Comparison of Cu, Ti and Ta Interlayer Explosively Fabricated Aluminum to Stainless Steel Transition Joints for Cryogenic Pressurized Hydrogen Storage

Salvador M. Aceves*, Francisco Espinosa-Loza, John W. Elmer, Robert Huber

* Corresponding author, (925) 422 0864, saceves@llnl.gov

Lawrence Livermore National Laboratory

7000 East Avenue, L-792

Livermore, CA 94550

Abstract

Fabrication of a bimetallic joint to facilitate a material transition from a 6061-T651 aluminum pressure vessel liner to stainless steel tubing in cryogenic pressure vessels is explored using three different dissimilar metal interlayers with an explosive welding (EXW) process. Due to difficulties in directly EXW joining aluminum (Al) 6061 to 304 stainless steel, interlayers are used to prevent interaction of the aluminum and stainless steel, thus minimizing brittle intermetallic phase formation. Titanium (Ti) copper (Cu) and tantalum (Ta) were selected as the dissimilar metal interlayer materials; each having advantages and disadvantages. Titanium is a commonly used interlayer for this joint, but can microcrack during EXW if the bonding parameters are not correct. Copper has the advantage that it is compatible with hydrogen, but is also known to form brittle intermetallics with aluminum. Tantalum is ductile and bonds well to both Al and stainless steel, and is a high temperature metal that does well to prevent interdiffusion and intermetallic phase formation. However Ta is the most expensive metal of the three. Results of the characterization of the three interlayer bonds showed that Ti produced the highest strength joints, Ta produced the most ductile joints, and Cu produced a joint that failed with low ductility at the Al/Cu interface. Based on these results, the Cu-interlayer joint is not recommended for this application, while the Ti and Ta interlayer bonds both appear to have sufficient strength and ductility for the intended use.

Introduction

Cryogenic pressurized hydrogen (H₂) storage is being considered for safe [1], long-range automobiles [2,3] with essentially zero venting losses under regular use scenarios [4]. Cryogenic pressure vessels comprise a high-pressure inner vessel made of a metallic liner reinforced with carbon fiber (type 3, similar to those used for storage of compressed gas), a vacuum space filled

with numerous sheets of highly reflective metalized plastic (for high performance thermal insulation), and a metallic outer jacket for long-term vacuum stability (Figure 1). Considering that the pressure vessel liner is frequently made of aluminum – a very thermally conductive, relatively weak metal – it is often optimum to use stainless steel inlet and outlet tubes to minimize wall thickness and heat transfer into the vessel. A stainless steel to aluminum transition joint is therefore a critical component for structurally efficient and well-insulated cryogenic pressure vessels. Compact joint design is vital for achieving high storage density, and joint strength and ductility are necessary for durable and impact resistant cryogenic hydrogen storage.

Figure 2a illustrates a typical design of this transition joint, which has a cylindrical plug shape and 4.57 cm diameter. This particular plug is made of 3.18 cm thick 6061-T6 aluminum bonded to 0.95 cm thick 304 stainless steel. The aluminum portion is threaded so that it can be screwed into the aluminum vessel boss, thereby minimizing overall vessel length and enabling cryogenic pressure vessels with high (over 40 grams of hydrogen per liter) *system* storage density [5,6]. The plug is finally arc welded to the vessel liner for hermetic, high-pressure sealing. Four holes are drilled through the plug, allowing four stainless steel tubes to be welded to it, using conventional arc welding techniques. The stainless steel to aluminum dissimilar metal joint presents a welding challenge due to the difference in melting points and densities between the two metals, and due to brittle intermetallic phases that rapidly form during fusion welding [7]. Because of these difficulties, the transition joint was made using an explosive welding (EXW) technique that has been selected as the preferred joining method.

Explosive welding (EXW) is commonly used to fabricate transitions between dissimilar metals that are otherwise difficult to join [8]. Nearly all dissimilar metal combinations can be joined by EXW providing that they have sufficient ductility and toughness at high strain rates to undergo the EXW process. In particular, stainless steel to aluminum joints have been fabricated in the past [9-11]. However, the EXW process is somewhat non-standard, and the bonding parameters are often considered trade secrets that are not discussed in detail. This is partly due to the specialized nature of the process, whereby only a few explosive fabricators perform this work, and partly due to the variety of materials and geometries that are bonded, each requiring setups that have been developed over a period of time by trial and error methods. Since many dissimilar metals are not compatible with each other, thin interlayer materials are required to provide sufficient strength and ductility to the final “bimetallic” EXW joint. These interlayers are rarely discussed in the open literature, but are an important and essential part of the joint. A literature survey revealed one publication that discussed the need for metallic interlayers and suggested candidate interlayer metals [12]. However, it did not reveal any performance data for

such joints. It is therefore imperative to research optimum approaches for building stainless steel-aluminum transition joints that may yield compact cryogenic vessels with the necessary strength and ductility.

