



Ultrasonic Assisted Soldering of Metallized Polymer Current Collectors for Batteries

Joints prepared with ultrasonic-assisted soldering were analyzed for mechanical and electrical performance by varying three major process parameters

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Abstract

In this work, a novel ultrasonic-assisted soldering (UAS) process was demonstrated for joining large stacks (64 films) of aluminum metallized polymer current collectors (MPCCs) to a tab for lithium-ion batteries (LIBs). When integrated into cells, MPCCs can act as a passive safety mechanism for thermal runaway and increase the gravimetric energy density. Joining MPCCs is difficult due to their composite nature, and traditional processes may be insufficient to enable integration of MPCCs in LIBs at scale. This experimental study investigated the impact of three key process parameters of through-hole UAS (welder cylinder pressure, ultrasonic vibration time, and vibration amplitude) on the electrical resistance and mechanical strength of the resulting joints. Varying the cylinder pressure had an insignificant effect on joint quality, except that at high cylinder pressures, film damage occurred at the joint area. Inconsistent joint quality was observed with short ultrasonic vibration times. For long ultrasonic vibration times, over-soldering occurred where the solder penetrated through the tab. The amplitude of vibration significantly impacted the mechanical strength and electrical resistance of the joints. The best performing joints were prepared within the low-to-middle range of vibrational amplitude tested (7.8–14.4 μm). A more thorough layer-by-layer resistance analysis was also performed on joints produced with near-optimum conditions. These joints had an average layer resistance of 0.0045 ohms. Optical microscopy and SEM imaging revealed that joints

made with near-optimum conditions had deeper solder penetration between MPCC films, compared to joints made with a lower amplitude.

Keywords

- Metallized Polymer Current Collectors
- Current Collectors
- Lithium-Ion Batteries
- Ultrasonic-Assisted Soldering
- Soldering
- Ultrasonics

Introduction

Current collectors (CCs) in lithium-ion batteries (LIBs) are a crucial component that provides an external electrical connection and supports the active material. While CCs play an essential role in battery cells, they are a passive component and contribute to the “dead weight” of a cell (Ref. 1). It has been estimated that CCs account for 15% of the weight of a LIB, although this number can vary depending on the cell format (Ref. 2).

Metallized polymer current collectors (MPCCs) are composed of a composite structure where a thin layer (~1 micron thick) of aluminum or copper is coated on both sides of a thin polymer substrate (~6 micron thick). MPCCs have been proposed and implemented as a way to increase the safety of LIBs. MPCCs can act as a passive safety mechanism in LIBs by isolating internal short circuits, significantly reducing the risk of thermal runaway (Ref. 3). When an internal short circuit occurs, the initial heat generation causes thermal degradation in the MPCC polymer substrate (Ref. 3). The MPCC will then “retreat” from the internal short location, eliminating the short circuit and preventing thermal runaway escalation (Ref.

3). MPCCs also provide benefits, such as reduced mass and material costs (Ref. 3). Additionally, MPCCs have improved ductility compared to foil CCs at similar thicknesses, due to the polymer substrate (Ref. 3); however, they also exhibit increased electrical resistance and decreased heat dissipation ability.

In LIBs, CCs must be electrically connected and mechanically bonded to an arrester tab. Depending on the cell format and design, this joint can include a single CC in a tab or many CCs in a tab. The quality of this CC(s)-to-tab joint affects the cell's internal resistance and is important for long-term cell health (Ref. 4). Integrating MPCCs into LIBs is challenging due to their unique composite nature, which presents joinability issues. The ultrathin (~1 micron) metallized layers have insufficient material to weld to each other and to tabs with sufficient mechanical and electrical properties. Additionally, the polymer substrate within MPCCs acts as an electrical insulator in the thickness direction, isolating each metallization layer.

The challenges in joining MPCCs for pouch cells have been documented by Gruhn et al. (Ref. 5), wherein the authors evaluated ultrasonic metal welding (UMW), laser welding, and mechanical clamping for joining MPCCs. Each process had its own limitations, and the authors concluded that further research should focus on developing new processes and adapting existing ones to integrate MPCCs. In their overview of MPCCs, Zhang et al. (Ref. 4) briefly discuss welding and improving weld quality. They suggest adding metal foils between MPCC layers to improve UMW, as well as double-sided ultrasonic roll welding. Pham et al. (Ref. 3) and Ye et al. (Ref. 1) did not detail the specific welding process they used. Works such as those aforementioned have demonstrated that it is possible to create LIBs with MPCCs on a laboratory scale; however, developing a robust joining solution for mass manufacturing remains a challenge.

For this work, soldering was considered a process of interest for joining MPCCs because solder could flow between layers of MPCCs at a temperature below the melting point of the polymer substrate. In conventional soldering, flux is necessary to remove surface oxides and contaminants, promoting wetting (Refs. 6, 7). In this work, only aluminum MPCCs were studied because aluminum is less solderable than copper (Ref. 8). The corrosive flux used to solder aluminum can totally consume the very thin aluminum layer in an MPCC, as well as a susceptible polymer substrate, such as polyethylene terephthalate (PET), the polymer substrate used in this work. Additionally, residual flux can cause corrosion issues, which may affect long-term reliability (Ref. 9). Fluxless soldering is possible through ultrasonic-assisted soldering (UAS) (Refs 6, 10, 11), which is explored in this work. Fluxless soldering is also possible through the use of "active solders" (Ref. 12). Active solders include elemental additions (In, Hf, Ti, Zr) that enable bonding of a non-wetting solder to a material by forming an interfacial reaction layer (Ref. 12). However, active solders are more expensive than traditional solders, require mechanical agitation to clear the molten solder oxide layer, and do not flow on their own (Ref. 12). The necessary mechanical agitation and lack of flow can both be addressed through the use of ultrasound (Ref. 12). Future work will explore the use of active solders for joining MPCCs.

