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LANL Progress on U-Mo Fuel Fabrication Process Development

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ABSTRACT

Recent progress at LANL in process development, prototype fabrication and alternate process development for monolithic LEU-10wt% Mo fuel will be discussed. Master alloy manufacture using vacuum arc re-melting (VAR) has produced several homogenized DU-Mo ingots. The effects of mold design are demonstrated for several designs of the triple plate mold. The feasibility of using HIP cans fabricated by sheet forming is demonstrated and the fabrication cost advantages are discussed. Comparison of a finite element model with experimental HIP can measurements is made. A new fixture for solid state bonding of electron beam welded aluminum cladding is shown and results of its use are discussed. The effect of foil annealing temperature on the metallography of Zr/U-Mo interfaces is shown. The as-cast surfaces of DU-Mo machined by electric discharge machining are shown and possible advantages in material utilization are discussed.

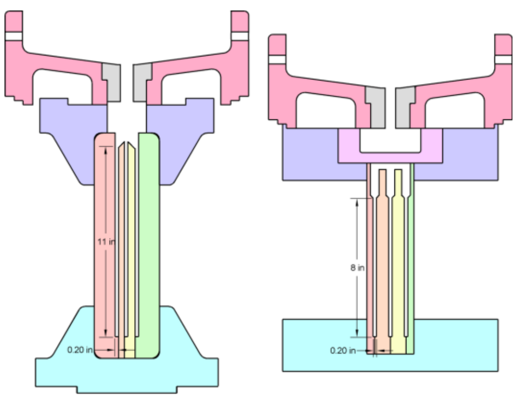
Introduction

This paper provides an overview of work at the Los Alamos National Laboratory (LANL) in U-10Mo fuel fabrication process development. Process development was oriented towards producing full scale prototypes, resolving scale up issues, and developing process variations such as canless HIP (Hot Isostatic Pressing) which have potential for fuel production cost savings. The process development overview will include casting mold optimization, use of the vacuum arc re-melt process (VAR) for master alloy preparation, preparation of rolling coupons using electric discharge machining (EDM), foil annealing, HIP can fabrication, and canless HIP. Plasma spraying, use of x-ray fluorescence (XRF) to measure zirconium diffusion layer thickness, bond strength measurements and residual stress measurements will be discussed in separate posters and papers.

Vacuum Induction Melting (VIM) Casting Optimization

LANL has been working in conjunction with the Y-12 complex to optimize the casting hot top, and material yield for two different mold geometries used for the U-10Mo fuel. The first geometry was a single rectangular mold known as a billet mold. The second geometry was a mold consisting of three thin plate molds linked together at their tops; this mold is called a triple plate mold. The original triple plate mold design is shown on the left side of Figure 1. The original triple plate mold as assembled and as instrumented with thermocouples is shown In Figure 2a. Castings from the original triple plate mold are shown in Figure 2b.

A DU-10Mo casting was made in the initial Y-12 designed Triple Plate Casting mold. This mold consisted of three mold cavities, each cavity being 254 mm tall x 203 mm wide x 5 mm thick (10.8 in. x 8 in. x 0.2 in.). The mold was instrumented with 10 thermocouples in the mold and 4 thermocouples in the casting cavity). The resulting castings were radiographed and showed numerous areas of porosity. Process modeling showed good qualitative agreement with macroscopic features of the castings and predicted a significant number of areas with isolated shrinkage and porosity. In addition, the temperature measurements and modeling showed that initial triple plate mold has effectively no hot-top, and that the mold was quite cool. This resulted in very fast solidification time. This in turn resulted in significant solidification before mold filling was complete, which can be an indication that incremental bands of solid metal (cold laps) are being formed in the casting. It was observed that the interconnected hot top makes removal of casting from mold (without breaking inner mold parts) very difficult.



a. b.

Figure 1 Original triple plate mold design (left) and optimized design (right). R. Aikin

a. b.