Ductile non-ferrous metals frequently used for EXW interlayers are aluminum, copper, nickel, titanium, and refractory metals such as tantalum and niobium. Pure metals, rather than alloys, tend to be softer and have higher ductility at high strain rates, and are generally preferred as interlayer materials. In this work, three different interlayer materials were considered: grade 1 (G1) titanium, commercially pure tantalum, and oxygen-free high thermal conductivity (OFHC) copper. Components were fabricated using these interlayers following similar EXW procedures and testing used at Lawrence Livermore National Laboratory in the past to join niobium to aluminum [13, 14]. This paper discusses the evaluation of bonds made between 304 stainless steel and 6061 T651 aluminum with these three interlayers, and shows that G1 titanium and Ta interlayers can both produce good bonds between these materials for cryogenic pressure vessel applications.

Experimental Procedures

Materials and Explosive Bonding

The EXW joints were made by High Energy Metals, Inc. (HEMI, Sequim, Washington), who developed a procedure for binding high strength aluminum alloys to stainless steel using two additional interlayer metals. These two interlayers are illustrated in Fig 2b, where a soft (annealed) aluminum interlayer is bonded to the thicker 6061-T651 Al base metal as the first step. The thin dissimilar metal interlayer (Cu, Ta, or Ti) is then bonded to the soft aluminum, and finally the 304L stainless steel is bonded to the thin dissimilar metal interlayer to complete the transition. The explosively bonded plates were fabricated with dissimilar metal interlayers of different thicknesses, as summarized in Table 1.

The surfaces to be bonded were ground to a surface finish of 3.2- μm root mean square (RMS) or better and cleaned in order to remove any oxide, dirt, or other foreign objects prior to each bond being made. The development of the bonding parameters was based solely on the experience of the vendor (HEMI), who employed explosive energies in the range of 0.5 to 3 MJ/bond using an ammonium nitrate/fuel oil (ANFO)-based explosive. This level of explosive was designed to produce interface velocities of approximately 2000 m/s to accelerate the flyer plate onto the substrate below and form the bond. Following standard procedures, the flyer plate was positioned above the base metal substrate prior to explosive detonation using small standoff preforms, with a distance approximately equal to the thickness of the sheet or plate

being bonded. Each of the plates met a flatness tolerance of better than 0.2 mm over 0.5 m in length, in order to maintain the uniform stand-off distance between the clad and base metals. The detonation was initiated near the mid-point of one end of the plates being bonded using smaller-capacity detonators for the thin interlayers, and larger capacity detonators for the thick 304 SS plate. Each bond was performed separately using an extra thin shielding sheet when required to prevent the surfaces of intermediate bonds from being damaged. Grinding was performed between bonding steps when necessary to prepare the surface of a bonded plate for the next bond. Preliminary ultrasonic testing was performed at HEMI to insure adequate bonded area in the final product prior to shipping the bonded plates to LLNL.

Ultrasonic testing (UT)

During EXW, an unbounded region is created around the perimeter of the plate, which is caused by edge effects of the explosive wave front. To determine the extent of this unbounded region, each plate was ultrasonically prescanned at HEMI, and then the plates were rough machined to remove the central portion of the bonded plate that contained the well-bonded region. The initial UT scans of the as bonded plates are shown in Figure 3 prior to machining away the unbounded regions. The HEMI scans are performed in a water tank if the bonds are flat enough, and with a hand-held probe if the bonded plates are deformed too severely for automatic scanning. In the case of the Cu bonded plate shown on the top of Fig. 3, the hand held probe was required, where the yellow outline on the plate indicates the transition from poor to good bonding (good bonding inside the yellow outline). The Ti and Ta-bonded plates were flat enough for automated scans as indicated in Fig. 3 middle and bottom. Note that the Ta bonded plate was also co-bonded with an adjacent Ti interlayer that consumed one-half of this plate. The Ti-interlayer portion of the plate was later machined away and set aside for other purposes.

After the good portions of each plate were removed, the plates were sent to LLNL and were rescanned using higher resolution UT after a light machining of the surface of the plates to make them flat. At LLNL, pulse-echo, immersion ultrasound testing was employed whereby a single transducer transmits and receives the ultrasound. The part is placed in a tank of water, and the part and/or transducer is scanned. In this case, the transducer was scanned. However, because of curvature of the parts making a surface following scan difficult, a modified water-tank C-scanning ultrasound technique was employed. For this technique, a flat surfaced ultrasound transducer was placed in contact with the part. The transducer was held by a fixture, which allowed it to follow the surface contour of the part as it scanned along the part. For these scans, the transducer was placed on the stainless steel face of the plates. The acoustic pulses of interest are easily identified by their arrival time. A transducer having a

central frequency of 5 MHz was employed. Acoustic waveforms were captured at various locations on each part, and the peak amplitudes of certain signals at each location were then assembled into images. Since the part was immersed in water, there was ample coupling of the acoustic energy between the transducer and the plates.

Optical Metallography

Cross sections of the explosive bonds were taken perpendicular to the EXW wave front, so that the wave formation could be clearly observed. The samples were polished and etched using conventional metallographic techniques, and were photographed using conventional optical light microscopy. Due to the dissimilar nature of the bonds, optimum etching of each layer was not always possible. However, the results to be presented later clearly show the individual layers and the nature of the different EXW interfaces. The formation of intermetallic compounds was observed at some of the interfaces, particularly at the Cu/Al interface.

