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Development of Aluminum-Clad Fuel Plate Processing Through Canned and Canless Hot Isostatic Pressing (HIP), and Studies of Aluminum Cladding Grain Growth during HIP

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ABSTRACT

The optimization of HIP processing of aluminum cladding for LEU-10Mo monolithic fuel plates is discussed. Canned HIP process optimization focuses on reduction of materials usage and processing effort, while improving the final HIPed product. Small scale formed sheet-steel HIP cans have been modeled, designed, and produced that significantly reduce production costs and resources. A new canless HIP approach to fuel-foil production is being developed at LANL, avoiding the need for protective stainless steel canisters. This process involves hermetically sealing the outer perimeter of individual fuel foils using electron beam welding. Details of this process and discussion of the challenges encountered will be given. Grain growth across the aluminum clad-clad interface is desirable. It was characterized using electron backscatter diffraction as a function of process modifications including macroscopic grooving to enhance Al flow, changing the cleaning method, and adding cold work. The results of these initial studies will be presented.

1. Introduction

The purpose of the Department of Energy's National Nuclear Security Administration's Office of Global Threat Reduction's Conversion program, formerly known as the Reduced Enrichment for Research and Test Reactors (RERTR) program, is to work with research reactors operators worldwide in an effort to convert reactors from the use of highly enriched uranium (HEU) fuel to the use of low enriched uranium (LEU). In support of this effort, the Convert program is working to develop new fuels to allow for the conversion of high performance research reactors worldwide.

In order to provide LEU fuel plates to meet the requirements of research reactors, monolithic fuel foils are used in place of dispersion fuels, which significantly change the manufacturing processes required to produce fuel plates. A significant step in the process is the bonding of aluminum cladding (alloy 6061) to an LEU-10Mo fuel foil that has been co-rolled with a thin zirconium diffusion barrier.

The purpose of the current work is to optimize the HIP process for producing monolithic fuel plates, and examine the resulting aluminum clad-clad interface, produced at the edges of the fuel plates and critical to fuel plate performance. The results of initial studies will be presented in three sections: canned HIP optimization, canless HIP processing, and Al-cladding interface grain growth enhancement. Canned HIP process optimization focuses on reduction of materials usage and processing effort, while improving the final HIPed product. Small scale formed sheet-steel HIP cans have been modeled, designed, and produced that significantly reduce production costs and resources. A new canless HIP approach to fuel-foil production is being developed at LANL, avoiding the need for protective stainless steel canisters. This process involves hermetically sealing the outer perimeter of individual fuel foils using electron beam welding. Details of this process and discussion of the challenges encountered will be given. Grain growth across the aluminum clad-clad interface is desirable. It was characterized using electron backscatter diffraction as a function of process modifications including macroscopic grooving to enhance Al flow, changing the cleaning method, and adding cold work.

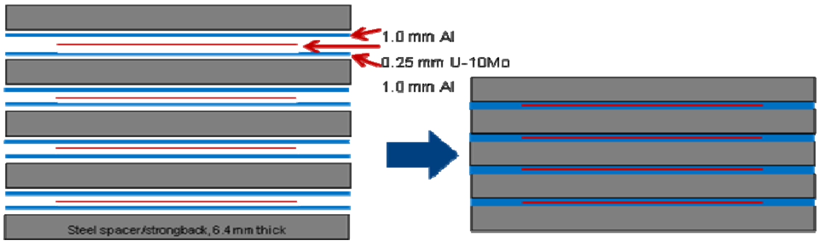
2. HIP Can Optimization

The initial experiments for hot isostatic pressing for the production of monolithic fuel plates were performed at the Idaho National Laboratory [1, 2]. The TIG-welded steel can used to contain the fuel plates during HIP processing was initially designed for and successfully used to produce sub-sized plates. The original HIP can design has recently been successfully scaled-up to produce high-quality full-scale aluminum-clad U-10Mo monolithic fuel plates [3], as shown in Figure 2.1(a). The goal of the current work is to further develop and optimize the HIP can design with the following primary goals:

- Maintain or improve HIP processing effectiveness (stress distribution).
- Reduce material usage, cost, and waste.
- Improve accuracy and repeatability of can dimensions for ease of assembly, improved manufacturability, and best possible fuel plate dimensions.
- Eliminate machining of HIP can parts.
- Minimize amount of welding required.