Figure 2 a. Original triple plate mold instrumented b. resulting casting (right). R. Aikin

The triple plate mold was re-designed with three concepts in mind: use of a distributor to get controlled simultaneous filling of the 3 plates, use of an overflow system to get controlled sequential filling of the 3 plates, and a horizontal geometry to maximize temperature gradients. Two designs were modelled, one using the molten metal distributor at the top of the mold and the other with a molten metal overflow system. Both designs incorporated a horizontal geometry and incorporated a proper hot top for control of the temperature at the top of the mold. Both designs were compared to the original Y-12 design. Based on simulation results the molten metal distributor redesign was selected and is shown on the right side of Figure 1. A mold was fabricated and an instrumented casting was done with 10 thermocouples in the mold and 4 in the casting cavity. A comparison of thermocouple readings from both the original design casting and the revised horizontal design casting are shown in Figure 3. The revised design provided higher mold temperatures as expected and premature freezing was not observed. The castings from the revised horizontal triple plate mold design are shown in Figure 4. The castings are all different sizes, indicating that the molten metal distributor did not function as expected, and further refinements are under investigation. The revised design did yield an optimum hot top for temperature control in the center plate position.

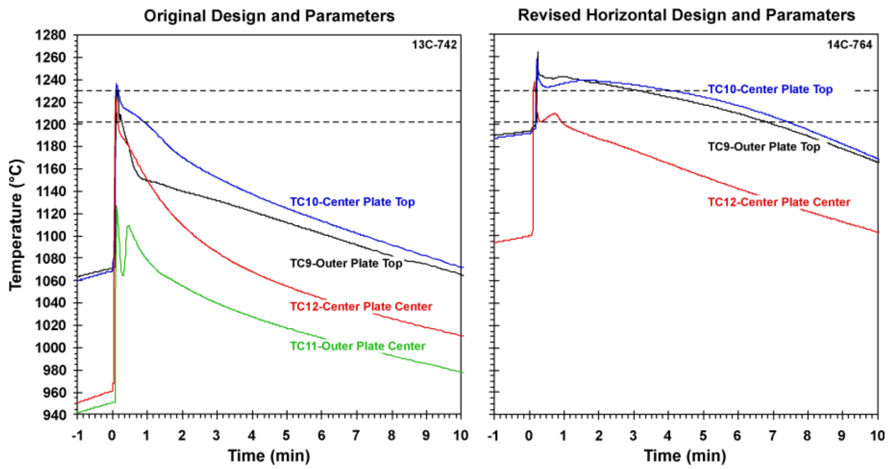


Figure 3 Comparison of thermocouple readings from original mold design and revised horizontal design. R.Aikin

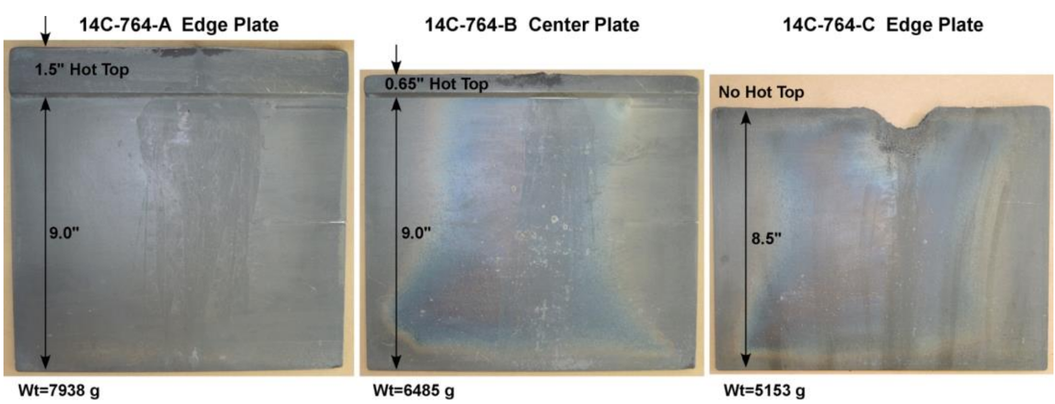


Figure 4 Castings obtained from the optimized horizontal triple plate mold design. R. Aikin

