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D4.2 Microstructural and chemical evolution

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DOCUMENT SUMMARY

Results from destructive examinations on irradiated fuel pins of the MARINE experiment. This includes fuel pin cutting and preparation of samples for optical and electron microscopy. SEM images and radial actinide distributions are presented.

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Summary

The goal of the MARINE irradiation was 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 was made between the pellet fuel (pin 1) and sphere-pac fuel (pin 2) forms in order to assess their performance.

MARINE was successfully irradiated for 359 Effective Full Power Days (EFPD) comprising 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. Average 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.

Non-destructive testing, including cladding profilometry and axial gamma spectrometry, as well as fuel pin puncturing were completed earlier and reported in PATRICIA deliverable D4.1.

Afterwards, the fuel pins were transported to the hot cell line for destructive analysis and cut to produce a number of specimens for optical microscopy and SEM.

Macroscopic observations show limited swelling of the pellets, without a closure of the fuel-clad gap. This observation is consistent with results, reported in D4.1, i.e. (1) the absence of PCI induced strains observed by profilometry and (2) the total (released) gas in the plenum found with puncturing of the fuel pin.

SEM images on the pellet fuel specimens reveal increased porosity surrounding particles with dimensions in the order of the UO_2 beads ($\sim 200\text{ }\mu\text{m}$) that were pressed and sintered during fabrication. Finally, in the case of higher magnifications of SEM images, it was possible to distinguish individual grains which were of order of $10\text{-}20\text{ }\mu\text{m}$.

Generally, the concentrations of uranium, plutonium and americium, obtained from fuel pellet specimens by SEM EDS, agree well with the predicted composition as calculated by FISPACT, which confirms that about half of the americium has been transmuted. The heterogeneous nature of the fuel is reflected in areas of increased Am-content of up to 10 wt%HM and the profile and quantity is accurately followed by its main transmutation product Pu across largest part of the fuel radius. Only towards the edge of the fuel pellet, roughly from 0.7R to 1R, does the Pu-content start to deviate from the Am-profile with the Pu/Am ratio gradually increasing to a factor of 2, where the thermal neutron flux was highest.

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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 was 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 a 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 was 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) comprising 12 cycles in the High Flux Reactor in Petten, between January 5th, 2016 and May 7th, 2017. More information is available in the dedicated irradiation report [3].

The post irradiation nuclear analysis (or burn-up analysis), dismantling of the experiment, clad strain measurements, gamma scanning and plenum gas measurements (fuel pin puncturing) on the fuel pins were completed and reported earlier in PATRICIA deliverable D4.1 [9].

After, fuel pins were transported to the hot cell line for destructive analysis and cut to produce a number of radial specimens for optical microscopy and SEM.

This report contains the summarized results of the destructive post-irradiation examinations.

2 Experimental description

2.1 Fuel pin specifications

MARINE experiment 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. Both fuels were fabricated by JRC-Karlsruhe via two different routes. The pellets were fabricated 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 1 provides details on the two fuel pins. Figure 1 shows the schematic drawings of the pins, including dimensions of the most important features.

Table 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
Bulk fuel density (g/cm ³)	10.46	7.45
Percentage T.D. (%)	94.61	55.36
Pellet O.D. (mm)	5.368	n.a.
Pellet height (mm) (/ # pellets)	6.185 (/ 9)	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

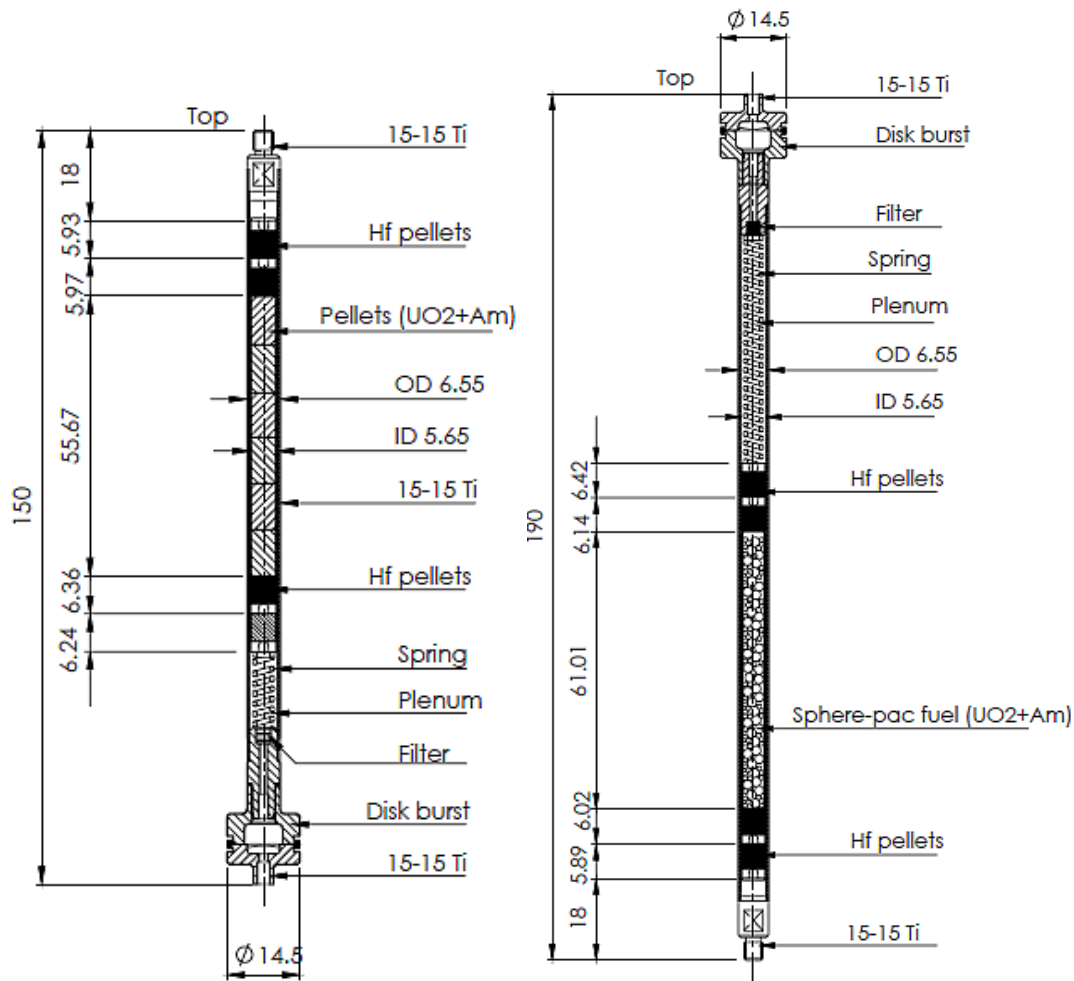


Figure 1 Schematic drawings of the two fuel pins (Left: pin 1 with pellet fuel. Right: pin 2 with Sphere-Pac fuel).

2.2 Estimation of maximum fuel temperatures with 2D ANSYS

In order to interpret the PIE data, it is important to know the fuel conditions at peak power, which in this irradiation experiment occurred at the end of irradiation. An estimate on fuel temperatures at EOI was provided earlier [9] based on nuclear and thermal calculations which were performed during the design phase. However, after a detailed nuclear analysis was performed after completion of irradiation, a review of this calculated temperature profile was done with a (2D) ANSYS thermal-mechanical model, using the (more accurate) final linear heat rating and recorded thermocouple values.

The main results on the power history and irradiation targets, earlier reported in D4.1, are additionally presented in Appendix A.

Material properties such as thermal expansion and thermal conductivity are taken into account and are the same as used during design and safety calculations [7]. This revised calculation is still considered indicative as physical and chemical processes in the fuel during irradiation are not taken into account. An uncertainty remains on the fuel's (changing) thermal conductivity, as in reality processes such as helium production, migration and release, result in altered porosity structures and changes of the composition of the gas in the fuel-clad gap (i.e. gap conductance).

Further, the sphere-pac fuel in pin 2 has variations in bead geometry, stacking density and contact surfaces, each affecting the fuel's overall thermal conductivity. In the used model, this fuel is modelled as a solid with specified bulk fuel density (see e.g. Table 1) with a thermal conductivity for sphere-pac

fuel that was also used during experiment design phase and reported earlier in the associated safety report [7].

The radial temperature profile in fuel pin 1 (and in the sample holder) is shown for BOI- and EOI- conditions in Figure 2, disregarding evolution of the fuel material properties. As the experiment is operated with an actively monitored and controlled thermocouple value of 390°C (pin 1), BOI power conditions of 60 W/cm lead to a fuel centre temperature of 750°C and a fuel edge temperature of 570°C. At EOI, the fuel pellets produced a power of 101 W/cm leading to fuel temperatures of 1004°C and 670°C in the centre and at the edge of the pellet, respectively.

Additionally, a final state is also calculated with a closed fuel-clad gap (representing swelling of the fuel) to have an indication on the effect of gap closure on the fuel's final temperature. In this case, only the fuel diameter has been changed, which doesn't affect the linear heating, though the calculation is carried out with a slightly reduced power density in W/cm³, compared to the EOI-calculation with an initial fuel-clad gap. With the closed gap, a fuel centre temperature of 714°C is reached.

Table 2 summarizes the temperature values in the fuel and cladding for the EOI irradiation conditions, as recorded by thermocouples and as calculated with the 2D ANSYS model. Here the case without gap closure is presented, as this is expected because of absence of clad strains [9]. The sphere-pac fuel, modelled as a porous solid without initial gap, has only a slightly higher estimated central temperature of 1039°C at a final linear heating of 79 W/cm.

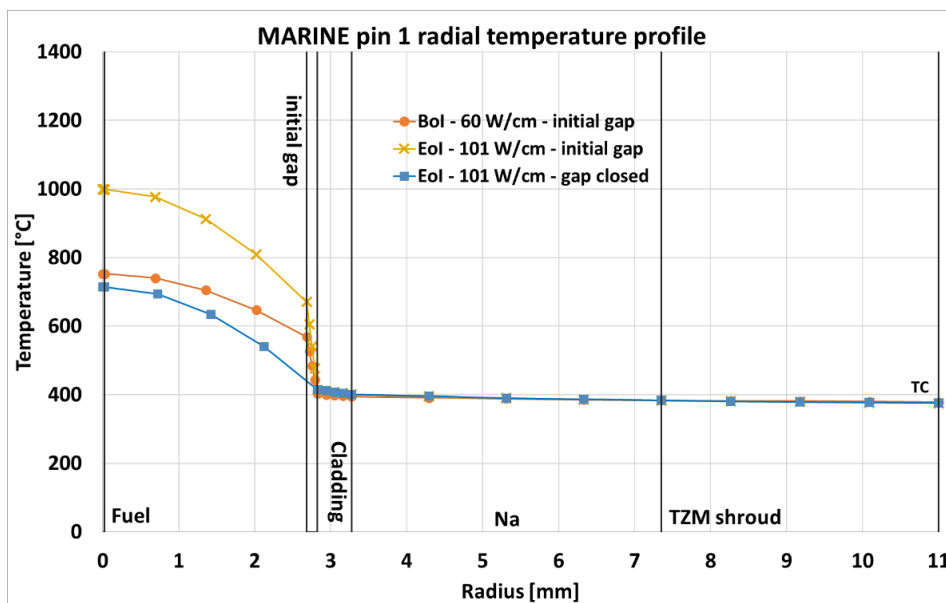


Figure 2 Calculated radial temperature profiles in the fuel pin for different fuel powers with as-fabricated and closed fuel-clad gap, related to the recorded thermocouple (TC) temperatures in the shroud.

Table 2 The measured thermocouple temperatures and calculated temperatures in fuel and cladding with 2D FEM for the last irradiation cycle, based on linear heating from post irradiation nuclear calculations

	Temperatures in last irradiation cycle [°C]			LHR @EOI [W/cm]
	Thermocouple max/ avg	Cladding ID / OD	Fuel Centre / Edge	
Pin 1	391/ 376	419 / 410	1004 / 670	101
Pin 2	379/ 367	409 / 399	1039 / 409	79

More detailed assessments of temperature profiles, taking into account a.o. physical and microstructural changes to the fuel are executed with a range of fuel performance codes (FPC), within task 5.3.

3 Fuel pin cutting

3.1 Fuel pin cutting - pin 1

The fuel pin was cut in order to obtain several radial and axial sections for microstructure and element analysis. Radial samples were cut at a thickness of ~2 mm to reduce the risk of breakout of fuel. A segment with a length of ~8 mm was cut, which was then embedded and cut to obtain an axial pellet interface. The locations of the cuts and samples are indicatively shown in Figure 3 and presented in Table 3.

Initial cuts on pin 1 showed that the fuel pellet(s) have cracked substantially (during irradiation and/or during cutting), see Figure 4. Approximately 5 mm fuel column (almost a full pellet height) fell from the cladding. A small amount of a fixating Epoxy resin was poured into the pin to fix the fuel in place. The broken fuel fragments are collected for future studies (e.g. dissolution).

A resin-fixed radial and axial sample were cut and embedded into a sample holder for further preparation for metallography. The first section (0242-07) contains a negligible amount of fuel, and the second section (axial) shows a piece of emptied clad tube with a cracked pellet (0242-19, produced from 0242-09), Figure 5. Preserving a longer central section of the fuel pin, finally a second radial sample was produced (0242-17). Produced specimens are related to original location/pellet number in the fuel stack, see Table 4.