UAS can be used to combine materials that are traditionally difficult to solder, including glasses (Refs. 13, 14), ceramics

(Refs. 15, 16), and various metals (Refs. 17–19), including aluminum (Refs. 20–25). Cavitation has long been considered a vital mechanism behind the UAS of aluminum (Refs. 23–25). When cavitation occurs near a solid surface, the collapse is nonspherical, and high-speed liquid jet streams drive into the solid surface, which fragments and clears oxides, thus facilitating wetting (Ref. 26). Acoustic streaming, where fluid flow is induced by an acoustic pressure gradient (Ref. 27) is another important phenomenon in UAS. Acoustic streaming can improve heat transfer in liquids and provides non-contact stirring (Ref. 27). Ultrasonics can also cause faster and greater filling of capillaries, pores, and voids — a phenomenon known as the ultrasonic capillary effect (UCE) (Refs. 15, 28, 29). Cavitation has been proposed as the driving mechanism behind the UCE (Ref. 30). However, Xu et al. (Refs. 28, 29) recently demonstrated that cavitation is not essential for the UCE.

In this work, a novel UAS process, which we refer to as "through-hole UAS," was developed to join MPCCs to themselves and to a tab, achieving high consistency with strong mechanical joints and low electrical resistances. Figure 1A presents a top-view photo of a through-hole UAS joint (64 films to a tab) and Fig. 1B illustrates a cross-section side view of a joint. In previous work (Ref. 31), we showed that through-hole UAS was capable of joining 64 aluminum MPCCs to a tab, while UMW could only join 32 aluminum MPCCs to a tab. Additionally, through-hole UAS produced stronger joints with lower electrical resistance and higher consistency compared to UMW.

In LIBs, CCs must maintain good chemical and electrochemical stability (Ref. 32). Corrosion of CCs can lead to debonding of the active material (Ref. 32). Corrosion products can also reduce the electrical capacity of LIBs and pose safety issues (Ref. 32). Although the CC-to-tab solder joint does not contain active material, the joint area is housed within the cell casing and can thus be exposed to battery electrolyte. For joining MPCCs, a low-melting-point solder must be used, where tin (Sn) based alloys are a common (non-lead [Pb]) option for low-melting-point solders. Preliminary experiments by collaborating researchers at the University of South Carolina (Ref. 33) has shown that Sn-based solders corrode within a LIB cell environment when directly exposed to electrolyte. Therefore, a solder joint used to join MPCCs must be protected from the electrolyte. To isolate a solder joint from the electrolyte, the joint would need to be fabricated outside of the battery housing or hermetically sealed from the electrolyte.

As research and development continue to increase the energy density and power of LIB, technology to prevent thermal runaway is becoming increasingly critical. MPCCs have the unique ability to act as a passive safety mechanism while also increasing energy density. However, the integration of MPCCs into LIB on a manufacturing scale is currently limited by the inherent joining issues of MPCCs. New and novel joining methods may be necessary to realize the benefits of MPCCs. UAS and through-hole UAS are potential solutions for joining MPCCs. Through-hole UAS can create mechanically strong, low-resistance joints with large stacks of MPCCs, which has not been possible with traditional welding processes, such as UMW.

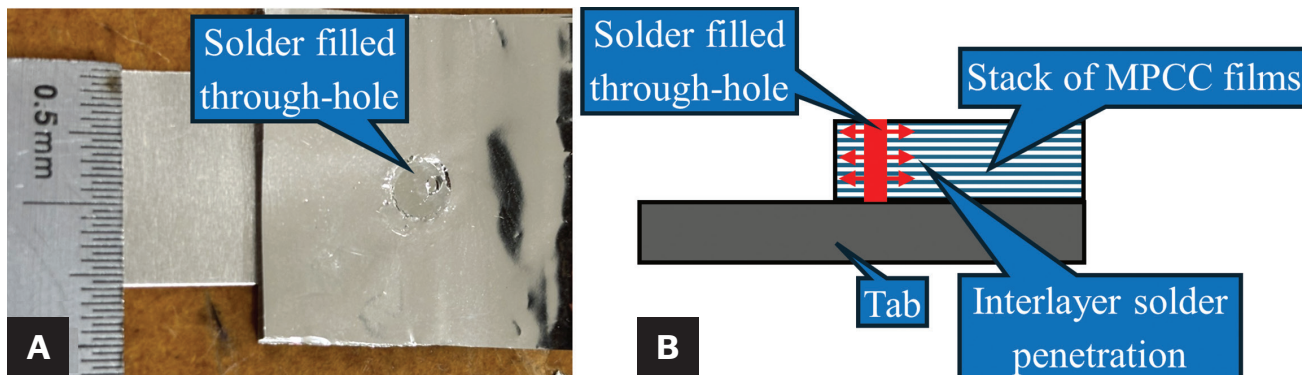


Fig. 1 – A – Through-hole UAS joint; B – cross-section side view of through-hole UAS joint.

In this work, through-hole UAS was experimentally studied to determine its ability to form MPCC/tab joints. An experimental evaluation was conducted to investigate the impact of primary process parameters (welder cylinder pressure, ultrasonic vibration time, and vibration amplitude) on the electrical and mechanical properties of MPCC/tab joints. Additionally, joints were produced with “near-optimum conditions” and used for a layer-by-layer analysis to measure the electrical resistance from MPCC to tab, for each MPCC in a stack. Optical microscopy and SEM techniques were also used to observe solder penetration between MPCC films, microstructural differences under low and high vibration amplitudes, and the interface between the solder and the tab.

Experimental Procedures

Materials

The MPCC used in this work consisted of a 1-micron-thick layer of aluminum deposited on both sides of a 6-micron-thick PET substrate. The MPCC material was obtained from Soteria Battery Innovation Group. The tab material used in this work was 0.1 mm-thick 1000-series aluminum. The solder used was a 96.5Sn-3.5Ag alloy (221°C eutectic temperature) purchased in rod form (1/8-in. diameter). Sn-Ag solder was chosen because its eutectic temperature is below the melting temperature of PET. Sn-Zn solder has an even lower eutectic temperature; however, Sn-Zn has worse corrosion resistance than Sn-Ag (Ref. 34). Prior to soldering, the solder rod was cut to a weight between 0.09 and 0.11 grams and hammered into a flat disc with a diameter of 3.5-4 mm.

Sample Geometry

A standard geometry was utilized for creating MPCC/tab joints. This work utilized stack sizes of 64 films (total film stack thickness of ~512 μm). The tabs had dimensions of 15 mm by 60 mm. The films had dimensions of 25 mm by 35 mm. To create the stacks of MPCC films, a five-meter length of MPCC film was folded on itself until it was 64 layers thick. After folding, the film was cut into 16 individual MPCC stacks. The through-hole was formed using a 4

mm diameter leather hole punch, and it was located 10 mm from one of the 25 mm edges in the film stack. As shown in Fig. 2, the tab was centered under the stack in the width direction, with one end aligned with one 25 mm edge and running the entire length of the stack and beyond. A small paper binder clip held the stack and tab together.

