DEVELOPMENT OF U-MO MONOLITHIC PLATES WITH GRADIENT FOILS

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ABSTRACT

Within the framework of the European HERACLES initiative, Framatome-CERCA and Technical University of Munich (TUM) are developing U-Mo monolithic fuel. A pilot U-Mo monolithic fuel manufacturing and inspection line is currently being implemented at the Framatome-CERCA R&D workspace. This entails U-Mo alloying followed by ingot, foil and plate production.

Some high-performance research reactors, such as TUM's FRM II reactor, may require U-Mo foils of variable thicknesses, known as gradient foils, to maintain core coolability when converting to lower enriched uranium fuels. Framatome-CERCA and TUM are working together to evaluate the possibilities for producing U-Mo monolithic plates with gradient foils, including manufacturing gradient foils, industrializing the TUM PVD coating process and industrializing the Framatome-CERCA C2TWP plate fabrication process. The aim is to test the robustness of the process on inert material before switching to U-Mo gradient foils and plates

In this report, we present the development work conducted on U-Mo gradient foils by Framatome-CERCA and TUM. The work is funded by the European Commission through the H2020 project LEU-FOREvER.

1. Introduction

Within the framework of international initiatives to minimize the use of Highly Enriched Uranium (HEU), the conversion of research reactors worldwide from HEU to Low Enriched Uranium (LEU) is a major focus of nuclear fuel cycle R&D.
Backed by the European Commission, R&D on the conversion of European High Performance Research Reactors (EU-HPPR), through the H2020 project LEU-FOREvER, is supported through the European HERACLES consortium. The HERACLES consortium is composed of Framatome-CERCA™, the CEA (Commissariat pour l'Energie Atomique), the ILL (Institut Laue-Langevin), the SCK•CEN (Belgian Nuclear Research Center) and TUM (Technical University of Munich).

Such conversion requires studies on new design, materials and fuel to compensate for the changes induced by lower enrichment. The Uranium-Molybdenum (U-Mo) alloy is a promising candidate among those currently being studied for a new, high density research reactor fuel, in dispersed or monolithic form.

The FRM II reactor (Forschungs-Neutronenquelle Heinz Maier-Leibnitz, Garching, Germany) is an EU-HPPR that uses a disperse cermet of highly-enriched Uranium (HEU) U3Si2 alloy fuel clad in aluminum and takes the form of a single cylindrical fuel element with involute-shaped fuel plates. Due to the specific cylindrical design, the fuel plates have a non-uniform uranium density distribution along the arc of the involute. Increasing interest in U-Mo monolithic fuel for European reactors such as FRM II (Forschungs-Neutronenquelle Heinz Maier-Leibnitz, Garching, Germany) has led to intensive development of this type of fuel in Europe.

First proposed as an alternative in 1996 [1, 2], the U-Mo alloy was used by CERCA in the 1960s for the first generation of French nuclear power reactors. In a monolithic Uranium-Molybdenum alloy fuel form with 10% of molybdenum, it allows for stabilization of the bcc γ-phase under irradiation, rather than the anisotropic expansion of the α-phase [3, 4].

Since 2005, some members of HERACLES have supported R&D and the industrialization of this type of fuel and product [5, 6]. Also, since 2008, CERCA™ and TUM, through the ALP program and with the support of the INL (Idaho National Lab), have worked on the development of monolithic plate fabrication [7].
This research has led to the development of a U-Mo fuel with Zirconium as a diffusion barrier and aluminum cladding.
In charge of the coating, TUM is developing a PVD process, while Framatome, with the C2TWP process [8, 9], is developing an AlFeNi cladding [10, 11].

In this report, we present the work conducted by TUM and CERCA on the development of gradient foils and plates and the industrialization of the Framatome-CERCA C2TWP plate fabrication process using inert material. The aim is to test the robustness of the process on inert material, linked to the fabrication of gradient foils and to the industrialization of the TUM PVD coating process before proceeding with U-Mo gradient foils and plates.

