LA-UR-13-22779

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Intended for: Report

Issued: 2013-04-18

Canless HIP Development for Aluminum Cladding of LEU Fuel Foils using Electron Beam Welding

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Summary Report for FY12

MST-6
Los Alamos National Laboratory
March 13, 2013

INTRODUCTION

The conventional canning method for HIP processing of fuel foils requires that foil assemblies be stacked within a stainless steel can, which is then welded shut and the internal air evacuated. A few of the detracting aspects associated with this method include i) the time required to hand construct and disassemble each can, ii) the possibility for bonding between adjacent foil assemblies and backing plates, and iii) the generation of large volumes of contaminated waste (can material and backing plates) that must be disposed. The new "canless" HIP approach discussed in this report involves the hermetic sealing of individual foil assemblies by electron beam welding their outer perimeter. A foil assembly consists of a zirconium-clad LEU (U-10Mo) foil sandwiched between two aluminum 6061 plates. Hermetic sealing is accomplished by making 4 intersecting aluminum-to-aluminum, full-penetration lap welds as shown in Figure 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. Multiple foil assemblies welded in this manner can then be placed in a HIP fixture and batch processed.

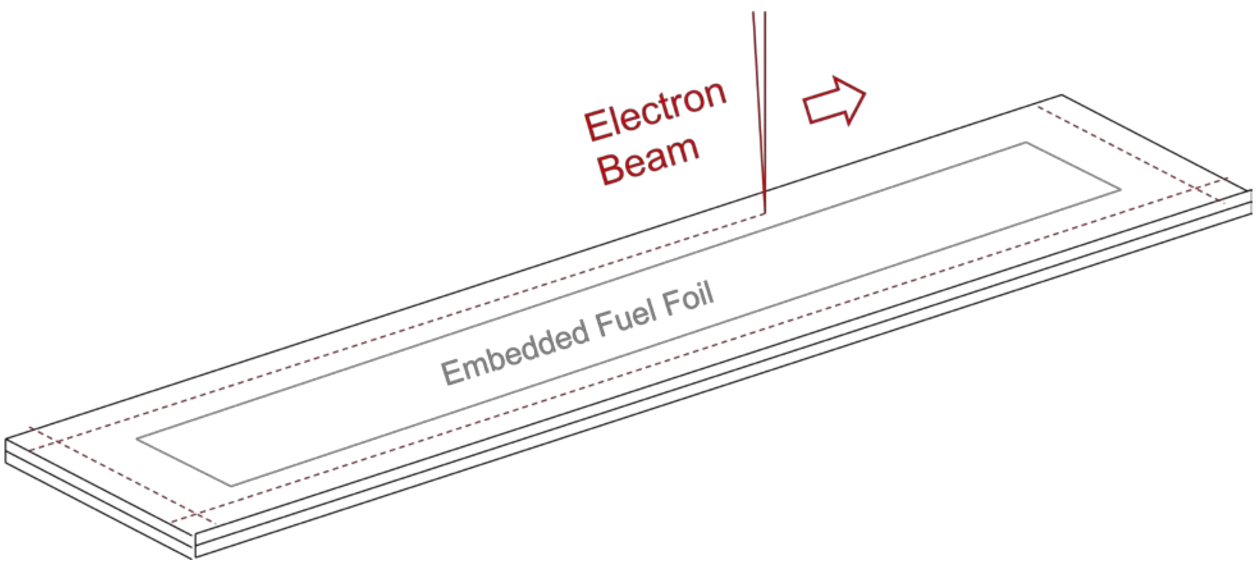


Figure 1: Schematic showing 4 intersecting electron beam lap welds made on aluminum plate along outer perimeter of fuel foil assembly. The LEU fuel foil sits within a pocket machined in one of the aluminum plates.

INITIAL GOALS

Studies performed on small aluminum coupons (3.5 inches square) in FY11 demonstrated the feasibility of using the canless HIP approach. The goal for FY12 was to scale-up the process to full sized (24 inch long) fuel assemblies, first using surrogate stainless steel foil in place of uranium, followed by the use of uranium foil. Specifically, four canless surrogate assemblies were to be welded, HIPed, and inspected for bonding using ultrasonics and metallography. The welding of surrogates, when proven successful, was to then be applied to Zr-clad DU-10Mo foils.

SUMMARY OF WORK PERFORMED

Fixture Development

The first task was to arrange for a weld fixture and traversing table suitable for welding 24 inch long coupons. An Aerotech X-Y positioning table (Figure 2) was utilized that requires computer programming in basic CNC machine code to permit the tracking of a desired rectangular pattern (Note: the electron beam remains stationary while the part to be welded is made to move under the beam). The size of the rectangular pattern to be sealed is 2.5 x 22 inches.

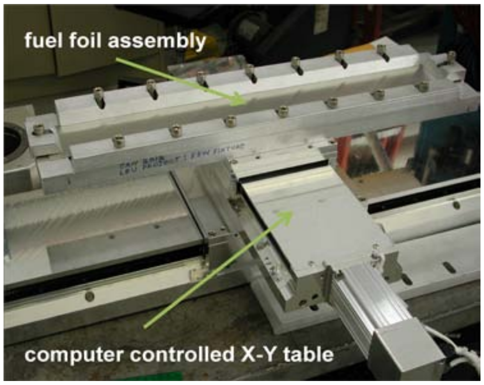


Figure 2: Photograph of EBW fuel foil welding fixture mounted to Aerotech X-Y linear motion table.

