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**Scale-Up of the HIP Bonding Process for Aluminum Clad LEU Reactor
Fuel**

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ABSTRACT

A combined experimental and modeling approach was applied to the scale-up of the LEU reactor fuel plate bonding process. In this process Zr/U-10%Mo/Zr fuel foils are placed in 6061 aluminum cladding and HIPed to form LEU reactor fuel plates. The HIP can geometry and HIP process parameters will be reviewed and recent results for scale-up of this technique will be discussed. Metallographic information for the HIP bonded fuel plates and preliminary results for the mechanical properties of the bonds formed during the HIPing process will also be presented.

1. Introduction

This work reports on the scale-up of HIP cladding, one of the key steps in the manufacture of monolithic LEU reactor fuel[1-4] developed by Idaho National Laboratory (INL). Monolithic LEU reactor fuel is being developed as part of the Global Threat Reduction Initiative U.S. High Performance Research Reactor (HPRR) Conversion Program. The HPRR Conversion Program is managed by U.S. Department of Energy (DOE).

Figure 1 is a schematic of the INL HIP can design for mini fuel plates. A typical HIP can contains an assembly of 5 strongbacks and 4 aluminum clad LEU fuel foil assemblies arranged as shown in Section A-A and Detail B of Fig. 1. Figure 2 is a schematic of the aluminum cladding and monolithic LEU foil fuel assembly, which is HIPed to create a fuel plate. Figure 3 shows a typical HIP can, number 12, after HIPing. Four cans are shown ready to HIP in Fig. 4.

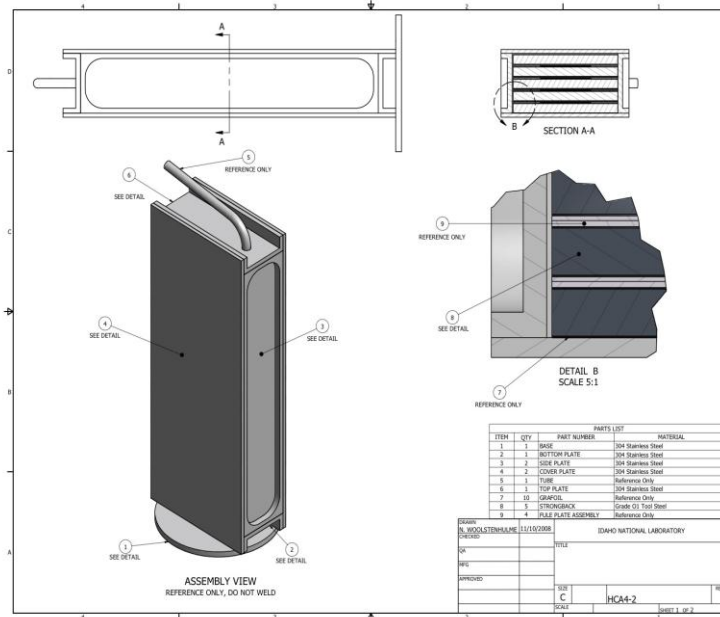
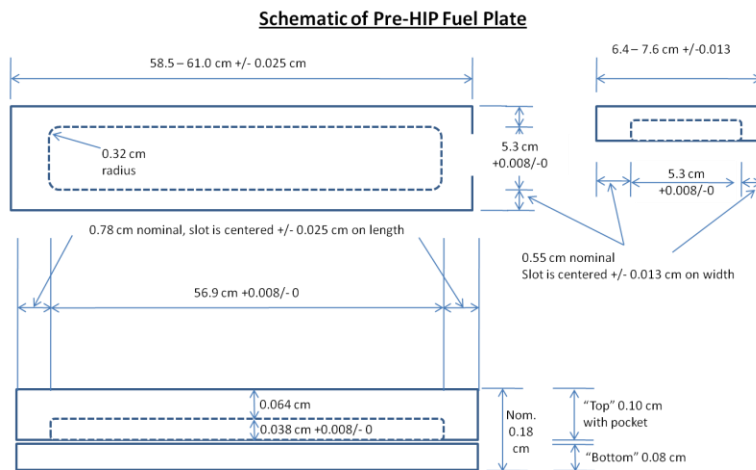


Figure 1. Diagram of the INL mini fuel plate HIP can.