Fabrication of DU-Mo Master Alloy using VIM/VAR

Laboratory scale sample production of the U-10Mo alloy has usually been done by alloying the uranium with molybdenum by arc melting 1-2 kg of elemental material together into a button. Castings are then made by vacuum induction melting anywhere from two to ten of these buttons together. Arc melting small buttons is not feasible for full scale production of U-Mo fuel. A combination of vacuum induction melting and vacuum arc remelting (VIM/VAR) is used in commercial production of titanium and nickel superalloys to provide large quantities of homogeneous alloys. The proposed approach is to make a DU-12.5Mo master alloy by VIM/VAR in a DU facility and then perform the final down blending of the HEU with the DU-12.5Mo master alloy in an HEU facility to make LEU-10Mo. Considerable time and money can be saved if the functions of alloying uranium with molybdenum and down blending highly enriched uranium (HEU) to low enriched uranium (LEU) are done in separate operations. The principal advantage is realized if the alloying of uranium with molybdenum can be done in a depleted uranium facility rather than in a more expensive HEU facility.

The first VIM/VAR trial for DU-12.5Mo was performed in two steps. First a composite electrode was made by casting 202 kg of molten uranium around 25 kg of molybdenum strips held in an asterisk pattern as shown in Figure 5. This yields the composite electrode shown in Figure 6a. This composite electrode is then machined for uniformity to fit into the VAR as shown in Figure 6b. The machined composite electrode is then melted in the VAR with a 4000 ampere setting to produce VAR ingot #1 as shown in Figure 6c. A centerline slice has been removed from VAR ingot #1 for analysis by macroscopic x-ray fluorescence. Chemical analysis samples have also been drilled out of this slice. The VAR run chart for VAR ingot #1 is shown in Figure 7a. Due to failure of the VAR controller, three re-starts were required and the current was higher than planned. A second VAR run (#2) was performed with some adjustments. The run chart is shown in Figure 7b and somewhat more stable operation is shown. VAR ingot #2 has been cast and is being machined for analysis.