Hardness Measurements

Hardness measurements were made on polished cross sections through the joint using a Leco LM 300 AT hardness tester using a diamond Vickers indenter. The tests utilized either a 25 gram or 100 gram weight for indentation that was calibrated on a standard block of known hardness. Arrays of indents were made in base metal layers mounted in epoxy. Indentation size was manually measured using the Amh43 software standard on the Leco test equipment. A 50x objective was used during measurement of the indent diameters. Base metal indents were made 500 μm or farther from the sample edges and interfaces. Indent arrays were spaced 1000 μm apart.

RAM tensile testing

Ram tensile testing was performed on each of the three interlayer bonds. The RAM tensile sample consists of a plug of material containing the EXW bonds in a reduced diameter cross section of the plug. Figure 4 shows a drawing of the test sample, where the stainless steel portion of the plate and the three bonds are located in the reduced section (bottom) portion of the sample. Figure 5 shows photographs of the RAM tensile sample and the loading fixtures that are placed in a standard crosshead driven tensile testing machine. Testing was performed by inserting a ram into the bored out portion of the sample, and pressing on the ram to load the bond zone in tension. The specimens are loaded while monitoring crosshead displacement and load, until failure or significant load drop is observed.

The RAM testing was performed on the Sintech electromechanical test frame, with a Sintech load cell S/N 1159 calibrated to $\pm 1\%$. As shown in Fig. 5, a 2.5 cm diameter spherical shaped load button was attached to the load cell to apply the compressive load to the ram pusher. A small steel platen was inserted between the load button and pusher to distribute the load and protect the surface of the pusher. Specimens were seated into a cylindrical base which had a shoulder around the inside diameter (Area = 1.77 cm²). The cylindrical base was positioned in the center of a spherical seat.

The tests were conducted at a crosshead speed of 0.050 cm/min. Data acquisition rate was 20 Hz, to capture time (s), crosshead deflection (mm), and load (N). The crosshead was manually lowered to approximately 0.063 cm above the surface of the steel platen prior to test to avoid preloading of the sample. Testing was performed using Military Specification MIL-J-24445A(SH) [15], and ASTM B898 [16] as references for sample design, loading and measurement.

Results and Discussion

Optical Metallography

Figure 6 shows the microstructure of the Cu-interlayer bonded plate. A low magnification overview of the bonds is shown in Fig. 6a, where the stainless steel (SS) is located on the top of the photo, and the three successive bond lines appear between the stainless steel and the 6061-T651 aluminum base plate. The bonds are clearly visible, showing nicely formed waves at the SS/Cu interface, a relatively flat Cu/Al interface, and large wave formation at the Al/Al interface. A higher magnification micrograph of the Cu/SS interface is shown in Fig. 6b that indicates good wave formation, amplitude on the order of 100 μm , and good bonding with no pores or cracks. A higher magnification micrograph of the Cu/Al interface (Fig. 6c) shows some notable features. First, the interface is very flat, with amplitude on the order of only about 10 μm . Second, there appears to be an intermetallic phase forming between the Al and Cu layers which etches darker than the interlayer metals. Third, there appear to be some very dark etching inclusions about 20 μm in diameter appearing near the interface. All three of these indications can lead to poor joint properties, particularly under fatigue or impact loading conditions. The third bond is shown in Fig. 6d at higher magnification, and indicates a deep wave pattern at the Al/Al interface with amplitude on the order of 500 μm . This Al/Al bond looks very good with no pores or cracks present in the cross section.

Figure 7 shows the microstructure of the Ta-interlayer bonded plate. A low magnification overview of the bonds is shown in Fig. 7a, where the stainless steel (SS) is located on the top of the photo, and the three successive bond lines appear between the stainless steel and the 6061-T651 aluminum base plate. The bond lines are clearly visible, showing smaller amplitude waves than on the Cu-interlayer joint in general. A higher magnification micrograph of the Ta/SS interface is shown in Fig. 7b that indicates well-formed waves between the two materials with amplitudes of approximately 50 μm . No cracks or pores were observed at this interface, and it appears to be a well-formed joint. A higher magnification micrograph of the Ta/Al interface is shown in Fig. 7c, which is relatively flat, with wave amplitude of approximately 20 μm . One obvious feature at this bond line is the appearance of a darker etching aluminum phase, sandwiched in between the original Al interlayer and the Ta. Within this darker etching Al phase are small fragments of Ta, which is evidence that this layer may have melted during the EXW bonding, thus allowing the complete entrapment of small Ta fragments. The third bond is shown in Fig. 7d at higher magnification, and indicates a wave pattern at the Al/Al interface with amplitude on the order of 200 μm . This Al/Al interface again looks very good with no pores or cracks present in the cross section.