For: HIP Can HCA-4-X-1
 Date: 5-10-2010
 Revision Number: 1
 By: K. Clarke

Material: 6061-T6 aluminum sheet

Figure 2. Schematic of aluminum cladding configuration for scaled-up HIP can used by LANL. LEU fuel foil is inserted into the dotted area.



Figure 3. A typical scaled-up HIP Can (#12) after HIPing.

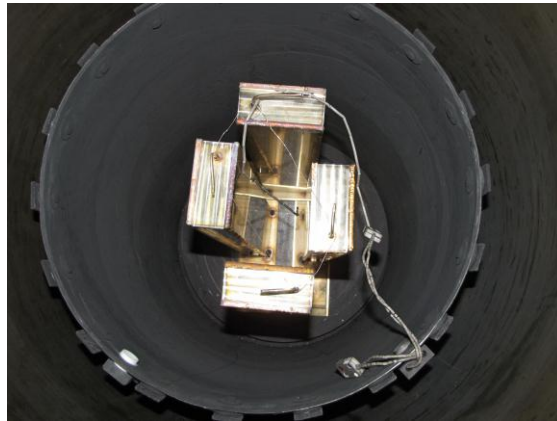


Figure 4. HIP cans 8, 9, 10 and 11 loaded into the HIP.

2. Initial Scale-up Experiments

LANL's initial philosophy was to scale-up the HIP process to produce full size fuel plates keeping everything identical to INL practice. The HIP can and fuel cladding were simply expanded 4 times. All HIP cans prepared by LANL except 13, 19, 20, 21 and 22 have internal dimensions of 61 cm long x 7.6 cm wide x 3.8 cm thick. Cans 13, 19, 20, 21 and 22 have internal dimensions of 66 cm long x 12.7 cm wide x 3.8 cm thick. The same stainless steel (304L) and aluminum alloys (6061-T6) were used for the can and cladding. The same parting agent, Grafoil^ζ was also used. The only appreciable changes to the INL practice for the initial experiments were the use of 304L stainless steel strongbacks instead of type O1 tool steel and the thickness of the Grafoil parting agent. For the first 4 experiments HIPing was performed at 138 MPa and 560°C with a 90-minute hold time. The heat-up and cool-down rates were approximately 280°C/hr.

^ζGrafftech International, Ltd., Cleveland, Ohio 44101.

Starting with experiment 5, the HIP pressure was reduced to 103 MPa; all other HIP cycle parameters remained the same.

The space between the HIP can sides and the aluminum cladding was also scaled-up from 1 to 6 mm on the sides and to 1.27 cm on the top and bottom. 1.27 mm thick Grafoil was used for the first HIP run, however, this did not allow sufficient room to stack five 0.64 cm thick strongbacks and 4 sets of aluminum cladding and fuel foils. Thinner 0.254 mm thick Grafoil was used for the next 3 HIP cans which were completed in FY10. After HIPing cans 2, 3, and 4 several problems were identified. First it was difficult to keep the cladding/fuel foil assemblies from moving around within the as-assembled HIP cans. Second, if the entire assembly of strongbacks, cladding/fuel foils and Grafoil was not thick enough, the fuel foils could move out of the pocket cut for it in the aluminum cladding. Third was the tendency of the Grafoil to react with the aluminum cladding. This caused problems separating the cladding from the Grafoil and caused surface finish problems on the cladding, which necessitated extensive sanding prior to machining. The fourth and last problem was that the edges of the strongbacks, at the 6 mm gap between the cladding and the can wall were deformed during HIPing. This last observation leads us to suspect that pressure was not uniform within the HIP can, likely as a result of the empty space in the can from fuel plates that are narrower and shorter than the strongbacks. Uneven pressure distribution within the can, has significant negative implications for dimensional uniformity of the finished plates, as well as, overall bond uniformity and quality.