A decision was made to form a hermetic seal by making four separate intersecting linear welds in a rectangular pattern (recall Figure 1), similar to what was done in FY11 preliminary development. The approach taken was to write two separate X-Y computer tracking programs; one program to track the two long sides of the foil assembly, and another program to track the two short sides of foil assembly as depicted in Figure 3. Once a tracking program is selected and initiated, the table is made to follow the desired beam path; tracking first one side of the foil and then the opposite side in repetitive, non-stop succession (usually 10 repetitions). This cyclic track repetition allows the operator to optically observe and adjust the alignment along the desired path, before manually initiating the beam. Once the operator is convinced there is good alignment, the beam is initiated just before reaching the foil and is stopped just after traversing its full length (i.e. a “running” start and stop). Once the two long sides have been welded in this manner, the second tracking program is downloaded and the two short sides are welded.

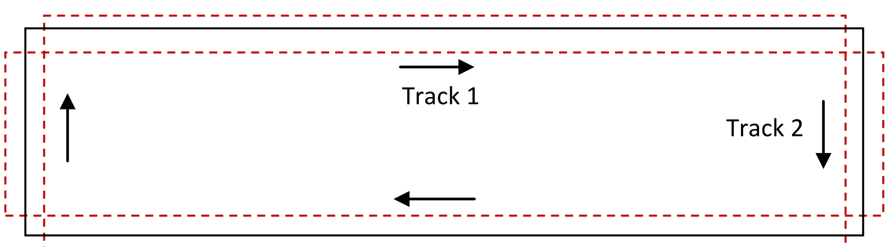


Figure 3: Schematic showing two tracking paths (dashed lines), where Track 1 is used to make two long welds and Track 2 is used to make two short welds. Arrows indicate where welds are made and the direction of welding. Solid line represents top view of foil assembly (not to scale).

A welding fixture was designed and fabricated consisting of an aluminum base plate plus aluminum hold-down bars that apply pressure around the foil's outer perimeter within close proximity to the weld path (within 0.030 inch of the weld) as shown in Figure 2. Pressure was adjusted by tightening bolts threaded into the base plate, usually set by hand with only moderate applied force. There was a concern that the clamping should not be too tight so as to prohibit air evacuation between plates during EB chamber pump-down. There was no pressure applied to the inner side of the weld path (i.e. directly over the fuel). By using this arrangement, the entire 4 welds can be made consecutively with only one chamber pump-down.

Weld Development

FY11 electron beam weld development on small coupons was performed in a 3 x 3 x 3 ft chamber, high voltage/high vacuum, Leybold-Heraeus machine. Since this chamber was not suitable for welding a part requiring weld travel of 26 inches (i.e. over-shoot of 24 inch long foil of about 1 inch on both ends), a larger chamber Leybold-Heraeus machine (5 x 5 x 5 ft) was used instead for this study. It was assumed that weld parameters and corresponding weld behavior would transfer from the smaller machine without major difficulty. The welding parameters that were transferred from the small machine and used to weld the first set of surrogate foil assemblies are provided in Table 1. Part of the argument for using a high travel speed (developed in FY11) was that this avoids problems with solidification cracking for which the Alloy 6061 is highly susceptible.

Table 1: Electron beam welding parameters developed in FY11 and transferred to large chamber machine for welding the first two surrogate foil assemblies.

Accelerating Voltage	100 kV
Beam Current	6.3 mA
Beam Focus	sharp focus (at surface)
Travel Speed	78 ipm
Vacuum	5×10^{-5} torr

In preparation for surrogate foil assembly and welding, Alloy 6061 aluminum plates were sheared from 0.037 inch thick rolled stock to dimensions: 3¾ x 24⅛ inches. This thickness was selected because thinner material (e.g. 0.032 inch thickness) was found in FY11 to be more prone to weld defects. Also, the thicker material is easier to hold flat for machining pockets. Some plates had pockets machined in them (2.076 wide x 22.366 long x 0.010 inches deep) designed to accept 0.010 inch thick 304 stainless steel foils with tight clearance (see Figure 4 for pocket layout). Pockets were machined using multiple passes of a small diameter (3/32 inch radius) end-mill so as to provide relatively tight radius corners to accommodate the stainless steel surrogate foil having 1/8 inch radius corners. This resulted in the pockets having multiple machine ridges on the order of a thousandth of an inch.

Plates not having pockets were used as cover plates in assemblies. Pre-weld cleaning of aluminum was done using a Babcock & Wilcox procedure consisting of degreasing followed by immersion in a pickling solution to remove oxidation: 19.5 vol.% HNO_3 (70%) + 2.4 vol.% HF (49%) + 78.1 vol.% H_2O. Stainless steel foils were cleaned using a conventional Oakite alkaline degreaser. Assembly for welding was done within 24 hours of cleaning using lint-free cotton gloves.