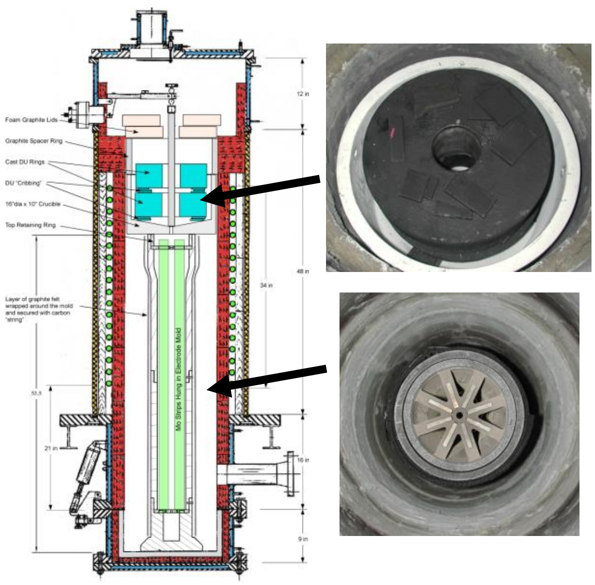


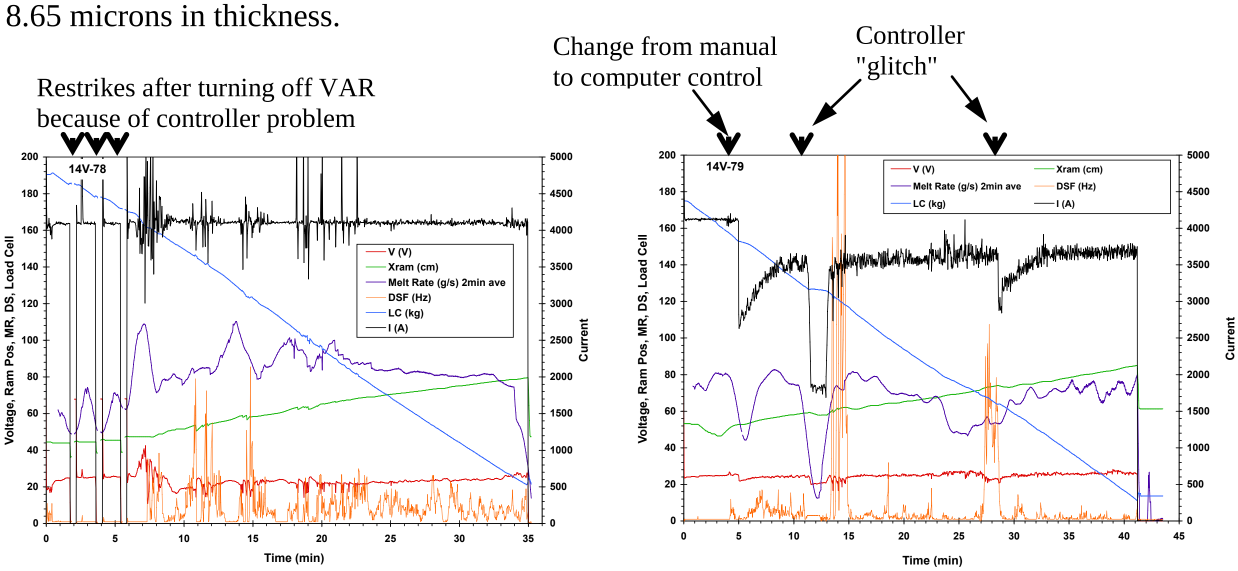
Figure 5 Position of DU charge in crucible (top right) and Mo strips hanging in asterisk shape (bottom right) in furnace. Top view is shown of Mo strips. The long axis of the strips is along the furnace axis. R. Aikin

a. **b.** **c.**

Figure 6 a. 227 kg DU-12.5Mo composite electrode (right) b. 190 kg machined electrode. c. 171 kg VAR ingot #1. R. Aikin