Finite element modeling of the stresses in the can during HIPing was undertaken. Fig. 5 is a diagram of the calculated stresses and predicted displacement directions. Fig. 5 confirms that stress is not distributed uniformly within the HIP can. Force applied during HIPing is not evenly distributed in the empty space between the edge of the aluminum cladding and the can wall. Figure 6 is an end view of the strongbacks and cladding from can 5. The strongbacks are deformed as predicted by the model. On the basis of the finite element model and observations on the as-HIPed strongbacks and cans, some fundamental process changes were made.

3. FY11 HIP Experiments

Table 1 is a listing of all of the HIP experiments performed between October 1, 2010 and September 30, 2011, which will be referred to as FY11. Starting with HIP can 7, free space within the HIP can was minimized. The cladding was cut to full size so that it fit as tightly as possible within the HIP can. The full height of the can was filled using 304 stainless steel spacers when necessary to take-up any excess space. This improved the transmission of pressure and the uniformity of its distribution within the can. Additionally, cutting the cladding to full can size prevented it from moving around in the HIP can during handling and filling the full height of the HIP can prevented fuel foils from moving out of the pocket in the aluminum cladding solving these two problems. Filling as much free space as possible resulted in very even pressure transmission and flat and straight cladding and strongbacks. Some representative aluminum clad fuel plates and accompanying strongbacks are shown in Fig. 7.

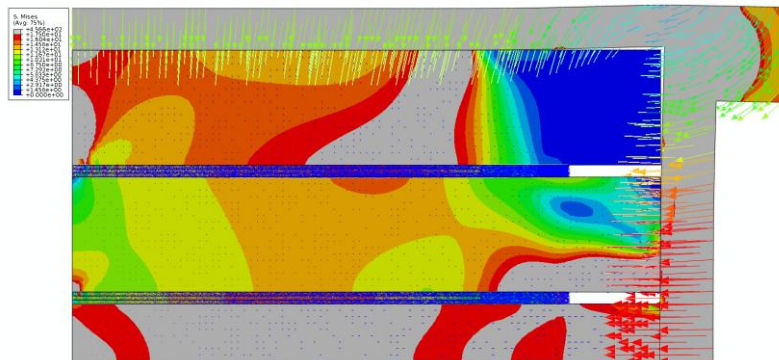


Figure 5. Diagram of calculated stresses on a HIP can during HIPing. Note bending stresses in corner of HIP can. The key is at the top right corner. Gray is the highest stress level; blue the lowest. Arrows indicate the displacement forces on the can.

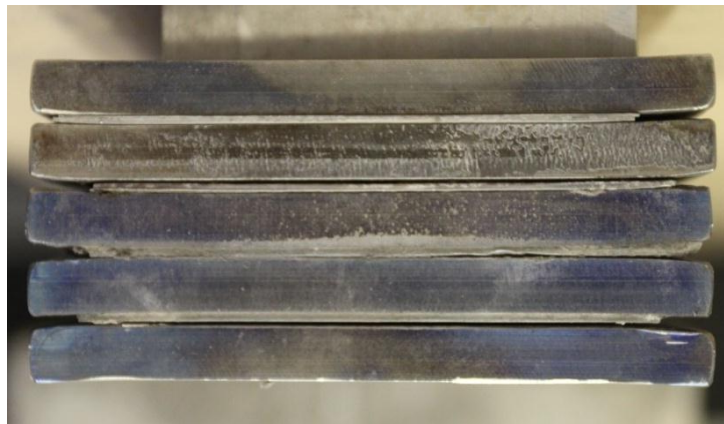


Figure 6. End view of strongbacks from HIP can 5. Note distorted edges of strongbacks where they are not supported by the aluminum cladding.

It was readily apparent from examining as-HIPed aluminum clad fuel plates that the bonding mechanism is extrusion of molten metal. The HIPing temperature is 560°C, which is 20°C below the liquidus temperature of the 6061 aluminum alloy used for the cladding. This temperature is sufficient so that the molten aluminum is extruded into any remaining free space. Figure 8 shows the extrusion of aluminum into the evacuation tube and into space between the strongbacks and HIP can.