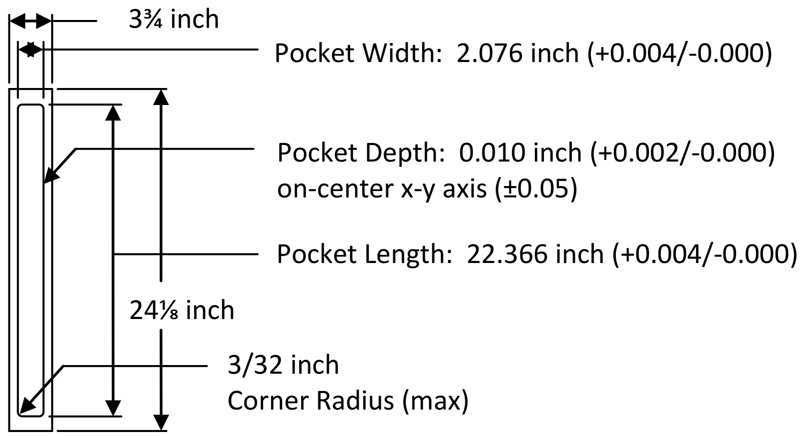


Figure 4: Aluminum plate dimensions and pocket layout.

Divot Weld Defect

Following the welding of 24-inch long practice coupons plus the first two surrogate foil assemblies, welds were observed to have full-penetration similar to the small coupon welds made in the small chamber machine in FY11. Also, there was no weld centerline cracking observed at weld-start regions as was sometimes observed in initial FY11 development. However, it was noticed that there were several surface irregularities (referred to here as "divots") on the weld crown as depicted in Figure 5a. Their frequency appeared random at roughly 1-2 divots per 24 inches of weld. Divots can be characterized as localized regions where the weld pool was disrupted, resulting in a wider weld pool. Associated with divot formation, the ejection of material from the weld pool was sometimes (but not always) observed by the weld operator. Accordingly, some divots had surface depressions (i.e. material removed) and some did not. Furthermore, the weld under-bead corresponding to divots always showed a localized absence of full penetration.

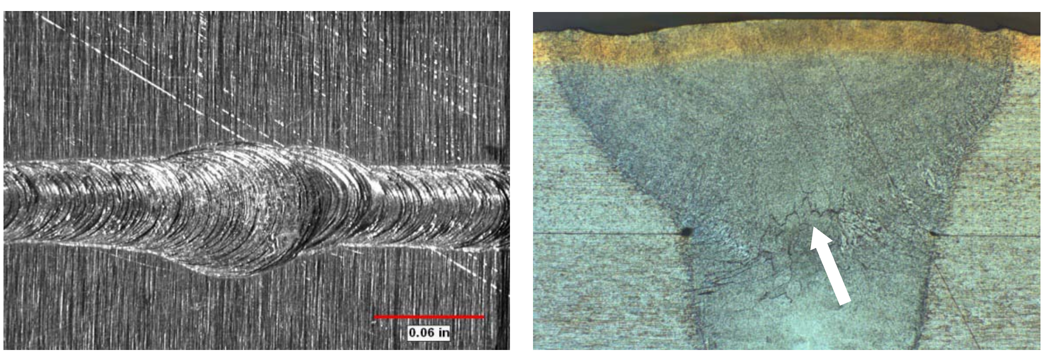


Figure 5: a) Top surface of weld bead showing divot defect and b) cross-section of divot showing solidification cracking across lap interface (see arrow).

The metallographic cross-section of divots (see Figure 5b) showed the presence of solidification cracking. These cracks are oriented in the plane of the lap joint, which is an abnormal occurrence. Normally such cracking is observed along the weld centerline, originating from solidification shrinkage stresses. This suggests that other imposed stresses may be at play here due perhaps to translational movement of plates relative to one another. The sudden translation of plates (e.g. due to inadequate hold-down force and clamping) could also explain the weld pool disturbance observed from the surface.

Another explanation for weld pool expulsion sometimes observed in electron beam welding (referred to as “blow-outs”) involves the explosive vaporization of volatile inclusions. Although, the material being welding appears from metallography to be very homogeneous without any massive inclusions. Problems with electron gun arc-outs (i.e. temporary loss and re-establishment of the beam) can also sometimes cause a disturbance in the weld pool. Arc-outs originate from material deposited on the gun or beam column, such as contamination from handling or vaporization from welding. Since Alloy 6061 consists of both Al and Mg (both elements have low melting temperature and high vaporization pressure), the possibility for this type of contamination does exist. Keyhole stability is another possible source of defect formation, whereby the liquid column surrounding the keyhole collapses causing momentary loss of penetration. This can occur when the liquid head periodically builds up around the key-hole cavity, and gravity topples the liquid into the key-hole. Normally, welds made with sharp focus are more susceptible to this occurrence.