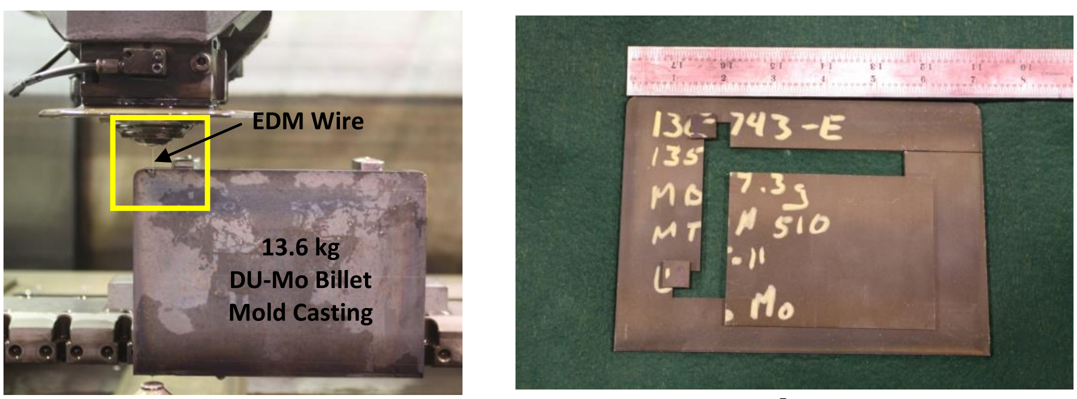
Rolling Coupon Preparation by EDM

Presently there is a large amount of U-Mo material lost to conventional machining to get coupons from U-Mo castings, first to saw coupons from castings and second to machine sawed surfaces into smooth surfaces. Electric discharge machining (EDM) uses much thinner cuts so less material is lost. The surfaces after an EDM cut are very smooth and may not require further machining. Use of EDM to machine U-10Mo castings may help minimize downstream material loss if no further machining of slices is needed. Figure 8a shows a 13.6 kg, 178 x 127 x 30 mm DU-10Mo casting which is being sliced along its central axis by an EDM wire into six 3 mm wide slices. One slice is shown in Figure 8b. The small plasma generated between the EDM wire and material being cut often generates a thin layer of re-solidified metal which is called the as-cast layer. As shown in Figure 9, the as-cast layer in the present trial varies between 3.14 and 8.65 microns in thickness.



a. b.

Figure 7 VAR run chart a. VAR Ingot #1 b. VAR ingot #2



a. b.

Figure 8. a. Casting being sliced by EDM. b. EDM slice cut into rolling coupon and metallographic samples. M. Hill

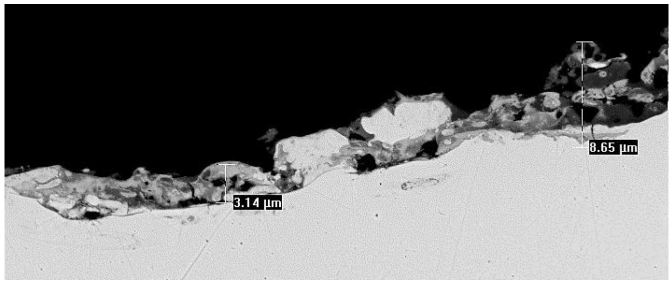


Figure 9 As-cast layer on EDM cut edge. M. Hill

Foil Annealing

In order to assess the effectiveness of post cold-rolling annealing of fuel foils, several levels of cold work were chosen, as shown in Table 1. The 4 higher levels of cold work were selected to have strains increasing in equal amounts, with targeted values of true plane strain of 0.4, 0.8, 1.2, and 1.6. A fifth very low level of cold rolling was added, to see if foils could be produced with a minimal level of cold rolling. Five corresponding levels of hot rolling were then calculated, so that the same starting and finishing thicknesses would be maintained for all samples. Five annealing temperatures, from 550 to 750°C, in 50°C increments, were also chosen. The cold-rolled foils were trimmed to size, and placed between copper sheets. Vacuum annealing for 1 hour at the chosen temperatures was followed by an oil quench.

Table 1. Target rolling reductions and strains for hot and cold rolling

% Cold Roll	**Final Foil Thick-ness (in.)**	**Hot-Rolled Foil Thick-ness (in.)**	**True Strain from Cold Rolling[1]**	**True Strain (plane strain)[2]**	**True strain from Hot Rolling[1]**	**True Strain (plane strain)[2]**	**Total Strain**	**Total Plane Strain**
5	0.015	0.0158	0.051	0.059	2.376	2.745	2.427	2.804
29	0.015	0.021	0.342	0.396	2.085	2.408	2.427	2.804
50	0.015	0.030	0.693	0.801	1.734	2.003	2.427	2.804
65	0.015	0.043	1.050	1.213	1.378	1.592	2.428	2.805
75	0.015	0.060	1.386	1.601	1.041	1.203	2.427	2.804

- all rolling is % reduction of thickness, strains are negative; negative sign omitted for ease of presentation

1 True strain calculated from thickness change only

2 True strain calculated assuming plane strain; i.e. no change in foil width during rolling; true plane strain = true strain x 1.155

Figure 10 shows the cold rolled foils placed on a copper plate in preparation for annealing. The bows and ripples in typical rolled foils are readily apparent in this view, although larger cold-rolling reductions appear to improve overall flatness (cold rolling increases from front to back in Figure 10). Another Cu plate, with an additional weight on top, was placed on top of the foils, and the vacuum annealing process was performed. The total mass of the Cu plate and the extra weight was 40 kg (90 lb). Figure 11 shows the same foils after the annealing treatment. The Cu plate has been distorted by the annealing procedure, but the foils are much flatter and more uniform.