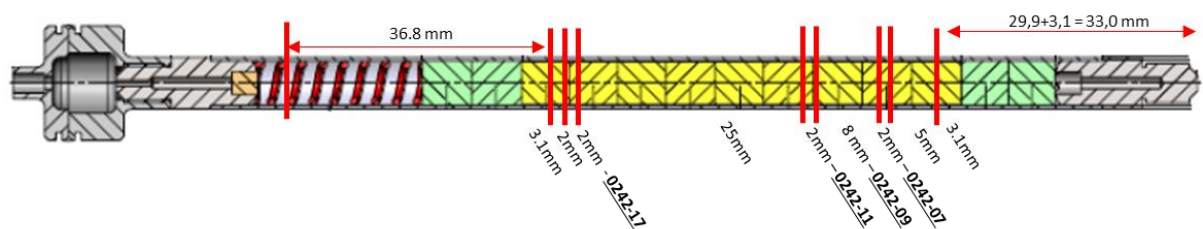


Figure 3 Schematic drawing of MARINE pin 1 with the locations and indicative lengths of the cuts.

Table 3 Location of the produced samples from pin 1 for metallography.

Height from pin bottom (mm)	Cut segment (mm)	Height from bottom of fuel column (BFC) (mm)	Description	sample ID
49.5	8	19.6	axial specimen	0242-09/-19
52.0	2	22.1	radial specimen	0242-11
77.5	25	47.6	middle segment	
80.0	2	50.1	radial specimen	0242-17

Table 4 Fuel specifications from the fabrication report [4] with originating pellets of the metallography samples indicated.

From Fabrication report							
	weight	height	diam	V	rho	TD	Sample
Pellet	g	mm	mm	mm ³	g/cm ³	%	
2	1.49	6.288	5.362	141.98	10.49	94.89	
3	1.471	6.275	5.369	142.05	10.36	93.63	0242-09 / 19
4	1.464	6.193	5.357	139.58	10.49	94.84	0242-09 / 19
5	1.498	6.296	5.373	142.77	10.49	94.87	0242-11
6	1.462	6.155	5.366	139.17	10.5	94.98	
7	1.476	6.237	5.364	140.96	10.47	94.67	
8	1.445	6.128	5.362	138.39	10.44	94.41	
9	1.442	6.069	5.377	137.81	10.46	94.61	0242-17
10	1.434	6.031	5.38	137.08	10.46	94.59	
Average						94.61	

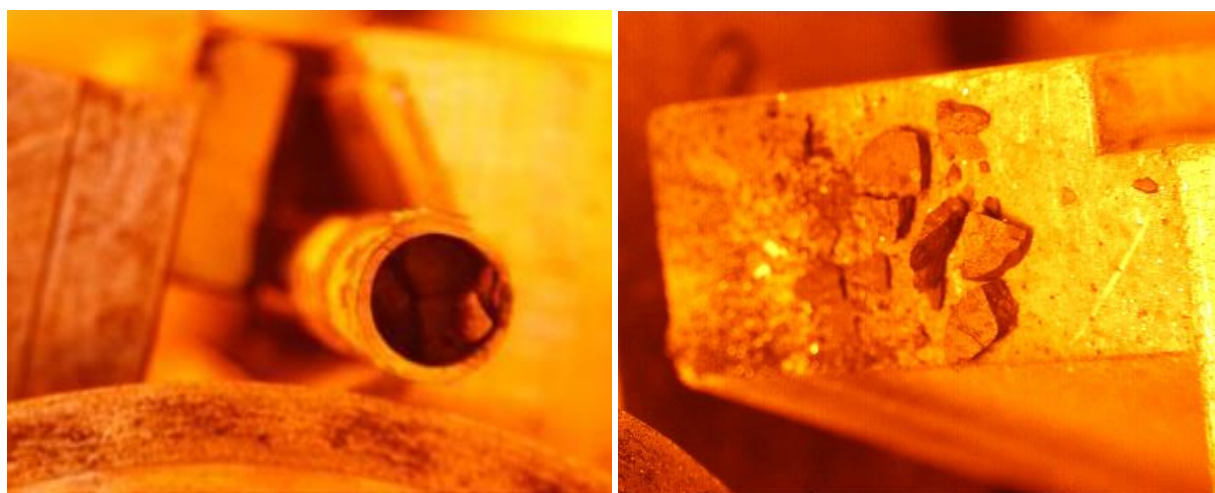


Figure 4 Impression of the first cut (0242-07) on the fuel column with fuel broken out (left). Fragments of the fuel were collected and stored for future dissolution experiments (right).



Figure 5 Impressions of a prepared axial sample (0242-19; left) and of a radial sample (0242-17; right) of pin 1.

3.2 Fuel pin cutting - pin 2

The fuel pin was cut in order to obtain several radial and axial sections for microstructure and element analysis. Radial specimens were cut at a thickness of ~3-4 mm to reduce the risk of breakout of fuel. A segment with a length of ~8 mm was cut (0254-09), which was then embedded and cut to obtain an axial pellet interface (0254-19). The locations of the cuts and samples are indicatively shown in Figure 6 and listed in Table 5.

As the Sphere-Pac fuel was expected to be loosely stacked in the cladding, to fixate it the cladding was cut partially at the locations where samples were to be taken. The goal was to keep these first cuts superficial to affect the fuel as little fuel as possible, Figure 7.

After fixating the fuel by pouring Epofix resin into the cuts, the pin was cut completely and the fuel showed to be fully fixed in the obtained samples. Resin-fixed radial and axial specimens were cut and embedded into a sample holder for further preparation for metallography, Figure 8.

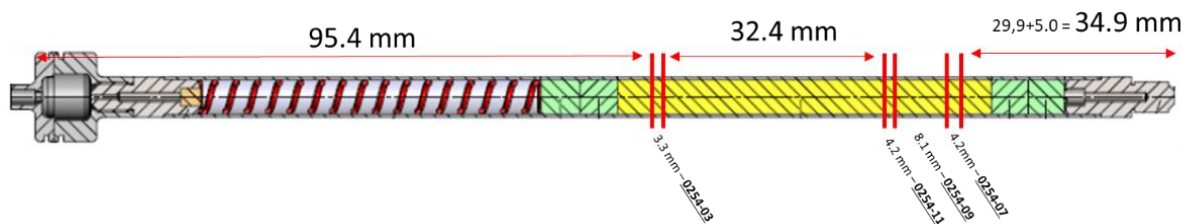


Figure 6 Schematic drawing of MARINE pin 2 with the locations and indicative lengths of the cuts.

Table 5 Location of the produced samples from pin 2 for metallography.

Height from pin bottom (mm)	Cut segment (mm)	Height from bottom of fuel column (mm)	Description	sample ID
39.7	4.2	9.8	radial specimen	0254-07
48.3	8.1	18.4	axial specimen	0254-09/-19
53.0	4.2	23.1	radial specimen	0254-11
85.9	32.4	56	middle segment	0254-12
89.7	3.3	59.8	radial specimen	0254-03



Figure 7 Images of first two cuts of MARINE pin 2, to allow fixing the sphere-pac fuel with resin.



Figure 8 Impression of a prepared radial and axial specimen from pin 2.

3.3 Polishing and coating

A selection of radial and axial cuts was embedded in Epoxy in aluminum sample holders, and ground and polished to a fuel thickness of about 1 mm, using Struers OP-U non dry suspension (0.04 μm colloidal silica) in the last polishing step.

After optical microscopy imaging, a conductive coating (~ 10 nm Au) was applied prior to the SEM analysis, using a JEOL Smart coater (DII-29010SCTR), modified for use in the hot-cell.

4 Ceramography

4.1 Optical microscopy

Images of the radial fuel pin specimens were made using a Leitz MM5 RT optical microscope. Image processing (stitching) was performed with Microsoft Image Composite Editor and dimensional analysis was performed with ImageJ software [8].

Fitting of cladding inner and outer diameter, as well as the fuel outer diameter were done by manual ellipsoid fitting, which have an intrinsic systematic uncertainty of $\sim 2 \mu\text{m}$, based on the original image resolution of $\sim 1000 \text{ px/mm}$.

4.2 MARINE Pin 1 – Macrography

First observations of cut specimens of pin 1 show a pellet with thermal stress induced cracks, as well as substantial porosity and large channels (Figure 9). The fuel-cladding gap has not closed, which agrees with negligible strains observed in profilometry. Basic geometric measurements using elliptical fits, were carried out for pellet and cladding diameters on two radial specimens and show an average diametral increase of the fuel of about 1.1% (Table 6) and a remaining variable fuel-cladding gap of 58-249 μm .

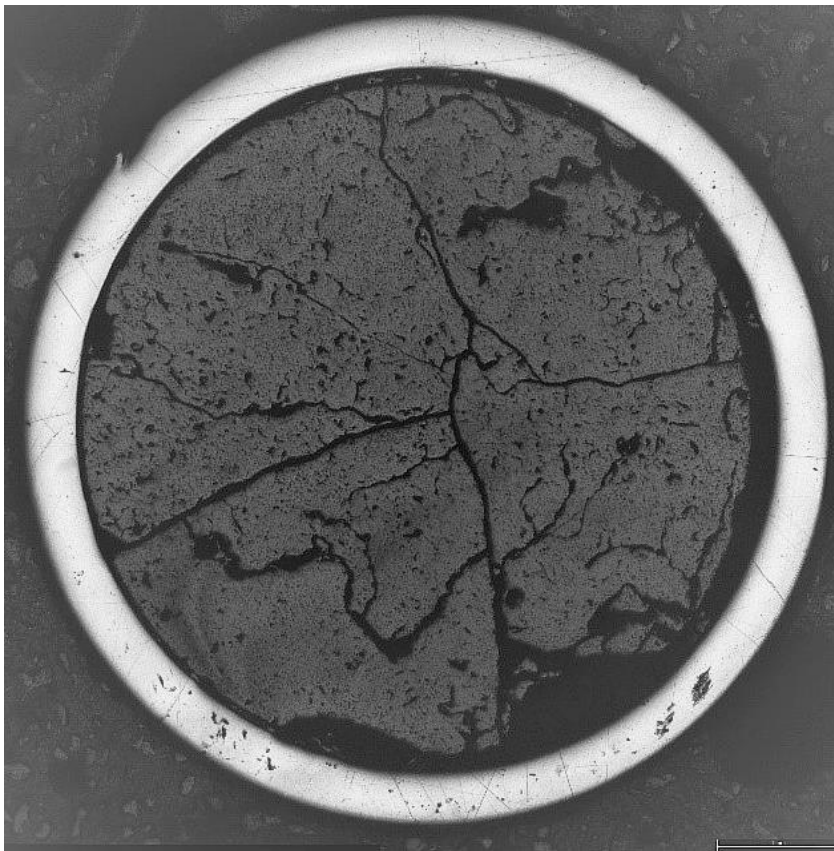


Figure 9 Macrographic image of a radial section of pin 1 (0242-17 / pellet 9).

Table 6 The as-fabricated pellet diameters and the diameters measured in the cut specimens after irradiation.

ID	Pellet #	Pellet diameter (mm)		
		As fabricated	PIE	Relative increase
0242-11	5	5.373	5.44 ± 0.01	1.2%
0242-17	9	5.377	5.42 ± 0.01	0.9%

Volumetric swelling

An estimate for the volumetric swelling was obtained by combining the results of image analyses from ceramography and neutron radiography. The latter was reported in D4.1 [9], but the data for the relevant pellets is repeated in Table 7 below. To determine and estimated swelling (rate), the height increase (and propagated uncertainty) is derived from the two neutron radiography images, as they have the same offset to the fabrication report value. The initial heights of the dedicated pellets are used in volume calculations.

Table 7 Calculated pellet volumes and relative increase, based on the analysis of ceramography and neutron radiography images before (BOI) and after irradiation (EOI).

		Pellet height (mm)			Volume (mm ³)		Volume increase (%)
	Pellet #	As fabricated	Neutron radiography		BOI *	PIE	
ID			BOI	EOI			
0242-11	5	6.296	6.34 ± 0.05	6.45 ± 0.09	143.7 ± 1.1	149.8 ± 2.2	4.2 ± 1.7
0242-17	9	6.069	6.14 ± 0.07	6.44 ± 0.06	139.3 ± 1.6	148.7 ± 1.4	6.7 ± 1.6

* Height at BOI is based on neutron radiography measurement

Using the macroscopic ceramography observations on the pellet fuel and the increase measured from neutron radiography, volumetric swelling is determined as presented in Table 7. A best estimate on the swelling rate, using the calculated burn-up and He-production numbers, repeated in Appendix A: A range of 2.7-4.3 vol% per %FIMA or 0.8-1.3 vol% per mg He/cm³ is found for the presented AmBB blanket fuel, irradiated under representative conditions in terms of power and temperature.

Using the relative increase of the full stack height (instead of the individual pellet) to calculate the volumetric increase, the maximal swelling rates are slightly higher: 3.2-4.9 vol% per %FIMA and 0.9-1.43 vol% per mg He/cm³, as published earlier [11].

Porosity analysis

Appendix B briefly explains the methodology and provides results, with porosities ranging from 8% close to the pellet's centre to 17-25% towards the fuel edge. The results are considered indicative as the available, useable images were limited.

4.3 MARINE Pin 2 – Ceramography

Despite the fixation of the fuel during the cutting process, grinding and polishing of cut specimens of pin 2 proved challenging due to break out of the highly fragmented fuel. Figure 10 shows a radially cut specimen.

Part of the fuel has likely displaced during the first part of the cut (the top left of Figure 10), to allow fixation of the fuel by inserting resin. Stitched macrographic images of two other radial specimens and one axial specimen, are presented in Appendix C.

Part of the large bead fraction (average fabrication diameter of 745 μm) is intact, frequently with large, elongated pores. Some large beads are cracked, while in other cases only fragments are visible in the specimen. Figure 11 shows magnifications of a large bead which shows a high density of small pores, with few larger pores. Cracks (as observed in the pellet fuel) are absent, as this has likely led to full fragmentation of beads, which is observed.