Through-Hole UAS

Through-hole UAS was performed using a Branson 2000ae ultrasonic plastic welder with a 40 kHz converter, 1:1.5 gain flat-bottomed titanium horn, and a hot plate set on the welder base. The horn surface was stadium-shaped, where the long axis was 12.7 mm, and the short axis was 11 mm. To achieve a wide range of ultrasonic amplitudes, three different boosters were used with gains of 1:1, 1:1.5, and 1:2.5. The hold time (0.1 s), trigger pressure (7.5) (unitless), and down speed control (2.5) (unitless) were all kept constant in this work. A low-down speed was utilized to prevent solder splashing.

To perform through-hole UAS, the stack of MPCC was placed on a hot plate (set point 240°C), with the horn centered over the through-hole. Generally, since PET has a melting point of 250–260°C, which is below the hot plate set point, the film was not expected to melt. Additionally, because the molten solder is much more compliant than the PET films, the strain amplitude will be much higher in the molten solder, which would limit viscoelastic heating in the PET. Next, the solder consumable was placed in the through-hole. A piece of Kapton film was placed between the horn and the sample to prevent solder from sticking to the horn. Then, the horn was brought down and back up, without the use of ultrasonics, to flatten the solder consumable and ensure centering. After the solder melted, which takes approximately 45 to 60 seconds (s), the horn was brought down, and ultrasonic vibration was applied. The joint was then removed from the hot plate and allowed to cool to near room temperature for approximately 5 minutes.

Resistance Measurements

Despite the importance of multi-layer CC to tab joints, there is a lack of reproducible standard test methods for measuring

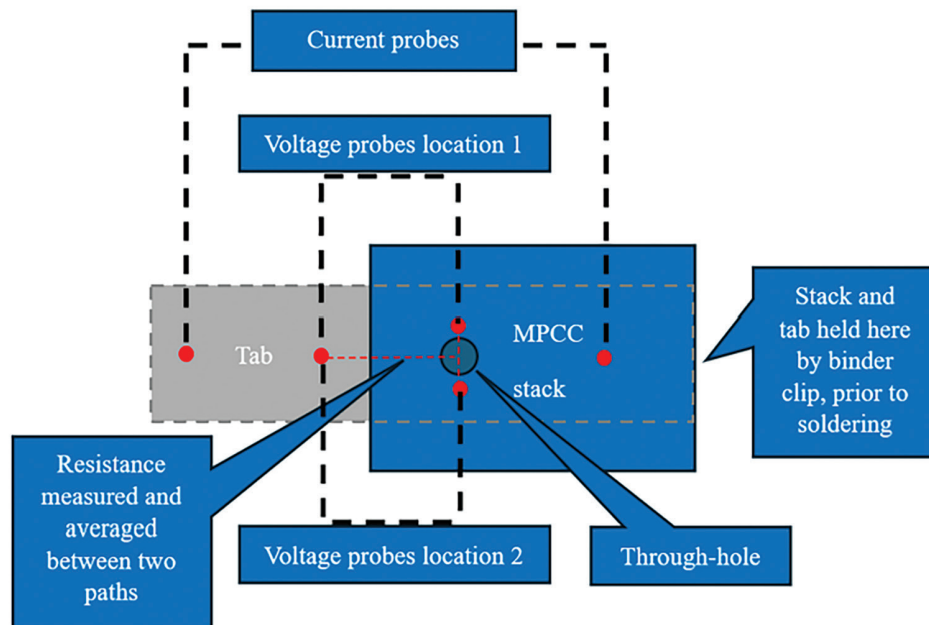


Fig. 2 — Four-point-probe schematic for electrical resistance analysis.

electrical resistance in this geometry (Ref. 35). Therefore, for this work, a four-point-probe approach, similar to that proposed in Grabmann et al. (Ref. 35), was used. Resistance measurements were performed using the four-point-probe function of a Hewlett-Packard 34401A multimeter. As shown in Fig. 2, the four-point-probe function applies a test current through the “outer” current probes and measures the voltage drop across the “inner” voltage probes. The multimeter automatically calculates and displays resistance using Ohm’s law. In our work, the current probes were alligator-style clips, with the front of the jaw being placed at the requisite locations. The voltage probes (Probe Master 8150) were gold-plated and spring-loaded with a 0.025-in.diameter rounded tip. The “reading hold” setting was used, which provided a resistance value when three consecutive readings within a 0.10% sensitivity band were found.

The four probes were arranged with approximately 15 mm between each probe, as shown in Fig. 2. For each film, two resistance readings were taken, one with the MPCC current probe in location 1 and one at location 2. Both locations are five mm away from the through-hole, on either side. The two resistance values were then averaged. Because of the difficulty of measuring resistances on the underside of each film, only the top metallization layer of each MPCC was analyzed. In the parameter evaluation, resistance was measured on the top, middle, and bottom MPCC films of a sample stack. From these three values, an average resistance for the sample was calculated. For the middle and bottom measurements, this was done by lifting the stack of films above the measurement layer and inserting the probe near the joint. In the layer-by-layer evaluation, resistance was measured for every MPCC film in a stack. This was performed by measuring the resistance of the top film, then completely peeling it off and discarding it to expose a fresh layer, and repeating the process.

Mechanical Testing

Mechanical testing was performed by measuring the peak force during pull testing of the full stack of films against the tab. The pull test was performed with a 50 lb force transducer (Omega LCF-50) fixed to the top grip of an Instron 4468 tensile tester. To perform the mechanical test, the portion of the tab under the stack of MPCCs was bent backward near the joint area, exposing the film stack. The films were then clamped in a fixture attached to the force transducer, and the tab was clamped in the bottom jaw of the tensile tester. The crosshead speed was set at 200 mm/min, and the peak pull force was measured using a Lebow Products Inc 7541 meter. Mechanical testing was performed on the samples from the parameter evaluation study after the resistance measurement was completed. The failure mode was also recorded; all failures fell into three categories: tab failure, film failure, and failure at the interface between tab and the through-hole solder, as shown in Fig. 3.

