Study on the bonding mechanism of multilayer copper with nickel sheet in ultrasonic welding process

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Abstract

The metal-plastic flow and bonding strength of three-layer copper (Cu) with single-layer nickel (Ni) joints via ultrasonic welding were investigated in this study. With the increase of welding time from 0.3 to 0.5 s, the weld bonding density between different layers’ interface increased gradually and approached about 95 ± 1.5%. Electron backscatter diffraction (EBSD) analysis of different regions of the welded joint revealed that the metal grains in the 1st layer became relatively small and elongated, while the 2nd and 3rd layers tended to be equiaxed grains. The joint bonding strength of T-peel tests changed with welding time and had two kinds of fracture models (nugget pullout and interfacial fracture). Compared with the bonding strength of the 1st-2nd and 3rd-4th interface, the 2nd-3rd interface of joints had the highest bonding strength of 400.6 N with nugget pullout fracture model at welding time of 0.4 s. In addition, the much higher hardness of the 1st layer is attributed to the strong plastic deformation of the 1st layer, while the dynamic recovery and dynamic recrystallization of the metal of the 2nd layer resulted in the decrease of hardness.

1. Introduction

In recent years, electric vehicles, hybrid electric vehicles, or plug-in hybrid vehicles have received widespread attention because of their great potential for reducing greenhouse gas emissions [1, 2]. The battery pack used in these vehicles is generally composed of a large number of circuit-connected and structurally fixed battery cells [3, 4]. However, because of the need to connect multilayer stacks with highly reflective and conductive materials, the production of these connections is not straightforward [5]. Therefore, a suitable connection technology is required to meet manufacturing battery packs. Generally, batteries in electric vehicles are exposed to harsh driving environments with vibration, different load conditions, and harsh temperatures [6], so the strength of the joint is a critical criterion for successful connection [7]. The joint strength should be sufficient to withstand all shock loads and vibration loads [8, 9]. In addition, the conductivity of the weldment should be high enough while ensuring that the welding area should not be too large, ensuring low resistance and avoiding the reduction of current density so that the energy loss is as small as possible [5, 8].

In order to meet the above requirements, a variety of connection technologies such as welding for battery pack manufacturing are being studied [9, 10]. However, the traditional fusion welding used for battery-grade sheets causes a series of problems, such as the formation of brittle intermetallic compounds and the deformation of the weldment [11, 12]. In addition, Luo et al [13] also rely on the concentration of heat at the interface to produce welds, making it very sensitive to the gap between the sheets, and the ability is limited to non-highly reflective and non-thermally conductive materials. For the fusion welding methods, the welding joint firmness was low for the formation of intermetallic compound and welding defects. As a method to solve these problems, solid-phase welding has the advantage of eliminating metallurgical defects such as the formation of intermetallic compounds (IMC), brittle phase and porosity [14–16] in the liquid phase reaction of the fusion zone, and it has
received wide attention for its unique advantages. The study found that ultrasonic welding (USW) is one of the promising technologies for these multilayer stacks for automotive battery manufacturing [11, 12]. USW avoids melting materials, diffuses, and bonds softened metals through interfacial friction to achieve material connection [4, 17]. While ensuring the necessary strength, there is little or no brittle intermetallic compound along the weld, ensuring a small resistance [18, 19]. This process is suitable for connecting soft metals with high conductivity and reflectivity, such as Al, Cu, Ni, Ag, and Au [20]. Therefore, USW has become a technology suitable for thin sheet welding in various electric vehicle batteries, electrical and electronic industries.

However, the lack of understanding of the welding behavior of each layer in the multilayer stack structure hinders the large-scale production in the manufacture of electric vehicle batteries. Although the current research on USW is very active, most focus on the mechanics and mechanism between bilayer similar or dissimilar metals [21–25]. Such as the research of Bakavos and Prangnell [21] reported interface to curl, vortex, ripple, and micro-bonding are the mechanisms behind the weld. The effect of the interlayer on the bonding strength and the evolution of the IMC were also investigated [24, 25]. However, the joining application of multilayer dissimilar metals is more common in actual production, and few studies have evaluated the feasibility of USW in multilayer stacking. The well-known research by Kang [26] and Lee et al [27] on the fabrication and simulation of multilayer lithium-ion battery tabs shows that it is necessary to understand the behavior of each layer in a multilayer stack configuration during the process. Zhao et al [28] and Lee et al [29] have tried to measure temperature rise through pluggable thin-film thermocouples to understand the welding mechanism better and develop real-time monitoring programs for the process. In addition, Zhao et al [30] used numerical simulation methods to predict fatigue life, while Lee et al [27] tried to predict welding energy, deformation, and workpiece temperature distribution.