The small bead fraction (fabrication diameter of 50 μm) cannot be identified as such cylindrical, but very small fragments in the order of $\leq 50 \mu\text{m}$. The small fragment fraction occupies the space between the large beads. Indeed, the fabrication report describes crushing and sieving to obtain a small 'bead' fraction with particles up to 50 μm in diameter. Magnifications of the small fragments (Figure 12) show similar high density of small pores, while the fragments are too small to host bigger pores, as is observed in the large bead fraction.

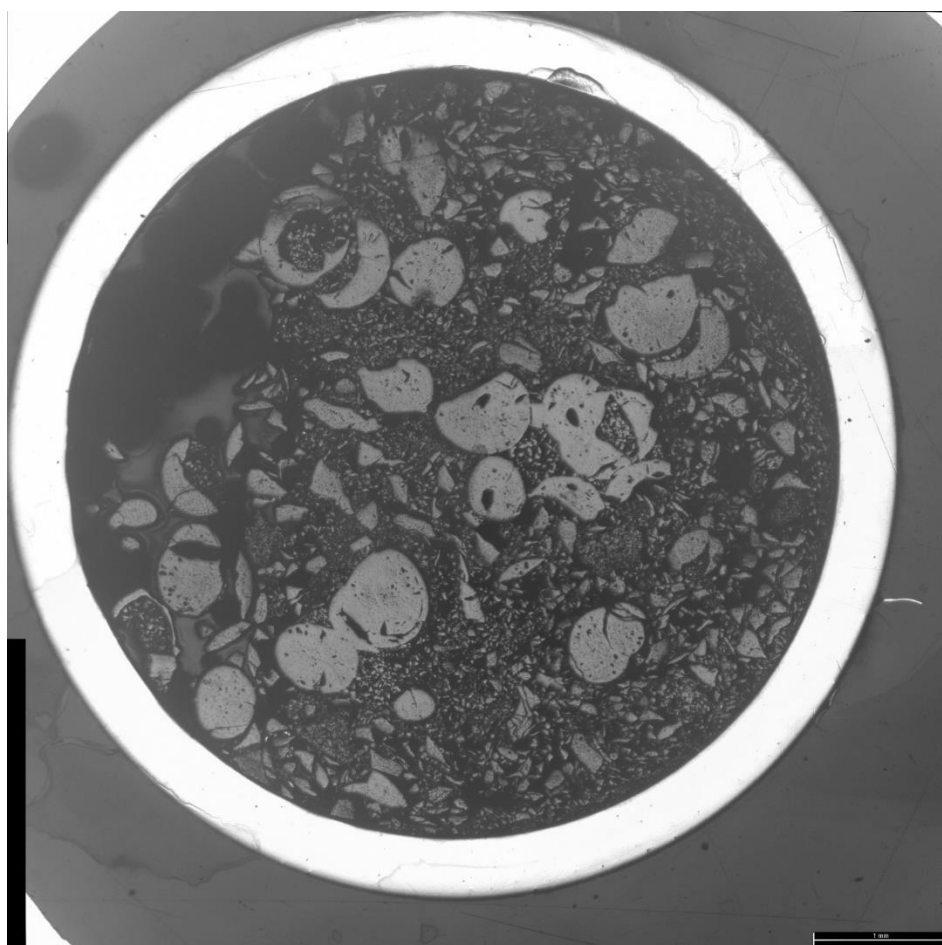


Figure 10 Stitched macrograph of radial specimen from MARINE pin 2, 0254-11. Part of the fuel (top left) was displaced in the first phase of cutting.

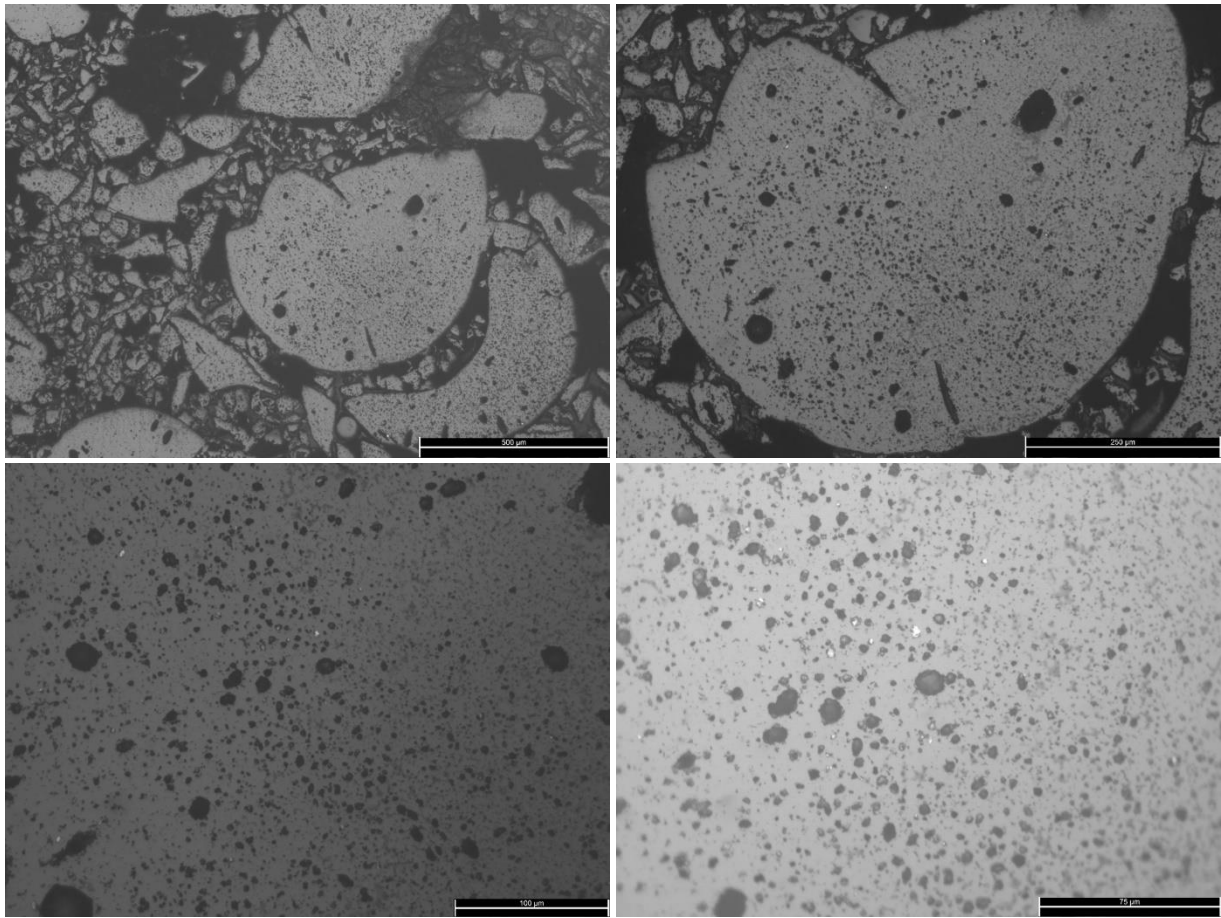


Figure 11 Magnifications of a large bead in specimen 0254-11, MARINE pin 2.

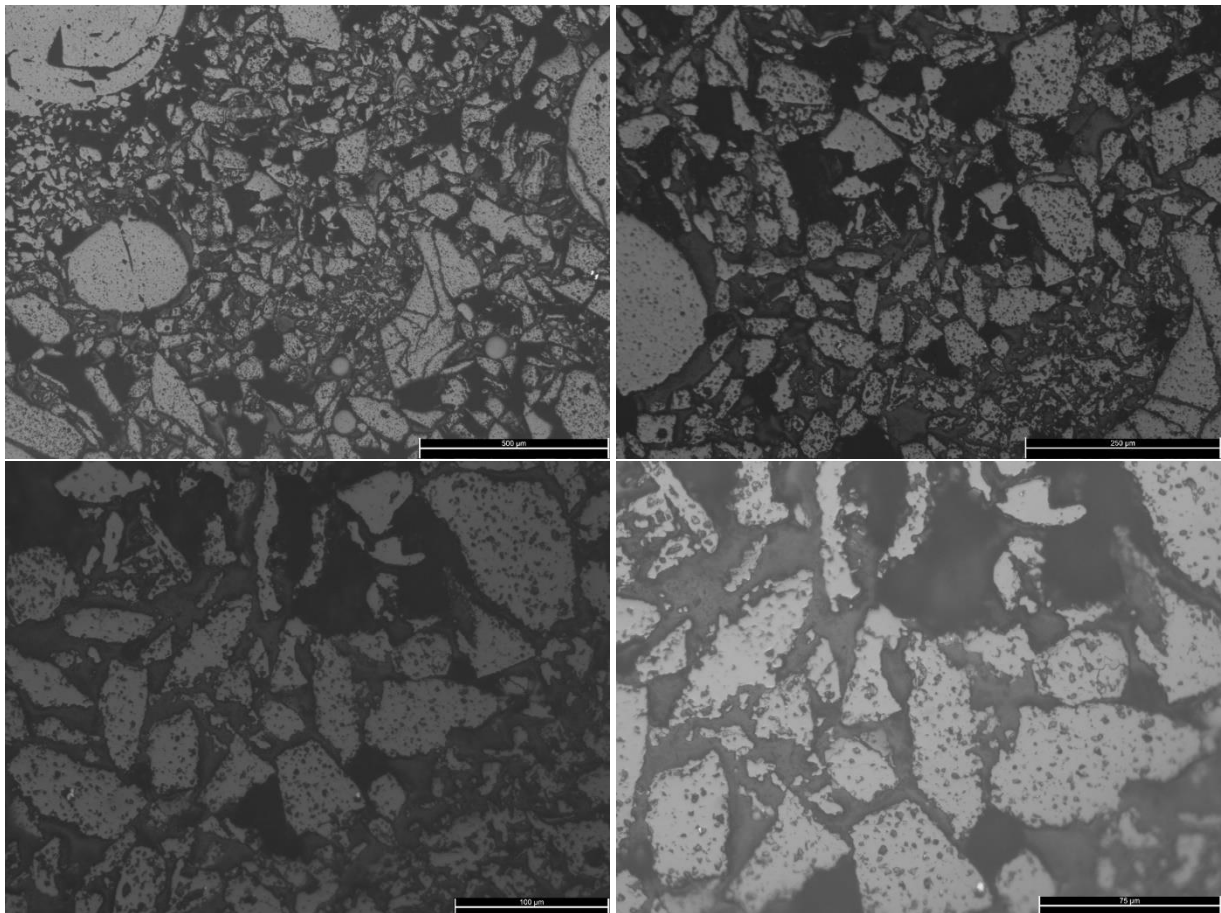


Figure 12 Magnifications of a small bead region in specimen 0254-11, MARINE pin 2.

5 Scanning Electron Microscopy

The Scanning Electron Microscope (SEM) used was a JEOL 6490 LV located inside an alpha-tight hot-cell line. Several images were taken with different magnifications from each of investigated radial sections using Secondary Electron Imaging (SEI).

An Oxford X-plore EDS (Energy Dispersive Spectroscopy) detector was installed onto the SEM and data acquisition and post processing were done using the Aztec software package. The EDS line measurements were carried out with an acceleration voltage of 25 keV to allow generation of higher energy L_{α} lines for the relevant actinides (besides the higher yield, lower energy M-lines) and working distance of ~ 10 mm. The line scans across the fuel radius were carried out with ~ 200 points from the fuel central hole to periphery with the cumulative dwell time of 1s/point. In a single case only 100 points across the line were acquired, which is mentioned explicitly in the section.

Two other detectors, WDS and EBSD, were not available during the experimental campaign.

5.1 Microstructure MARINE pin 1 (specimen 0252-11)

Poor surface condition of this specific specimen required several repetitions of grinding and polishing steps, which removed about half of the effective surface of the radial specimen.

Specimen 0242-11 was brought into the SEM where initial overview images show the high amount of cracking of the fuel, with some displacement locally in fuel-clad regions (Figure 13).

Across two radial lines, SOI2 and SOI3 (Figure 14), ranging from the pellet centre to the periphery, magnifications were made revealing some features of the microstructure.

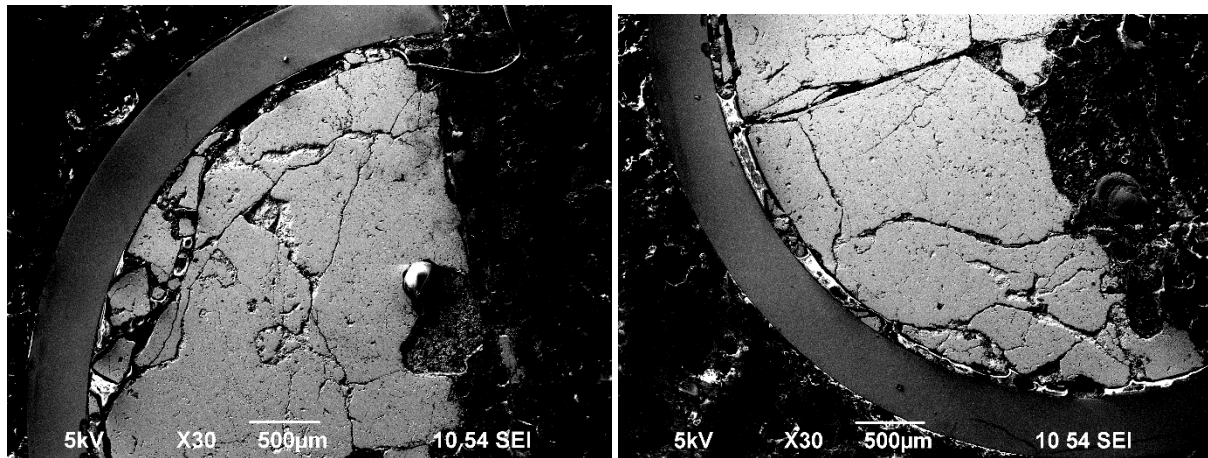


Figure 13 Overview of specimen 0242-11.