Amplitude of Vibration Measurement

Amplitude of vibration at the horn surface (peak-to-peak) was measured using a laser vibrometer (Polytec HSV 700) with a digital oscilloscope (Tektronix TDS 2024C). For each booster used (1:1, 1:1.5, 1:2.5), measurements were taken at 20%, 40%, 60%, 80%, and 100% vibrational amplitude. The measured values were compiled to develop a regression equation from which the amplitude of vibrations used during the parametric study was determined (see Table 1).

Table 1 — Parameter Ranges Utilized in Parameter Evaluation Study, Underlined/Bold Indicate Values for Parameters Held Constant while Third Parameter was Varied

Parameters						
Weld Cylinder Pressure (lb/in. ²)	15	<u>25</u>	40	55	70	85
Ultrasonic Vibration Time (s)	0.2	0.4	<u>0.6</u>	0.8	1.0	1.2
Peak-to-Peak Amplitude of Vibration (μm)	1.74	8.41	<u>14.71</u>	24.86	33.41	

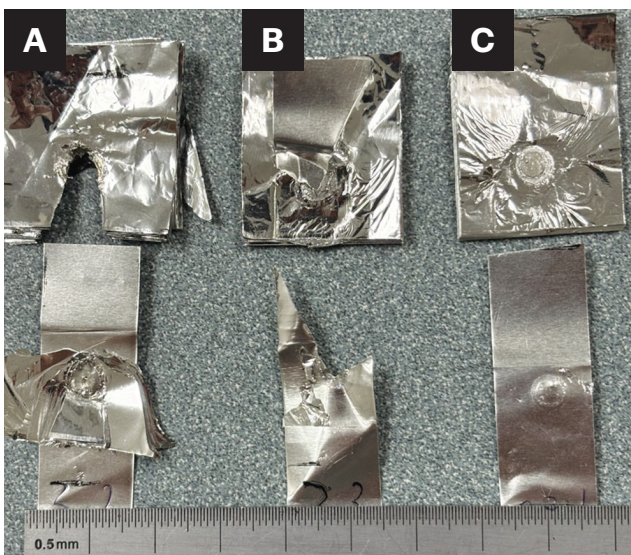


Fig. 3 — MPCC (top) to tab (bottom) joints showing three failure modes: A — Failure in films; B — failure in tab; C — failure at solder/tab interface. Film stacks for interface failure and tab failure are flipped to show the bottom of the stacks.

Parameter Evaluation Study

The one-factor-at-a-time (OFAT) method, also known as the single-factor design of experiments (DOE) method, was used to evaluate the effects of process parameters on resistance and strength. While OFAT is inefficient and does not accurately capture factor interactions, it is simpler, and it provides a better understanding of the effect of each parameter on the outputs than multi-factorial DOE approaches. The “near-optimum conditions” that were previously found by trial and error (Ref. 30) were used as a baseline for this OFAT parameter evaluation. In this work, welder cylinder pressure, ultrasonic vibration time, and amplitude of vibration were adjusted one parameter at a time, while the other two parameters were kept constant at the baseline values near-optimum

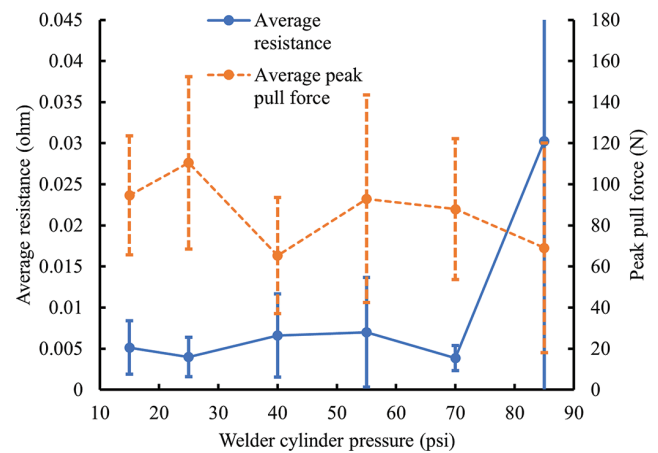


Fig. 4 — Combined average electrical resistance (ohm) and average peak pull force (N) with varying welder cylinder pressure (14.71 μm amplitude of vibration, 0.6 s ultrasonic vibration time).

parameters. Table 1 shows the range of parameters used and the near-optimum parameters held constant while varying the other parameters. For each parameter set tested, five joints were produced. As mentioned in the “Resistance Measurements” section of the Experimental Procedures, these joints were tested electrically to measure the resistance of the joint for the top, middle, and bottom MPCC films. From these three values, an average resistance for the sample was calculated. Next, the sample was mechanically tested, and the type of failure was recorded.

Layer-by-Layer Resistance Analysis

To provide a more detailed analysis of the electrical resistances of the joint between MPCC films in a stack and the tab, a layer-by-layer resistance analysis was conducted. Five joints were produced using the following near-optimum parameters: ultrasonic vibration time of 0.6 s, a cylinder pressure of 25 lb/in.², and a vibrational amplitude of 14.71 μm. In the

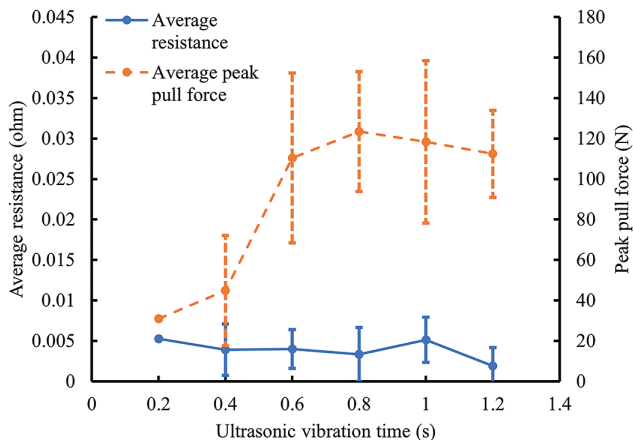


Fig. 5 – Combined average electrical resistance (ohm) and average peak pull force (N) with varying ultrasonic vibration time (s) (14.71 μm amplitude of vibration, 25 lb/in.² welder cylinder pressure).