2. Overall design and fabrication considerations

The preliminary design selected for the development of the gradient plates matches the geometry of HERACLES European irradiation tests, with foils of 45 mm in width, 762 mm in length and variable thickness from 0.60 to 0.17 mm (Figure 1). For the preliminary tests presented in this report, small-size gradient foils are preferred, with width 19 ± 0.1 mm, length 82.5 ± 1 mm and thickness between 0.60 and 0.22 ± 0.05 mm. In the future, the process could be adapted to the specific geometry needed for FRM II fuel plates, for example, specifically regarding the width.

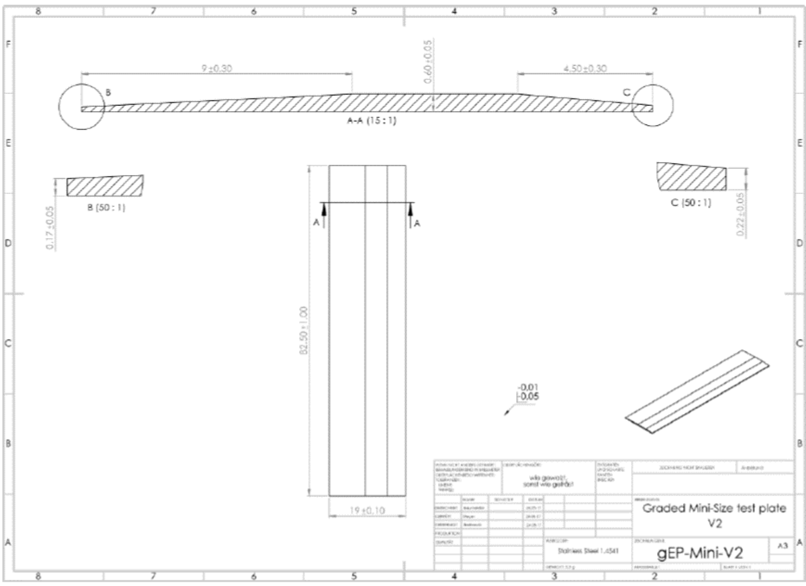

Figure 1: Dimensions and section view of HERACLES gradient fuel plate

The complete chain of production for this fuel is ongoing, shared between CERCA™ and TUM. Figure 2 presents the way in which production will be divided following development using inert material. The major steps are procuring the raw materials, preparing the alloy feedstock, casting the coupon or the rod, rolling the coupon into a bare foil or producing the alloy powder, machining the foil or producing a foil by SLM, coating this foil and, last, cladding to obtain a fuel plate.

During the tests with inert material, the foils and methods used to obtain gradient foils were developed by TUM at the R&D stage [12]. Two general types of material-forming processes were considered for this study: subtractive processes and additive processes. The results of these developments show that both milling and selected laser melting are suitable for producing stainless steel graded mini-size foils at the R&D stage. However, both solutions required a post-process treatment prior to PVD, such as annealing in the case of the subtractive solution and shot peening and HIP for the additive solutions.
Development of the production process for the bare foils started at CERCA™ mid-2019, in collaboration with TUM [13]. Studies on producing powders and foils by additive manufacturing are also ongoing at CERCA™ [14].
A zirconium coating is then applied on the foil surfaces to prevent diffusion and the reaction between the aluminum cladding and the alloy used for monolithic fuel [15].
The C2TWP process developed at CERCA™ [16] is then used to apply an aluminum cladding around the coated foil.
This process is now in an optimization phase for flat full-size uranium-molybdenum foils and for inert mini-size gradient foils.