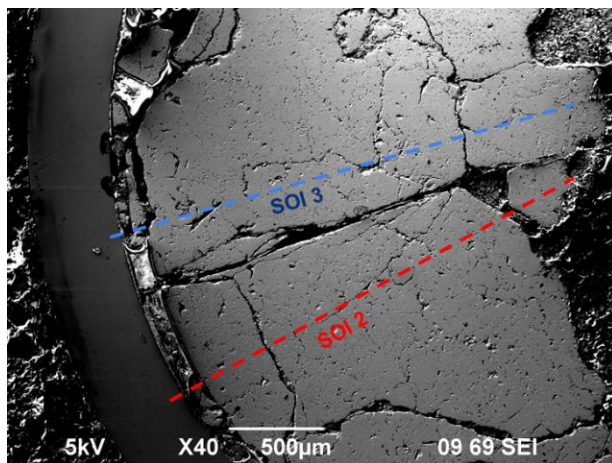


Figure 14 SEM image of specimen 0242-11 with the locations of two performed line scans.

Line scan 0242-11 SOI3

Figure 15 shows details across the line of SOI3. The grains can be distinguished already at moderate magnifications, highlighted by the rectangles in the top left overview image. The grains are best visible in the magnification near the pellet centre (purple rectangle), shown in the top right purple-lined image, and are qualitatively measured to be in the range of 5-10 μm , up to a maximum of $\sim 20 \mu\text{m}$ in some cases.

In some locations, grains appear to be etched along the boundaries or lined by a different phase, see e.g. the yellow magnifications of Figure 15.

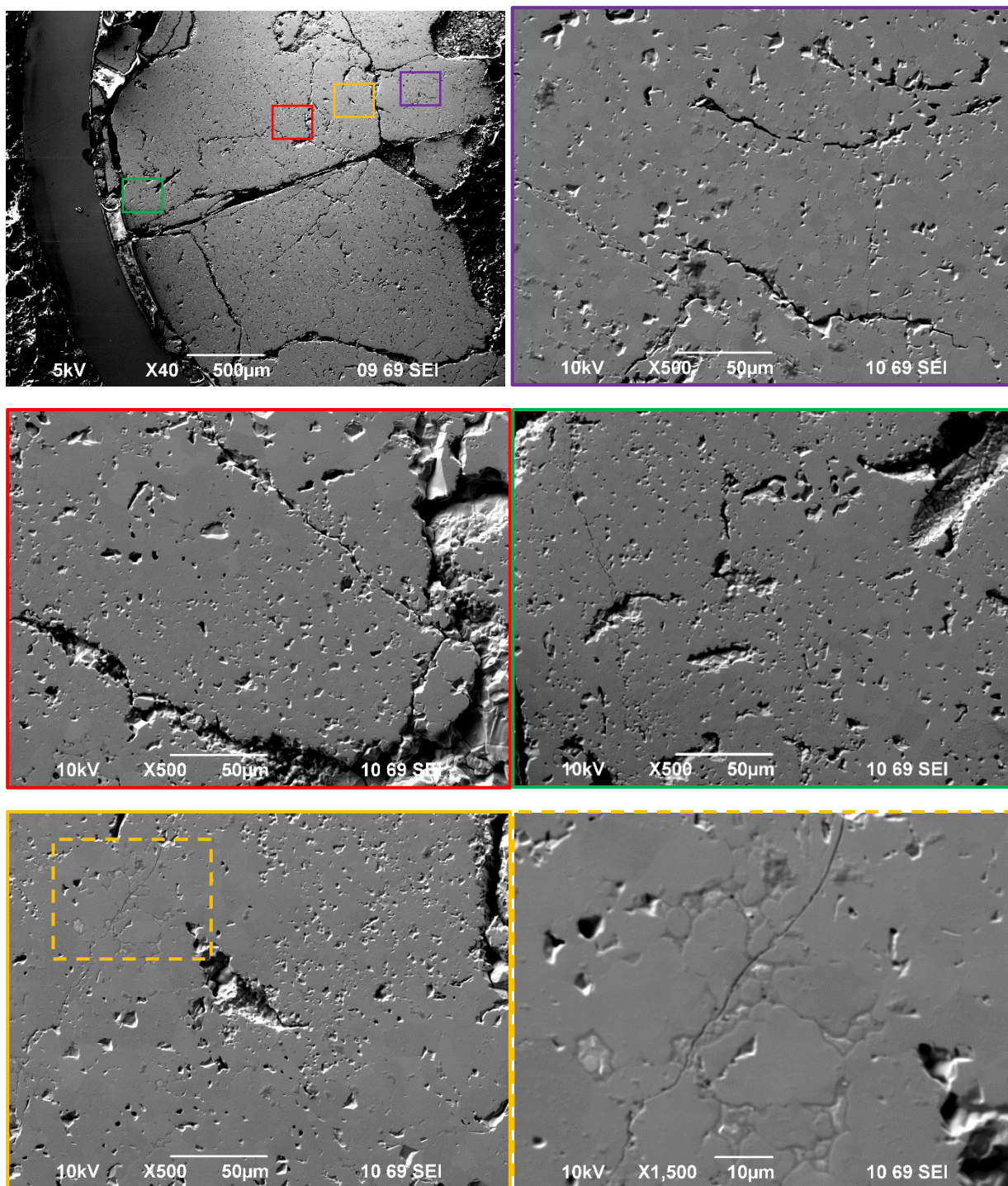


Figure 15 Magnifications of different locations across the SOI3 line. Individual grains can be observed by contrast differences (top left, purple edge) or when grain boundary seem slightly eroded and could be occupied by a different phase (bottom right, yellow edge).

Line scan 0242-11 SOI2

Figure 16 shows the region of SOI2, with a moderate magnification of the central region, highlighted by the blue rectangle, which is shown in Figure 17. Here, indicated by red arrows, larger areas of $\sim 200 \mu\text{m}$ are lined by porosity and are seemingly remnants of the initial (UO_2) beads that were pressed and sintered as part of the fabrication process.

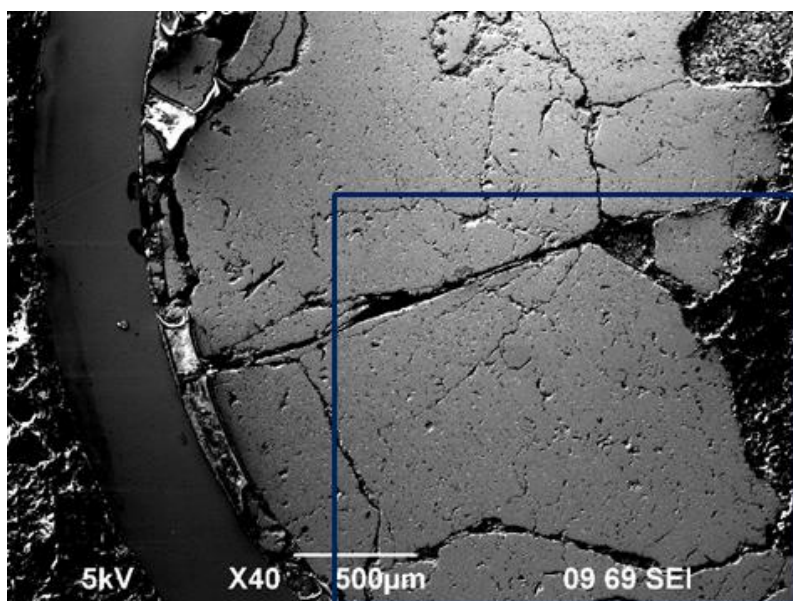


Figure 16 Region of SOI2 of 0242-11. The blue rectangle indicates the location of the magnification in Figure 17 and Figure 18.

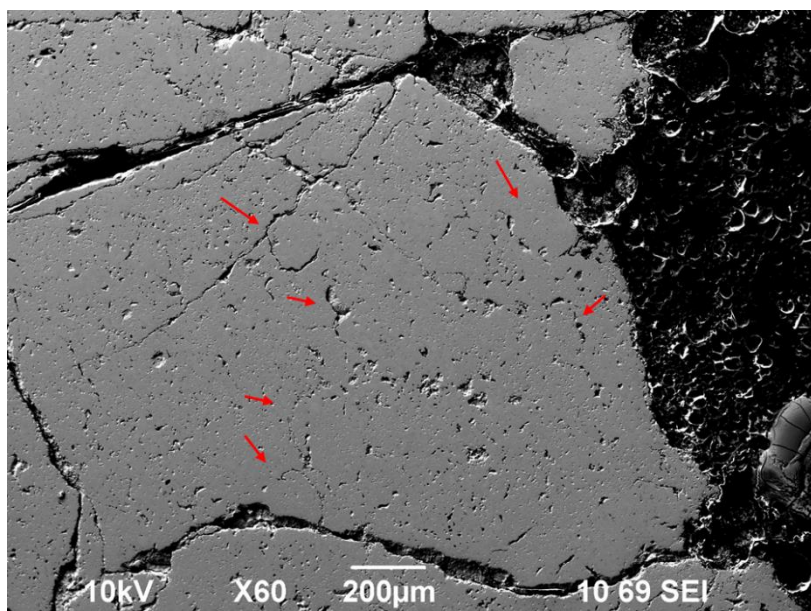


Figure 17 SEI image of fuel central region of 0242-11. In several locations remnants of the original beads that were pressed and sintered.

Figure 18 shows various magnification in the pellet central region showing individual grains, ranging from 5-20 µm that can be distinguished by their contrast differences (i.e. blue rectangle, bottom left image).

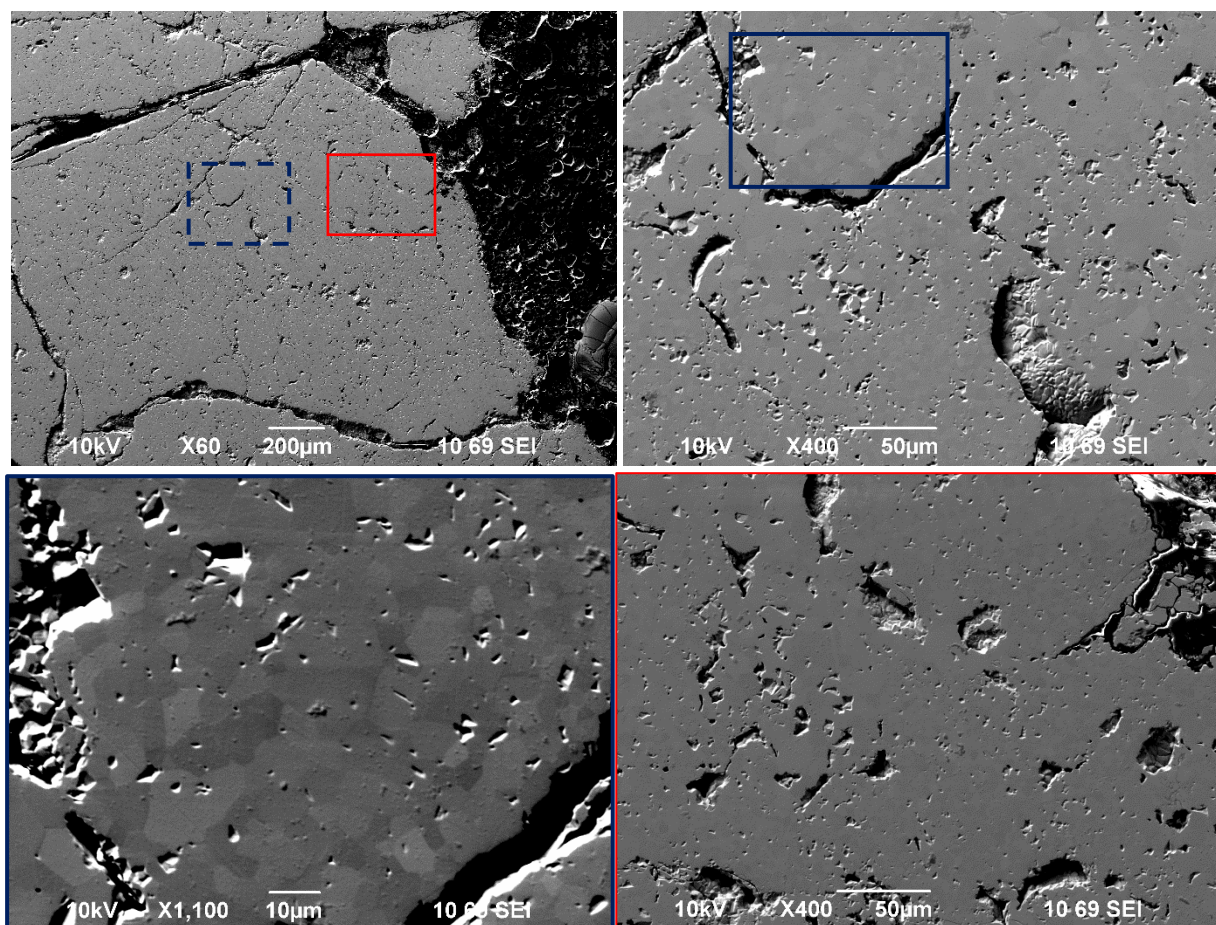


Figure 18 Magnifications of different locations across the SOI2 line. Individual grains can be observed by contrast differences (bottom left, blue edge).

5.2 Actinide distributions in MARINE pin 1

The presented data provide, global results on actinide distributions in the two radial specimens produced from pin 1 0242-11 and 0242-17. The setup and/or its detectors were eventually not available to do a more thorough analysis of the microstructure or local distributions of elements. Also analysis of the specimens from pin 2 was not achieved. The following figures are plotted with absolute distance on the x-axis. Appendix D presents the same plots with relative distance (r/R) on the x-axis.