Thus, in summary, there are at least four possible explanations for this observed defect:

1) Insufficient clamping force allowing material movement during welding
2) Inhomogenieties in material (compositional variations or inclusions)
3) Erratic electron gun behavior (e.g. gun arc-out).
4) Unstable EBW keyhole

HIP Processing of Fuel Foils with Divots

Since, at the time, it was not known if cracking was involved with divots or if divots would cause problems with HIP processing, a decision was made to HIP two canless surrogate fuel foil assemblies containing divots. The assemblies were HIPed using a new HIP fixture whereby they were hung from the top and allowed to hang freely (see Figure 6). It is important that foils remain free to expand and contract lengthwise during the HIP thermal cycle to avoid buckling. In previous work (FY11) it was demonstrated that foils will buckle under their own weight if supported from beneath during HIPing.

Figure 6: New HIP fixture made specifically for canless HIP processing of 24-inch long foils. Foils are clamped at the top of the fixture and hang freely.

After HIPing, the fuel foils (with divots) were examined and found to be intact with little difference in appearance or flatness from before HIPing. Also, there was no indication of problems with creep. When foils were evaluated using scanning ultrasonic (UT) inspection, however, it became apparent that there was no bonding and that the welds had leaked during HIPing. In particular, water was found to leak into and out of the HIPed plates during UT testing, where water is used as the transducer couplant. The UT technique uses a pulse-echo method whereby a strong reflectance signal represents an unbonded interface. Where there is bonding (or water), more of the signal gets transmitted through the foil as opposed to being reflected, thus resulting in a weaker reflection signal. The strong reflectance signal in Figure 7, as represented by the light yellow color in the middle of the foil, suggests a condition of non-bonding. The darker color around the perimeter is likely due to the presence of water.

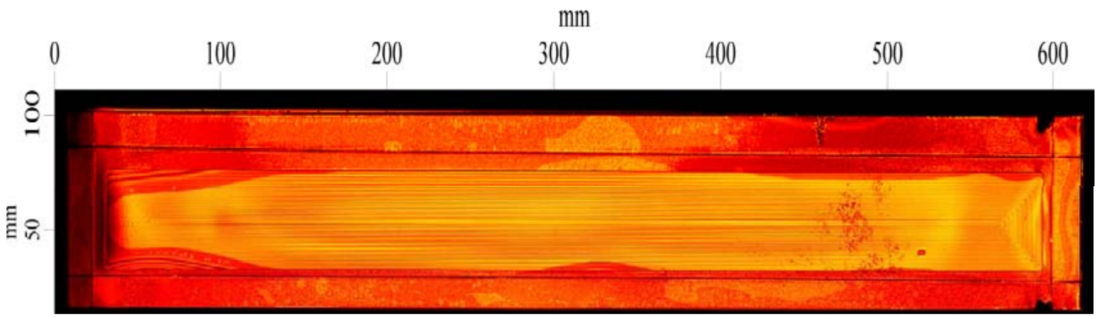


Figure 7: Ultrasonic scan of canless HIPed surrogate fuel foil having weld divot defects.
Light color (yellow) indicates high reflectance signal in pulse-echo UT technique.

After UT inspection, one of the foils was cross-sectioned in the vicinity of a divot and examined at varying depths via multiple grind and polish iterations (see Figure 8). It is clear from Figure 8a that i) there was no Al-Al or Al-SSt bonding, and ii) there is a large gap between plates, indicative of pressure buildup between aluminum plates during HIP depressurization. The nature of the weld in this macrograph (with outward flow of weld metal at the lap interface) suggests that there may have been some gap even before HIP processing. This provides added evidence of a weld fixture problem, with insufficient clamping force to completely close the gap prior to welding.

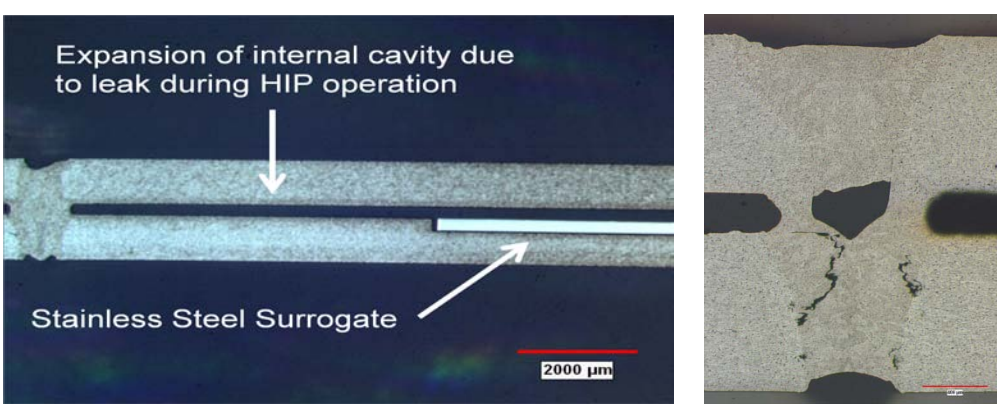


Figure 8: Cross-sections of surrogate fuel foil assembly in vicinity of weld divot processed with canless HIPing.