In order to better understand the stress distribution in the original can design, finite element (FE) modeling was performed [4]. Figure 2.1(b) shows a transverse section the stackup inside the original can design, with sturdy gray steel plate strongbacks, blue aluminum cladding, and red Zr co-rolled U-10Mo fuel foils. It is desirable to produce a uniform stress across the aluminum-aluminum and aluminum-fuel plate interfaces to create consistent bonding conditions. Figure 2.1(c) shows the calculated stress distribution in a transverse section (half shown for symmetry) of the original can design, and suggests that the stresses across the stackup are generally very consistent, varying only approximately 10% from the center of the fuel plate (left side of the image) to the edge of the fuel foil (arrow). The stress distribution toward the edge of the can suggests that the 3 mm-thick can wall and significant weld tabs at the corners of the can may be too rigid, and results in stress shadowing at the edge of the fuel plates, where the critical aluminum-aluminum bond is produced. An optimized version of the current can would be designed with less-rigid walls, which would also reduce the amount of material required to produce the can. In addition to the possibility of improving the stress distribution, it would be advantageous to significantly reduce the amount of welding required to produce the can, which would reduce time and cost, but would also improve dimensional repeatability.

(a)



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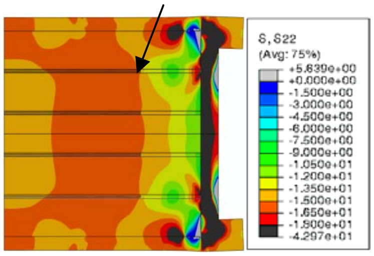
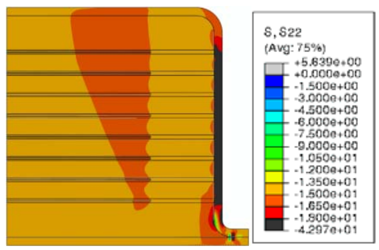


(c)

Figure 2.1. (a) Full-scale (610 mm tall) original design HIP cans prior to HIP processing. (b) Schematic illustration of strongback-fuel plate stackup used within HIP cans. (c) Finite element (FE) analysis results of stress distribution at HIP temperature and pressure (560°C, 103 MPa).

To achieve these goals, a formed HIP can design is proposed. An example of a scaled-down (15 cm long) formed HIP can is shown in Figure 2.2(a). The forming process allows the use of a single sheet of mild steel to produce the base of the can, which can be filled with an appropriate stackup, and evacuated and sealed by welding a flat lid. This method eliminates welding before filling the can, which reduces dimensional variations, and minimizes the amount of welding required to seal each can. In addition, the forming method allows for the use of much thinner steel sheet, which both reduces waste and makes it possible to use alternative welding techniques which can improve productivity. First, however, the proposed can configuration was modeled by FE analysis to determine if the stress distribution would be acceptable relative to the original can design. Figure 2.2(b) shows the results of the FE modeling, which predicts an improved stress distribution, which may lead to improved final HIPed fuel plates, but will at the very least result in a more repeatable process.

(a)

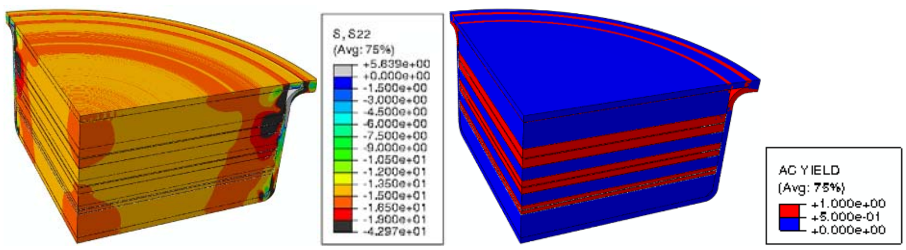


(b)

Figure 2.2. (a) Scaled-down (150 mm long depression) version of proposed formed HIP can. (b) Results of FE analysis of stress distribution in a formed can, showing reduced stress gradients relative to the original can design, and suggesting the formed can is a viable option for HIP can optimization.