5.2.1 Actinide distribution in specimen 0242-17

Specimen 0242-17 was brought into the SEM and two regions were identified, coarsely ranging from pellet centre to the edge. This sample is deemed suitable for examination based on the state of the surface of the specimen, see Figure 19. In these regions several EDS scans were performed, each marked as a numbered site of interest (SOI x).

The scans SOI2, SOI6 and SOI1 are taken in the direction where a substantial gap exists between pellet and cladding. SOI7 is the orientation where a minimal gap is observed.

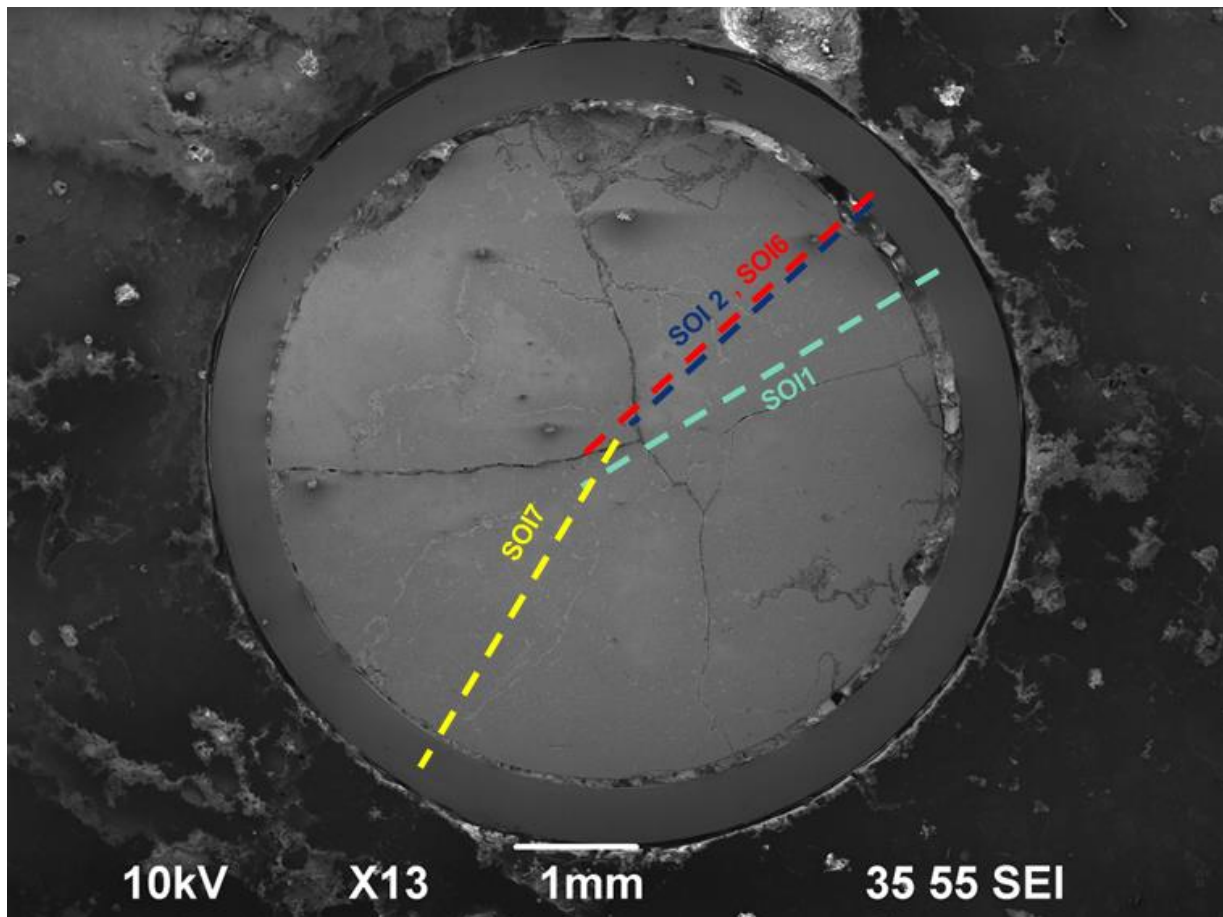


Figure 19 Overview of specimen 0242-17 in the SEM with the sites of interest (SOI) highlighted that were examined.

Line scans 0242-17 SOI2 and SOI6

Line scans SOI2 and SOI6 are taken at the same location, practically across the same radius/orientation of the fuel pellet, to confirm, to check the repeatability of the measurement after some fine tuning of the acquisition settings.

The top image of Figure 20 and Figure 21 show the range of the line scans across the surface of SOI2 and SOI6, respectively. The bottom trends show the found contributions of uranium, plutonium and americium (normalized to 100wt% HM) across that radius. The dotted line indicates the actinide composition (fuel stack average) as calculated by MCNP FISPACT calculations, reported in [9] and summarized in Table 9. Note that oxygen was not measured by the detector and the presented normalized wt% is an addition to ~88.5% (using the original O/M of 1.93).

Aside from local inhomogeneity, resulting in lower concentrations, the bulk uranium concentration, largely unchanged by the irradiation, approximates the calculated FISPACT value of 77 wt% across parts of the pellet radius.

The transmutation of over 50% of the americium is shown by measured concentration of Am, as well as the substantial concentration of its transmutation product, i.e. plutonium. Nominally, the calculated concentrations of 5-6% of both Am and Pu agree well with the measured values.

The heterogeneous nature of the fuel is visible in various areas, most extremely observed between 800-1300 μm in SOI2 (consistent with values at 1200-1700 μm in SOI6, Figure 21) where the americium and plutonium concentrations have rather constant values of ~15 wt%. As there was no reference material

available (non-irradiated) on which similar measurements could be made, it is hard to comment on the achieved heterogeneity of the fuel, but the fabrication process of filling highly porous UO_2 beads with an Am nitrate solution would certainly allow large variations in concentration, especially with the oxide phase reduced to Am_2O_3 during fabrication.

At the fuel periphery ($\sim 200\text{-}300\ \mu\text{m}$ measured from the fuel periphery), the relative uranium concentration drops and the relative concentration of Pu increases to $\sim 10\ \text{wt}\%$, while Am remains at its nominal value of 5-6%.

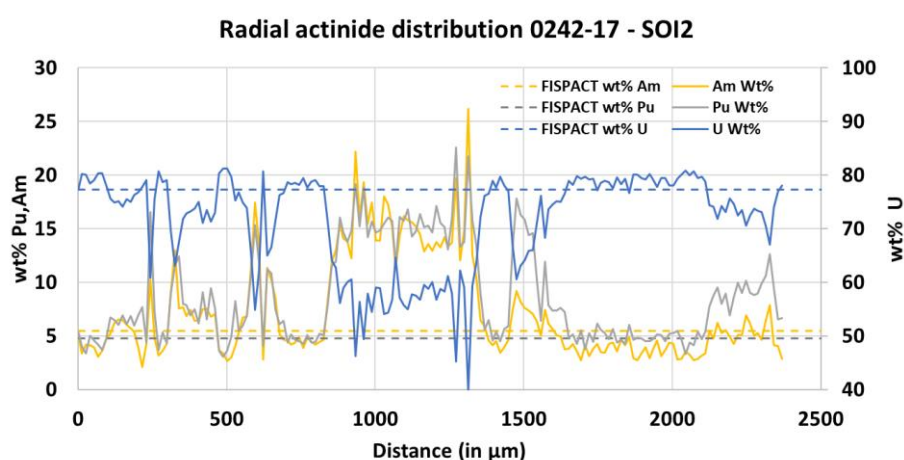
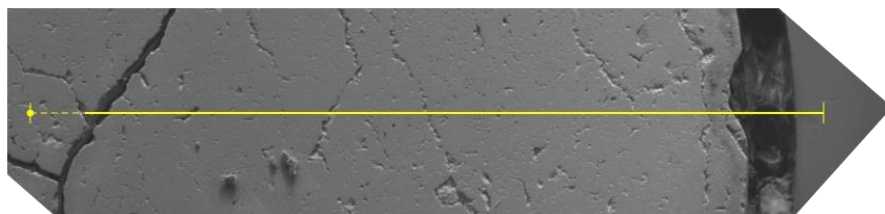


Figure 20 Radial distribution of U, Pu and Am concentrations across the indicated line of SOI2 of specimen 0242-17.

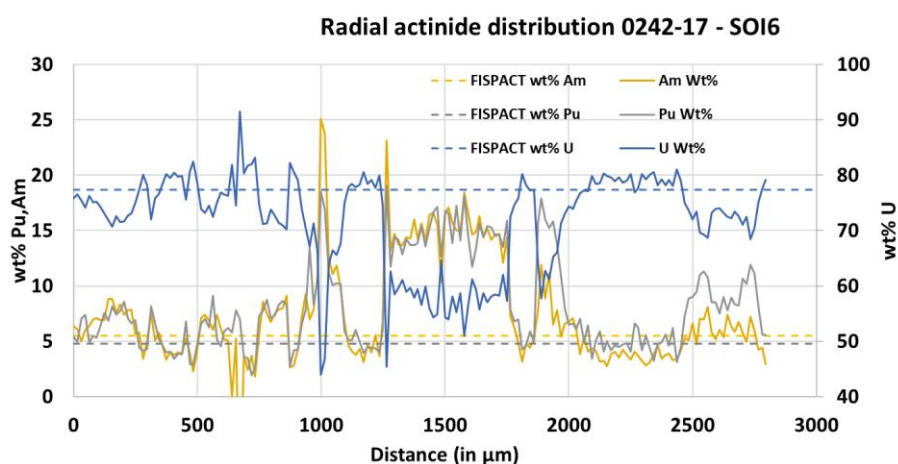
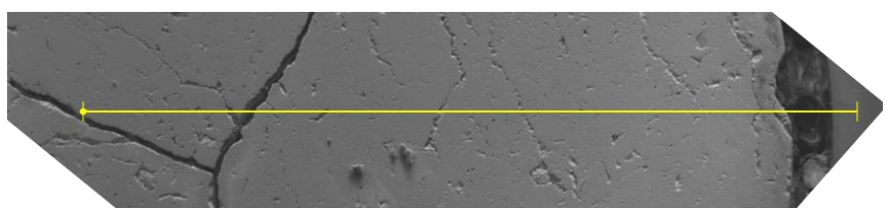


Figure 21 Radial distribution of U, Pu and Am concentrations across the indicated line of SOI6 of specimen 0242-17.

Line scan 0242-17 SOI1

As with other line scans from the same region (SOI2 and SOI6), SOI1 shows normalized concentrations that are nominally consistent with calculated FISPACT values and the heterogeneity of the fuel is visible in local variations of Am and Pu, Figure 22. This scan was produced with 100 points on the line, hence the slightly lower resolution.

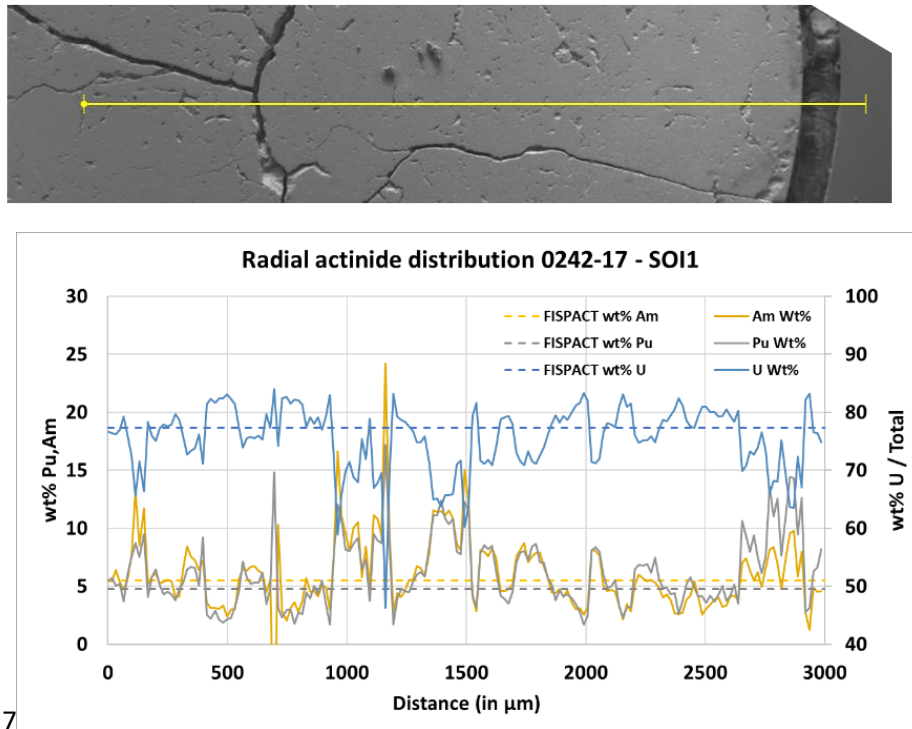
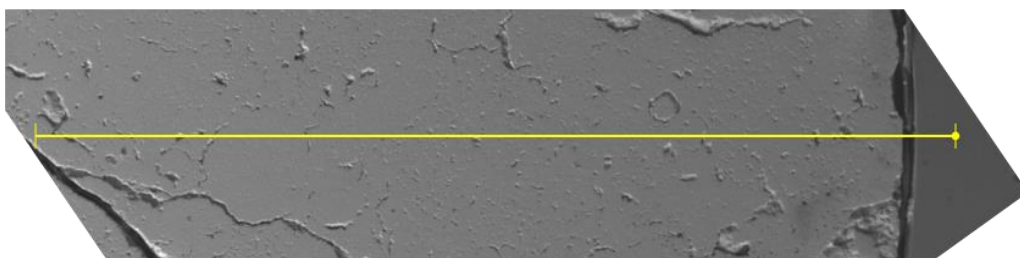


Figure 22 Radial distribution of U, Pu and Am concentrations across the indicated line of SOI1 of specimen 0242-17.