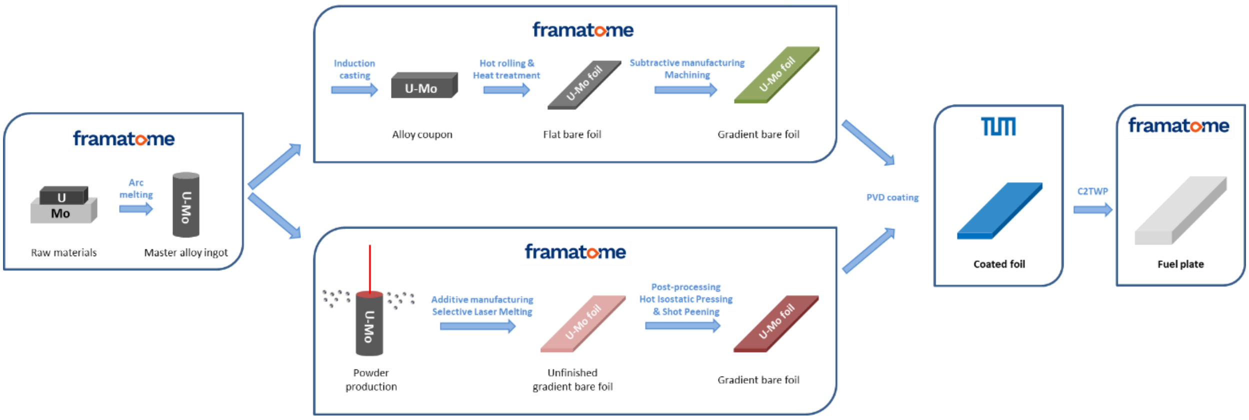

Figure 2: Fabrication process for U-10Mo fuel plates

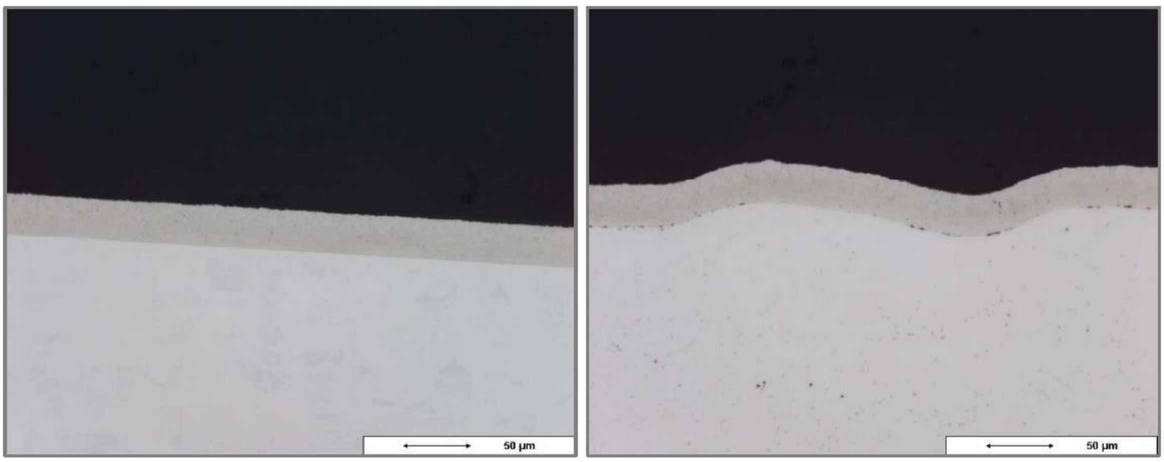

Figure 3: Cross-section of a Zr-coated HPM foil (left) and of a Zr-coated SLM foil (right). The coating follows the surface topography of both foils.

The influence of the gradient foil production process on the characteristics of the foil are non-negligible for the foil itself and for the C2TWP process. The machined foils present a smooth surface with a regular Zr layer but induce loss during processing. Such losses, once a U-Mo alloy is used, must be taken into consideration. On the other hand, SLM foils show irregular surfaces despite post-processing, together with slight bending in the large-scale design (Figure 3). However, while it presents a better yield than machining, it does involve a 3-step process.

It is important to note that this development is at the R&D stage and industrialization of these processes has yet to be assessed.

3. C2TWP process

Following preparation of the Zr-coated stainless-steel gradient foils at TUM, 13 mini-size foils are selected for cladding with AlFeNi alloy using the C2TWP process with four sets of parameters, as described in Table 1.

Batch	Trial number	Inert GRADIENT Foil No.	Foil manufacturing process	Parameter 1	Parameter 2	UT results
1	1	#207-SS316L-SLM	SLM + Shot peening	C2TWP 1	P1	NOK
	2	#217-SS321-M+	Machining	C2TWP1	P2	NOK
	3	#218-SS321-M+	Machining	C2TWP2	P2	NOK
2	4	#237-SS321-M+	Machining	C2TWP3	P3	OK
	5	#220-SS321-M+	Machining	C2TWP3	P3	NOK
	6	#239-SS321-M+	Machining	C2TWP3	P3	NOK
	7	#210-SS316L-SLM	SLM + Shot peening	C2TWP3	P3	NOK
	8	#115-SS316L-SLM	SLM + Shot peening	C2TWP3	P3	NOK
	9	#109-SS316L-SLM+HIP	SLM + Shot peening + HIP	C2TWP3	P3	NOK
	10	#301-SS316L-SLM	SLM	C2TWP3	P3	NOK
	11	#302-SS316L-SLM	SLM	C2TWP3	P3	NOK
	12	#303-SS316L-SLM	SLM	C2TWP3	P3	NOK
	13	#234-SS321-M+	Machining	C2TWP4	P3	NOK