Since the research on multilayer USW is still in its early stages, the application of USW in the manufacture of multilayer lithium-ion batteries is still limited. The formation of bonds at the interface and the vibration transmission through different interfaces are unclear. This work investigates the microstructure, mechanical properties, and fracture behavior of ultrasonic spot-welded multilayer Cu/Ni joints ubiquitous in lithium-ion battery assembly. It systematically analyzes interfacial bonding mechanism and change mechanism of metal grains in different layers. Manufacturers can be guided to achieve good quality in battery pack manufacturing, electronics, and automotive parts based on scientific understanding obtained from this work.

## 2. Materials and methods

In this study, Cu and Ni sheets (purity ≥ 99.9%) with dimensions of 100 × 25 × 0.2 mm were selected as the weld materials. Their properties obtained by tensile test and hardness are given in Table 1.

| Material | Tensile strength (MPa) | Elongation (%) | Hardness (HV) |
|----------|-----------------------|----------------|---------------|
| Cu       | 247 ± 5               | 9 ± 0.3        | 108 ± 2.0     |
| Ni       | 541 ± 8               | 3 ± 0.2        | 157.9 ± 3.1   |

The welding sample is composed of three layers of Cu sheets placed on the upper part and one layer of Ni sheet placed on the bottom. And each layer of sheets is denoted as 1st, 2nd, 3rd, and 4th layer from top to bottom.

The USW process was performed using a BWX-2017A machine with a sonotrode tip size of 7 × 9 mm, which operates at 20 kHz frequency. Its schematic diagram of the USW system is shown in figure 1(a). Figure 1(b) shows the shape of the sonotrode tip and anvil. The sonotrode tip consists of 3 × 4 gridding knurls with 2 mm spacing and 0.5 mm depth. The long edge of the sonotrode tip is perpendicular to the vibration direction. The welding time is critical and changes every 0.1 s step from 0.2 s to 0.5 s. During the ultrasonic welding process, the constant input parameters value is the amplitude of 60% and clamping pressure of 3.0 bar. Before the USW process was carried out, the sheets were ultrasonically cleaned with acetone and dried to remove surface contaminants.

The mechanical property evaluation was evaluated via T-peel test performed by a universal testing machine (WDW-20) at a 10 mm min⁻¹ displacement rate under normal room temperature, and their configurations for joints are demonstrated in figures 2(a)–(c). Metallographic samples of the joint cross-section were obtained by wire cutting. Then, silicon carbide sandpaper and diamond suspension were used to grind and polish the metallographic samples to mirror-like surface. Optical microscope (OM, OLIMBUS-GX71) and scanning electron microscope (SEM, S-4800) were used to observe the weld structure and to measure the weld bonding.
density. Microstructural analysis was performed by field emission scanning electron microscope (FE-SEM, JSM-7800F) equipped with electron backscattered diffraction (EBSD) systems. X-ray diffraction (XRD, D8 ADVANCED) technology was used to scan the welded joint interface to analyze new phases, wherein scanning angle was $20^\circ$–$80^\circ$, the step length was 0.05°, and the speed was 0.5 s/0.05°. The hardness of samples was measured by Huayin 2000A hardness tester. The fracture morphology was observed and analyzed, and its phase
composition was identified by scanning electron microscope, energy dispersive spectrometer (EDS, X-MAX20), and XRD.

3. Results and discussion

3.1. Microstructure and weld bonding density
As shown in figures 3(a)–(b), when the welding time was 0.2 and 0.3 s, a more obvious gap could be seen at the interface (figure 3(e)), and only a tiny gap can be observed when the welding time was increased to 0.4 s (figure 3(c)). However, as time increased, the gap change was not apparent. To scientifically analyze the influence of welding time on the interface gap between samples, the concept of weld bonding density is introduced for quantitative analysis, which is the ratio of the interface bonding length between the weld joints to the total length of the weld joint.

