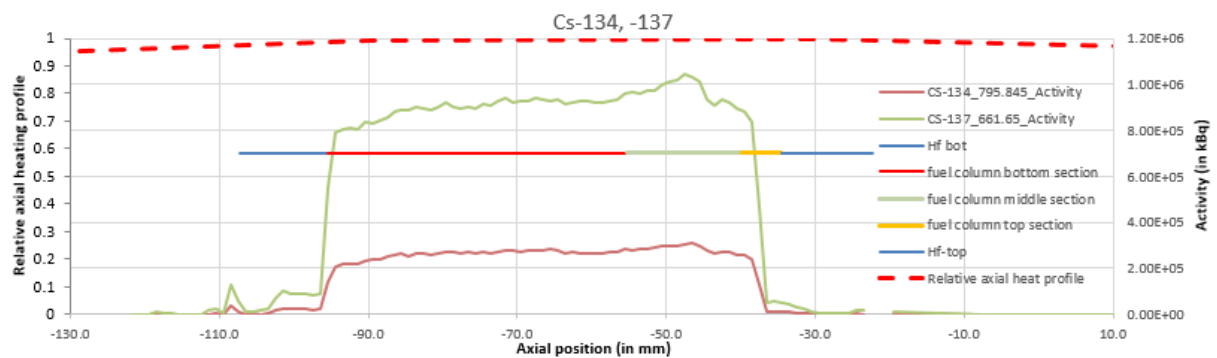


Figure 6.9 The activity distributions of Cs-134 and -137 in the sphere pac pin. The relative axial heating profile is indicated as a relation to the axial neutron flux profile and the fuel columns of various densities are indicated.

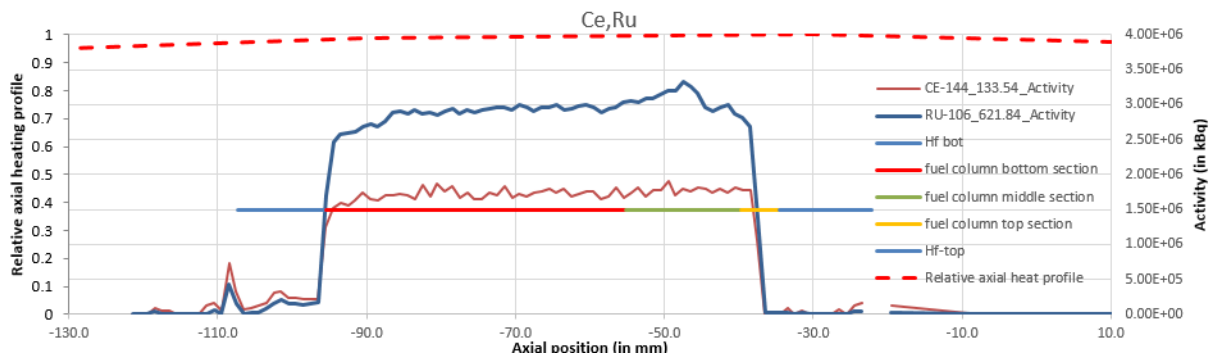


Figure 6.10 The Ce-144 and Ru-106 activity distributions in the Sphere-pac pin. The relative axial heating profile is indicated as a relation to the axial neutron flux profile and the fuel columns of various densities are indicated.

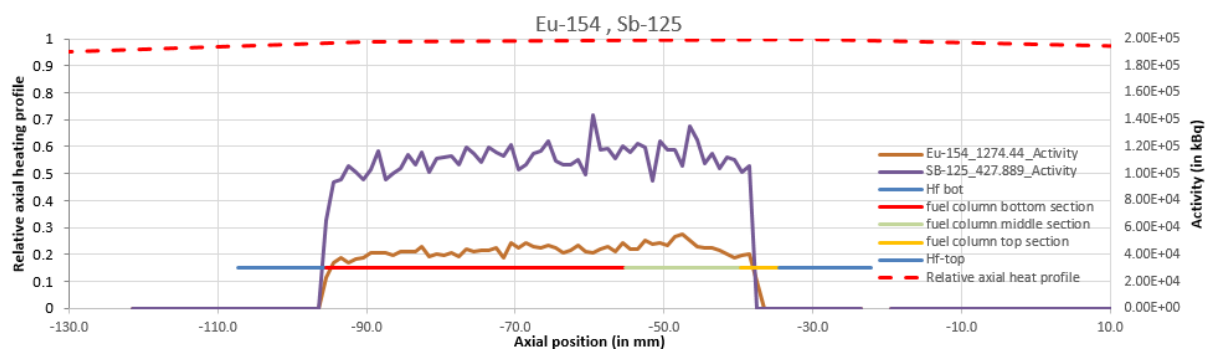


Figure 6.11 The Eu-154 and Sb-125 activity distributions in the Sphere-pac pin. The relative axial heating profile is indicated as a relation to the axial neutron flux profile and the fuel columns of various densities are indicated.