The encouraging results from FE modeling of the formed HIP can led to the first experiments, which were performed on round formed cans (90 mm diameter) using relatively thin (1.5 mm) steel sheet. Figure 2.3(a) shows the formed can and stackup (left image) after the application of the parting agent (MoS_2 aerosol spray) and ready for assembly and welding. Three of these round formed HIP cans have been successfully processed. Figure 2.3(b) shows the first formed HIP can, which was sealed by electron-beam welding, after HIP processing. The can deforms freely to compress the internal stackup, resulting in excellent final dimensions of the three plates in each can, which varied less than 0.025 mm in thickness in any of the cans processed. Further modeling of the round can design with the same internal stackup and gaps resulting from forming radii, as shown in Figure 2.3(b), still shows excellent stress distribution (left image), and results in plastic deformation of the aluminum plates, but only elastic deformation of the strongbacks, as is desired (right image). In addition to testing the round can design concept, TIG welding and electron-beam welding was used to seal a formed round HIP can, with similar results. Finally, various thicknesses of strongback were used, some as thin as 3 mm, and were found to work suitably and be reusable with the formed HIP can design, likely because of the improved stress distribution. Thinner strongbacks will result in the increase in the number of plates that can be processed per can, further reducing cost and waste. Based on the initial modeling and experiments, we find that the formed hip can approach is an excellent way to achieve the goals of improved stress distribution, considerably reduced cost, greater dimensional control, elimination of machining, and a significant reduction of welding, while improving the overall product.

(a)



(b)

Figure 2.3. (a) A round formed HIP can and stackup (9 cm diameter) immediately prior to assembly and welding (left), along with the same can after HIP processing. The stackup consists of two 6 mm thick strongbacks and two 3 mm thick strongbacks separating three plates of Al-Zr-Al with total thicknesses of 4.5, 3.0, and 1.8 mm. (b) FE analysis of the can shown in (a), indicating excellent stress distribution (left) and the desired plastic deformation (right) of the aluminum cladding and elastic deformation of the strongbacks.

Further testing is underway to examine the performance of formed HIP cans with wall thicknesses of approximately 0.5 mm, cans with a rectangular shape with improved proportions relative to the plates used in research reactors, and scale-up to produce full-sized fuel plates.

3. Canless HIP

Some of the detracting aspects of associated with the traditional canned HIP approach include i) excessive time required to hand construct and disassemble each can, ii) the possibility for bonding between foils and backing plates, and iii) the generation of large volumes of contaminated waste (can material and backing plates) that must be disposed. The latter factor has very important cost implications, where it has been estimated that there will be between 1,500 and 4,200 liters of waste generated per year with millions of dollars in disposal cost.

The new “canless” HIP approach discussed here avoids altogether the need for canning prior to HIP processing. It involves the hermetic sealing of individual foil assemblies by electron beam welding around their outer perimeter. This is accomplished by making 4 intersecting aluminum-to-aluminum, full-penetration lap welds as shown in Figure 3.1. Since welds are made in a hard vacuum (10^{-5} torr), there is no need for a separate air evacuation step. The fuel itself sits within a pocket machined into one of the aluminum plates.

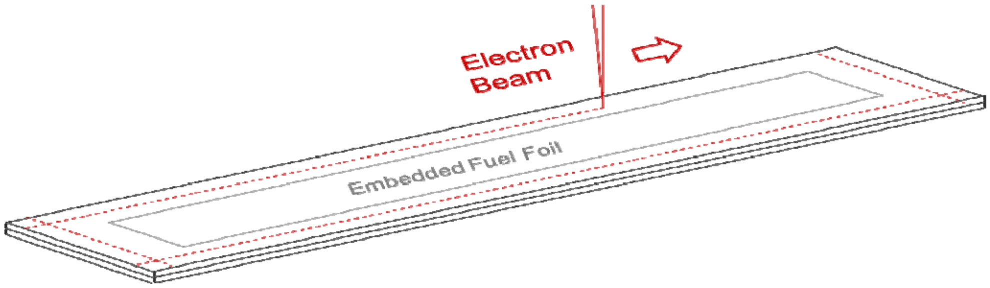


Figure 3.1. Schematic showing 4 electron beam lap welds made on aluminum plate along outer perimeter of fuel foil assembly.