Initially Grafoil was used as a parting agent. Grafoil was found to be totally unsuitable for this purpose as it reacts with molten aluminum. For HIP cans that used Grafoil, separating the aluminum cladding from the strongbacks was found to be a very difficult task requiring the use of prying tools and hammers. The use of prying tools resulted in gouged and bent cladding. Extensive sanding was also required to obtain a surface good enough for subsequent ultrasonic testing and finishing.

TABLE 1. FY11 HIP Experiments

HIP Can	Cladding (mm)	Foil	Strongback	Parting Agent	Status	Comments	Parameter
5	.76/1	.25 mm SS	As-received 304	BN, APA-3, APA-1, APA-2	Analyzed	Undersized cladding	Parting Agents
6	.76/1	.25 mm SS	As-received 304	Grafoil, MoS ₂ , Fiberfrax, Mo Neolube	Analyzed	Undersized cladding	Parting Agents
7	.76/1	none	Type 01 tool steel	Neolube	Analyzed	First use of Full-sized cladding	Full-sized cladding Strongback
8	.76/1	none	Straightened, Annealed 304	MoS ₂	Analyzed		
9	2.29/2.29	.50 mm dU	Type 01 tool steel	Neolube	Analyzed	Forming Test Samples	Foil/cladding thickness
10	1/1	.25 mm dU	Type 01 tool steel	Neolube	Analyzed	Forming Test Samples	Foil/cladding thickness
11	.76/1	none	As-received 304	Neolube	Analyzed		
12	.76/1	none	H13 tool steel	MoS ₂	Analyzed		
13	.76/1.27	SS	Type 01 tool steel	MoS ₂	Re-canned	Bulged during HIPing	Can Size (OSU Demo Can) Parting Agents
14	.76/1	none	Straightened, Annealed 304		Awaiting de-canning		
15	.76/1	.25 mm SS	Straightened, Annealed 304	MoS ₂	Awaiting de-canning	Bulged during HIPing	Reproducibility
16	.76/1	.25 mm SS	Straightened, Annealed 304	MoS ₂	Awaiting de-canning		Reproducibility
17	.76/1.27	dU	As-received 304		Awaiting Bake-out	Test Samples	Tack-welded Cladding Reproducibility
18	.76/1	.25 mm SS	As-received 304	MoS ₂	Ready to HIP		
19	.76/1.27	dU	Type 01 tool steel	MoS ₂	Awaiting Bake-out	OSU test plates	Reproducibility
20	.76/1.27	dU	Type 01 tool steel	MoS ₂	Awaiting Bake-out	OSU test plates	Reproducibility
21	.76/1.27	dU	Type 01 tool steel	MoS ₂	Awaiting top welding	OSU test plates	Reproducibility
22	.76/1.27	SS	Type 01 tool steel	MoS ₂	Awaiting top welding	OSU Demo Can Re-Do	Size
23	.76/1.52	.76 mm dU	S7 tool steel	MoS ₂	Awaiting top welding	Forming Test Samples	Foil/cladding thickness

The next parting agent evaluated was Neolube^δ. Neolube was found to be an improvement as hammers were no longer necessary. However, the improvement was limited, as prying tools were still needed to separate the cladding from the strongbacks. The as-separated cladding was still gouged and bent but to a lesser degree. Boron Nitride^ξ was found to be the best parting agent but was abandoned after only the initial trial because of the neutronic properties of boron. The next best parting agent was

^δ Huron Industries, Inc., Port Huron, MI 48060.

^ξ Combat Boron Nitride Aerosol Spray, Saint-Gobain Advanced Ceramics, Amherst, NY 14228.

found to be molybdenum disulfide^β (MoS₂). MoS₂ is easily applied as an aerosol and soon became the standard parting agent. We were consistently able to separate aluminum cladding from strongbacks without using tools; hence, the cladding was free of gouges and was not bent.