Table 1: Fabrication and C2TWP parameters of the inert mini-size gradient foils

The aim of the test was first to test the feasibility of the cladding process with gradient foils and then to select the best process conditions for the C2TWP and material preparation to be applied given these foil geometries.

The work on inert foils must make it possible to select standard parameters, which will be qualified using mini- and full-size DU coated foils.
All the inert mini-size plates were inspected by ultrasonic testing (UT) and one was cut for destructive examination. The results are presented below.

4. Inspection results

4.1 First batch

A first batch of 3 mini-size gradient foils was prepared by C2TWP processing to demonstrate the feasibility of the process for this specific design.
The Figure 4 presents the results of UT analysis of the three inert mini-size fuel plates with gradient foils.

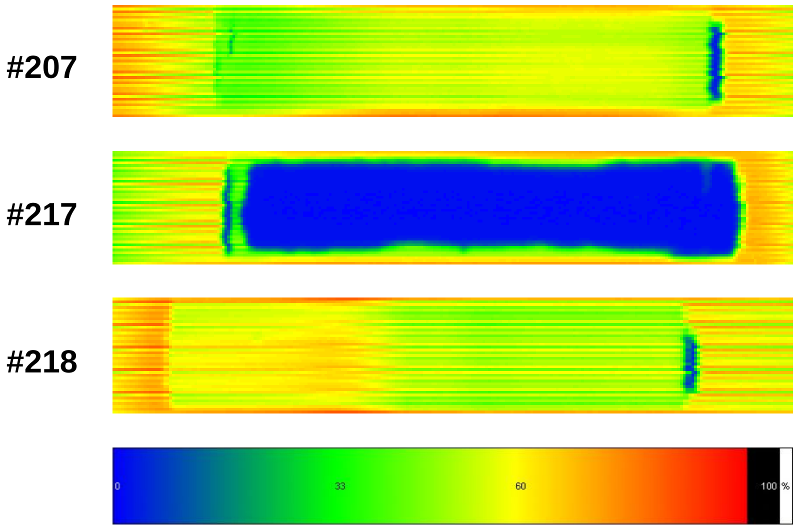

Figure 4: Results of UT inspections on inert mini-size gradient foils

Plate 217 highlights a large debonding area between the cladding and the foils, illustrated by the blue color. This full decohesion reveals issues in the foil itself and/or the C2TWP parameters used. Observing Plates 218, prepared using a foil machined in the same way as that in Plate 217, the first hypothesis is invalidated.

The UT results obtained on Plates 207 and 218 are thus more encouraging, showing better bonding, in green/yellow on the UT pictures, with some decohesion bands in the lengthwise direction, at the interaction surface between the foils and the cladding. On both, a radial debonding defect can be seen at the end of each plate, qualified as an end-gap, showing a lack of bonding between the two sides of the cladding.

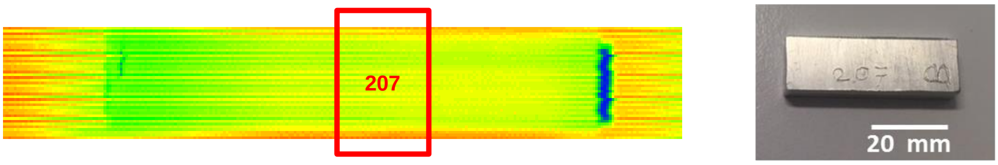

Figure 5: Position of Sample B on Plate 207

Following these UT observations, a metallographic analysis was conducted on a sample of Plate 207 (Figure 5). The aim was to check the quality of the bonding in an area where the UT signal shows good results and on the foil with the least favorable surface appearance (see Figure 3). Areas with defects were considered irrelevant and were excluded from the tests.