6.4 Stack length elongation based on gamma spectrometry

A coarse estimation of the fuel stack height of pin 1 (pellets) was made by determining the activity distributions and cut-offs of non-mobile fission products in the fuel pellet column.

Using the distributions on Ce-144, Cm-243 and Ru-106, the reconstructed fuel stack length is estimated to be 58.51 ± 0.05 mm. This implies a stack height increase of 5.1% compared to the fabrication report [4] and 4.8% compared to the BOI neutron radiography data.

7 Helium and fission gas analysis

After profilometry and gamma scanning, MARINE fuel pin 1 was punctured in order to reconstruct the internal pin pressure and to analyse the plenum gas composition, i.e. helium and fission gases. A short description of the setup used for puncturing is given below.

7.1 Instrumentation

The puncturing setup has been designed to investigate retention and release of helium and fission gasses in fuel pins, generally from irradiation experiments in the HFR. It consists of a glove box with a number of volumes that can be vacuumed by a turbo molecular pump, the pressure continuously recorded by a number of pressure gauges, see Figure 7.1. Gas can be fed to a Hiden gas mass spectrometer to determine the gas isotopic composition.

The glove box is connected via a gas tube to a chamber that is located in the hot cell, and accommodates the irradiated fuel pin, see Figure 7.2. The chamber can be vacuumed once the pin has been loaded. The chamber carries a spindle with a sharp knife or needle, which is lowered to punch through the fuel cladding. The released gas is expanded into the vacuum system in the glove box where it is stored for mass spectrometric analysis. After puncturing the pin and storage of the plenum's gas, the pin's internal volume can be determined by expanding helium from a well-known volume to the vacuum chamber with the punctured pin.

With expanded pressure and determined plenum volume, the pin's internal pressure (at 3 years after EOI) can be reconstructed.



Figure 7.1 Puncturing setup in the glove box.

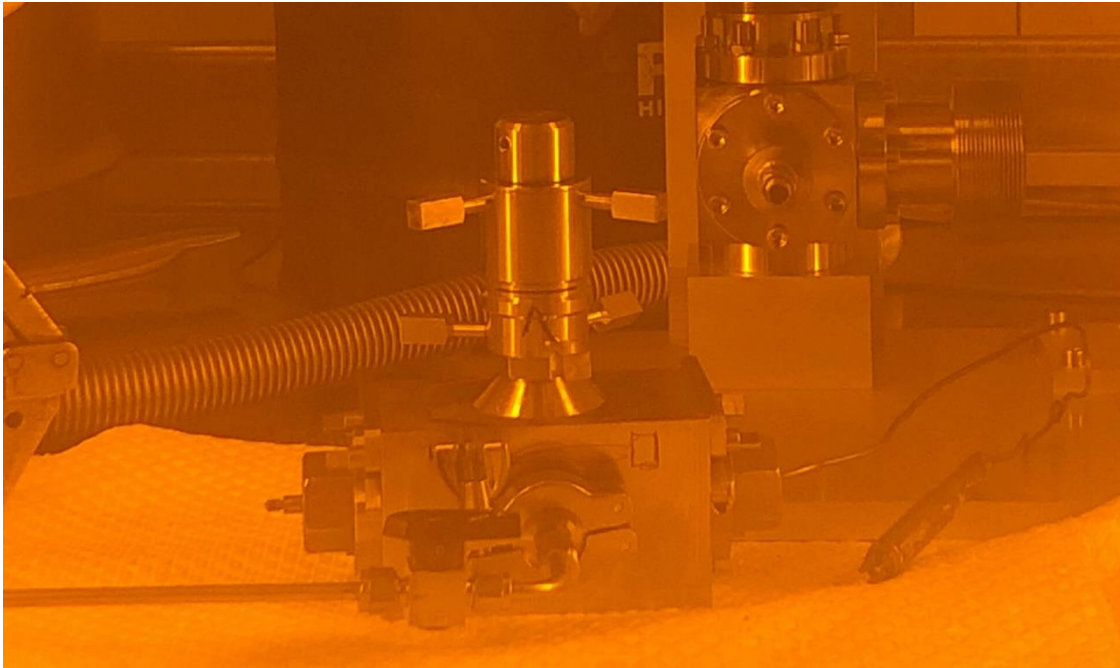


Figure 7.2 MARINE pin 1 loaded into vacuum chamber in the hot cell

The results of the puncturing of the MARINE pins is reported in detail in [11].