One of the problems encountered when making these welds has been a divot defect involving an occasional disruption in weld pool continuity and solidification cracking across the lap joint (see Figure 3.2). Divots can be observed from the top of the weld, with the weld bead appearing broader and having less penetration. Its origin is not yet understood, nor is its affect on HIP bonding. However, ongoing work is being performed to address this concern and mitigate or avoid its formation. Initial results suggest that this problem can be mitigated simply by welding over the defect. The nature of the problem may be related to inadequate clamping force in the welding fixture.

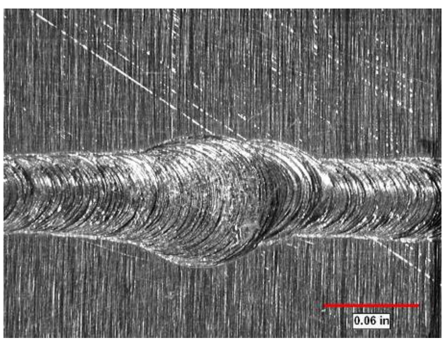


Figure 3.2. Weld divot as seen from top view of electron beam weld (left) and cross-section showing solidification cracking across the lap joint (right).

Multiple canless foil assemblies can be placed in a HIP fixture and batch processed. It is necessary that foils hang freely during the high temperature HIP cycle to allow unimpeded thermal expansion. A fixture proven to do this is shown in Figure 3.3. Foils that have been HIPed in this manner have considerable distortion after being HIPed as shown in Figure 3.3. This distortion is likely due to stresses associated with differences in thermal expansion between aluminum and stainless steel for these 610 mm x 95 mm plates.

In summary, the canless approach to fuel foil HIPing shows considerable promise for cost savings in labor and contaminated waste disposal. This process has been successfully scaled-up from 90 mm x 90 mm square coupons to 610 mm x 95 mm plates. A weld defect (divot) is occasionally encountered that can be mitigated by rewelding. Despite welding issues, the examples that have been HIPed to date suggest the e-beam process can produce hermetically sealed plates. HIPed foils have considerable distortion, which is a problem that must be addressed either with modified HIP fixtures or post-HIP processing.

Figure 3.3. Canless foil top-mounted to HIP fixture prior to HIP processing (upper left and image) and after HIP processing (upper right image), also showing two round formed HIP cans on base plate. Lower images show plates after HIPing and details of e-beam weld. Plates are approximately 610 mm long.