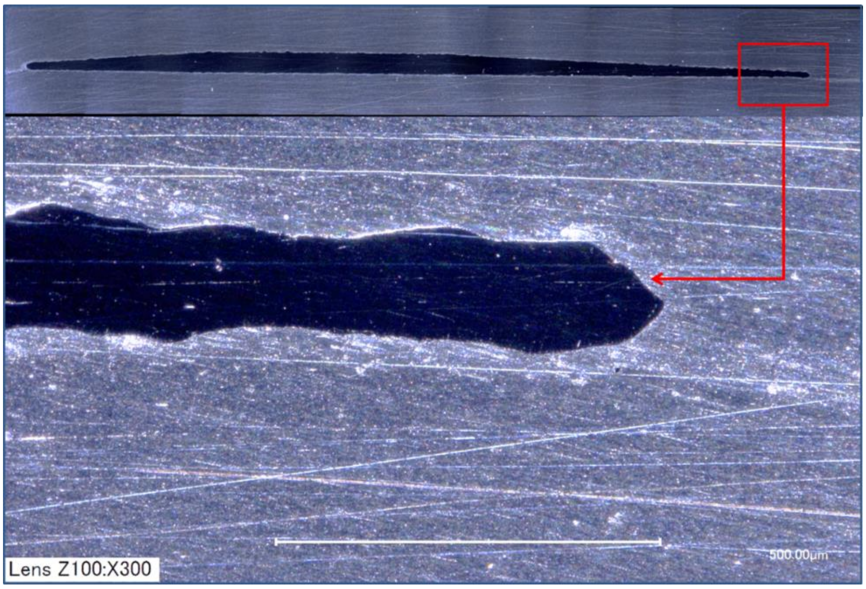

Figure 6: Optical micrography (x300) of Sample B of Plate 207

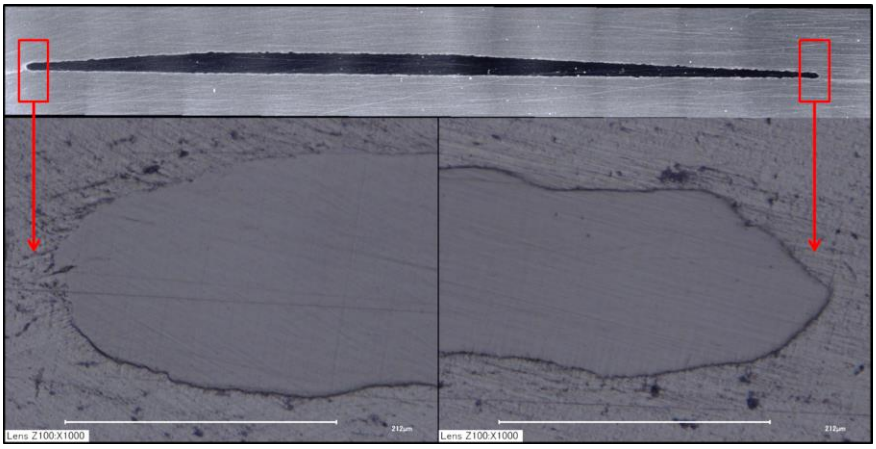

Figure 7: Optical micrography at higher magnification (x1000) of Sample B of Plate 207

As illustrated in Figure 6 for the bonding of cladding on a rough surface, and in Figure 7 for the bonding at the lateral edges of the foil, the metallographic characterizations of Plate 207 highlight satisfactory bonding on promising UT areas.

Both machining and SLM + shot peening processing result in satisfactory bonding between the cladding and the foil, even with variations in surface quality, and so seem to be acceptable as a basis for the C2TWP process.

These preliminary results enable us to further investigate the C2TWP process and material preparation parameters in order to improve and resolve the various defects in future trials. Tests on a second batch have therefore been conducted to improve the quality of the cladding around the mini-size gradient foil.

4.2 Second batch

The objective of this second batch was to select the best process parameters for the C2TWP and material preparation applied to the mini-size gradient foils. After applying the cladding, the inert mini-size plates were examined by UT (
Figure 8) and radiography, and one plate was cut for destructive examination.