Figure 8b, taken closer to the center of the divot, shows solidification cracking that has opened up considerably due to pressure build up between aluminum plates during HIP depressurization. The large cavity located at the weld center is abnormal in appearance. It appears to have formed as a result of liquid feeding around a cavity, perhaps a result of pool ejection or keyhole collapse. There are also clear signs of partial cavity fill-in (vertical lines along cavity wall) where oxidation prevented wetting (i.e. cold shut).

Modification of Weld Parameters and Weld Fixture

Before welding a second set of surrogate fuel foil assemblies, some minor corrections in weld parameters and fixturing were made in attempt to address the weld divot problem. Weld parameters were adjusted to broaden the weld pool by means of defocusing (i.e. broadening) the electron beam. Weld keyholes formed using sharp focus (original parameters) are inherently less stable. By running a series of welds on scrap plate, a set of parameters giving full penetration and a smooth over-bead was selected as listed in Table 2. By defocusing the beam (i.e. focus slightly above plate surface), the weld width is increased (i.e. decreased power density) and, accordingly, a higher current is needed to maintain penetration. The weld cross-section corresponding to these new parameters is given in Figure 9. Note that the EB weld is now more like a gas-tungsten arc weld in shape (not size), eliminating the characteristic nail-head apparent in Figure 5. Also note a large gas pore originating at the lap joint, often observed in these cross-sections.

Table 2: Revised Electron Beam Welding Parameters to Improve Keyhole Stability.

Accelerating Voltage	100 kV
Beam Current	8.0 mA
Beam Focus	sharp focus above plate surface (+ 5 units)
Travel Speed	78 ipm
Vacuum	5 x 10^{-5} torr

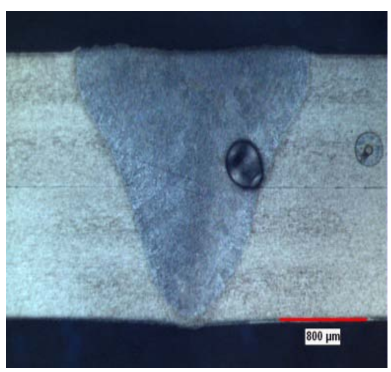


Figure 9: Weld cross-section from practice coupon corresponding to revised EBW parameters of Table 2. Large gas pore is observed originating from lap interface.

Weld fixture modifications were made to provide clamping on both sides of the beam path as depicted in Figure 10. In essence, a bar extending over the center of the foil assembly provided hold-down pressure on the inner side of the beam path. The amount of force exerted, however, was limited due to the associated moment arm. By the nature of this new configuration, fuel foil assemblies now require two separate pump-down and weld cycles, where the foil assembly must be rotated 180° in the fixture between cycles.

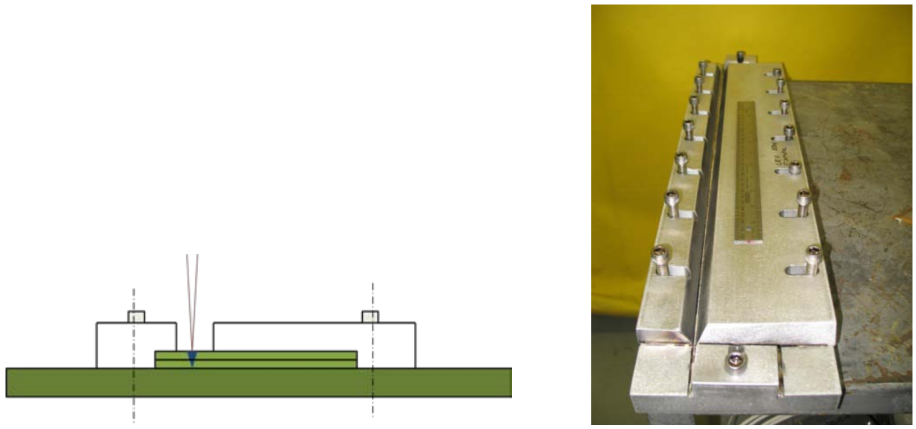


Figure 10: Modified EBW fixture providing clamping on both sides of the beam path.

Welds made on two surrogate fuel foil assemblies using the above modified weld parameters and fixture again resulted in divot defects. It was decided at this point to attempt weld repair, knowing that divots previously resulted in failed HIP runs. This was accomplished by welding directly over the divots using the same established weld parameters (Table 2). A 2-inch (run and stop) weld length was employed. The good bead appearance of the repair welds suggested that this approach may serve to seal the joint. Although it is known from general experience that welding over defects does not always eliminate them.

HIP Processing of Repair-Welded Fuel Foils

The HIP processing of two surrogate fuel foil assemblies, with repair-welded divots, resulted in foils having extensive distortion (both twisting and bending). This behavior is unlike the previous two assemblies that leaked, did not bond, and did not distort from HIPing. This distortion is characterized in Figure 11 where before-HIP and after-HIP photographs are compared. This suggests that some bonding between aluminum and stainless steel must have occurred.

Figure 11: HIP fixture with hanging canless surrogate fuel foil assemblies a) before HIPing and b) after HIPing. Foils are observed to have both bend and twist distortion as a result of HIP operation.