Figure 4 shows the weld bonding density of different interfaces of each joint under different welding times. When the time was 0.2 s, the weld bonding density: the 1st-2nd interface (100%) > the 2nd-3rd interface (64.14%) > the 3rd-4th interface (46.67%). When the time increased to 0.3 s, the welding bond density: the 2nd-3rd interface (92.92%) > the 1st-2nd interface (79.76%) > the 3rd-4th interface (77.94%). It means that the
interface with the highest weld bonding density changed from the 1st-2nd interface to the 2nd-3rd interface as
the welding time increased from 0.2 to 0.3 s. Combined with the principle of ultrasonic welding equipment
analysis, the sonotrode directly contacts the 1st layer of metal and squeezes and rubs it during the welding
process. It is considered that the 1st layer of metal produces relatively large relative sliding with the 2nd layer of
metal under the action of the sonotrode, and the sliding removes the contaminant and oxidized layer on sheet
surface and generates heat to soften the metal. It would form a virtual bond in short welding time between the
1st-2nd interface. With the increase of welding time, the virtual bond would tear firstly and join again under the
action of the sonotrode pressure and shear force, and then produces material bonding, interlocking to form a
robust bond region, which leads to the increase of the joint bonding strength. The relative sliding between the 1st
and 2nd layers reduced, and the relative sliding between the 2nd and 3rd layers increased due to the partial bond
between 1st and 2nd in a short time. Therefore, the weld bonding density rises steadily and continuously under
the continuous action of the sonotrode, and the bonding strength also continues to increase. Since the force and
relative sliding required to form a bond between the 3rd and 4th layers need to be transferred through the 1st and
2nd layers, there will be a certain hysteresis and attenuation. And comparing with the three layers of Cu, the 4th
layer of Ni has higher hardness and less susceptible to deformation so that the weld bonding density between the
3rd and 4th layers is lower. In a word, with the increase of welding time from 0.3 to 0.5 s, the weld bonding
density between different layers’ interface increased gradually and approached about 95 ± 1.5%.

3.2. Plastic deformation and EBSD analysis
The joint formation is an essential basis for studying the effect of welding time on the welding process and
understanding the bond mechanism of welding materials. Figures 5(a)–(d) indicates images at the valley regions
of the knurl pattern of sonotrode tip for the joints performed at varied welding time from 0.2 to 0.5 s, exhibiting
the filling behavior of the top sheet extruded into the spaces between the knurl teeth during USW. With
increased welding time, the Cu was squeezed into the valley regions of the knurl pattern of sonotrode tip, therefore the depth of the sonotrode tip penetrating into the Cu sheet increased, as indicated in figures 5(a)–(d).
It can be concluded that the sonotrode exerts pressure on the Cu layer by friction. The high-frequency vibration
generated by the sonotrode causes relative friction between the sonotrode and the top sheet and between sheets,
the metal in bonding regions was softened due to friction heat and was squeezed to the surrounding movement.
However, the time was too short to produce sufficient energy, and the plastic flow of the material was not enough
to completely fill the space. Therefore step-shaped protrusions with a lower height were formed (figure 5(b)), and
there was generally a gap between the layers under the protrusions. When the time increases to 0.4 s, the
metal below the sonotrode rolling flower tooth bulge continues to flowed plastically under the vibration friction
of the sonotrode, the thickness of the metal layers directly below the sonotrode tip decreased. The thickness of
the bumps formed by the flow of metal increased and gradually formed a peak-like morphology (figure 5(c)). As
time continues to increased, it can be observed in figure 5(d) that the shape of the surface metal Cu layer change
was not apparent because as the metal gradually filled the space between the sonotrode tips, material plastic flow
and deformation became more difficult even if the welding energy input continues to increased.
Thus the plastic flow of material under ultrasonic vibration can be summarized into the following three processes: 1. The sonotrode exerts a shear force on the material’s surface by friction; 2. The repeated vibration and friction generated by the sonotrode cause plastic deformation of the surface of the material near the sonotrode knurled pattern and flow outwards to the sonotrode knurling pattern depression; 3. With the expansion of plastic deformation area, the acoustic roll pattern bugles more sink into the material, and the material gradually fills the roll pattern depression [11]. The rolling pattern sinks more profoundly in the initial stage, then slows down. The explanation for this phenomenon is that the initial shape change is large because there is enough space to accommodate the deformed material flow. However, with the continuation of the USW process, there is no more space available to accommodate the flowing material, which makes it more difficult for the material to flow. At the same time, material flow occurs due to a large amount of plastic deformation. Plastic deformation improves the clamping of the specimen and thus increases the friction work along with the weld interface. However, excessive material flow can lead to thinning and softening, which negatively affects the strength of the joint [16].