Figure 10 A series of cold rolled foils prepared for annealing at 650°C. have been placed on a Cu sheet. D. Alexander, K. Clarke

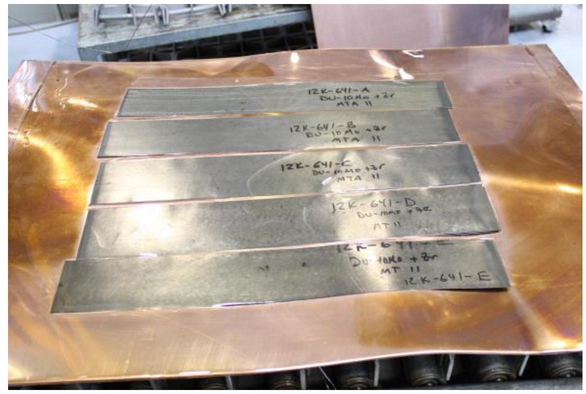


Figure 11. The same foils as Fig 10 after vacuum annealing for 1 hour at 650°C, followed by an oil quench. D. Alexander, K. Clarke

One concern about an annealing process is whether the heat treatment has any effect on the interface between the Zr and the U-10Mo. There is particular interest in the possible presence of a Mo depletion zone between those two primary phases. Sections were taken from the middle of the annealed foils and and prepared for microprobe line scans. Figure 12 shows a small dip in Mo concentration inside the uranium rich zone at approximately step 12.

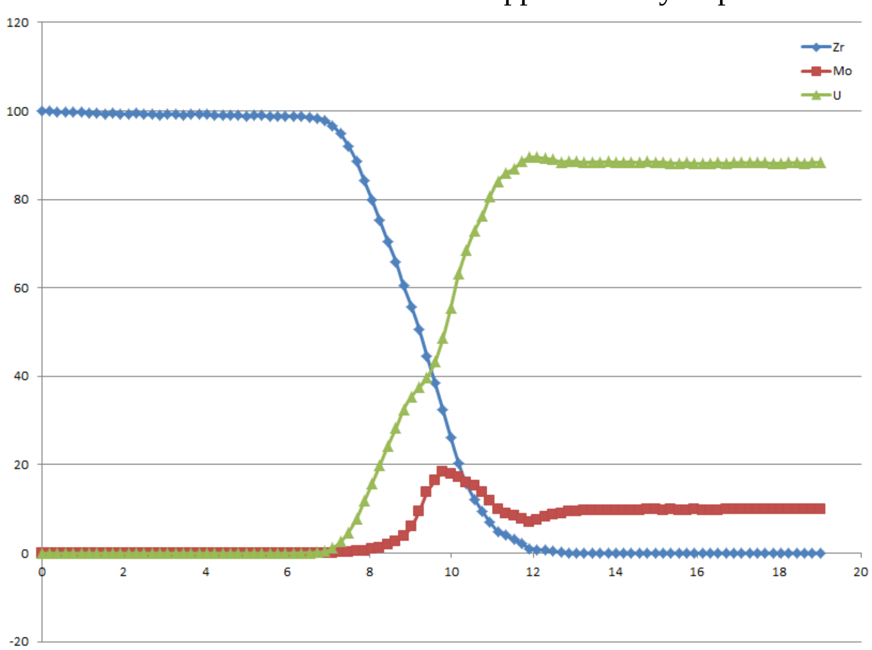
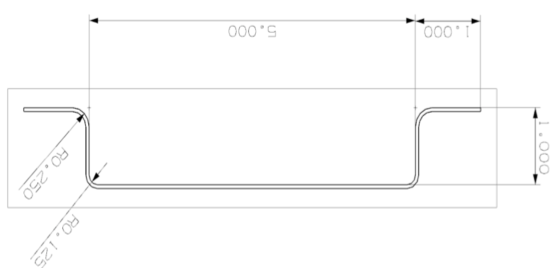


Figure 12 Microprobe beam scan type line scan with 0.2 micron step size. Sample was hot rolled 75%, then cold rolled 65% to final thickness. It was annealed for 1 hour at 700°C and oil quenched.