In addition to this distortion, the position of the embedded stainless steel foil could clearly be seen in each assembly (e.g. Figure 12a) due to indentations caused by aluminum flow around the stainless steel foils during HIPing. In one assembly, the foil was found to reside outside the pre-machined pocket meant to hold the foil in place (Figure 12b). The camber associated with the stainless steel foil (machined from coiled stock) makes it act like a spring during assembly and prevents it from laying flat in the machined pocket. This spring action no-doubt also influenced the foil distortion behavior observed in Figure 11b.

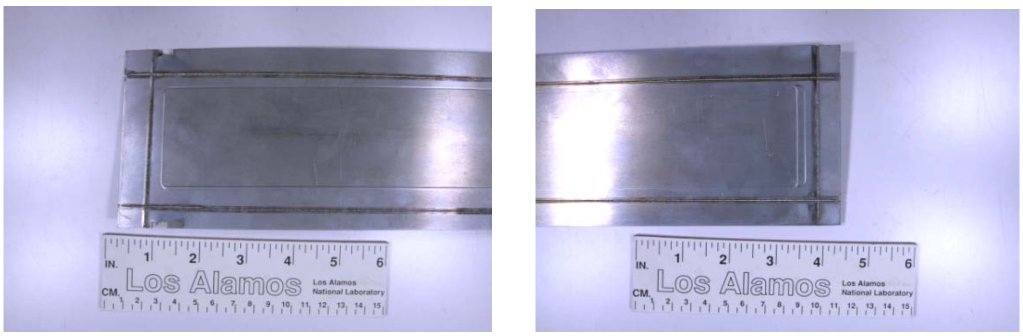


Figure 12: Post HIP foil assemblies showing a) outline of encased stainless steel foil and b) indication that stainless steel foil slipped out of pocket.

Applying scanning ultrasound inspection after HIP processing was made particularly difficult due to foil distortion. In order to achieve efficient send/receive transducer performance, the transducer must remain perpendicular to the plate surface. When not perpendicular, much of the signal may be lost. Thus, the foil assemblies were examined in segments, aligning the transducer as best as possible within each segment. One of the foils showed evidence of good bonding on one side only (see Figure 13). Light colors indicate strong reflectance, which indicates an interface and possibly a poor bond. In Figure 13, the outer perimeter (outside the hermetic seal) is the brightest indicating non-bonding. The black region just inside the hermetic seal indicates good Al-Al bonding with little if any reflectance. The lighter region in the middle of the foil represents a good Al-SSt bond, where the dissimilar metal interface is expected to reflect a weak signal even when bonded.

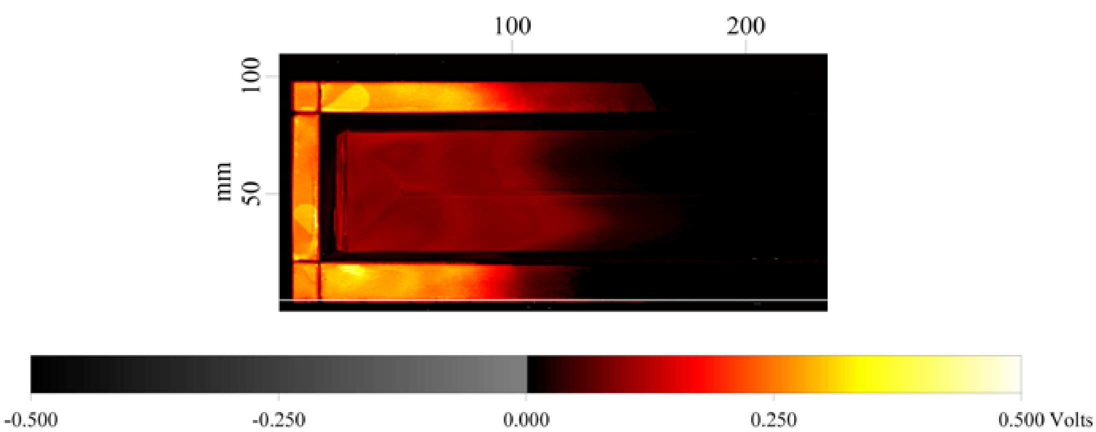


Figure 13: Scanning ultrasound image take from the end of a HIPed surrogate foil assembly.

A cross-section taken from a HIPed assembly is characterized in Figures 14-16, specifically from a region that UT inspection predicted there to be Al-SSt bonding. Figure 14 confirms that there is Al-Al bonding inside the hermetic seal, but not outside. This is to be expected, since outside the hermetic seal there will be heavy oxidation at faying aluminum surfaces (formed at elevated temperature) preventing solid-state bonding. The dark etching regions of aluminum appear to correspond to regions of compression strain (e.g. around the inside of the weld).

