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Improved thermal joining of aluminum and aluminum-polymer-composites for battery applications through laser surface structuring

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Abstract

Lithium-ion batteries are the major electrochemical energy storage solution for electromobility applications because of their advantageous characteristics (e.g. specific energy and energy density) compared to other battery technologies. Currently, mostly three cell designs (prismatic hardcase cells, prismatic pouch cells and cylindrical cells) with different advantages and disadvantages are present in the market. Novel cell concepts were developed to combine the strengths of the available cell designs, e.g. the good mechanical stability of hardcase cells and the high gravimetric energy density of pouch cells. Hybrid cell designs based on aluminum-polymer-composite foils and aluminum are a promising approach. In this paper, a laser structuring process is presented, which significantly improves the adhesion between thermally joined aluminum-polymer-composite foils and solid aluminum. Microscopic structures were formed on the aluminum surface by nanosecond laser pulses. The results of a laser parameter study were analyzed optically in terms of surface roughness by laser scanning microscopy and mechanically by adhesion strength tests. Based on that, strategies for the laser structuring process were derived.

Keywords: laser structuring; lithium-ion batteries; battery casing; aluminum; aluminum-polymer-composite

1. Introduction

Battery cell components like the electrodes, the separator and the electrolyte are enclosed by a casing, which should protect them from external influences such as moisture or dirt. At the same time the cell-internal components, which are partly toxic and flammable, must not be released into the environment. Additionally, a cost-efficient and high-throughput production of the housings with a low scrap rate is required for reasons

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of competitiveness in the battery market. Currently, three major cell designs dominate the market. Prismatic hardcase cells and cylindrical cells with a rigid aluminum casing as well as prismatic pouch cells with a soft aluminum-polymer foil casing. Each casing design has advantages and disadvantages regarding various aspects, e.g. weight, mechanical strength and fabrication costs. A novel cell concept with hybrid elements offers the potential to combine advantages and reduce drawbacks of the existing designs. This paper presents a battery cell casing, consisting of a hardcase battery housing joined with a pouch foil as a top cover, which allows producing a battery housing with good mechanical strength and reduced weight as it can be seen in Fig 1. To produce a hybrid battery casing, a solid aluminum surface has to be joined with the pouch foil (aluminum-polymer-composite).

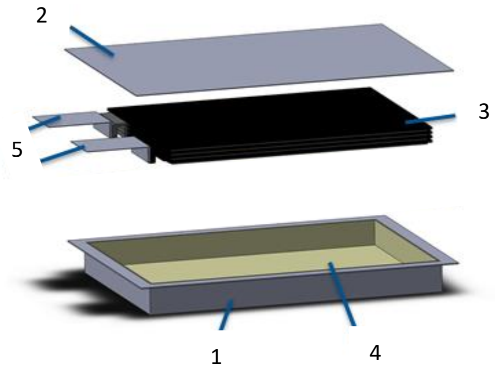

Fig. 1. Set-up of a concept for a hybrid battery casing

2. State of the art

Rigid aluminum and aluminum-polymer-composite foils can be joined by adhesive bonding or micro-mechanical interlocking. Both techniques are promising because of their integrability into a cost-efficient series production (Steinert et al., 2018). During adhesive bonding, a chemical and mechanical joint is generated (Goering et al., 2017). For mechanical interlocking, a polymer is used as the adhesive while infiltrating the metallic surface. In the literature, several joining methods are described which are used to produce the mechanical interlocking. Normally, the metal and polymer surfaces are located on top of each other under pressure and the contact area is heated until the polymer melts (Saborowski et al., 2019). The thermal energy for melting can be induced by a laser beam, induction heating, ultrasonic oscillation or direct heat conduction (Saborowski et al., 2019). For mechanical interlocking, it is beneficial if the surface is rough. This can be explained by the formation of numerous micro-sized, shape-connected joints in the surface structure (grooves, pores and surface roughness structures) (Temesi and Czigany, 2020). Several structuring methods such as grit blasting, etching, thermal spraying and laser structuring were applied to obtain advantageous surface features (Saborowski et al., 2019). The main advantage of laser structuring is the high degree of freedom in designing the roughness features, which results in the highest obtained adhesion strength (Saborowski et al., 2019).

The pre-treatment of the metal surfaces with laser radiation for an enhanced joining with polymers was investigated by several authors. Roesner et al., 2011 showed that laser structuring of stainless steel influences the shear strength of the joint with a thermoplastic material. It was observed that especially if small areas were joined, microstructuring with laser radiation and subsequent thermal joining can result in a better joint strength compared to an adhesive bond. Heckert and Zaeh, 2016 used laser surface pre-treatment to improve the strength of aluminum and glass fiber-reinforced thermoplastics joints. Macroscopic (structure height > 200 µm), microscopic (structure height: 100 nm – 200 µm) and nanoscopic (structure height < 100 nm) structures were generated and the influence on the shear tensile strength was examined. For short and long fiber-reinforced thermoplastics, microscopically structured aluminum showed the highest joint strength. For endless fiber-reinforced materials, nanoscopically structured aluminum reached the highest joint strength. Schricker et al., 2019 examined the temperature- and time-dependent penetration of surface structures during thermal joining of polypropylene and aluminum. It was shown that the penetration depth of the surface structure is the main parameter which is responsible for achieving a high binding strength between metal and plastic. Woitun et al., 2020 showed that the connection of aluminum and a thermoplastic polymer can be improved in terms of higher tensile shear strength through laser pre-structuring prior to injection molding. It was discovered that the joint strength depends on the surface area as well as on the groove geometry. Grooves with a depth of 100 µm showed an advantageous behavior compared to deeper grooves. Wunderling et al., 2020 thermally joined steel samples to carbon-fiber-reinforced thermosets. To improve the joint strength, the surface of the steel was structured using a circular laser beam oscillation. Thus, productivity and adaptability to complex geometries were increased in comparison to processing strategies with linear laser beam guidance.