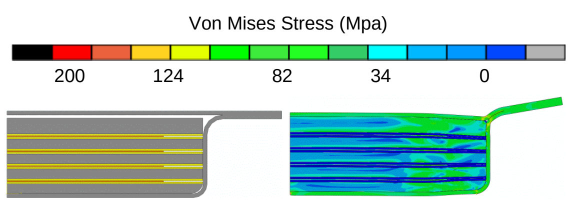
HIP Can Development

Several initiatives have been started to reduce the cost of HIP can fabrication, including use of formed HIP cans and canless HIP. The present baseline method of HIP can fabrication in the program is to have a trained welder build each six sided HIP can by hand from six plates of stainless steel. This is a labor intensive process and the weld volume and location often leads to significant dimensional distortions even before the can is loaded with foils and cladding. One method of minimizing the weld volume and labor time is to fabricate a can by mechanically forming a thin steel sheet into five sides of a HIP can. The contents can be loaded from the open top, and only the top has to be welded on to complete the HIP can. Welding to date has been performed under vacuum in an electron beam welder.

Figure 13a shows the latest design iteration in long formed HIP cans, version 3 (V3). It has a minimal draft angle on the HIP can sides and is designed to be less than 45 kg (100 lb) when loaded with four sets of strongbacks, aluminum cladding and U-10Mo fuel foils. It has a depth of 33 mm, an internal width of 125 mm, and an internal length of 610 mm. The flange radius is 6.3 mm. Finite element analysis (FEA) of the V3 can design under HIP conditions (560C, 103 MPa) is shown in Figure 123b. No critical stresses are shown. The feasibility of this design was tested by loading the formed HIP can solely with stainless steel strongbacks and electron beam welding under vacuum. HIP was then performed. As shown in Figure 14, the V3 HIP can design meets requirements of HIP can. The HIP can collapsed well around internal stackup and remained flat. This was performed without parting agent for the sake of simplicity. Next trials will include bakeout of as-sprayed (MoS_2) strongbacks prior to can assembly.



a.



b.

Figure 12 Long formed HIP can version 3 a. Schematic cross section b. FEA modeling of Von Mises stresses during HIP . K. Clarke, L. Tucker

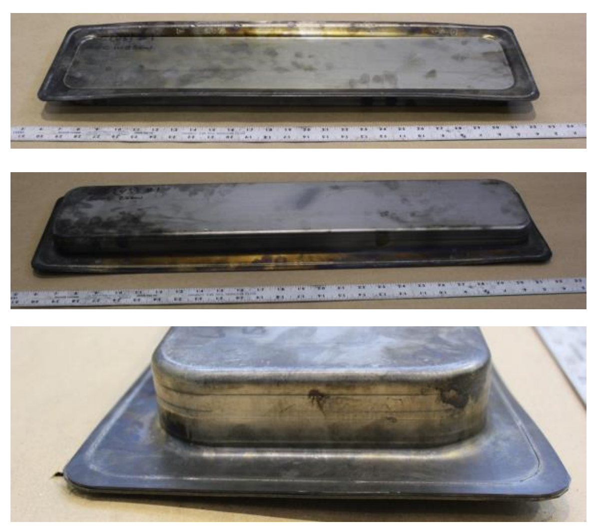


Figure 13 Long formed HIP can version 3 after HIP. K. Clarke

The FEA model for the baseline TIG welded, rectangular HIP cans was compared to measurements made on several cans after HIP. Results are shown in Table 2. FEA and experimental measurements differed by 0.04 to 0.21 mm, less than 0.5% difference.