To further understand the plastic deformation of different layers, four specific regions of the joint performed at 0.4 s were analyzed via EBSD (figure 6(a)). Figures 6(e), (h), (k), and (n) show the grain boundary (GB) maps, in which the crystal grains in the range of $<15^\circ$ are marked as small-angle grain boundaries, and the crystal grains in the range of $>15^\circ$ are marked as high-angle grain boundaries. Lots of low angle grain boundaries (LAGBs) that consist of high-density dislocations and substructure are shown in figure 6(e), which indicates that strong plastic deformation has occurred in the metal of region C, and the direction in which the grains were elongated was consistent with the plastic flow direction of the material. Thus, the concentrated (111) orientation of grains in region C (figure 6(c)) is attributed to sheer plastic deformation under the action of sonotrode. Besides, region F also has many LAGBs. However, the number of LAGBs in other regions has been dramatically reduced, especially few LAGBS can be observed in region I (figure 6(i)), mainly composed of high angle grain boundaries (HAGBs), a few LAGBS are mainly concentrated at the Cu–Ni interface in region L, which is associated with the dynamic recovery and dynamic recrystallization during ultrasonic welding. The dislocations and substructure migrated and disappeared, and some combined to form HAGBs under the sharp temperature rise in USW. The equiaxed grains with random orientation observed in figure 6(i) confirm the recrystallization in region k, a variation of grain size resulted in inhomogeneity in mechanical properties [31, 32]. Figures 6(d), (g), (j), and (m) display the difference of deformation degree of four regions: region C > region F > region L > region I, which also reflects the process of plastic flow and bulge formation. The crystal grains of the 2nd layer below the valley are more similar to equiaxed crystals, indicating a high degree of dynamic deformation.

Figure 5. Images at the valley regions of the knurl pattern of sonotrode tip for the joints performed at varied welding time: (a) 0.2 s, (b) 0.3 s, (c) 0.4 s, and (d) 0.5 s.
recrystallization because plastic deformation starts from the center of each knurled pattern increased with the welding process.

As shown in figure 7, the EDS line sweep was carried out in the vertical direction of the contact interfaces (i.e. 3rd-4th) was performed at 0.2 s, 0.3 s, 0.4 s, and 0.5 s to analyze the change of diffusion depth between layers. It was found that the diffusion depth of joint was 0.5 s > 0.4 s > 0.3 s > 0.2 s. It indicates that the mutual diffusion depth of each layer of metal atoms increased as the input welding energy increased, therefore a better metallurgical bonding was formed. Moreover, Since the formation of IMC will affect the bonding strength of the

Figure 6. EBSD results of a joint performed at 0.4 s welding time: morphology of typical peaks and valley (a), color-coded grain orientation legend of Cu (b), IPF map of C (c), F (f), I (i) and L (l), KAM map of C (d), F (g), I (j) and L (m), GB map of C (e), F (h), I (k) and L (n).
interface [31, 33], XRD analysis was performed of the Cu/Ni interface, which showed no IMC between the Cu and Ni during the ultrasonic welding process, and the microscopic bonding is only the diffusion of atoms according to the analysis of figure 8.

3.3. T-peel strengths and fracture analysis
Figures 9(a)–(c) shows a summary of the mechanical performance of the joint. The T-peel strengths of the 2nd-3rd interface are bigger than that of the other two weld interfaces (figure 9(a)). It can be observed from figure 9(b) that the T-peel strength between the 1st-2nd interface gradually decreased with the increased welding time, which is mainly attributed to the thinning of the 1st layer metal (as indicated in figure 3(d)) [34]. Figure 9(c) shows that as the welding time increased, the T-peel strengths increased to the maximum value (307.5 N) at 0.40 welding time and then decreased. The fracture images of the T-peel test were observed further to analyze the bonding characteristics of different weld interfaces. The main failure types of the 1st-2nd interface and the 2nd-3rd interface at different times are the through-thickness tearing (pullout fracture) that occurred outside the weld area along its perimeter (figure 10), indicating that the actual nugget was developed at the 1st-2nd and 2nd-3rd interfaces.

However, the failure types of the 3rd-4th interface at different times show a significant difference. To further analyze the failure mechanism of the 3rd-4th interface, SEM was performed to conduct systematic research on the 3rd-4th interface. The result is shown in figures 11–14. Figure 11 shows the fracture morphology of the joint
Cu side performed at 0.2 s welding time. There are many scratches on the Cu surface of the joint edge region and metal adhesion phenomena, but some base metal regions are not welded. When the time is 0.3 s (figure 13), although the Cu surface is still relatively flat, the welding region is significantly larger, and it is difficult to observe the existence of base metal. However, the interface bond strength is still low, and the fracture mode is still interface fracture. The vertically fractured patterns were found inside the welding spot in these photographs, indicating a ductile fracture. There is a large amount of plastic deformation in the welding region on the copper surface, a large amount of metal adhesion and the existence of dimples can be observed when the time increases to 0.4 s, which is a typical ductile fracture surface [35]. However, when the time increased to 0.5 s, the morphology of the dimples is not apparent, and the existence of cracks is observed. This is one of the reasons for the decrease of the bonding strength, which is consistent with the results of the T-peel test.