Line scan 0242-17 SOI7

Finally, in the radial orientation where a minimal fuel-clad gap width exists, a line scan (SOI7) was performed, Figure 23. Largely, the same general observations hold for this region, i.e. nominally consistent with FISPACT-calculations and variations in concentration due to the fuel's heterogeneity. The profiles are largely flat across the first 1000 μm of the scan after which it hits a more americium (and thus plutonium) rich region between 1000 and 1700 μm. In this scan, the increasing Pu-concentration at the periphery is less pronounced than in previous lines and only starts to deviate from the Am-profile in the outermost 100-150 μm.



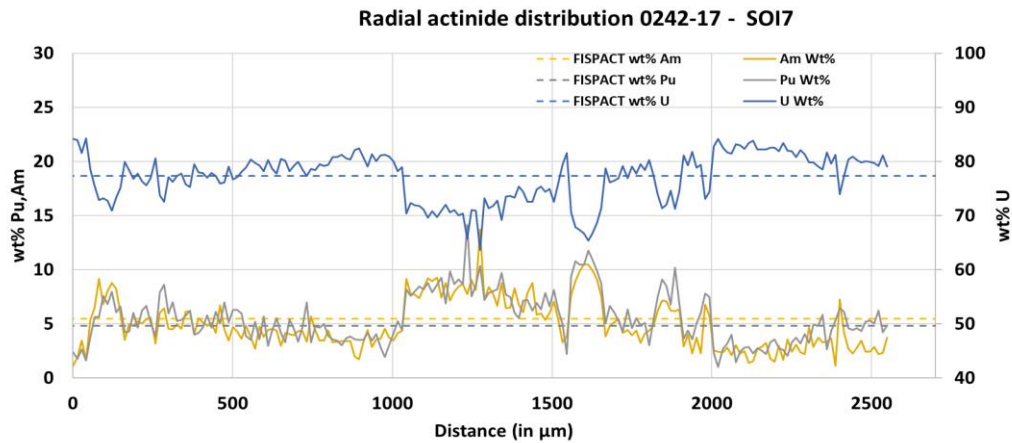


Figure 23 Radial distribution of U, Pu and Am concentrations across the indicated line of SOI7 of specimen 0242-17.

Radial Pu/Am ratio in sample 0242-17

Looking closer at the profiles of Am and Pu, it is clear that in the innermost 1700μm of the line scans, they largely coincide both in shape and intensity. Beyond the first 1700μm, the Pu content increases gradually, compared to the Am-concentration up to the fuel edge, most clearly visible at SOI2 and 6.

Figure 24 shows the ratio of the Pu and Am concentrations, where the relative Pu concentration gradually increases from near 1 in the pellet central region, to a value of 1.5-2 towards the fuel edge in the outer region of the pellet ($> 0.7R$). This radial transmutation profile is most likely caused by the irradiation conditions, i.e. a predominant thermal neutron spectrum and a high capture cross section at low neutron energy.

With the Pu-increase and U-decrease in the fuel periphery, it could be considered there is an additional cause for this relative Pu-increase, i.e. by transmutation of U-238 to Pu-239. This U-238 cross section is orders of magnitude lower than neutron capture by Am-241 for thermal neutrons, making an original Am-rich periphery region the most likely cause for this local increase. Additionally, substantial local U-transmutation would be observed as a deviation from the linear trend observed in Figure 24.

In case of SOI 7, the increase of Pu concentration towards the edge is less pronounced as in SOIs 1,2 and 6 and is only distinguishable in the outer region of the pellet ($> 0.9R$), Figure 25.

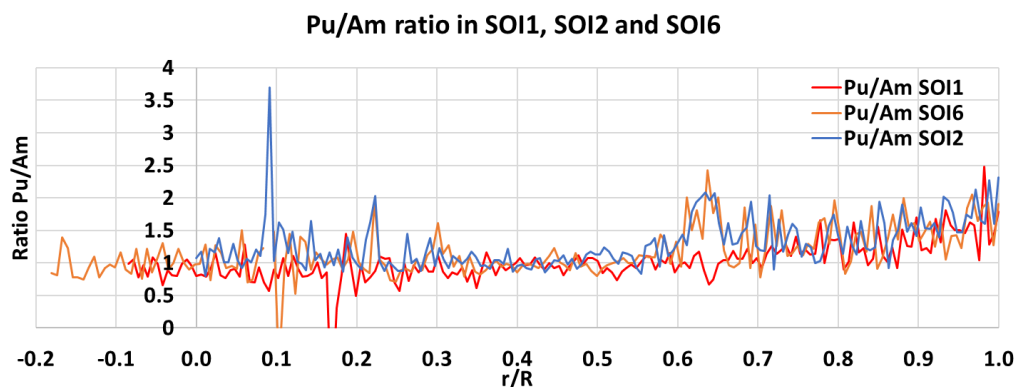


Figure 24 The ratio of Pu and Am across the radius of SOI1, SOI2 and SOI6, showing a constant Am/Pu in the inner 0.6R (or ~1500μm) of the pellet, and the Pu/Am ratio (i.e. transmutation fraction) gradually increasing towards the fuel edge from 0.65-0.7R onwards.

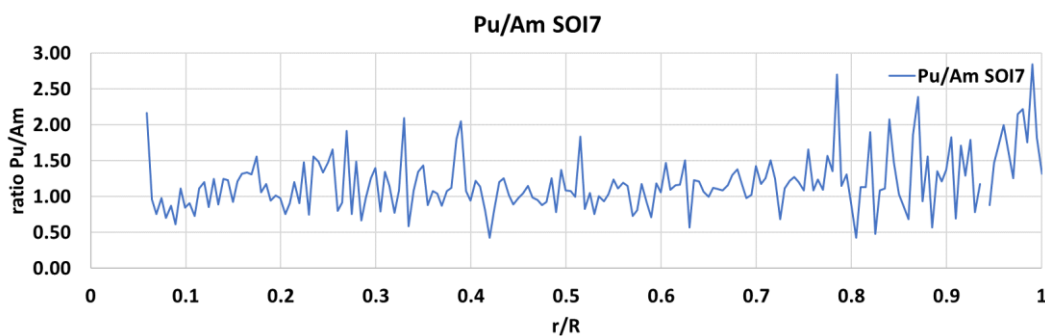


Figure 25 The ratio of Pu and Am across the radius of SOI7, showing a varying but in average a constant Am/Pu ratio across large part of the radius. An increasing ratio is only clearly distinguishable in the outer region $> 0.9R$.

5.2.2 Actinide distribution in sample 0242-11

EDS line scans were carried out along two lines, marked as sites of interest in Figure 14, where also the microstructures were examined. Figure 26 shows the location of the line scan, SOI2, and its results. The following figures are plotted with absolute distance on the x-axis. Appendix D presents the same plots with relative distance (r/R) on the x-axis.

Overall, the uranium concentration, largely unchanged by the irradiation, approximates the calculated FISPACT value of 77 wt% and shows a flat profile in large parts across the radius.

The transmutation of over 50% of the americium is visible in the nominal concentration of Am (drop from $\sim 13\%$ to $\sim 6\%$), as well as the substantial concentration of its transmutation product, plutonium. Nominally, the calculated concentrations (FISPACT) of 5-6% of both Am and Pu agree well with the measured values.

Line scan 0242-11 SOI2

This line scan at SOI2, was performed with only 100 points across the line (instead of 200) resulting in a reduced resolution, Figure 26. The heterogeneous nature of the fuel is visible in various areas, with locally Am-rich regions, e.g. at 1300-1400 μm , where the americium and plutonium concentrations peak to a concentration of ~ 15 wt%.

In the outer region of ~ 700 μm of the fuel pellet, the relative concentration of Pu increases to 8-10 wt%, while Am remains drops slightly across this distance to 5%.

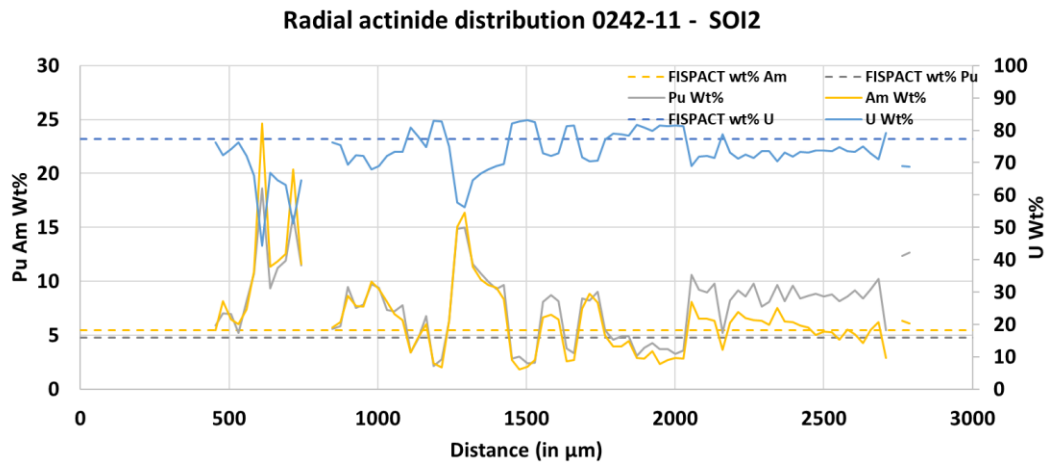
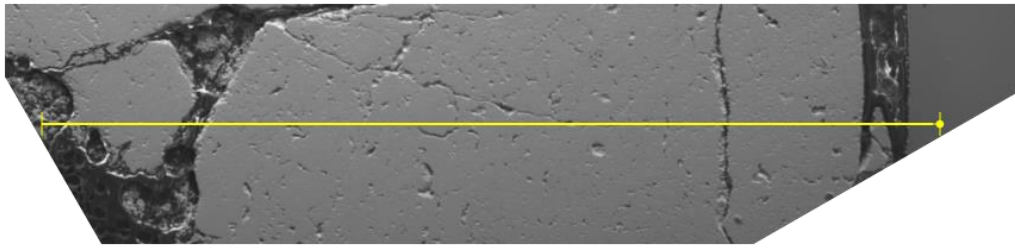


Figure 26 Radial distribution of U, Pu and Am concentrations across the indicated line of SOI2 of specimen 0242-11.

Line scan 0242-11 SOI3

The line scan at SOI3 seems to show the most least heterogeneity with a rather flat uranium concentration across the radius, consistent with calculated values, Figure 27. Variation exists in Am and Pu concentrations, but do not exceed 10 wt% locally.

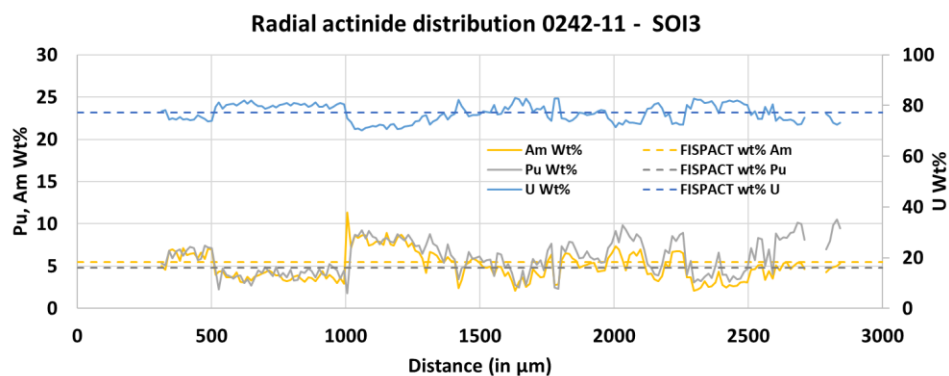
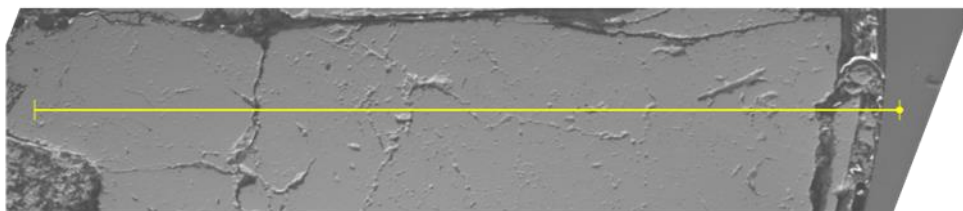


Figure 27 Radial distribution of U, Pu and Am concentrations across the indicated line of SOI3 of specimen 0242-11.

Pu/Am ratio in sample 0242-11

Similar observations as for specimen 0242-17, with Am and Pu showing identical profiles in shape and height across the inner most 70% of the pellet radius (up to 0.7R), where their ratio is close to 1, Figure 28. Also in specimen 0242-11, the relative Pu concentration gradually increases and the Pu/Am ratio deviates from 1 in the pellet central region, to a value of 1.5 - 2 towards the fuel edge in the outer 700 μ m (or > 0.7R) of the pellet.