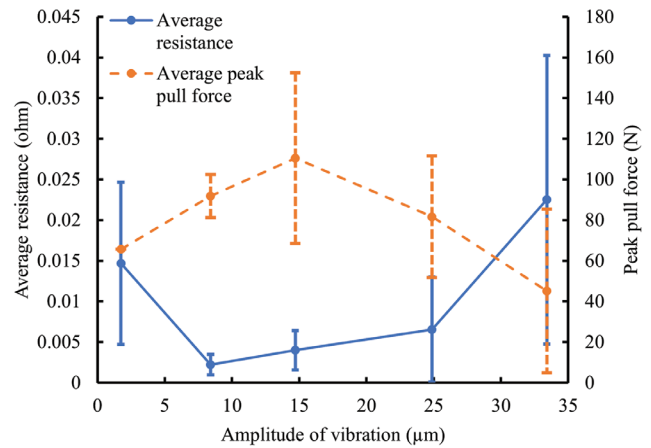


Fig. 6 – Combined average electrical resistance (ohm) and average peak pull force (N) with varying amplitude of vibration (0.6 s ultrasonic vibration time, 25 lb/in.² welder cylinder pressure).

Table 2 – Failure Modes of Mechanically Tested Samples at Varying Welder Cylinder Pressures

Welder Cylinder Pressure (lb/in. ²)	15	25	40	55	70	85
Failure Mode and Number of Respective Samples	3 Tab 2 Film	3 Tab 1 Film 1 Interface	4 Film 1 Interface	2 Tab 2 Film 1 Interface	Tab 2 Film 1 Interface	1 Tab 3 Film 1 Interface

Table 3 – Failure Modes of Mechanically Tested Samples at Varying Ultrasonic Vibration Time (s)

Ultrasonic Vibration Time (s)	0.2	0.4	0.6	0.8	1.0	1.2
Failure Mode and Number of Respective Samples	1 Film 1 Interface Note: 3 fell apart before testing.	1 Tab 1 Film 3 Interface	3 Tab 1 Film 1 Interface	1 Tab 4 Film	3 Tab 2 Film	5 Film

layer-by-layer evaluation, resistance was measured using the four-point probe method for every MPCC film in the stack, as mentioned previously.

Microscopy

Six through-hole UAS joints were analyzed with optical microscopy and SEM imaging. Three strong joints were produced with the near-optimum parameters of a cylinder pressure of 25 lb/in.², an ultrasonic vibration time of 0.6 s, and a vibration amplitude of 14.71 μm . Three weak joints were produced using the same cylinder pressure and ultrasonic vibration time as the strong joint, but with a much lower vibration amplitude of 1.74 μm . The weak joints exhibited poor bonding between the film stack and tab, and in each joint, the tab detached immediately following soldering.

Optical microscopy was performed using an Olympus GX51 microscope. Prior to optical microscopy, the samples were cross-sectioned (cut parallel to the short axis of the MPCC stack) near the center of the through-hole of the solder joint and mounted with acrylic. The standard metallographic procedure was used for grinding and polishing. A vibratory polisher (Buehler Vibromet 2) was used for the final polishing step. The sample was placed in the vibratory polisher on the lowest amplitude setting (10%) for 25 min using a 0.05 μm colloidal silica suspension. The colloidal silica also provided selective etching, showing the joint's microstructure. These mounted samples were also used for SEM imaging on a Thermo Scientific Apreo FEG SEM. Prior to imaging, the samples were carbon-coated (10 nm) for improved visibility. SEM was used in backscatter electron imaging mode. The parameters used are included in the SEM images. Energy-dispersive x-ray spectroscopy (EDS) was utilized to analyze the interface between

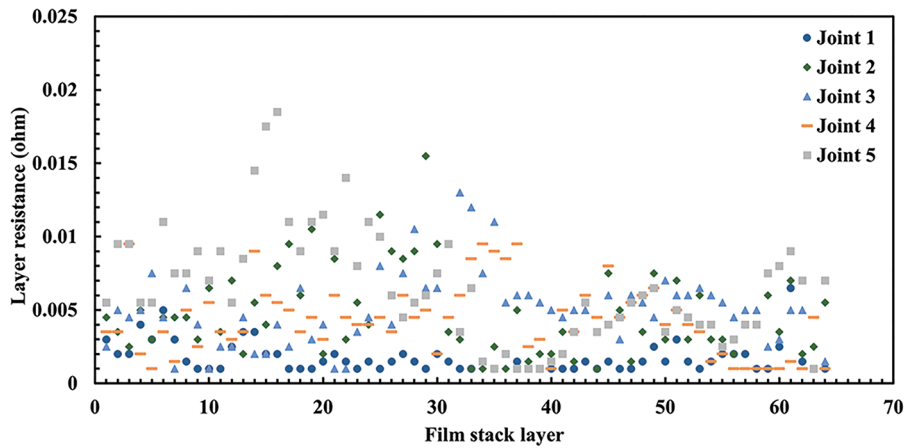


Fig. 7 — Layer-by-layer resistance of through-hole UAS joints (25 lb/in.² weld cylinder pressure, 0.6 s ultrasonic vibration time, 14.71 μm amplitude).

Table 4 — Failure Modes of Mechanically Tested Samples at Varying Amplitudes of Vibration (μm)

Amplitude of Vibration (μm)	1.74	8.41	14.71	24.86	33.41
Failure Mode and Number of Respective Samples	1 Film 1 Interface Note: 3 fell apart before testing.	5 Tab	3 Tab 1 Film 1 Interface	2 Tab 3 Film	4 Film 1 Interface

the solder and tab for a strong joint (made with 14.71 μm amplitude). EDS was performed on the Apreo SEM using an Octane Elect 30 mm² Energy Dispersive Spectroscopy silicon-drift detectors (SDDs).

Results

Parameter Evaluation

Figure 4 presents the average film resistances and average peak pull forces of samples fabricated with varying welder cylinder pressure. Error bars show $\pm$ one standard deviation from the plotted averages for Figs. 4–6 (outliers were included in the calculation of standard deviations and averages).

Table 2 shows the failure modes of the mechanically tested samples at varying welder cylinder pressures.