7.2 MARINE Pin 1

MARINE pin 1 was punctured on 9 November 2020. With an internal plenum volume of 3.286cc, the internal plenum pressure was reconstructed to be 14.7 ± 0.3 bars (2σ). Figure 7.3 and Figure 7.4 show the calculated history of the gas production and pressures, respectively.

From calculations, summarized in Table 3.3, the maximum internal pressure is estimated to be 24.60 bars assuming 100% release of produced helium and fission gases at the date of puncturing ('EOI+3yrs'). Comparing the calculations and the measured pressure, after deducting the filling pressure of 2.75 bars, between 53% and 56% of the produced gas would then have been released to the pin plenum in this case.

However, assuming the gas produced after EOI is retained in the fuel, due to its cold state, the measured pressure would be effectively the same pressure as at EOI (16.9 bar, assuming full release up to EOI, see

Table 3.4 Table 3.4), in which case the released fraction of the produced gas is between 82% and 87%.

Finally, if it is considered that only He has been released during irradiation (and fission gas release (FGR) is negligible) due to the low fuel temperature, the measured pressure represents 95% to 97% release of the helium produced during irradiation.

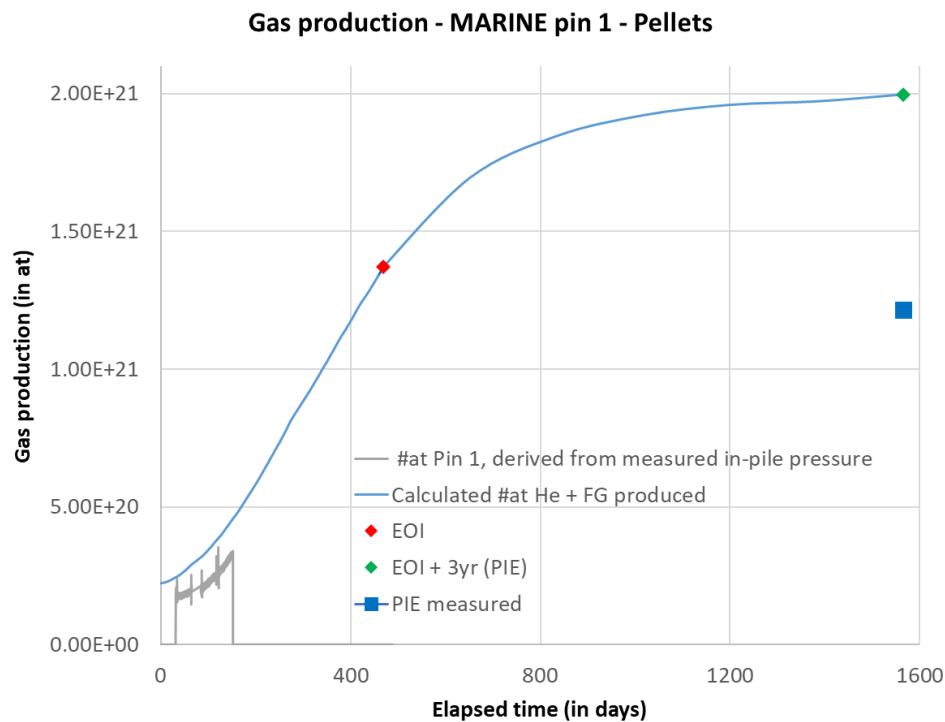


Figure 7.3 The blue line shows the calculated gas production from MARINE pin 1, with EOI (red) and date of PIE (green) indicated. Also the measured pressure at puncturing is shown, converted to # atoms. In grey the in-pile pressure reading is shown, converted to # atoms.