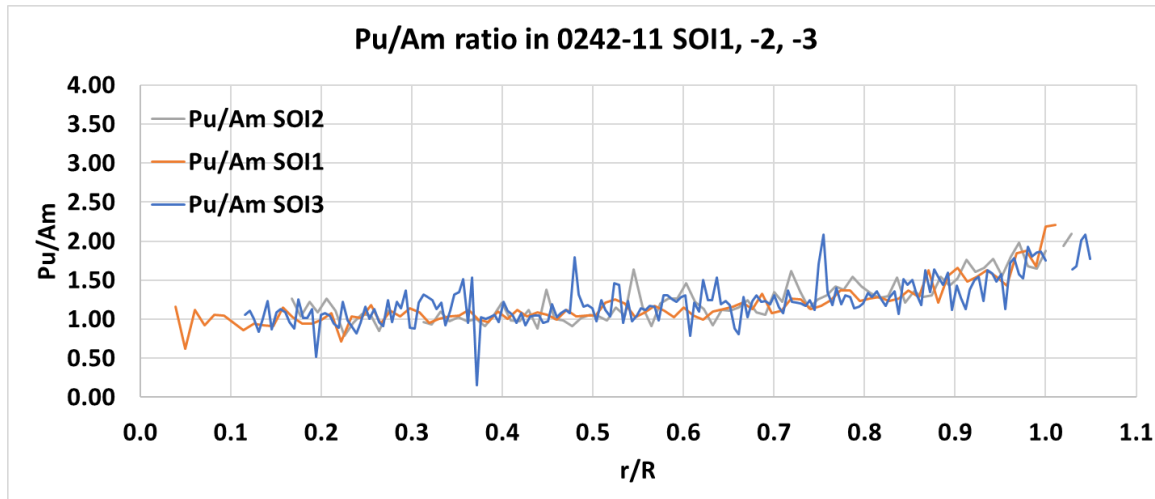


Figure 28 The ratio of Pu and Am across the radius of SOI1, SOI2 and SOI3 of specimen 0242-11, showing a constant Am/Pu in the inner 0.6R (or ~1500 μ m) of the pellet, and the Pu/Am ratio (i.e. transmutation fraction) gradually increasing to 1.5-2 from 0.65-0.7R towards the fuel edge.

6 Conclusions

MARINE experiment was successfully irradiated for 359 Effective Full Power Days (EFPD) comprising 12 cycles in the High Flux Reactor in Petten, between January 5th, 2016 and May 7th, 2017 and non-destructive examinations were completed in 2021 and reported in PATRICIA deliverable D4.1 [9]. MARINE's two fuel pins were transported to the alpha hot cell line for destructive analysis, and were cut to produce a number of radial specimens for optical microscopy and SEM. Delivered results were less than planned due to a long non-availability of the required hot cell line /equipment.

Fuel swelling (rate)

Two radial specimens were prepared from MARINE pin 1. Image analysis of macroscopic ceramography on these pellet fuel specimens yield a diametral increase of 0.9-1.2%. Combining these measurements with axial increase as measured from neutron radiography images gives a volumetric swelling of 4.8 vol% to 6.7 vol%.

The swelling as function of burn-up, or rather helium production, is then derived using the values provided by nuclear calculations (and gas quantities confirmed by puncturing), that were reported in D4.1.

Finally, a swelling rate can be established of 3.2-4.9 vol% per %FIMA or 0.95-1.43 vol% per mg He/cm³ for the presented AmBB blanket fuel, that was irradiated under representative operational conditions (i.e. fuel temperature and power density).

Microstructure

Macrography of specimens from pin 1 revealed that there are induced cracks in pellets due to thermal stresses, as well as substantial porosity and large channels. The fuel-cladding gap has not closed, which agrees with negligible strains observed by profilometry. SEM images on these specimens reveal increased porosity surrounding particles with dimensions in the order of the beads (~200 µm) that were pressed and sintered during fabrication. Finally, in some cases higher magnification of SEM images allows to distinguish individual grains to be in the order of 10-20µm. More detailed information on grain size distribution could not be obtained due to the unavailability of the EBSD detector.

A limited study on porosity was carried out across the radius of a single specimen from the pellet fuel. This first result shows the lowest porosity of ~7% near the centre of the pellet ($r/R \approx 0.1$), while porosity of 15-25% is observed in the examined locations in the outward direction (at r/R of 0.5, 0.7 and 0.9). In all cases, measured pore areas are translated to a frequently used unit of Equivalent Circular Diameter (ECD) of which the median value (often reported as D50) is used as figure of merit in particle and pore size distributions. In the examined cases, these range from 2.1-2.7µm, indicates a level of homogeneity in the porosity.

Actinide distributions

Several EDS line scans were produced across different radii of two prepared specimens of pin 1, bearing the pellet fuel. Generally, the found concentrations of uranium, plutonium and americium agrees well with the predicted composition as calculated by FISPACT, which confirm that about half of the americium has been transmuted. The heterogeneous nature of the fuel is reflected in areas of increased Am-content of up to 10 wt%HM and the Am-profile and -quantity is accurately followed in by its main transmutation product Pu across largest part of the fuel radius. Only towards the edge of the fuel pellet,

roughly from $\sim 0.7R$ to $1R$, does the Pu-content start to deviate from the Am-profile, with the Pu/Am ratio gradually increasing to a factor of 2 in the outer 30% of the fuel radius.

As there was no reference material available (non-irradiated) on which similar measurements could be made, it is hard to comment on any evolution of the heterogeneity of the fuel during irradiation, but the fabrication process of filling highly porous UO_2 beads with an Am nitrate solution would certainly allow large variations in concentration, especially with the oxide phase reduced to Am_2O_3 during fabrication.

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Appendix A Power history and achieved transmutation targets

This section presents the post irradiation power calculation (Figure 29) and eventual fuel burn-up and transmutation fraction and He production (Table 8) for both fuel pins of MARINE, that was presented more elaborately in D4.1 [9].

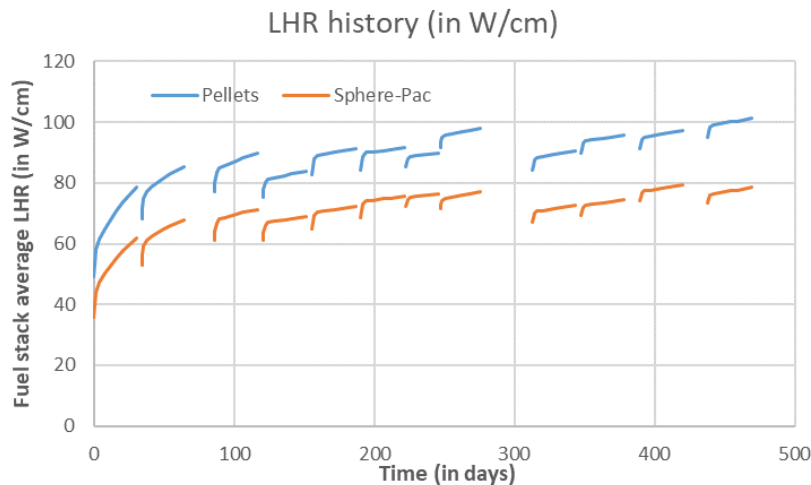


Figure 29 The linear heat rate of both fuel stacks during irradiation. The nuclear analysis did not yield statistically significant axial gradients across the fuel stacks.

Table 8 Main irradiation results as summarized in D4.1

Pin Nr.	1: Pellets	2: Sphere-Pac
# Full Power Days in HFR (elapsed days)	359 (469)	
LHR _{max} (W/cm)	101	79
He production at EOI (mg/cm ³)	5.34	3.57
Am-transmutation fraction (%)	55%	64%
Burn-up (% FIMA)	1.57%	1.98%

Table 9 shows the main constituents of the fuel in pin 1 at BOI and EOI. These values serve as a reference to later measurements of the fuel composition by SEM EDS and shows most transmuted Americium, resides in the fuel as Plutonium.

Table 9 Relative constituents (actinides and oxygen) to the fuel composition at BOI and EOI, as calculated by FISPACT, average for the fuel stack.

	Wt% (normalized)	
	BOI	EOI
U	75.6	77.3
Pu	0.3	5.5
Np	0.8	0.7
Am	11.8	4.8
O	11.5	11.7

Appendix B Preliminary porosity analysis on MARINE pin 1 pellet fuel

Basic porosity analysis was done with ImageJ software on a selected set of magnifications, at the radial locations indicated in Figure 30, corresponding to approximate relative radii, r/R , of 0.1, 0.5, 0.7 and 0.9. The images cover an area of 0.08 mm^2 and have a resolution of $\sim 0.1 \mu\text{m}/\text{px}$. Threshold for pore identification was set manually, see Figure 31, and false positive/noise reduction was done by excluding pores with an Equivalent Circle Diameter¹ (ECD) of $1 \mu\text{m}$ (or area of $\sim 14 \mu\text{m}^2$), based on image quality and resolution.

Figure 32 shows the other magnifications of areas indicated in Figure 30. In all images scale bars are excluded from analysis, as well as the area of the fuel clad gap in the magnification at r/R 0.9 (bottom Figure 32).

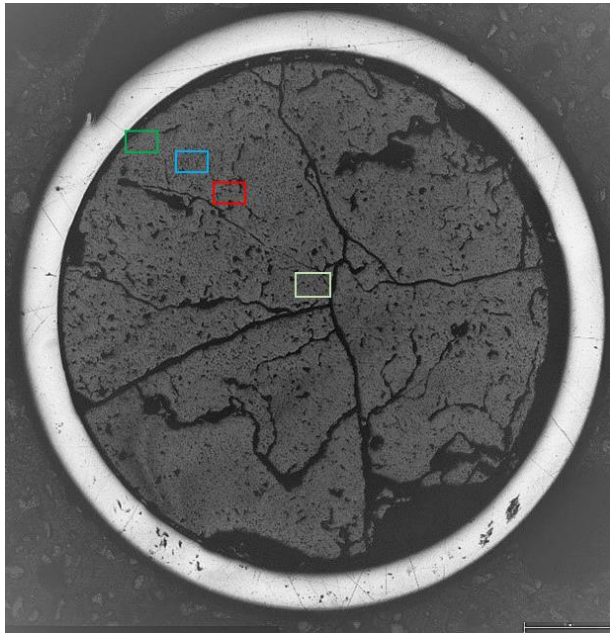


Figure 30 Radial section of MARINE pin 1, specimen 0242-17, with locations of magnifications for porosity analysis highlighted.

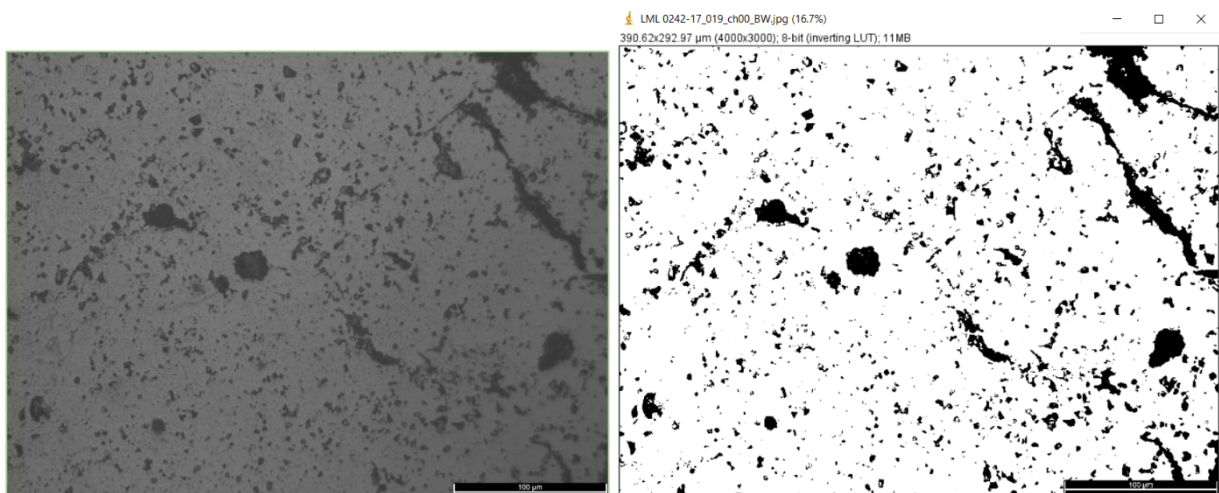


Figure 31 Example of resulting image after setting threshold in ImageJ to obtain porosity fraction. Magnification at radial location r/R of 0.1.

¹ Equivalent Circular Diameter, ECD is defined as $= 2\sqrt{A_{\text{pore}}/\pi}$

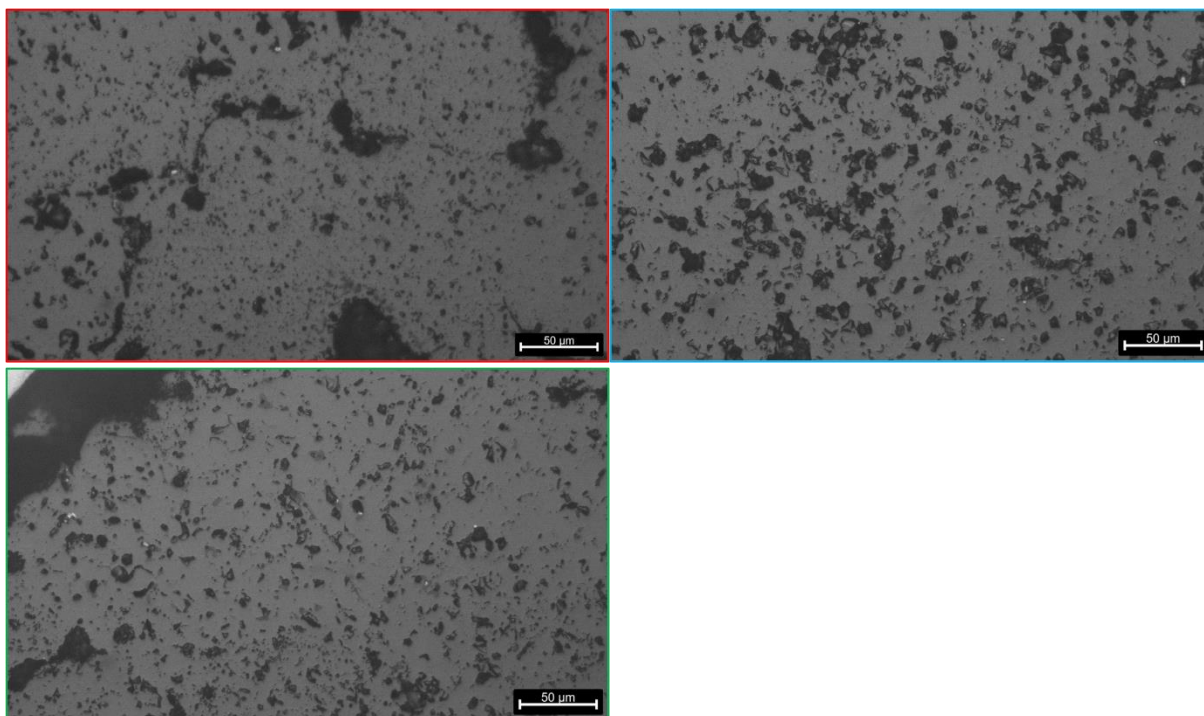


Figure 32 Magnifications of radial section of MARINE pin 1, specimen 0242-17, at radial locations r/R of 0.5, 0.7 and 0.9, on which basic porosity analysis was done.