The average resistances and peak pull forces with varying ultrasonic vibration times are shown in Fig. 5. For an ultrasonic vibration time of 0.2 s, two joints failed immediately after the joining process, and another failed after the resistance measurement; therefore, no standard deviation was calculated for this parameter set.

Table 3 shows the failure modes of the mechanically tested samples at varying ultrasonic vibration times.

Figure 6 illustrates the average electrical resistance and peak pull force for samples prepared with varying vibration

amplitudes. At the lowest amplitude of vibration (1.74 μm), three out of five samples failed at the solder/tab interface during resistance testing, prior to mechanical testing; therefore, no standard deviation was calculated for the peak pull forces at an amplitude of 1.74 μm .

Table 4 presents the failure modes of mechanically tested samples with varying vibration amplitudes.

Layer-by-Layer Resistance Measurement with Optimal Parameters

The layer-by-layer resistances of five through-hole UAS joints produced with the near-optimum parameters described previously are shown in Fig. 7. Film stack layer 64 represents the topmost film on the stack.

Through-Hole UAS Microscopy

For microscopy, three strong joints made using near-optimum conditions, and three weak joints were evaluated. In both cases, the near optimum cylinder pressure of 25 lb/in.² and ultrasonic vibration time of 0.6 s were used, while the ultrasonic vibration amplitude for the strong joints was 14.71 μm and for the weak joint it was 1.74 μm . Reducing the ultrasonic vibration amplitude resulted in poor bonding between the tab and the film stack, as well as high average electrical resistances. Both sets of joints were analyzed to

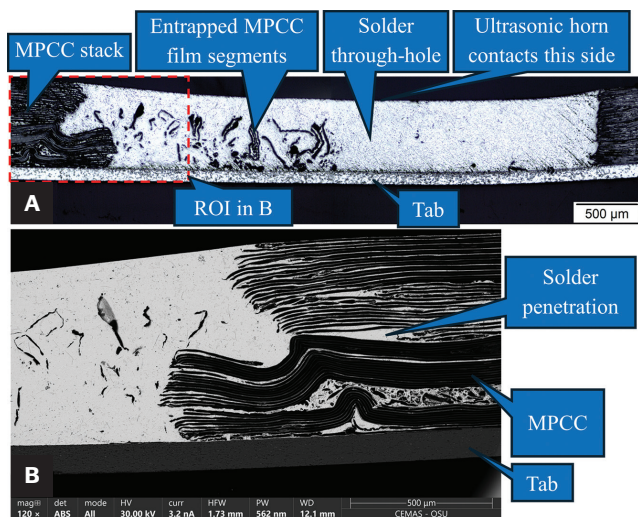


Fig. 8 — A — Stitched (two images) optical micrograph cross-section of strong through-hole UAS joint (50X mag); B — SEM micrograph showing region of interest shown in A (120X mag).

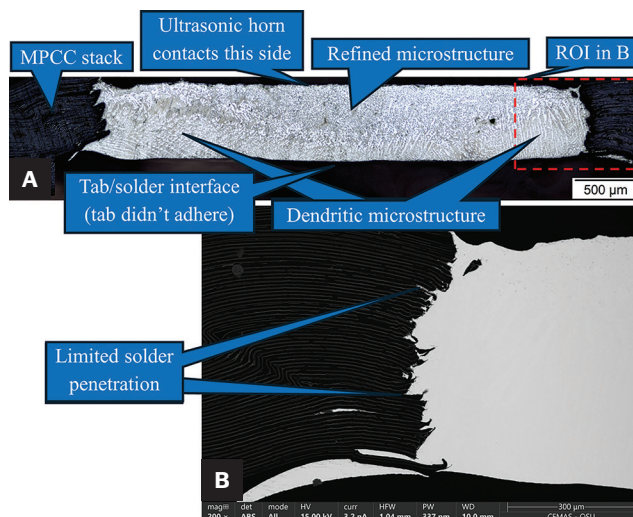


Fig. 9 — A — Stitched (two images) optical micrograph cross-section of weak through-hole UAS joint (50X mag); B — SEM micrograph showing region of interest shown in A (200X mag).

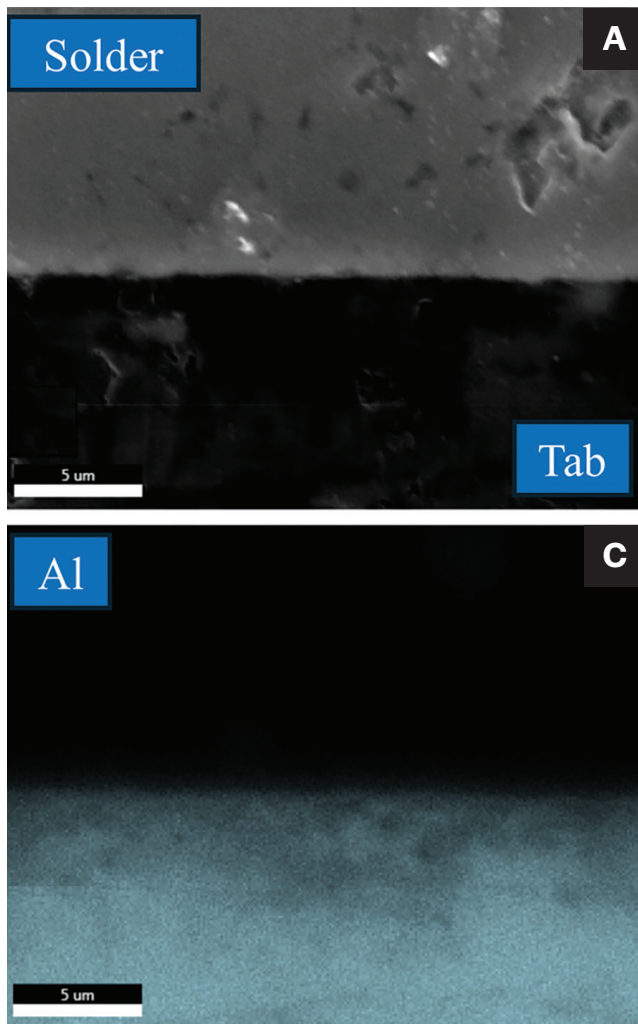


Fig. 10 — A — EDS ROI showing interface between solder and tab in the center of joint; from strong through-hole UAS joint (Fig. 8); B — Ag ROI showing possible intermetallic at interface; C — Al ROI; D — Sn ROI.