Figure 8 shows the microstructure of the Ti-interlayer bonded plate. A low magnification overview of the bonds is shown in Fig. 8a, where the stainless steel (SS) is located on the top of the photo, and the three successive bond lines appear between the stainless steel and the 6061-T651 aluminum base plate. The bond lines are clearly visible, and all three bonds show visible waves at their respective interfaces. A higher magnification micrograph of the Ti/SS interface is shown in Fig. 8b that indicates shallow and somewhat irregular wave formation with approximately 40 μm amplitude. Some small cracks appear to be present at the bond line that extends approximately 20 μm into the SS. A higher magnification micrograph of the Ti/Al interface is shown in Fig. 8c, which shows nicely formed waves with approximately 50 μm amplitude. No cracks or pores appear, and this interface appears to be the best Interlayer/Al bond of the three interlayers tested. The third bond is shown in Fig. 8d at higher magnification, and indicates a wave pattern at the Al/Al interface with amplitude on the order of 400 μm . This Al/Al interface again looks very good with no pores or cracks present in the cross section.

Ultrasonic Scans on Machined Plates at LLNL

After rough machining, the dimensions of the plates measured 20x25 cm for the Cu-interlayer plate, 12.5x43.2 cm for the Ta-interlayer plate, and 25x60 cm for the Ti-interlayer plate. These

plates were further tested using high resolution UT to look for defects or unbounded regions in what is believed to be the good bonded portions of each plate.

Figure 9 (top) shows amplitude versus time ultrasound trace from the Cu interlayer plate. The first signal (at $t \sim 4.5 \mu\text{s}$) is a very small blip corresponding to the ultrasound reflecting off the steel-Cu interface. The acoustic impedances of these two materials are well matched, and a small reflection indicates that most of the acoustic energy is transmitted through the interface. Directly behind this blip (at $t \sim 5 \mu\text{s}$) is a larger signal corresponding to the ultrasound reflecting off the Cu-Al interface (since the two aluminums are very similar, they are considered as a single acoustic layer). This signal is larger than the steel-Cu interface and the acoustic impedance mismatch is much larger. The Cu was the thickest interlayer, and this facilitated easy identification of the front and back surface reflections from this interlayer. The largest signal seen (at $t \sim 16 \mu\text{s}$) corresponds to the reflection off the Al back surface. Figure 9 (middle) is a C-scan image of the peak value of the interface signal (in this case Cu-Al). Figure 9 (bottom) is a C-scan image of the peak value of the back wall reflection. If there had been a complete lack of bonding at this location, the first signal would have been much larger, and there would have been no signal corresponding to the back surface of the aluminum.

Although there is a fair amount of amplitude variation seen in Figure 9 (middle), Figure 9 (bottom) is qualitatively very similar. Acoustic energy was reflected back to the transducer at all locations, indicating that the layers were bonded at all locations. Since the two images are similar, the ratio of the two amplitude images is similar indicating that the bond quality is similar across the entire part. A lot of the variation in the peak amplitude is likely caused by differences in the amount of acoustic energy getting into the part. Some of this may have been caused by surface roughness. Therefore, looking for differences in the interface reflection and the back surface reflection in each of the plates will help identify bond strength variation.

Figure 10 (top) shows amplitude versus time waveform from the Ta interlayer plate. The Ta interlayer was the thinnest of the three interlayers and it is not possible to distinguish separate arrivals from the front and back surfaces (at $t \sim 4.5 \mu\text{s}$). For this plate, the steel-Ta interface had a much greater acoustic impedance mismatch compared to the steel-Cu interface. Therefore, the back wall reflection (at $t \sim 16 \mu\text{s}$) is smaller relative to the interface signal. Figure 10 (middle and bottom) show peak amplitude images for the interface, and back wall respectively. As with the Cu interlayer plate, there is a fair amount of amplitude variation seen in these two images. However, the two images show a similar variation, and therefore indicate that the bond is fairly consistent throughout much of the plate. There is a strip along the left side and the bottom and a few small areas along the top that have very low amplitude return. Acoustic energy

makes it through all of these areas, but since the amplitude is so low, it is difficult to judge bond quality. The tensile samples were machined from the right side of the plate.

Figure 11 (top) shows amplitude versus time waveform from the Ti interlayer plate. Although the Ti interlayer was fairly thin, it is possible to distinguish separate arrivals from the front and back surfaces (at $t \sim 4.5 \mu\text{s}$), although the back surface signal arrives before the front surface signal has diminished. For this plate, the steel-Ti interface had a much lower acoustic impedance mismatch compared to the steel-Ta interface. Therefore, the back wall reflection (at $t \sim 16 \mu\text{s}$) is larger relative to the interface signal. Figure 11 (middle and bottom) show peak amplitude images for the interface and back wall respectively. As with the other plates, there is a fair amount of amplitude variation seen in these two images. However, the two images show a similar variation, and therefore indicate that the bond is fairly consistent throughout much of the plate. There is a strip along the left side of the plate that has lower amplitude, and this resulted from the back surface of the Al plate being deformed in that area. This meant that acoustic energy reflecting off this deformed part did not retro-reflect and was in fact reflected away from the transducer. Because of this, tensile samples were machined from the right side of the plate.