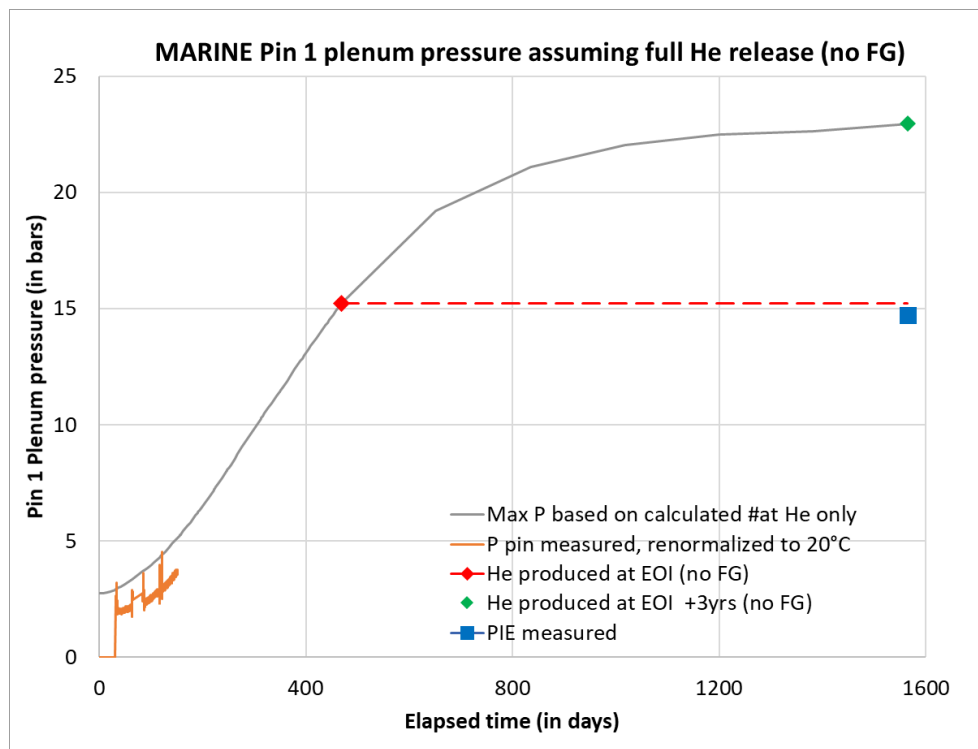


Figure 7.4 The grey line shows the calculated pressure build-up in MARINE pin 1, derived from the gas production amount, with EOI (red) and date of PIE (green) indicated. Also the measured pressure at puncturing is shown. In orange the in-pile pressure reading is shown.

After expanding the plenum gas to the storage volume, mass spectrometry is carried out on small quantities of gas that are taken from the volume and expanded to the mass spectrometer. The measured isotopic vectors of the released Xe and Kr are in good agreement with the predictions from the nuclear analysis at EOI + 3yrs, see Table 7.1.

Table 7.1 Comparison of the measured isotopic vectors of Xe and Kr with mass spectrometry and the results from the nuclear analysis (@EOI+3yrs)

	Mass spectrometry results [11]	Nuclear Analysis [5]
Xe131	11.17%	11.21%
Xe132	19.50%	18.64%
Xe134	25.89%	24.91%
Xe136	43.44%	45.25%
Kr83	16.52%	14.86%
Kr84	31.55%	28.48%
Kr85	5.98%	6.22%
Kr86	45.95%	50.44%

Determining gas species fractions in at least three samples, it is found that the Xe and Kr fractions are 0.25% and 0.03%, respectively, with the remaining 99.7% being He. When converting these partial pressures to number of atoms in the samples and extrapolating to original plenum pressure, this corresponds to released fractions of 94%, 2.3% and 4.6% of He, Xe and Kr, respectively, see Table 7.2.

Table 7.2 Results of gas species fractions in the gas samples and their released fraction, when compared to nuclear analysis

			Fraction in gas sample	Atoms in plenum (extrapolated from measured fractions)	Atoms produced (EOI) [5] & initial filling (2.75 bar He)	Released fraction
He			99.72%	1.16E+21	1.23E+21	93.97%
Kr			0.03%	3.75E+17	8.19E+18	4.58%
Xe			0.25%	2.95E+18	1.28E+20	2.30%
Total measured atoms in plenum, atoms				1.16E+21		
Total measured atoms in plenum, moles				1.93E-03		
Mass-spectrometer/pressure measurements				100.00%		

With a burn-up of ~2.0% FIMA (section 3.2) and an estimated maximum fuel temperature of 1019°C (section 3.4), the expected fission gas release (FGR) seems to be negligible compared to the FGR from PWR UO₂ fuel rods the same burn-up and are certainly less than 5% [12].