Table 10 presents the main results of the described porosity analysis procedure. Relative porosity was found to range between 8% and 25%, with the lowest porosity at the centre of the pellet. While (interconnected) pores reach apparent areas of $1018 \mu\text{m}^2$ and $1153 \mu\text{m}^2$

It is noted that these results are only indicative, as analysis in multiple radial orientations and from multiple specimens are required to obtain statistically representative results.

Table 10 Measured porosity features of the four presented magnifications.

r/R	Pore count (#)	Cumulative pore area (μm^2)	Pore area fraction (%)	Median ECD [D50] (μm)	Analyzed image area (μm^2)
0.9	914	11370	15%	2.30	74346
0.7	750	20284	25%	2.67	79788
0.5	922	13762	17%	2.13	79788
0.1	800	8889	8%	2.24	113264

Appendix C Pin 2 macrography

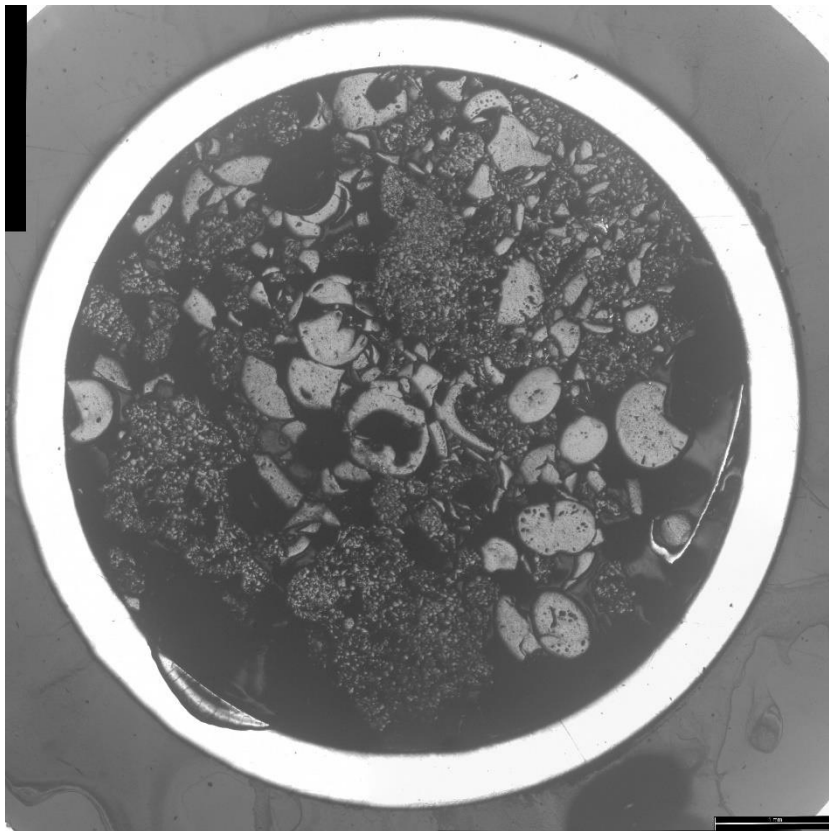


Figure 33 Stitched macrograph of specimen 0254-03.

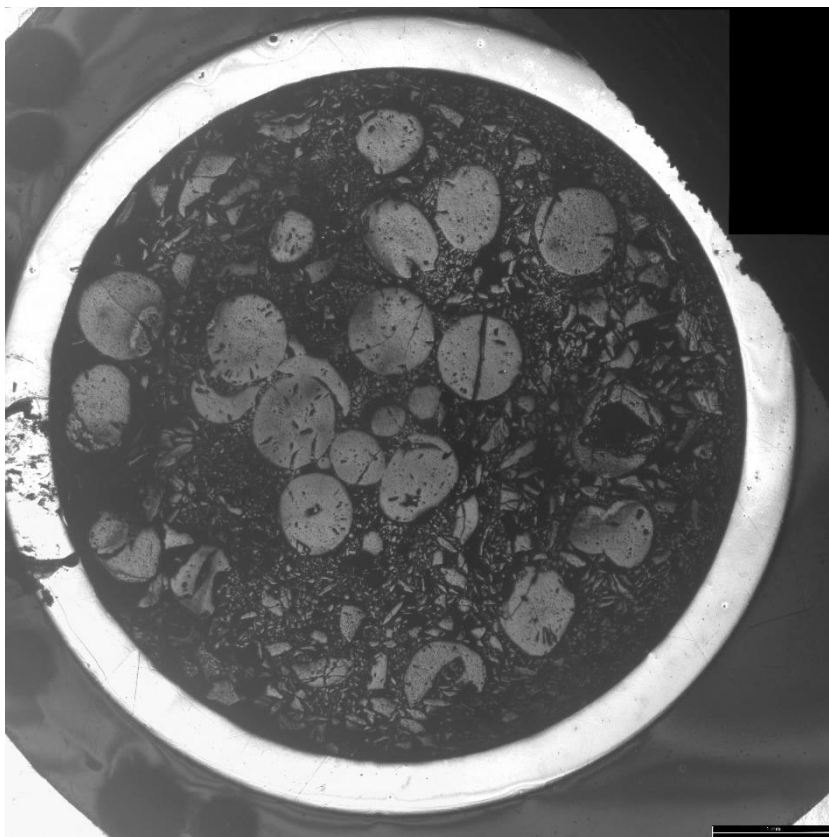


Figure 34 Stitched macrograph of specimen 0254-07.

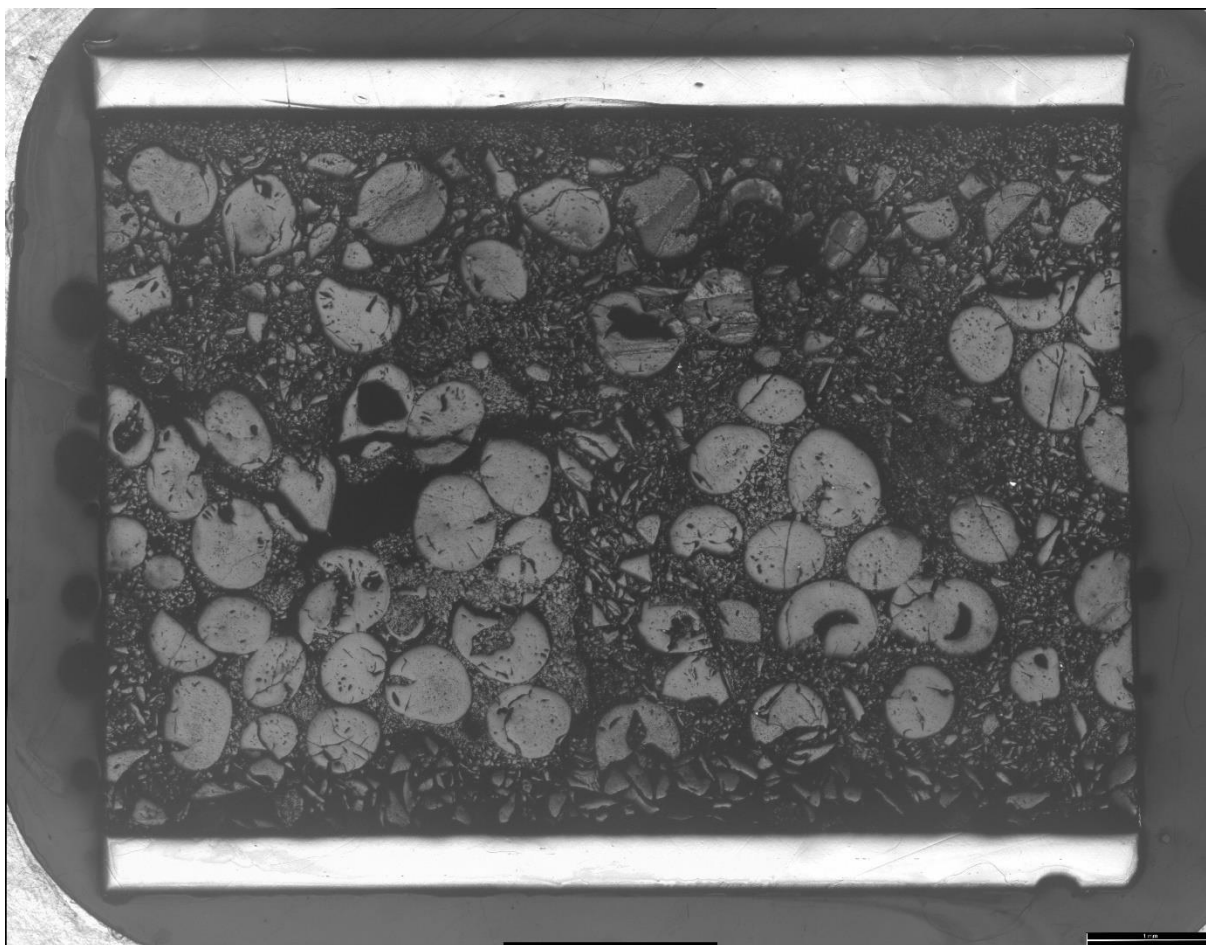
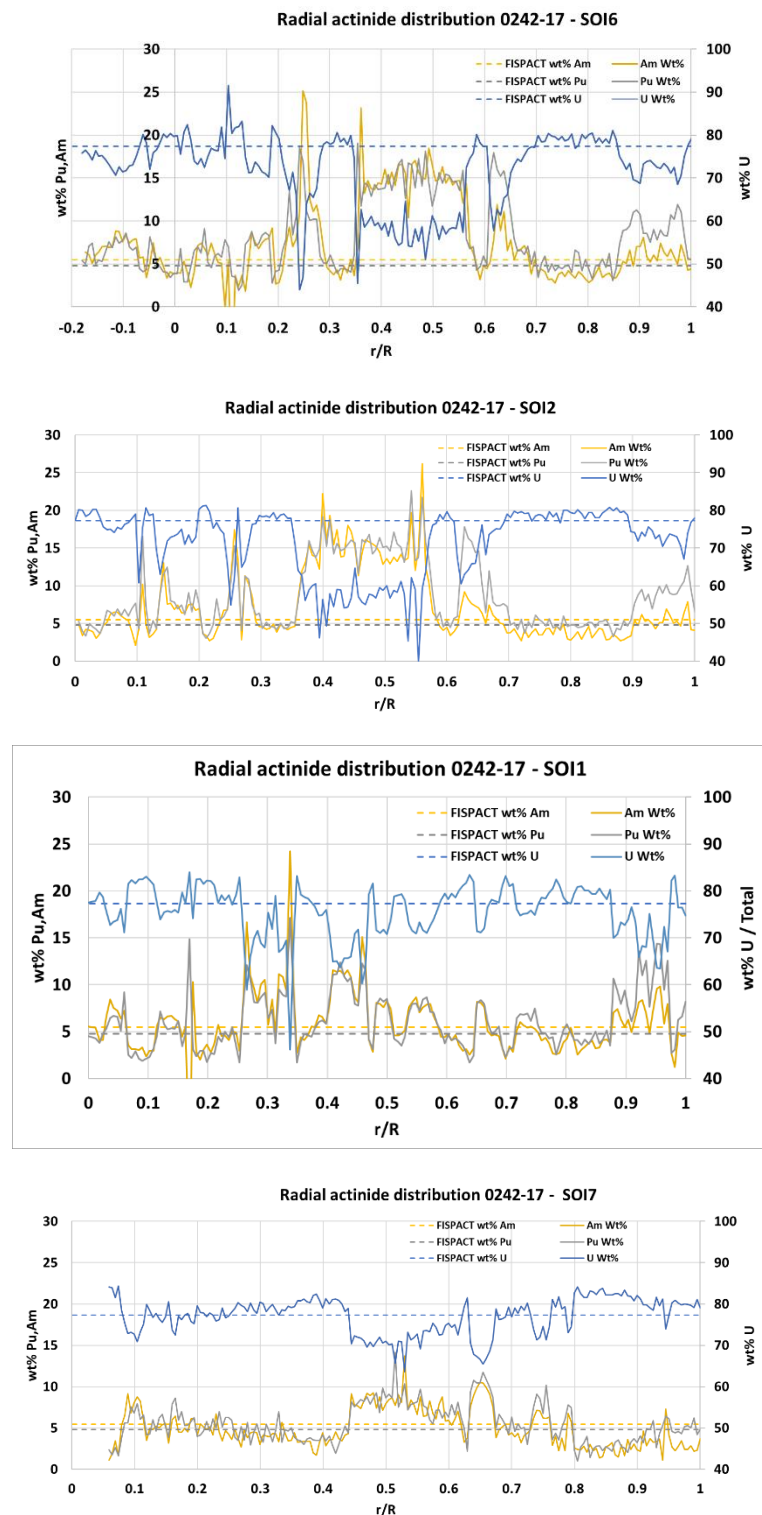


Figure 35 Stitched macrograph of axial specimen from MARINE pin 2, 0254-13.

Appendix D Radial actinide distributions

Performed line scans as presented in section 5, are shown here with relative distance across the fuel r/R .



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