Fig. 11 — MPCC/tab joint produced with 70 lb/in.² welder cylinder pressure, 14.71 μ m amplitude of vibration, and 0.6 s ultrasonic vibration time, showing damaged films around the joint area.

Table 5 — Average Peak Pull Forces (N) of the Three Observed Failure Modes

Failure Mode:	Average Peak Pull Force (N):
Films	90.22
Tab	107.26
Solder/Tab Interface	32.77

determine if the solder penetration between MPCC films and if the solder microstructure varied between samples made with high and low amplitudes.

Figure 8 shows a cross-section of a strong joint at different magnifications. Figure 8A shows the entire cross-section of the joint that was created by stitching together two optical micrographs taken at 50X magnification. Figure 8B is a SEM micrograph showing the solder-MPCC interface. Figure 9 shows a cross-section of a weak joint with both a stitched optical micrograph and a SEM micrograph.

EDS was performed on the strong through-hole UAS to investigate the intermetallic at the interface between the solder and tab. A voltage of 30 kV and a current of 3.2 nA were utilized to acquire the EDS maps. The mapping was performed near the center of the solder/tab interface at approximately 7000X magnification. Figure 10 shows the mapped region and the ROIs for Ag, Al, and Sn.

Discussion

Failure Mode

Table 5 shows the average peak pull forces for the three observed failure modes for the entire study. When failure occurred at the solder/tab interface, the bond between the solder and tab failed in shear. This may have been caused by insufficient wetting of the tab by solder or insufficient clearing of the oxides on the tab surface. Failure in the films occurred as the films tore around the area surrounding the through-hole. While the through-hole acts as a stress concentrator for initiating this failure mode, a portion of the films remained attached to the solder-filled through-hole following failure. The failure mode with the highest average peak pull force was failure in the tab.

Welder Cylinder Pressure

The average electrical resistance was constant across nearly the entire range of welder cylinder pressures, except at 85 lb/in.². The average peak pull force showed a slight decrease as pressure was increased, although this trend was not com-

pletely consistent. At the lowest cylinder pressure (15 lb/in.²), all the samples failed in either the tab or the films. Although one sample failed at the interface at the second lowest cylinder pressure tested (25 lb/in.²), this pressure level had the highest average peak pull force. No visible deleterious effects of cylinder pressure occurred until reaching 70 lb/in.², where one sample showed significant damage from the joining process. Figure 10 is a photograph of a typical sample produced with 70 lb/in.² cylinder pressure exhibiting significant damage around the through-hole area. At the highest cylinder pressure, 85 lb/in.², three of five samples showed significant visible damage. Two film layers prepared with 85 lb/in.² had outlier high resistances. These results indicate that joints produced with lower cylinder pressures have superior mechanical performance with similar electrical resistances.

Ultrasonic Vibration Time

Similar to welder cylinder pressure, the average electrical resistance remained low across the entire range of ultrasonic vibration times. However, joints produced with 0.2 s of ultrasonic vibration time were not consistent, as two fell apart immediately after soldering, and an additional joint fell apart after electrical testing. A slight increase in strength was observed from 0.2 s to 0.4 s; however, three out of five samples failed at the solder/tab interface. A large increase in strength was found from 0.4–0.6 s. At 0.6 s of ultrasonic vibration time, only one sample failed at the interface. The average peak pull force reached a maximum at 0.8 s of ultrasonic vibration time and then slightly decreased with increasing ultrasonic vibration time. With a time of ultrasonic vibration greater than 0.6 s, no joints failed at the interface between the solder and the tab. At 1.2 s of ultrasonic vibration time, two out of five samples exhibited “oversoldering,” where the solder penetrated through the tab. These results indicate that to create strong joints, a minimum duration of ultrasonic vibration (≥ 0.6 s in this case) is necessary to achieve the clearing of oxides and wetting on the tab surface. However, with longer ultrasonic vibration times (≥ 1.2 s) solder penetration through the tab can occur.

Vibrational Amplitude

The amplitude of vibration showed the strongest effect on mechanical strength and electrical resistance. Average mechanical strength was highest at a vibrational amplitude of 14.71 μm , while the lowest average electrical resistance occurred at 8.41 μm . Both amplitudes produced joints that were mechanically strong and exhibited low resistance. With an amplitude of 8.41 μm , all five joints failed in the tab. One instance of interface failure was found at an amplitude of 14.71 μm , despite this condition having the highest average peak pull force. Joints fabricated with an amplitude of 1.74 μm were not consistent, and three joints fell apart at the solder/tab interface immediately after soldering. The lack of robustness at this low amplitude may be caused by insufficient clearing of the oxides and solder wetting on the tab. Higher amplitudes produce higher acoustic intensity, which increases acoustic cavitation, streaming, and the UCE. Damage to the films around

the through-hole (similar to Fig. 10) began to occur at a vibrational amplitude of 24.86 μm , with three out of five samples showing damage. At the highest amplitude, 33.41 μm , all five samples showed film damage surrounding the through-hole.

Layer-by-Layer Analysis

In the layer-by-layer resistance analysis, it was demonstrated that the top metallization layer in each film of the MPCC/tab stack was electrically connected to the tab. Figure 7 shows that resistances are stable during testing of the top half (film 32–64) of the stack; however, higher resistances are seen within samples for the bottom half of films (films 1–31). This is despite the fact that each successive MPCC tested would have a ~ 8 μm (thickness of films) shorter path length for the resistance measurement. The test method utilized here relied on tearing away successive layers of MPCCs, and the act of tearing away each layer likely damaged the solder/MPCC bonding below, resulting in higher resistances as the test progressed.

When comparing these through-hole UAS layer-by-layer results with those of previous work (Ref. 31), the results presented here had lower average layer resistances. In the previous work, the average of all the layer-by-layer film resistances for ten 64-film aluminum MPCC/tab joints was 0.04 ohms, compared to 0.0045 ohms in this work. The reduction in layer-by-layer resistance in this work resulted from improvements in the through-hole UAS process, including letting the solder become fully molten before applying ultrasonics, improved through-hole punching, and better fixturing.