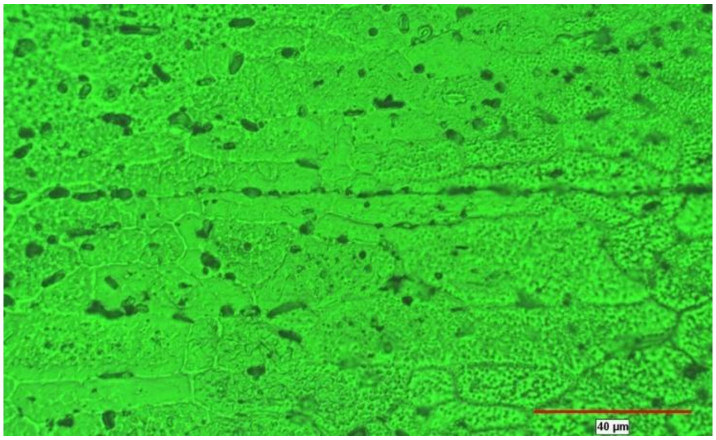
4. Al Cladding Grain Growth

The clad-clad bond in plate-type fuels provides for primary containment of fission products and is therefore important in safety analyses. The amount of grain growth (penetration) observed across the clad-clad interface in dispersion fuel plates fabricated by the roll bonding process has been a long-standing criteria used in manufacturing process qualification and periodic process inspection. The acceptance criteria for fuel plates manufactured by the roll bonding process varies from reactor to reactor, but in general, the "metallurgical bonding" requirement has historically been associated with the observation of anywhere from >20% to >50% grain growth across the bond line length, as determined by detailed metallographic inspection. These interface grain growth levels are routinely achieved when proper Al cleaning procedures are followed and sufficient thickness reduction ratios are employed in the hot rolling process.

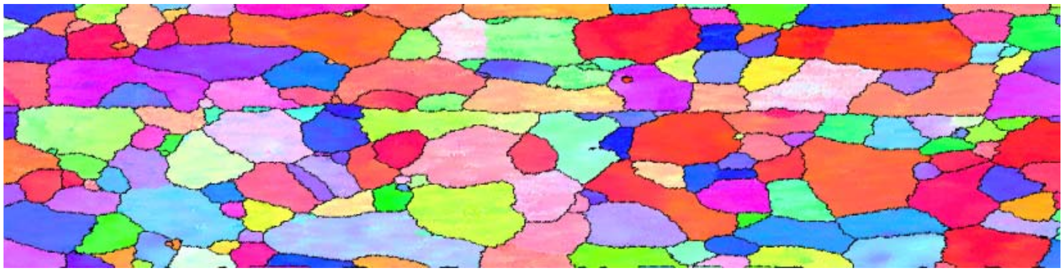
Monolithic fuels are fabricated by the HIP bonding process are characterized by a significantly different thermomechanical history compared to roll bonded dispersion fuels. Specifically, during HIP bonding there is no large scale thickness reduction to drive recrystallization and grain growth, and metallurgical bonding occurs primarily via diffusional and creep mechanisms. As might be expected, there is substantially less interface grain growth is present in HIP bonded fuel plates compared to roll bonded fuel plates, values approaching to zero percent of the bond line.

The purpose of this work is to increase the amount of grain penetration growth across the clad-clad interface in monolithic fuel plates fabricated by the HIP process. The grain penetration is considered to result from the *growth* of Al grains (initially present or recrystallized) that originated on either or both of the Al mating surfaces.

Orientation Imaging Microscopy (OIM) was selected as the "gold standard" for quantifying grain penetration, and was used to examine long stretches of bond line (5-25 mm). Light Optical Microscopy (LOM) does not reveal grain penetration well, but is helpful in that it shows second-phase particles at interface.



(a)



(b)

Figure 4.1. Bond line region from **control condition with no deformation and LANL cleaning**, imaged by (a) LOM and (b) OIM. The bond line runs horizontally across the center of both images, and is decorated by second-phase particles in (a). No grain growth either across or in the vicinity of the bond line is evident. The OIM image is 400 microns wide.

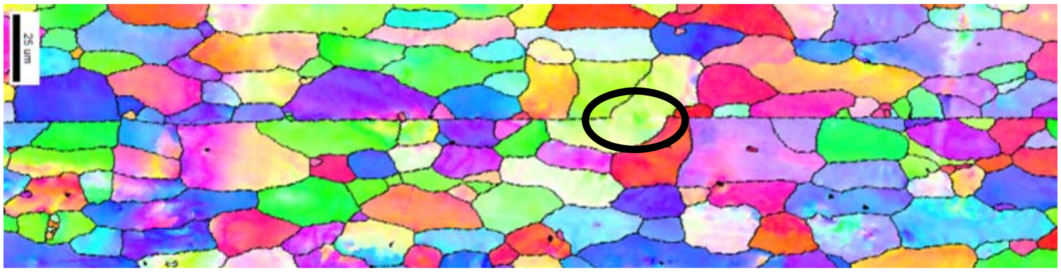


Figure 4.2. Bond line region from **control condition with no deformation and B&W cleaning**, imaged by OIM. One of the three of fields of view exhibiting isolated areas of grain are shown here; the other 15 OIM fields of view had no grain penetration. The OIM image is 400 microns wide.