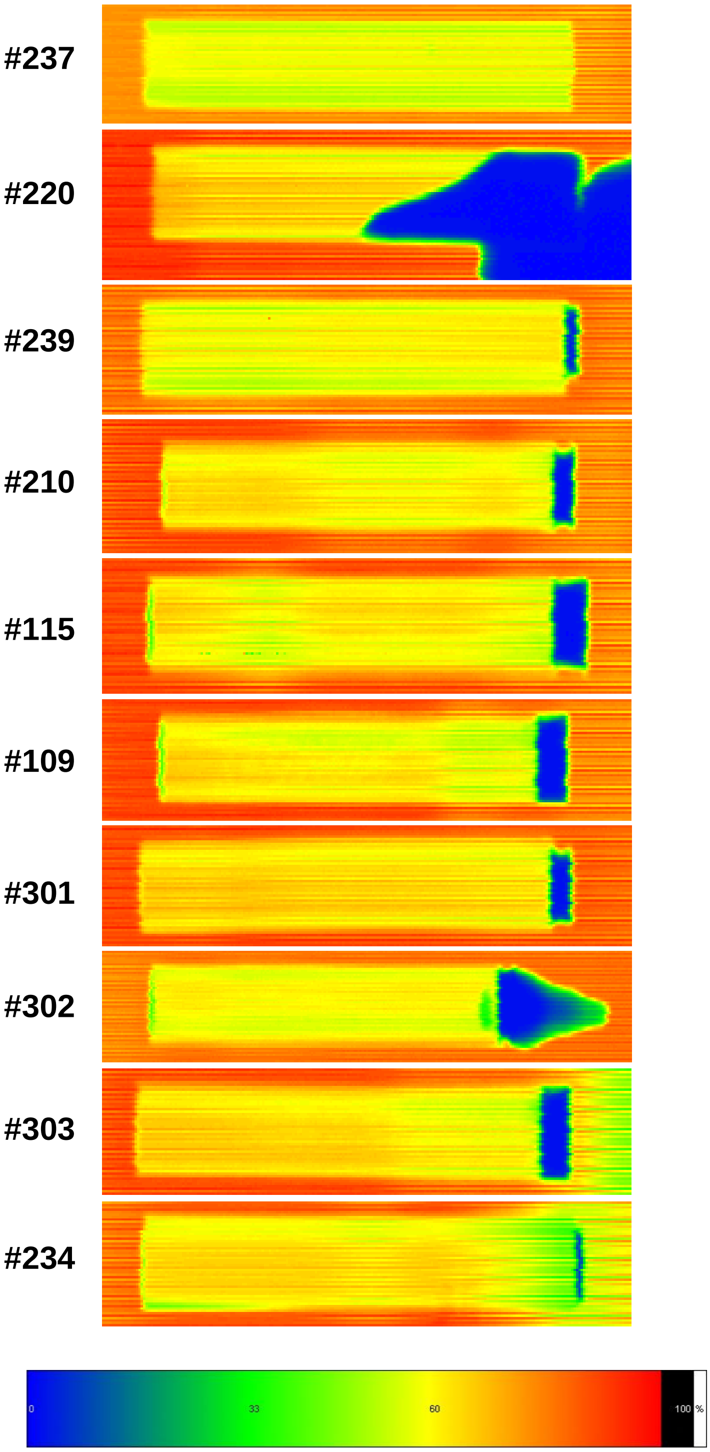

Figure 8: Results of UT analysis on inert mini-size fuel plates with gradient foils

As for the previous batch, ultrasonic testing showed that the foil manufacturing process chosen (i.e. machining, SLM or SLM + shot peening) has no impact on the cladding bonding.
As illustrated, and with the exception of one plate (#220), all plates show a similar result for the bonding, with good cohesion between the foil and the cladding. However, all plates show a UT defect at the plate ends, apart from Plate 237, which presents no defect across the length or breadth.
Whatever the manufacturing process used for gradient foils and the C2TWP process parameters, a slight gap is observed at the end of the process.

Through the results of the Plate 237, it is so technically feasible to reach mini-size plates without defect using a gradient foil. It is necessary to work on the process to repeat and confirm the result.

The larger gaps observed on Plates 115 and 109 are linked to the length of the gradient foil, which was shorter than expected. However, this does not explain the other cases.
Compared to flat foils [17], the differences in behavior between the materials used for the gradient foils and for the cladding have a more significant influence. The maximum thickness of the gradient foils is more than double that of the flat foils.