Hardness Measurements

Microhardness measurements were made in the base metals and in the thin interlayers. These data were compared to handbook values in order to get confirmation of the heat-treating of the materials and any additional changes that might have occurred through explosive bonding. The data are summarized in Table 2 for each of the bonds. Average readings are shown based on a minimum of 5 hardness measurements per location. For the 6061-T6 Al and 304SS base metals, measurements were made close to the bond (within 1 mm of the interface) and far from the bond (more than 6 mm from the interface) to see if any localized strain hardening could be detected from the EXW process.

The hardness values of the 6061-T651 base metal are essentially the same near and close to the interface at 127 HV. This value is higher than typical handbook values of about 110 Hv but is not unusual for peak hardness 6061-T651. The invariability of the hardness near and far from the interface is expected since the -T651 condition is already at peak hardness, and was not expected to increase in hardness by the EXW process. The hardness of the 304 SS base metal on the other hand, showed significant differences throughout the EXW plate. Average values for the three plates showed a hardness of 276 HV close to the interface, and an average value of 319 HV far from the interface. Both values are significantly harder than annealed 304 SS, which is reported to be about 129 HV. Clearly the EXW process strain hardened the 304 SS and

produced a gradient in hardness throughout the plate. Additional observations are that the hardness of the 304SS in the Cu-interlayer plate is higher than that of the Ta-interlayer or Ti-interlayer plates; presumably due to different EXW parameters that may have been used to bond the different interlayers.

Hardness measurements were also made on the thin interlayers, and these data are also reported in Table 2. In all cases the hardness values of the interlayers are significantly higher than the reported annealed handbook values. For the G-1 Ti interlayer, the hardness measured 209 HV, which is about 50% harder than reported annealed G-1 Ti. For the Cu interlayer, the hardness measured 129 HV, which is more than 2x the reported annealed value. For the Ta interlayer, the hardness measured 163 HV, which is about 50% higher than the reported annealed value. In all cases, the interlayer hardness is significantly higher than that of the soft Al interlayer, so failure of these bonds would be expected to be in the soft Al interlayer if the bonds are ductile, or at an interface if the bonds are brittle.

Ram Tensile Tests

Ram tensile testing was performed on two samples from each of the three interlayer bonds. In the results that follow, the stress was calculated from the load using the cross sectional area of the annular region of the bond line in tension for each sample as indicated in Table 3. Strain was not measured directly on these samples due to the multiple materials in the bonds, so the RAM tensile results are plotted as stress vs. crosshead displacement.

The Cu-interlayer bond results are shown in Fig. 12, where the two tests are superimposed and show very similar behavior. The peak stress of the joint averaged 1920 bar, which is significantly higher than the strength of the 6061-O Al interlayer, which is reported to have a yield strength of 552 bar and an ultimate strength of 1240 bar as summarized in Table 4. The increased strength of the joint over the properties of 6061-O Al is a combination of the strain hardening induced in the thin interlayer during the bonding process, and constraint produced by the stronger materials on either side of the soft interlayer which have ultimate strengths in excess of 2480 bar as summarized in Table 4. The fracture in both samples occurred quickly with an audible report, as the load dropped to nil with little additional strain during the fracture.

Figure 13 shows macrophotographs of the fractured Cu-interlayer parts. The SS side of the RAM tensile bar is shown in Fig. 13a, which clearly shows that separation occurred at the Al/Cu interface, with the entire Cu interlayer adhering to the SS portion of the sample. Fig. 13b shows the other side of the RAM tensile bar, which contains the entire thickness of the Al-O interlayer and Al-T651 base metal. Deformation of the soft Al is apparent where the EXW waves in the

harder Al stand proud at Al/Al interface. Fig. 13c shows the fracture surface on the Al side of the broken sample. The fracture surface is macroscopically flat and cleanly separated at the Al/Cu interface. The rapid fracture and flat fracture surface are indications that this joint may have notch and impact toughness concerns.

The Ti-interlayer bond results are shown in Fig. 14, where the two tests are superimposed and show very similar behavior to each other. The peak joint stress averaged 2570 bar, which is again significantly higher than the strength of the 6061-O Al interlayer for similar reasons as before. In addition, the Ti interlayer joint is stronger than the Cu-interlayer bond by about 40%. The fractures in the Ti-interlayer joints also occurred with an audible report, but not nearly as loud as those of the Cu-interlayer bonds. It is clear from the stress-displacement curves that the sample did deform a bit more than the Cu-interlayer bonds after peak load, thus showing some ductility during the fracture process.

Figure 15 shows macrophotographs of the fractured Ti-interlayer parts. The SS side of the RAM tensile bar is shown in Fig. 15a, which shows a thick AL layer bonded to the Ti-interlayer bonded to the SS base metal. The thick Al has a roughened and tapered appearance and is the result of a ductile “cup” and “cone” type fracture that occurred in the soft Al interlayer. Fig. 15b shows the other side of the RAM tensile bar, which also shows a substantial amount of the soft AL interlayer attached to the Al-T651 base metal. This is the “cup” portion of the failure as identified by the machining marks on the surface of the Al interlayer. Deformation of the soft Al is apparent where the EXW waves in the harder Al again stand proud at Al/Al interface. Fig. 15c shows the fracture surface on the Al side of the broken sample. Unlike the Cu-interlayer bond, this surface is macroscopically rough with shear lips forming the “cup” shape of the fracture, and indicating that the failure occurred in a ductile manner within the soft Al interlayer.