Though the fission gas release does not contradict with data of PWR UO₂, irradiated under conditions close to MARINE's (in terms of LHR and temperatures), it should not be assumed this comparison holds when this fuel (Am- bearing blanket) is irradiated to higher burn-up.

7.3 MARINE Pin 2

Puncturing pin 2 has not been performed due to a malfunction of the vacuum chamber's spindle after puncturing of pin 1. At the same time, the in-pile readings of the internal pressure of pin 2 indicated that it was likely to be leaking. It was attempted to confirm this leak by puncturing the complete sample holder, but results were not convincing. If opportunity allows within a reasonable timeframe, it will be tried to confirm this leak in a next campaign with the vacuum setup. Otherwise, the pin will be transported to the hot cell line for destructive analysis.

8 Conclusions

MARINE was successfully irradiated for 359 Effective Full Power Days (EFPD) in 12 cycles in the High Flux Reactor in Petten, between January 5th, 2016 and May 7th, 2017.

The fluence monitor sets, retrieved during dismantling, were measured and reporting of the results has been completed, after which the post irradiation nuclear analysis (or burn-up analysis) was made and has been completed. Burnups of 1.57% and 1.98% FIMA were calculated for pin 1 and pin2, respectively. Comparing Am-241 contents before and after irradiation, these calculations yield a reduction of 55% and 64%, respectively.

Axial gamma spectrometry was carried out, largely showing activity distributions as expected. Slightly increased activities of various species in the supposed low-density fuel section of pin 1 is not yet fully explained.

Clad strain (or profilometry) measurements were carried out on August 17-18, 2020. No irregular strains were observed and maximal diameter increases of 7µm and 4µm were found at the height of the fuel columns of pin 1 (pellets) and pin 2 (Sphere-Pac), respectively. It cannot be distinguished if these increases are strains or e.g. oxide layers on the cladding. Destructive examinations could yield more information on this.

Assuming the strains are related to swelling, a maximum on the volumetric swelling is 16vol% or, in relation to burn up 10vol% / % FIMA. After cutting of the pin and retrieving single pellets, a better measurement on the volume and density could be made.

Finally, pin 1 was punctured and the internal plenum pressure was measured to have been 14.7 bars, converting to a $\sim 86.5 \pm 1.0\%$ (2σ) release of the total gas produced (at end of irradiation), according to the post irradiation nuclear calculations or 96% if only full He-release is assumed.

Mass spectrometry on samples of the retrieved plenum gas shows isotopic composition of Xe and Kr that is consistent with results from the nuclear analysis. The average elemental fractions in the measured gas samples are 99.7% He, 0.25%Xe and 0.03% Kr, which correspond to released fractions from the fuel of 94.3%, 2.3% and 4.6% of He, Xe and Kr, respectively.

The release fractions are consistent with literature values of fuel irradiated under similar conditions (fuel power, temperature and burn-up).

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List of tables

Table 2.1 Fuel and fuel pin specifications	6
Table 3.1 E/C for reaction rates in MARINE	9
Table 3.2 Selection of actinide contents in fuel at BOI and EOI in atoms/cc as used in / obtained from the nuclear analyses	11
Table 3.3 Calculated gas production amount from filling and production (at EOI + 3 years), with a maximum estimated pressure build-up in case of total release of gaseous products to the pins' plenums	13
Table 3.4 Calculated gas production amount from filling and production (at EOI), with a maximum estimated pressure build-up in case of total release of gaseous products to the pins' plenums	13
Table 3.5 Estimations of maximum temperatures at cladding OD and fuel centre, derived from pre-irradiation calculations.....	14
Table 4.1 Pellet heights as indicated in the fabrication report [4], and as measured by analysis of the neutron radiographs at begin and at end of irradiation	15
Table 7.1 Comparison of the measured isotopic vectors of Xe and Kr with mass spectrometry and the results from the nuclear analysis (@EOI+3yrs).....	30
Table 7.2 Results of gas species fractions in the gas samples and their released fraction, when compared to nuclear analysis	30