To adapt the parameters to these variations in the mechanical properties and resolve the end-gap issue, other C2TWP process parameters must be tested.
However, any improvement or adjustment regarding the C2TWP parameters for this specific type of material will have to be adapted for U-Mo foils. It could be interesting to proceed with uranium alloy tests rather than develop a set of parameters that will not be applicable.

Figure 9 illustrates the radiography results for 4 of the 10 inert mini-size fuel plates with gradient foils used for Batch 2 following C2TWP. This examination was used in addition to UT analysis to inspect foil integrity following C2TWP processing (i.e. cracks, other defects etc.).
All radiography examinations of the fuel plates were similar, and none showed any cracks or other defects on the foils.

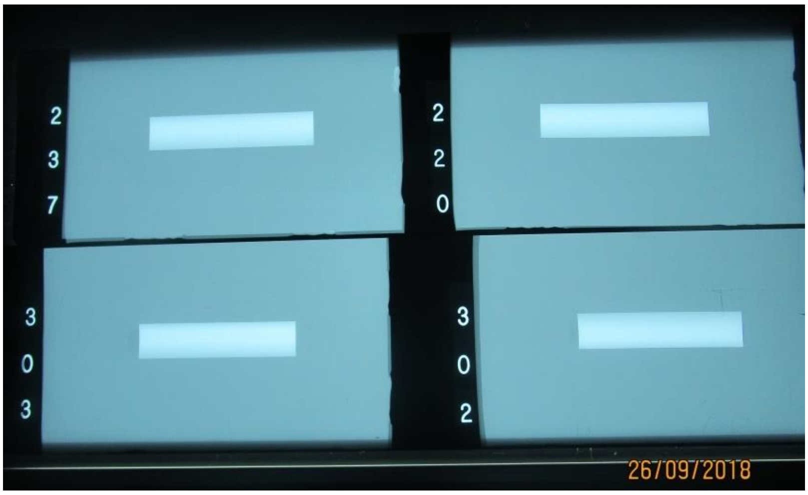

Figure 9: Radiography results for inert mini-size fuel plates with gradient foils No.237, No.220, No.303 and No.302

Further metallurgical observation did not provide any other conclusive information compared to that conducted on samples in Batch 1. Other than the end gap observed on all the foils, the bonding on the foils and at the lateral edges was consistent and without defect. No conclusive information could be drawn from these observations.

However, even with the radial decohesion defect observed by UT, the satisfactory enclosure of gradient foils in aluminum cladding by C2TWP process is feasible.

5. Conclusion

Framatome-CERCA™ and TUM are working together to implement a U-Mo monolithic fuel fabrication line in Europe. This project is funded by TUM and supported by the HERACLES consortium, and is included in the EU-HPPR conversion roadmap.

In this paper, we report on the status of development work on U-Mo gradient plates. The capability of the C2TWP process to produce plates from gradient foils has been demonstrated at the R&D stage. However, it has been shown that several improvements are required to obtain a foil without any cladding defects. This has been tested on mini-size plates, and transitioning to half-size or full-size plates, for inert or U-Mo materials, will require the parameters to be adapted.
It has been shown that, whatever the foil manufacturing technique used (i.e. machining, SLM with or without shot peening and HIP), the cladding bonding meets the specifications regarding UT analysis. The end-gap defect is mainly caused by the behavior of the material used for the foil. Development on full-size inert gradient foils is required before tests and validation on DU-Mo coated mini-size and then full-size gradient foils. Optimizing the C2TWP process should resolve the few residual bonding defects at the fuel plate ends.
The results obtained so far are promising. It has been showed that through numerous tests and modifications on the C2TWP process and parameters, mini-size plates that reaches specification are feasible, such as the plate #237. It is so technically feasible to obtain monolithic plates with gradient foils.
However, similar to the work done for plates with flat foils, transition from mini-size to full-size plates will require other studies.

In parallel, the difficulties involved in obtaining gradient foils using U-Mo, the coating of such gradient foils and the uncertainties regarding the behavior of coated U-Mo gradient foils means that it is necessary to continue working on these topics and on other solutions.

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