The Ta-interlayer bond results are shown in Fig. 16, where the two tests are superimposed and again show similar behavior to each other. The peak joint stress averaged 1960 bar, which is again significantly higher than the strength of the 6061-O Al interlayer, for the same reasons as the other two joints. The largest difference between the Ta interlayer joints and the other joints is the amount of deformation that occurs after the peak load is reached. In the case of the Ta interlayer joint, the displacement after peak load is approximately 0.25 mm, as compared to approximately 0.025 mm for the Cu and Ti interlayer joints. The ductile behavior resulted in a slow tearing of the sample through the soft Al interlayer such that the two halves of the RAM sample never completely separated.

Figure 17 shows macrophotographs of the fractured Ta-interlayer parts. This interlayer displayed the most ductility, and never completely failed. Fig. 17a shows the part after testing, which hinged about one part of the specimen. In this figure, the SS portion of the sample is shown on the top of the photograph. As with the Ti-interface bond, approximately half of the Al interlayer is attached to each side of the fracture surface, which shows a cup and cone macroscopic appearance. In the case of the Ta-interlayer bond, the cupped portion of the fracture surface is attached to the SS side of the joint and the cone portion is attached to the Al side of the joint. Deformation of the soft Al is again apparent where the EXW waves in the harder Al again stand proud at Al/Al interface. Figure 17b shows a close up photograph of the SS side of the joint, and upon close examination necking can be observed in the vicinity of the Ta interlayer. This behavior was not observed in either the Ti or the Cu interlayer joints and most likely contributed to the increased strain at failure and increased ductility of the Ta-interlayer joint compared to the others. Fig. 17c shows the fracture surface on the Al side of the broken sample. This surface is similar to that of the Ti-interlayer joint and is macroscopically rough with shear lips. However, the fracture on the Al side of the Ta-interlayer joint forms the cone rather than the cup shape of the fracture. This behavior again indicates that the failure occurred in a ductile manner within the soft Al interlayer.

Conclusions

Compact, ductile and strong stainless steel-aluminum transition joints are critical for cryogenic pressurized hydrogen storage. However, stainless steel to aluminum joints present a welding challenge due to the difference in melting points and densities between the two metals, and due to brittle intermetallic phases that rapidly form during fusion welding. Since aluminum and stainless steel are not compatible with each other, thin interlayer materials are required to provide sufficient strength and ductility to the final “bimetallic” EXW joint. In this paper we researched three interlayer metals (copper, titanium, tantalum) to determine best alternatives for optimum cryogenic pressure vessel performance.

All three interlayer metals produced EXW bonds between 6061 Al and 304 SS. Optical metallographic cross sections showed classic wave formation and what appeared to be good bonding at most of the interfaces with a few exceptions. Cu-interlayer bond: the Cu/Al interface was flat and appeared to have formed intermetallic compounds between the two metals. Ta-interlayer bond: some melting may have occurred in the aluminum at the Ta/Al interface. Ti-interlayer bond: some microcracking was observed in the stainless steel at the Ti/SS interface. RAM tensile testing of the bonds was performed to get a measure of failure

stress and to get a relative measure of ductility of each of the bonds. The results of the RAM tensile tests showed that all of the joints exceeded the strength of the weakest material (the soft 6061-O Al, UTS=1240 bar) by 50% or more due to the constraint of the surrounding stronger materials. Furthermore, Ti produced the highest strength joint, Ta produced the most ductile joint, and Cu produced a joint that failed with low ductility at the Al/Cu interface. Based on these results it is recommended that the bimetallic transition joints be produced from either the Ta-interlayer or the Ti-interlayer bonds. The higher ductility of the Ta-interlayer bond should provide more reliability from impact and fatigue than the Ti-interlayer bond. However, the Ti-interlayer bond should be able to withstand higher loads prior to failure. The only bond that is not recommended for this application is the Cu-interlayer bond due to its apparent brittle nature as a result of intermetallic compounds at the Cu/Al interface.

Acknowledgments

This project was funded by DOE, Office of Hydrogen and Fuel Cell Technologies, Jesse Adams, Technology Development Manager. This work performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344.

References

1. G Petitpas, SM Aceves, Modeling of sudden hydrogen expansion from cryogenic pressure vessel failure, *International Journal of Hydrogen Energy* 38 (19), 8190-8198.
2. SM Aceves, G Petitpas, F Espinosa-Loza, MJ Matthews, E Ledesma-Orozco, Safe, long range, inexpensive and rapidly refuelable hydrogen vehicles with cryogenic pressure vessels, *International Journal of Hydrogen Energy*, 38, 2480-2489, 2013.
3. SM Aceves, F Espinosa-Loza, E Ledesma-Orozco, TO Ross, AH Weisberg, TC Brunner, O Kircher, High-density automotive hydrogen storage with cryogenic capable pressure vessels, *International Journal of Hydrogen Energy*, 35, 1219-1226, 2010.
4. G Petitpas, SM Aceves, MJ Matthews, JR Smith, Para-H₂ to ortho-H₂ conversion in a full-scale automotive cryogenic pressurized hydrogen storage up to 345 bar, *International Journal of Hydrogen Energy* 39 (12), 6533-6547, 2014
5. A comparative analysis of the cryo-compression and cryo-adsorption hydrogen storage methods, G Petitpas, P Bénard, LE Klebanoff, J Xiao, S Aceves, *International Journal of Hydrogen Energy*, 39 (20), 10564-84, 2014.