Table 2 Comparison of FEA predicted HIP'd can thickness for TIG welded rectangular cans with experimental measurements

Location from edge [mm]	Can measurements [mm]	FEA dimensions [mm]	Can measurements minus FEA dimension [mm]
5.08	45.6311	45.7708	-0.1397
12.7	44.9453	45.1612	-0.2159
25.4	45.0342	44.89704	0.13716
38.1	44.9453	44.90212	0.04318
76.2	44.99102	44.88434	0.10668

Canless HIP

Canless HIP is an attempt to decrease HIP can fabrication costs and bonding force distribution issues by eliminating the stainless steel HIP can altogether. The concept is to seal the U-10Mo foil in aluminum cladding under vacuum before it is placed in the HIP. Electron beam welding is used to weld the perimeter of the top and bottom aluminum cladding together. Since the electron beam welding (EBW) process is performed in vacuum, there is a vacuum between the U-10Mo foil and the cladding after the aluminum cladding pieces have been welded together.

A fixture was fabricated which was designed to test several concepts for holding the aluminum clad foils flat during the cool down of the HIP after the HIP cycle. The HIP fixture was fabricated from Ni alloy for greater high temperature strength. Fuel plates can be weighted or not. A spring loaded front plate was used to further limit bowing and twisting. Prototypical 610 mm long fuel plates were made using aluminum 6061 annealed prior to assembly for 1 hour at 420C in vacuum between 6 mm (0.25 inch) thick 6061 plates. The plates were helium quenched to room temperature in ~12 minutes). Two different designs were used for the pocket in the cladding which fuel foils lie within. The first design was a typical asymmetric pocket machined in only one of the two cladding pieces. The second design, called the symmetric pocket design, machined an equivalent depth cladding pocket in both pieces of cladding. Figure 14a shows a schematic of the HIP fixture and Figure 14b shows a photograph of it.

The results of the HIP fixture test are shown in Table 3 and Figure 15. One weld apparently leaked, the source of the leak is being investigated. There was no twisting of the three other HIP'd fuel plates. All HIP'd plates were slightly bowed but much less than with the previous fixture design.

Acknowledgements

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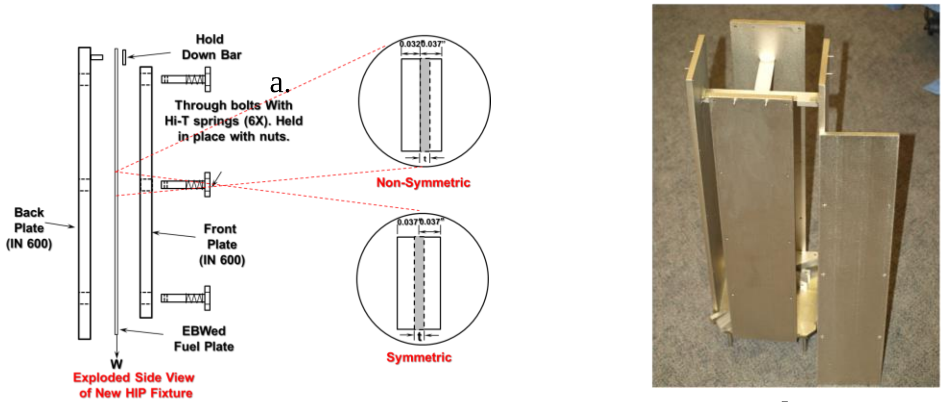

a. b.

Figure 14 Modified HIP fixture designed to hold 610 mm long canless HIP fuel plates flat during HIP cool down. a. Schematic b. Photograph. T. Lienert

Table 3 Plates, test condition in modified HIP fixture and result

Plate #	Thickness (mm)	Symmetric	Spring Loaded	Weighted	Result
536-1	0.51-0.56	N	N	Y	Leaked?
538-1	0.53-0.64	Y	Y	N	Bowed
547-1	0.53-0.58	Y	Y	N	Bowed
539-1	0.53-0.56	N	N	Y	Bowed

Figure 15 Photograph of plates after HIP. Plate 536-1 at the top has a bulge probably due to a leak in a weld during the HIP run. T. Lienert