List of figures

Figure 2.1 Schematic drawings of the two fuel pins	7
Figure 2.2 Schematic drawings of the two fuel pins	7
Figure 2.3 X-ray tomography image, made by JRC to show the difference in fuel smear density along the fuel column	8
Figure 3.1 The fuel power history calculated prior to irradiation (pre) and after irradiation (post) with spectrum corrections applied for MARINE pin 1.	10
Figure 3.2 The fuel power history calculated prior to irradiation (pre) and after irradiation (post) with spectrum corrections applied for MARINE pin 2.	10
Figure 3.3 The linear heat rate of both fuel stacks during irradiation. The nuclear analysis did not yield statistically significant axial gradients across the fuel stacks	11
Figure 3.4 The production of gaseous and volatile fission products in MARINE pin 1 during irradiation and the following period of cooling	12
Figure 3.5 The production of gaseous and volatile fission products in MARINE pin 2 during irradiation and the following period of cooling	13
Figure 4.1 The full neutron radiogram of the MARINE pins at begin of irradiation.....	15
Figure 4.2 Close-up of the fuel column at BOI with the pellet edges marked during analysis in ImageJ.....	15
Figure 4.3 Pellet heights as indicated in the fabrication report [4], and as measured by analysis of the neutron radiographs at begin and at end of irradiation (BOI and EOI).....	16
Figure 5.1 Left: the setup with a fuel pin (not MARINE) loaded for profilometry. Right: MARINE pin1 clamped in the chuck, ready to be loaded onto the setup.....	17
Figure 5.2 Axial scans along pin 1 (pellets) of MARINE at various angles.	18
Figure 5.3 Axial scans along pin 2 (Sphere-Pac) of MARINE at various angles.....	19

Figure 6.1 Left: Schematic cross section of the gamma spectrometry setup in the D-cell in the HCL. Right: Image of the setup in-cell; MARINE is loaded in the setup. The collimator can be seen in the rear wall.	21
Figure 6.2 This schematic serves only as clarification of the orientation of the various gamma spectra, presented in the following figures.....	22
Figure 6.3 The Am and Cm activity distribution along the pellet fuel column of MARINE pin 1. The relative axial heating profile is indicated as a relation to the axial neutron flux profile. ...	22
Figure 6.4 The Cs-134 and Cs-137 activity distribution along the pellet fuel column of MARINE pin 1. The relative axial heating profile is indicated as a relation to the axial neutron flux profile.	23
Figure 6.5 The Ce-144 and Ru-106 activity distribution along the pellet fuel column of MARINE pin 1. The relative axial heating profile is indicated as a relation to the axial neutron flux profile.	23
Figure 6.6 The Sb-125 and Eu-154 activity distribution along the pellet fuel column of MARINE pin 1. The relative axial heating profile is indicated as a relation to the axial neutron flux profile.	23
Figure 6.7 This schematic serves only as clarification of the orientation of the various gamma spectra, presented in the following figures.....	24
Figure 6.8 The activity distributions of Am-241 and Cm-243 along the Sphere-pac column in MARINE pin 2. The relative axial heating profile is indicated as a relation to the axial neutron flux profile and the fuel columns of various densities are indicated.	24
Figure 6.9 The activity distributions of Cs-134 and -137 in the sphere pac pin. The relative axial heating profile is indicated as a relation to the axial neutron flux profile and the fuel columns of various densities are indicated.	25
Figure 6.10 The Ce-144 and Ru-106 activity distributions in the Sphere-pac pin. The relative axial heating profile is indicated as a relation to the axial neutron flux profile and the fuel columns of various densities are indicated.	25
Figure 6.11 The Eu-154 and Sb-125 activity distributions in the Sphere-pac pin. The relative axial heating profile is indicated as a relation to the axial neutron flux profile and the fuel columns of various densities are indicated.	25
Figure 7.1 Puncturing setup in the glove box.	27
Figure 7.2 MARINE pin 1 loaded into vacuum chamber in the hot cell.....	28
Figure 7.3 The blue line shows the calculated gas production from MARINE pin 1, with EOI (red) and date of PIE (green) indicated. Also the measured pressure at puncturing is shown, converted to # atoms. In grey the in-pile pressure reading is shown, converted to # atoms. .	29
Figure 7.4 The grey line shows the calculated pressure build-up in MARINE pin 1, derived from the gas production amount, with EOI (red) and date of PIE (green) indicated. Also the measured pressure at puncturing is shown. In orange the in-pile pressure reading is shown.	29