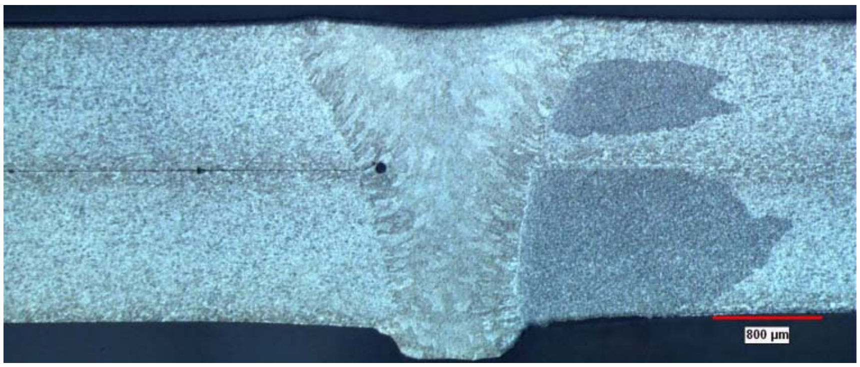


Figure 14: Weld cross-section of HIPed surrogate foil assembly showing Al-Al bonding on the inner (right) side of hermetic seal. Dark etching regions appear to relate to areas of compression strain experienced during HIPing.

When the encased SSt foil is observed at low magnification (Figure 15), the interface between Al and SSt is found to consist of a heavily fragmented band of intermetallic compounds (20-30 microns thick). The presence of this band confirms that there was bonding between Al and SSt allowing diffusion of Fe, Cr, Si and Al across the bond interface. The fracture surfaces observed within the intermetallic could have served to give indications of non-bonding noted during UT inspection. Aluminum appears to have extruded between the fractured intermetallic in some places.

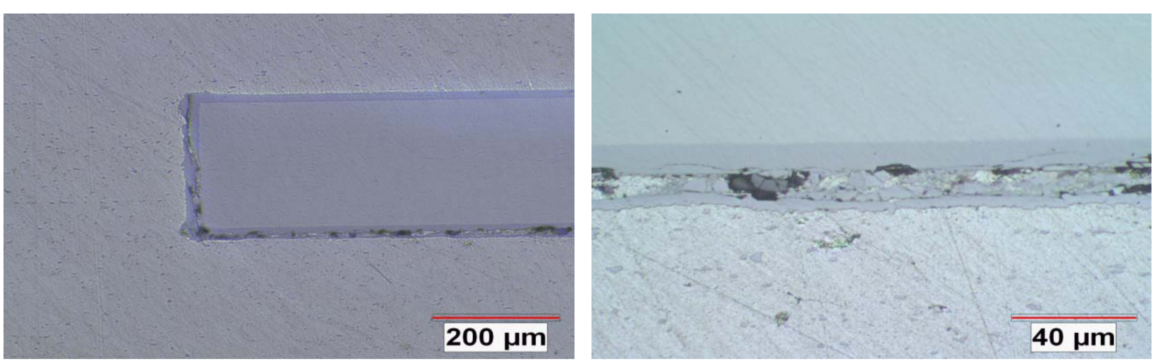
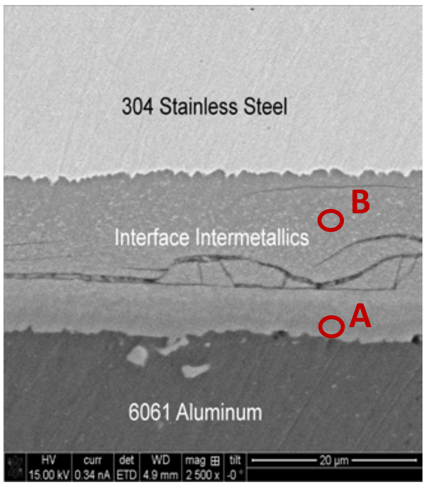


Figure 15: Cross-section of HIPed surrogate foil assembly showing a) stainless steel foil (center) surrounded by 6061 aluminum with intermetallic compounds at interface and b) higher magnification view of fractured interface intermetallics.

SEM/EDX analysis (Figure 16) confirmed that there exists a compositional gradient across the band of intermetallics: C, Fe, and Cr diffuse toward the Al; Al and Si diffuse toward the SSt. A uniform layer of intermetallic (2 µm thick) is found adjacent to the aluminum that appears to consist of the aluminum-rich intermetallic $FeAl_6$ (see Figure 16, point A). Further away from this layer there is a thicker layer (Figure 16, point B) that is richer in iron and chromium, closer to $FeAl_3$ stoichiometry. This thicker layer appears to consist of two different phases.



	A	B	(atomic %)
Carbon	1.62	3.39	
Aluminum	75.68	66.75	
Silicon	6.53	1.62	
Chromium	3.41	5.44	
Iron	11.68	21.15	
Nickel	1.09	1.64	

Figure 16: SEM macrograph of HIPed surrogate foil assembly showing Al-SSt interface intermetallics with corresponding EDX analysis at two different circled areas: A and B.

Redesigned Welding Fixture

In order to more positively secure the 24-inch fuel foil assembly during EB welding and expand upon the approach demonstrated earlier in Figure 10, a new welding fixture was designed and built. The goal was to prevent movement of aluminum plates during welding and ensure close contact between aluminum and fuel foil prior to HIPing. Several features were changed from the original fixture: 1) hold-down bars were made from stainless steel instead of aluminum (Al-Al contact tends to slide), 2) more leverage distance was provided on the hold-down bars between the clamping screws and pivot edge to give higher clamping force near the weld, and 3) toggle clamps were used to hold down a stainless steel bar placed in the central region above the fuel foil assembly. These features can all be seen in the photograph of Figure 17. One long and one short weld can be made in one pump-down cycle, after which the foil must be removed from the fixture and rotated 180° for the second cycle.