Bonding in Al-Al sandwiches were examined (no fuel or surrogate) as a function of modifications that were done to the 6061-Al *upstream* of the HIP cycle. The HIP cycle was left unchanged (560ºC for 90 minutes at 103 MPa). All modifications used the Babcock & Wilcox (B&W) cleaning process (Proceco cleaning, then HNO_3/HF acid pickling). As a control, we also examined the legacy LANL cleaning process (40% HNO_3 acid + 1% NH_4HF_2, then 20% NaOH, then repeat.) Three hermetic sealing methods: were used: 610-mm cans, 100-mm round cans, and 100-mm canless squares. These procedures were described in more detail earlier in this report.

Two control conditions were first examined. These involve Al that was cleaned by one of two aqueous processes but with the Al metal otherwise unaltered by deformation or other means. Figures 4.1 and 4.2 show the results. Very little grain growth was found in either instance (LANL – 0.4%; B&W – 1.8%). The second phase precipitates/dispersoids coating the bond line are unhelpful since they reduce metal-metal contact. These results indicate that improved aqueous cleaning is insufficient to generate adequate grain growth.

Table 4.1. Summary of results and other approaches currently under investigation

Strategy (physical mechanism)	**Processing modification**	**Results and status**
Cleaning oxide and debris off aluminum surface (increases metal-metal contact)	LANL cleaning process - no deformation	0.4% grain growth.
	B&W cleaning process - no deformation	1.8% grain growth.
	Transferred Arc (TA) cleaning + electrospark deposition of pure Al	In progress.
Macroscopic grooves (shearing breaks up oxides & deforms Al)	Machine grooves in Al plates at strategic locations. Informed by finite element modeling.	0% grain growth far from grooves. OIM of regions at grooves in progress.
Al metal flow at contact points of **engineered asperities**	Knurling	On hold for later.
	Steel wool (grade 0)	In progress.
	Coarse sanding (180 grit)	Not promising.
Recrystallization & grain growth from **stored energy**	Cold rolling (63% reduction)	In progress.
	Bead blasting (glass beads)	In progress. Early results promising.

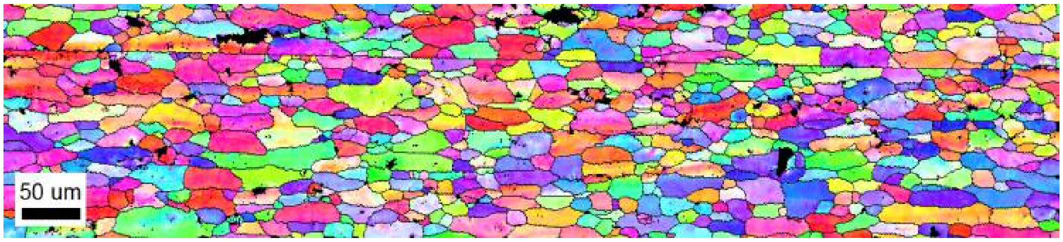


Figure 4.3. Bond line region from macroscopically grooved sandwich #1 (0.127-mm deep, 9.8-mm wide channels) imaged by EBSD. This fields of view was away from the grooved regions. No grain growth is visible in this or the other two grooved sandwiches.

Given the foregoing results, process improvement studies were undertaken. The focus was on changes that can be practically and economically implemented in the production environment. The strategies are summarized in Table 4.1. Of these, only limited results are available for the macroscopically grooved sandwiches (Figure 4.3). Among the strategies, we contend that imparting stored energy is the most promising approach, since it directly provides a driving force for grain growth (like that imparted during roll-bonding of dispersion fuels). Initial results from bead blasting are promising.

From these bonding studies, the following conclusions can be made:
1. OIM is ideal for quantifying grain penetration across clad-clad interfaces.
2. Changing the aqueous cleaning, by itself, is insufficient to increase the grain growth
3. Other strategies (grooving, asperities, stored energy) are in progress.

5. References

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6. Acknowledgements

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