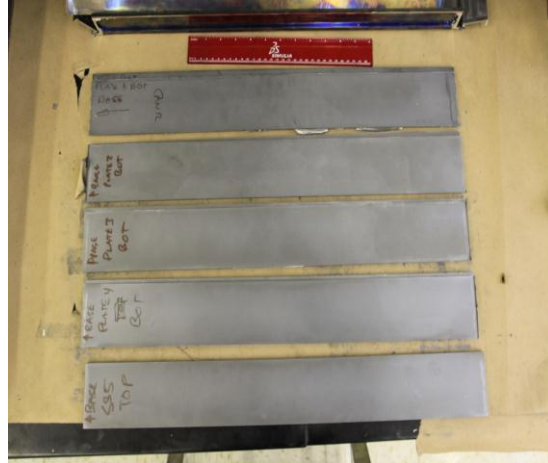


Figure 7. Surrogate aluminum fuel plates from HIP can 12.

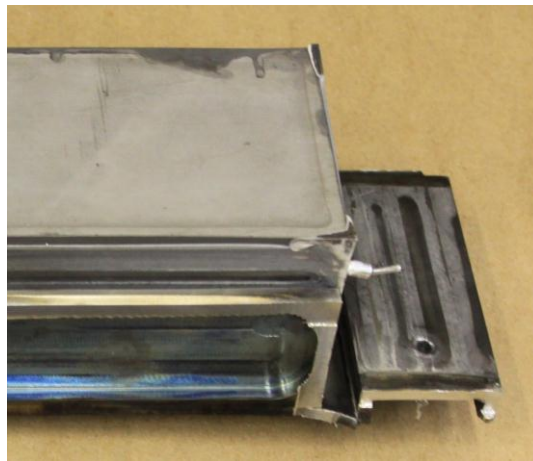


Figure 8. Side-view of strongbacks and aluminum cladding plates with top removed from HIP can 9. Note where molten aluminum was extruded into the HIP can evacuation tube on right side.

Numerous other parting agents were evaluated as well. Aerosol colloidal graphite lubricant from Alfa Aesar^φ was found to be promising and should be evaluated further. In addition, plasma sprayed coating of erbia, yttria and zirconia–24-wt% magnesia warrant further evaluation as strongback coatings. These plasma sprayed ceramics appear to work well, however, application is difficult and will need to be repeated before each use. Several parting agents were found to be totally unsatisfactory. These include

^β Molykote 321 Dry Film Lubricant Spray, Dow Corning, Midland, MI 48686.

^φ Alfa Aesar, Ward Hill, MA 01835.

a ceramic oxide felt from Fiberfax^γ and three oxide papers APA-1, APA-2 and APA-3 from Zircar^φ. Molten aluminum wets and infiltrates the oxide felt and papers making them impossible to separate from the cladding. Molybdenum foil was also evaluated as a parting agent and found to be totally unsatisfactory. The molybdenum reacted with the aluminum and could not be removed from the cladding.

Several types of strongbacks were evaluated. These included type O1 tool steel, S7 tool steel, H13 tool steel and 304 stainless steel in both the as-received and straightened and annealed condition. Prior to Can 5, all strongbacks were as-received 304 stainless steel. These early cans have a gap between the can sidewalls and the aluminum cladding of about 6 mm. It was common to find that strongbacks above and below the 6 mm gaps were deformed as shown previously in Fig. 6. After the switch to full-sized aluminum cladding the strongbacks did not deform significantly upon HIPing as shown in Fig. 9. The three types of tool steel (O1, S7 and H13) were received ground flat with square corners and an excellent surface finish. The finish of the 304 stainless strongbacks was much rougher and the corners are rounded. The 304 strongbacks tended to leave the as-HIPed cladding with “fishtailed” edges caused by the extrusion of aluminum into the space between the rounded corners. The “fishtailing” is shown in Fig. 9.