Initial trials were made to evaluate the new fixture and see if this had any effect on eliminating divots. Four 24-inch lap joint strips of 6061 aluminum (0.037 inch thick x 1½ inch wide) were welded using the parameters of Table 2. One strip had no divots, two strips had one divot, and one strip had two divots. This corresponds to approximately 1 divot per 24 inches of weld and does not appear to be a significant improvement over the original fixture.

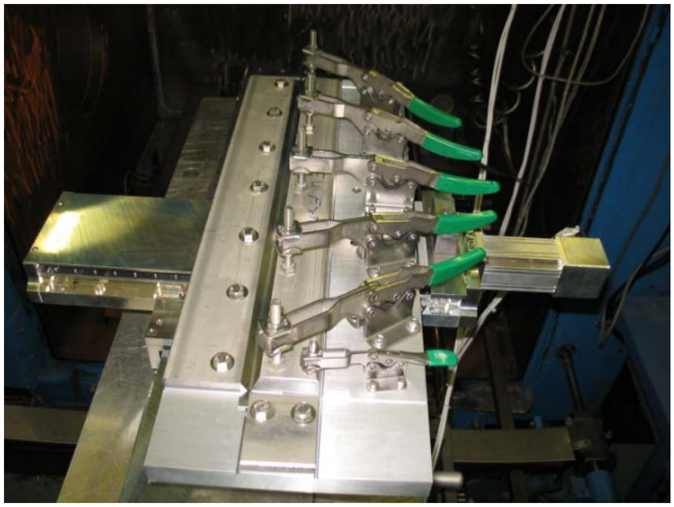


Figure 17: New EBW welding fixture incorporating improvements including stainless steel hold-down bars, higher clamping pressure, and use of toggle clamps for the central region.

Elimination of Divots: Gun Rebuild

By October 2012 there was increasing evidence of beam instability for the EBW machine used in this study (i.e. small variations observed in current and voltage even when welding ferrous alloys). It was decided at this point to rebuild and clean the electron gun. This involved replacing the filament with a new tungsten ribbon and polishing the gun itself, to remove metallic vapor deposits. This is a maintenance task that had not been done for over a year, as it is normally done only when needed. Practice welds were made on several 24 inch long strips of aluminum both before and after the gun rebuild. It was discovered that after the gun rebuild, there were no more divots observed. This provides strong evidence that the origin of weld divots is linked to arc-outs caused by deposits on the gun.

Future Work

There remain several topics of concern regarding Canless HIP processing that require further development, primarily involving pre-weld preparation and fixturing for both welding and HIPing. There is presently at LANL a concurrent study looking at optimum surface treatment and cleaning procedures to promote bonding during HIP processing. These improvements will need to be implemented and evaluated in the Canless HIP process. How bond strength is influenced by delay time between cleaning (i.e. oxide removal) and EBW hermetic sealing also needs to be examined, as does the degree of vacuum in the hermetic seal. If a cleaned part sits for too long a time, the possibility for surface contamination and oxide growth may degrade the resulting HIP bond. Also important is the magnitude of the hermetic vacuum, which is likely influenced by EBW pump-down time in addition to hold-down pressure applied in the weld fixture. This hermetic vacuum should be measured for various different conditions of pump time and clamping pressure, and then be correlated with bond strength. Establishing acceptable process limitations will be needed when implementing the Canless HIP process on a large scale. Finally, it is clear that more restraint is needed in the HIP fixture to prevent excessive bending and twisting associated with dissimilar metal thermal contraction.

SUMMARY

The concept of canless HIP processing of 24-inch surrogate fuel foils has been successfully demonstrated with Al-Al and Al-SSt bonding achieved in foil assemblies. This represents a positive advancement from the initial FY11 feasibility study involving the canless HIP bonding of small aluminum coupons.

Problems were encountered initially with pressure leaks during HIPing, believed related to weld divot defects. The divot defect has been characterized with metallography and has been shown to involve solidification cracking. The cause of this weld defect remains uncertain, although several possible sources have been proposed. It has been demonstrated that this problem can be mitigated by re-welding directly over the affected region, a process that has been found to result in acceptable surrogate foil HIP bonds between Al-Al and Al-SSt.

The Al-SSt HIP bond resulted in significant brittle intermetallic compound formation with severe cracking in the intermetallic. The presence of these fractures gave problems with indications of non-bonding during UT inspection, thus demonstrating that stainless steel is not an appropriate surrogate material for HIP bonding studies.

Weld fixturing has evolved during development of the canless HIP process, progressing through three different designs each giving improved clamping capability. However, improved clamping did not eliminate the divot defect problem. A fixture to hang 24-inch foils during HIPing was evaluated and found to work. Its free hanging design does not interfere with thermal expansion/contraction. Also, creep was not found to occur. However, there was significant distortion (twisting and bending) in the foils following HIP processing, believed related to the dissimilar metal bond.