6. Ahluwalia RK, Hua TQ, Peng JK. On-board and off-board performance of hydrogen storage options for light-duty vehicles. *International Journal of Hydrogen Energy*, 37(3), 2891-910, 2012.
7. Atabaki MM Nikodinovski M Chenier P Ma J Harooni M Kovacevic R, Welding of Aluminum Alloys to Steels: An Overview. *Journal for Manufacturing Science and Production*, 14(2) 59–78, 2014.
8. Banker JG Reineke EG Explosive Welding, *Metals Handbook*, Tenth Edition, ASM International, Vol. 6, 1993.
9. Acarer M Demir B An investigation of mechanical and metallurgical properties of explosive welded aluminum-dual phase steel, *Materials Letters*, 62, 4158-4160, 2008.
10. Tricarico L Spina R Sorgente D Brandizzi M Effects of heat treatments on mechanical properties of Fe/Al explosion-welded structural transition joints, *Materials and Design*, 30 2693-2700, 2009.
11. Young GA Banker JG Explosion welded, bi-metallic solutions to dissimilar metal Joining, *Proceedings of the 13th Offshore Symposium (Texas Section of the Society of Naval Architects and Marine Engineers)*, Houston, Texas, February 24, 2004.
12. Cutter D What you can do with explosion welding, *Welding Journal* 85(7), 38-43, 2006.
13. Palmer TA Elmer JW Brasher D Butler D Riddle R Development of an Explosive Welding Process for Producing High Strength Welds between Niobium and 6061-T651 Aluminum, *Welding Journal* 2006, 85(11), 252s-263s.
14. Elmer JW Terrill P Brasher D Butler D Joining Depleted Uranium to High-Strength Aluminum Using an Explosively Clad Niobium Interlayer *Welding Journal*, 81(8), 167s-173s, 2002.
15. Military Specification MIL-J-24445A(SH), "Joint Bimetallic bonded, aluminum to steel," Section 4.6.2, July 25, 1977.
16. ASTM B898-99 "*Standard Specification for Reactive and Refractory Metal Clad Plate*", 1999.

Tables

Table 1: Materials and thicknesses used to make each of the EXW plates.

Material	Cu-Interlayer plate (mm)	Ti-Interlayer plate (mm)	Ta-Interlayer plate (mm)
6061Al-T651	38.1	38.1	38.1
6061Al-O	1.57	1.57	1.57
Interlayer	1.57	0.508	0.178
304 SS	12.7	12.7	12.7

Table 2: Results of the hardness tests of the different metals and interlayers in the bonded plates. Data are compared to typical (handbook) data prior to EXW bonding.

Material	Al 6061 (-T651)	Al 6061 (annealed)	Ti G1 (annealed)	Cu OFHC-101 (annealed)	Ta (annealed)	304 SS (annealed)
	HV-25	HV-25	HV-100	HV-25	HV-25	HV-25
Typical handbook values	110	30	130	48	110	129
Ti-interlayer plate (close/far)*	128/129	61	209	N/A	N/A	300/256
Cu-Interlayer plate (close/far)*	126/128	49	N/A	129	N/A	345/317
Ta-interlayer plate (close/far)*	127/128	50	N/A	N/A	163	312/256
Average (close/far)*	127/128	53	N/A	N/A	N/A	319/276

* Base metal measurements were made close (<1mm) and far (>6mm) from the interface.

Table 3: Ram tensile test results for each of the three interlayers.

Specimen	Crosshead at peak load (mm)	Peak Load (kN)	Tensile Area (cm ²)	Ultimate Strength (bar)	Failure Location
Copper1	0.737	44.1	2.26	1950	Cu/Al, flat fracture
Copper2	0.685	42.3	2.25	1880	Cu/Al, flat fracture
Titanium1	0.914	57.3	2.25	2540	Al-O, cup and cone

Titanium2	0.914	58.1	2.24	2590	Al-O, cup and cone
Tantalum1	0.737	42.5	2.24	1900	Al-O, cup and cone
Tantalum2	0.737	45.3	2.25	2010	Al-O, cup and cone

Table 4: Typical handbook values of yield and ultimate strength of the EXW materials

Property	Al 6061 (-T651)	Al 6061 (annealed)	Ti G1 (annealed)	Cu OFHC-101 (H00)	Ta (RXL)	304 SS (annealed)
	(bar)	(bar)	(bar)	(bar)	(bar)	(bar)
Yield Strength	2760	552	2070	1930	2070	2070
Ultimate Strength	3100	1240	2760	2480	2410	5170

Figures

Figure 1. Cryogenic pressure vessel configuration [2]. The inner vessel is an aluminum-lined, carbon fiber-wrapped (type 3) pressure vessel similar to those typically used for storage of compressed gases. This vessel is surrounded by a vacuum space filled with numerous sheets of highly reflective metalized plastic (minimizing heat transfer into vessel), and an outer metallic jacket to preserve the vacuum.