Figure 9. (a) T-peel strengths of joint performed at 0.4 s welding time, (b) Effect of welding time on 1st-2nd joint interface T-peel strengths, (c) Effect of welding time on 3rd-4th joint interface T-peel strengths.

Figure 10. Typical failure type at (a) 1st-2nd and (b) 2nd-3rd weld interface of a joint performed at 0.4 s welding time.
3.4. Micro-hardness analysis

To understand the hardness distribution of each layer in multilayer ultrasonic welding, the hardness of each layer in the typical characteristic area of the welded joints with a welding time of 0.4 s was measured (figures 15(a)–(d)). The results are shown in figure 15(e) that the hardness of the joint on the Cu side is lower.
than that of the Cu base material. The hardness gradually increases in the direction away from the solder joint, approaching the hardness of the Cu base material. The hardness distribution of the Ni side shows the complete opposite law (figure 15(g)). The hardness of the Ni side is higher than that of the Ni-base material as a whole, and gradually decreases with the direction away from the joint to basically the same hardness as the base material.

Figure 13. Typical SEM images of fracture morphology of a joint performed at 0.4 s welding time: (a) overall view of Cu side, (b) magnified image of box B, (c) magnified image of box C, and (d) magnified image of box D.

Figure 14. Typical SEM images of fracture morphology of joint welded at 0.5 s welding time: (a) overall view of Cu side, (b) magnified image of box B, (c) magnified image of box C, and (d) magnified image of box D.
The hardness distribution in Cu and Ni exhibits such a different law because the recrystallization temperature of Ni is much lower than the Cu recrystallization temperature. When the temperature rise caused by friction at the entire solder joint is basically the same, the work hardening phenomenon of Ni is much greater than the work softening caused by recrystallization [36], so it is manifested as an increase in hardness. In contrast, the work hardening of Cu surface is less than work softening, so it is manifested as a decrease in hardness.

In addition, it can be seen from figure 15(f) that the hardness of the peak of the first layer of copper is much greater than that of the valley, while the hardness of the other two layers is not significantly different. In the vertical direction, the hardness of the first layer is greater than that of the other two layers, whether at the peak or valley, and the hardness of the third layer of copper is similar to that of the second layer. During ultrasonic welding, the metal undergoes softening due to friction heat and work hardening caused by cyclic load and back stress. Besides, the softening effect is associated with dynamic recrystallization during USW [37–39].
dislocations and substructure migrate and disappear, and some combine to form HAGBs under the sharp temperature rise in USW, which causes a decrease in hardness. The equiaxed grains with random orientation observed in figure 6(i) confirm the recrystallization in region k. The crystal grains of the 2nd layer below the valley are more similar to equiaxed crystals, indicating a high degree of dynamic recrystallization and a more substantial softening effect because plastic deformation starts from the center of each knurled pattern increases with the welding process. Progress gradually expands outward, so the valley does not have that high work hardening and appears as hardness: C > F > L ≈ K. The relationship between the amount of plastic deformation analyzed confirms this idea according to the kernel average misorientation (KAM) diagram (figures 6(d), (g), (i) and (m)).

4. Conclusions

This paper studied the ultrasonic welding of multilayer Cu–Ni joints. The influences of welding time on bonding strength, weld structure formation and microhardness distribution were studied. The bonding mechanism of multilayer Cu–Ni ultrasonic welding was analyzed by combining the welding bond density. Based on the results of this research, the following conclusions are drawn:

1) With the increase of welding time from 0.3 to 0.5 s, the weld bonding density between different layers’ interface increased gradually and approached about 95 ± 1.5%.

2) It was found that the strong plastic deformation of the 1st layer metal occurred as well as the grain became relatively small and elongated. In contrast, the grains of the 2nd and 3rd layers tended to be equiaxed grains, with minor plastic deformation and more dynamic recovery, which caused a lower fraction of LAGBs.

3) The T-peel experiment shows that the 2nd-3rd interface had the highest bonding strength. The 1st-2nd and 2nd-3rd interface was the nugget pullout failure. The failure mode of the 3rd-4th interface changed from interface failure to nugget pullout failure with the increase of time, and microcracks were observed in the copper metal at 0.5 s, which was one of the reasons for the decrease in bonding strength.

4) The hardness of the 1st Cu layer was much higher than the other two layers, while the 3rd layer hardness was slightly higher than that of the 2nd layer, which was caused by the strong plastic deformation on the surface of the 1st layer and the dynamic recovery and dynamic recrystallization of the 2nd layer of Cu metal.

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Data availability statement

The data generated and/or analysed during the current study are not publicly available for legal/ethical reasons but are available from the corresponding author on reasonable request.

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