Microscopy

Optical microscopy and SEM imaging were used to observe solder penetration between MPCC films under two different ultrasonic vibration amplitudes of 14.71 μm for the strong joints and 1.74 μm for the weak joints. For each amplitude, three joints were fabricated, cross-sectioned, mounted and polished, and observed under the microscope. Although micrographs for only one strong and one weak joint are included, the microscopic observations were consistent for each set of three joints. While cross-sections are an imperfect method for observing morphological differences, there were clear differences in the depth of solder penetration between MPCC films for the two vibration amplitudes.

For the strong joints, deep and clearly defined penetration of the solder between MPCC films was observed (see Fig. 8B). The penetration of the solder is a result of various factors, including the downward force of the horn, acoustic streaming, cavitation, and the UCE. Clearly defined penetration is not seen everywhere, for example, on the left-hand side of Fig. 8A. However, each of the three samples made under near-optimum conditions had one or both film/solder interfaces of the cross-section showing deep penetration. The lack of deep solder penetration in some areas of the solder/MPCC interface is attributed to the nature of cross-sections, where grinding down the sample could reveal a totally different morphology. Within Fig. 8, entrapped broken MPCC film fragments are

also observed, which could be the result of the through-hole punching process, or the UAS process itself (cavitation).

None of the weak joints displayed deep or clearly defined solder penetration between MPCC films (see Fig. 9B). The low amplitude of vibration used to create these samples resulted in a reduced effect of acoustic streaming, cavitation, and the UCE. Additionally, far fewer entrapped MPCC films were observed in these samples, which may indicate that amplitude can affect the number/volume of broken and entrapped film fragments. No tab is seen in the Fig. 9 micrographs, as the tab fell off following joining. The inadequate tab/solder joint is a result of poor wetting of the tab by the solder, due to the lack of cavitation.

The selective etching, due to the colloidal silica polish, revealed differing microstructures between the strong joints (Fig. 8) and weak joints (Fig. 9). The joint made with high amplitude (Fig. 8) shows a refined microstructure with no visible dendrites. The joint made with low amplitude (Fig. 9) shows two different microstructures. At the top of the joint, closest to where the ultrasonic horn contacts, a refined microstructure similar to the joint made with a high amplitude is observed. This microstructure forms a semioval shape. Towards the bottom of the solder joint, dendrites are clearly visible. Very fast cooling rates for Sn-3.5Ag alloys can result in developing a combination of Sn dendrites and a eutectic structure (Ref. 36). The dendritic microstructure seen in Fig. 9 is attributed to the low ultrasonic vibration amplitude used to make these joints, compared to the strong joints made with much higher vibration amplitude (Fig. 8). Ultrasonic treatment is known to enhance heterogeneous nucleation through cavitation and acoustic streaming, which breaks down dendrites and distributes the resulting fragments (Ref. 37). High pressures generated by cavitation can also increase undercooling, further increasing the heterogeneous nucleation and creating fine grains (Ref. 37). Therefore, the high vibration amplitude for the strong joints resulted in powerful acoustic streaming and/or cavitation that broke down dendrites to form a refined microstructure. However, the low amplitude of vibration used for the weak joints resulted in weak streaming and cavitation, thus yielding two distinct zones with differing microstructures.

EDS of the interface between the solder/tab of a strong joint (made with 14.71 μm amplitude) showed areas of high Ag content dispersed across the interface. These areas show Ag took the form of “islands” spread along the interface, rather than a cohesive layer. The areas where these islands of silver formed did not show elevated levels of Sn. Sn-Ag solder contains a dispersion of fine Ag_3Sn compounds (Ref. 38). When Sn-Ag alloy is soldered to Al, Ag_2Al and Ag_3Al intermetallics can form at the interface (Ref. 34). The lack of Sn at these Ag islands indicates that the islands may be Ag_2Al or Ag_3Al , rather than Ag_3Sn . Due to the ultrafast soldering time, the intermetallic may not have had time to grow into a cohesive layer. However, cavitation is known to accelerate the growth of intermetallics during soldering (Ref. 39). Characterizing the interface between solder and tab will be the subject of future work.

Conclusions

In this paper, through-hole UAS is shown to consistently form joints between a 64-film-thick stack of aluminum MPCCs and an aluminum tab. The effects of welder cylinder pressure, ultrasonic vibration time, and vibration amplitude on electrical and mechanical performance were analyzed. The specific conclusions are as follows:

1. Welder cylinder pressure affects the joint outcomes the least when compared to the amplitude of vibration and ultrasonic vibration time. At high cylinder pressure ($\geq 70 \text{ lb/in.}^2$), film damage begins to occur around the through-hole. At the highest cylinder pressure tested (85 lb/in.^2), three out of five samples exhibited severe damage around the through-hole, resulting in several samples having high resistances. Lower cylinder pressures (15, 25 lb/in.^2) produced the strongest joints without compromising electrical resistance.

2. With short ultrasonic vibration times (0.2 s), joining is inconsistent, resulting in some joints that fall apart at the solder/tab interface immediately after soldering. Mechanical strength increases with ultrasonic vibration times up to 0.6 s, after which it plateaus. At high ultrasonic vibration times (1.2 s), over-soldering can occur where the solder penetrates through the tab. Electrical resistance remained low throughout the range of times tested.

3. Amplitude of vibration showed clear trends for both electrical resistance and peak pull strength. At low amplitudes (1.74 μm), joining is inconsistent, and electrical resistances are elevated. Mechanical strength showed a positive relationship with amplitude up to 14.71 μm . Further increases in amplitude resulted in weakened joints that displayed damaged films around the through-hole. Additionally, joints produced with an amplitude of 33.41 μm had high outlier resistances. The lowest average resistance was observed at an amplitude of 8.41 μm , with average resistance increasing as the amplitude varied above or below this value.

4. Layer-by-layer resistance testing of joints prepared with near-optimum conditions showed that each (top) metallization layer is electrically connected to the rest of the stack. The average single film resistance for all five joints tested was 0.0045 ohms.

5. Optical microscopy and SEM imaging showed that joints made with near-optimum conditions had much deeper and clearly defined solder penetration between MPCC films, compared to joints made with low amplitudes. Joints made with the near-optimum parameter set showed a refined microstructure. Joints made with the low amplitude parameter set displayed two distinct microstructural regions: a refined microstructure near where the horn contacts the joint, and a dendritic microstructure near the anvil side of the joint.

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