Figure 2: Top-Schematic drawing of the transition joint where the explosive weld joins 304 SS to 6061-T6 aluminum. Dimensions in mm. Bottom—Details of the "explosive bond line" where thin soft aluminum and a dissimilar metal interlayer are used to make the bond between 304SS and 6061-T651 Al.

Figure 3: Initial HEMI ultrasound (UT) scans of the explosively bonded plates before machining. Top shows the Cu bonded plate which was hand scanned due to plate curvature, the middle shows the Ta and Ti interlayer bonded plate, and the bottom shows the Ti bonded plate. Regions of good bonding are indicated within the boxed regions. Dimensions are in cm.

Figure 4: RAM Tensile Coupon Specifications. Dimensions in mm.

Figure 5: Photographs of the RAM tensile test setup. The upper left shows the RAM tensile sample for one of the Cu-interlayer specimens. The upper right shows this same sample along with the ram (left) and die (right). The lower photo shows the parts assembled in the testing fixture with the load cell and centering fixture in place.

a

b

c

d

Figure 6: Optical metallographic cross section through the Cu-Interlayer bond. A) Overview of the three interfaces with 304 SS on top (marker=1000 μm), b) high magnification of the Cu/304L SS interface (marker=100 μm), c) high magnification of the Cu/Al interface showing intermetallic (light gray) phase formation (marker=100 μm), and d) high magnification of the 6061 Al-O/6061 Al-T651 interface (marker=400 μm).

a

b

c

d

Figure 7: Optical metallographic cross section through the Ta-Interlayer bond. A) Overview of the three interfaces with 304 SS on top (marker=1000 μm), b) high magnification of the Ta/304L SS interface, c) high magnification of the Ta/Al interface (marker=100 μm), and d) high magnification of the 6061 Al-O/6061 Al-T651 interface (marker=400 μm).

a

b

c

d

Figure 8: Optical metallographic cross section through the Ti-Interlayer bond. A) Overview of the three interfaces with 304 SS on top (marker=1000 μm), b) high magnification of the Ti/304L SS interface (marker=200 μm), c) high magnification of the Ti/Al interface (marker=200 μm), and d) high magnification of the 6061 Al-O/6061 Al-T651 interface (marker=400 μm).

Figure 9: Ultrasound testing scans of the Cu interlayer plate. The top figure shows amplitude versus time trace obtained from Cu interlayer plate, the middle shows peak amplitude image of Cu-Al interface, and the bottom shows peak amplitude image of Al free surface.

Figure 10: Ultrasound testing of the Ta interlayer plate. The top figure shows amplitude versus time trace obtained from Ta interlayer plate. The Ta layer is much thinner than the acoustic wavelength and therefore the reflection off the front and back surfaces of the Ta plate suffer superposition and are not clearly distinguishable. The middle figure shows peak amplitude image of steel-Ta-Al interfaces, and the bottom figure shows peak amplitude image of Al free surface.

Figure 11: Ultrasound testing scans of the Ti interlayer plate. The top figure shows amplitude versus time where the first signal shows that the reflection from the front and back surfaces of the Ti layer arrived very close in time to one another. The middle shows the peak amplitude image of steel-Ti-Al interfaces, and the bottom figure shows peak amplitude image of Al free surface.

Figure 12: Ram tensile test results for the two Cu-interlayer samples plotted in stress versus crosshead displacement.

Figure 13: Photographs of the fracture surface of the Cu-interlayer bond made at 30x magnification prior to reduction. a) SS side of the RAM tensile bar showing Cu/SS bond line and fracture at Cu/Al interface. b) Al side of the RAM tensile bar showing deformation at the Al/Al interface. C) Fracture surface detail on the Al side of the RAM tensile bar, with Al-O interlayer intact.

Figure 14: Ram tensile test results for the two Ti-interlayer samples plotted in stress versus crosshead displacement.

Figure 15 Photographs of the fracture surface of the Ti-interlayer bond made at 30x magnification prior to reduction. a) SS side of the RAM tensile bar showing SS/Ti/Al-O bonds and the “cone” portion of the fracture in the soft aluminum interlayer. b) Al side of the RAM tensile bar showing deformation at the Al/Al interface and the “cup” side of the soft aluminum interlayer. C) Fracture surface detail on the Al side of the RAM tensile bar.

Figure 16: Ram tensile test results for the two Ta-interlayer samples plotted in stress versus crosshead displacement.

Figure 17: Photographs of the fracture surface of the Ta-interlayer bond made at 30x magnification prior to reduction. This sample hinged but never separated into two pieces. a) SS side (top) contains the “cup” portion of the soft Al interlayer fracture. b) Close up of the SS side of the joint, showing evidence of deformation in the Ta interlayer. C) Al side of the fracture surface showing the “cone” portion of the soft Al interlayer fracture surface.