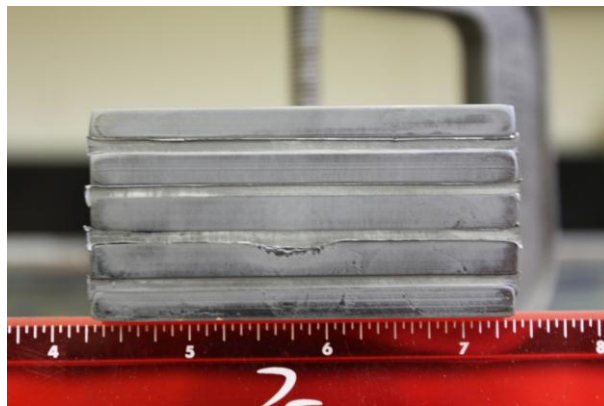


Figure 9. End-view of strongbacks and aluminum fuel plates from HIP can 8. Note fishtailing of fuel plates at edges.

All of the strongbacks tested were satisfactory and can probably be re-used, although re-use was not specifically evaluated. We did not determine any difference in strongback performance other than that due to surface finish. The differences in strength between the various types of tool steels and 304 stainless steel had no effect. The compressive strength of all of these steels is vastly greater than molten 6061 aluminum and when the free space in the HIP can is minimized the bending stresses responsible for deforming the strongbacks are eliminated.

^γ Unifrax, Inc., Niagara Falls, NY 14305.

^φ Zircar Ceramics Inc., Florida, NY 10921.

The bend test adopted by INL was performed on several as-HIPed surrogate fuel plates. The bend test consists of bending a strip of cladding 15.24 cm long by 0.95 cm wide, 90° over a 1.27 cm radius mandrel and then 180° in the other direction over another mandrel. The cladding strip is then examined for delamination. In all cases no delamination was observed. We then proceeded to bend the cladding strip back-and-forth 180° 18 times until it broke without delaminating. Figure 10 shows a bend test sample from HIP can 4, plate 1.

Extensive metallography was performed on fuel plates from HIP cans, 9 and 10, which contained 20 and 10 mil depleted uranium surrogate fuel foils respectively. Figure 11 is a composite of three micrographs merged to reveal a cross-section of plate 2 from HIP can 10. The cladding is flat and straight and the depleted uranium foil is well encased with no visible gaps. Figure 12 is a higher magnification micrograph of plate 3 from HIP can 10. In this micrograph the zirconium cladding is visible on the upper and lower edges of the dU foil. As in Fig. 11, the dU foils appears to be well bonded to the aluminum cladding; no gaps or voids are visible. The piece of aluminum in the bottom of the micrograph had the pocket into which the dU foil was placed; the top piece of aluminum was flat. Note the bond line on the right side of the micrograph extending from the top right corner of the dU foil in an arc.

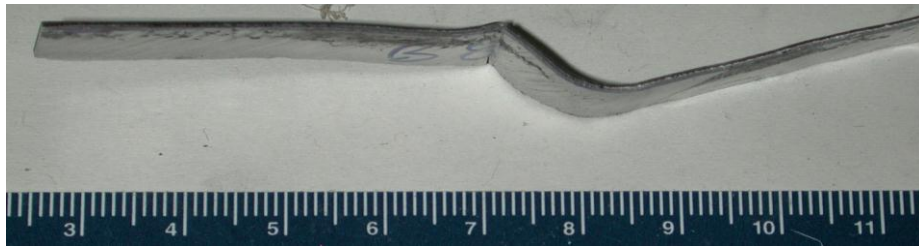


Figure 10. Bend test sample from plate 1 of HIP can 4 after performing 18 cycles of the INL bend test. Note that no delamination occurred prior to fracture through the entire thickness.

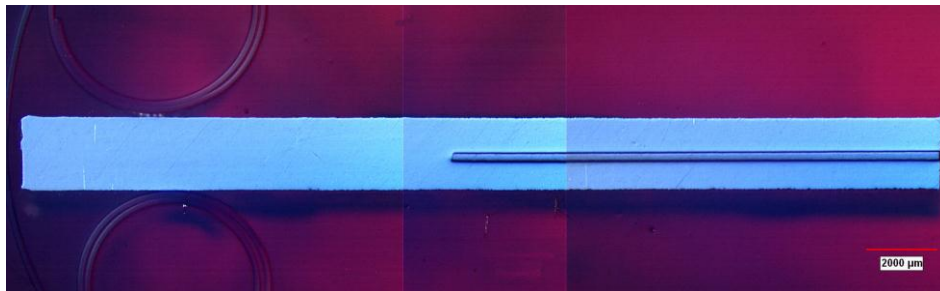


Figure 11. Composite micrograph of section from plate 2 of HIP can 10. 0.254 mm dU foil clad in 2.0 mm of aluminum. The cladding thickness, overall plate flatness and bonding are shown to be very consistent.

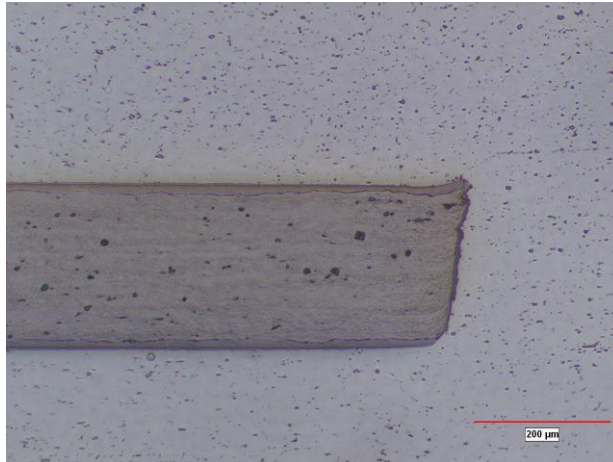


Figure 12. Micrograph from plate 3 of HIP can 10. 0.254 mm dU foil clad in 2.0 mm of aluminum. Note aluminum-aluminum bond line on right side.

4. Conclusions

The INL process was modified in several critical areas to yield a consistent technique for HIP bonding aluminum cladding to produce full-sized LEU reactor fuel. Results from the first four experiments, coupled with finite element modeling led us to minimize any free space in the HIP can. This development resulted in a more uniform stress distribution and better reproducibility and quality. Metallographic results show that bonding is good and that there are no gaps or voids between the aluminum cladding and the dU fuel foils. The INL bend test also indicates that the aluminum cladding is well bonded and does not delaminate. Bonding occurs through extrusion, molten aluminum is forced into any remaining free space during the HIP process. Effective parting agents are essential to the process as they allow recovery of the fuel plates without damage.

5. References

1. G.A. Moore, F.J.Rice, N.E.Woolstenhulme, J-F.Hue, B.H.Park, S.E.Steffler, N.P.Hallinan, M.D. Chapple, M.C. Marschall, B.L. Mackowiak, C.R. Clark and B.H. Rabin and D.E. Burks, "Monolithic Fuel Fabrication Process Development at the Idaho National Laboratory," Proceedings of RERTR 2009, Beijing, China, November 1-5, 2009.
2. G.A. Moore, F.J.Rice, N.E.Woolstenhulme, W.D.Swank, D.C.Haggard, J-F.Hue, B.H.Park, S.E.Steffler, N.P.Hallinan, M.D. Chapple and D.E. Burks, "Monolithic Fuel Fabrication Process Development at the Idaho National Laboratory," Proceedings of RERTR 2008, Washington, D.C., October 5-9, 2008.
3. G.A. Moore, J-F.Hue, B.H. Rabin and M.J. Nilles, "Full Size U-10 Mo Monolithic Fuel Foil and Fuel Plate Fabrication-Technology Development," Proceedings of RRFM 2010, Marrakesh, Morocco, March 2010.
4. J-F. Jue, B.H.Park, C.R. Clark, G.A. Moore and D.D. Keiser, Jr., Fabrication of Monolithic RERTR Fuels by Hot Isostatic Pressing, Nuclear Technology, Vol. 172, Nov. 2010, pp. 204-210.