3. Objective and approach

Increasing the specific energy on battery cell as well as on battery module level and decreasing production costs are essential steps to improve the competitiveness of electric vehicles. Batteries with novel casing designs can be a promising approach. To build a novel battery casing consisting of an aluminum hardcase with a pouch foil (D-EL408PH, DNP Co. Ltd.) top cover, solid aluminum has to be thermally joined with an aluminum-polymer-composite foil. To improve the adhesion strength between both components, a surface structuring method was applied. In this study, the influence of various process parameters of laser structuring (e.g. pulse repetition rate, structuring pattern, forward feed and number of repetitions) on the joint of aluminum sheets with aluminum-polymer-composite foils was evaluated by tensile shear strength tests. Based on this, process strategies and recommendations for the laser surface treatment were derived to enable the fabrication of a hybrid battery casing.

4. Experimental set-up

4.1. Laser structuring

The laser radiation was provided by a pulsed ytterbium fiber laser (YLPP-1-150V-30, IPG Laser GmbH). The specifications of the laser source are shown in table 1. The beam deflection was performed with a 2D scan head (Racoon 21, Novanta Europe GmbH). The laser structuring set-up is shown in Fig 2. The laser beam was directed with the mirrors of the scanning optics within the working area and focused by an F-theta lens (S4LFT0080/126, Sill Optics GmbH & Co. KG). The spot diameter of the laser beam at the focus point was approximately 30 µm. Structuring was conducted on aluminum sheets with a length of 10 mm and a width of 45 mm.

Table 1. Characteristics of the laser source

Laser system	IPG YLPP-1-150V-30
Type	Nd:YAG fiber laser
Operation mode	pulsed
Wavelength λ	1060 nm
Nominal laser power P	0 – 30 W
Pulse duration τ	0.15 ns, 1 ns, 2 ns, 5 ns
Pulse repetition rate f_R	60 – 1200 kHz
Spot diameter d_f	$\approx$ 30 µm
Beam quality factor M^2	2.0

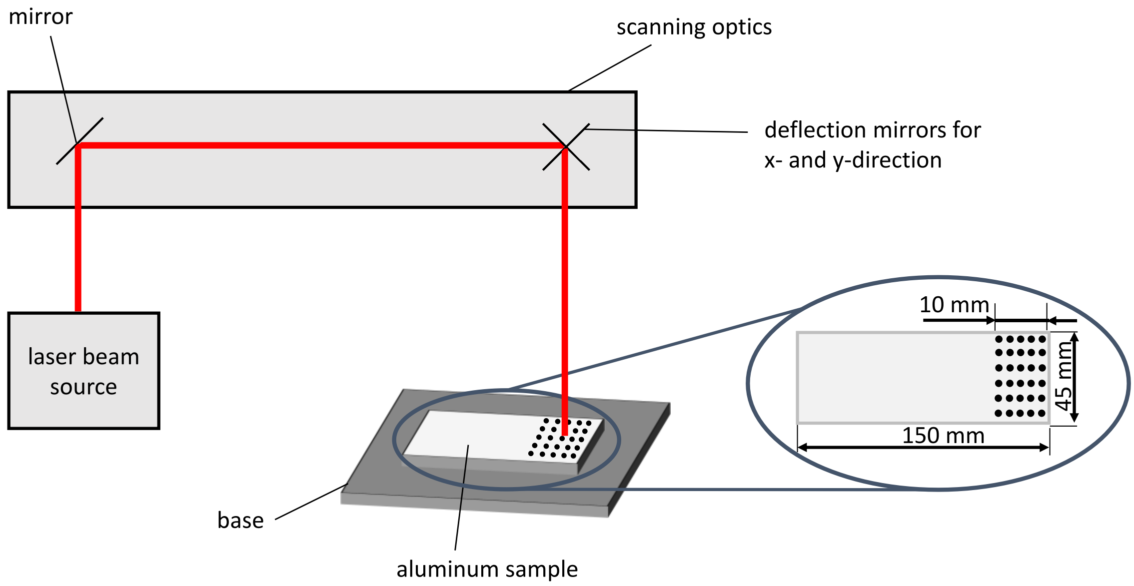

Fig. 2. Schematic set-up for laser structuring of aluminum samples

4.2. Thermal joining

After structuring, the aluminum samples were thermally joined to the pouch foils. During thermal joining, pressure and heat were applied by a manually operated hot press (Impulse Sealer W-2510HT, Wu-Hsing Electronics Co. Ltd). The experimental set-up is schematically shown in Fig 3. The press was heated to a temperature of 250 °C for joining. While heat and pressure were applied, the thermoplastic coating (polypropylene) of the pouch foil flew into the kerfs. After the heating process, the polypropylene cured and interlocking was achieved whereby the bond between the aluminum and the pouch foil was created. In preliminary tests it was identified that through a sealing time of 1.2 s the heat input can be optimized and the material damage can be reduced. To improve the bonding strength, the sealing procedure was repeated four times with pauses of five seconds between the individual steps.

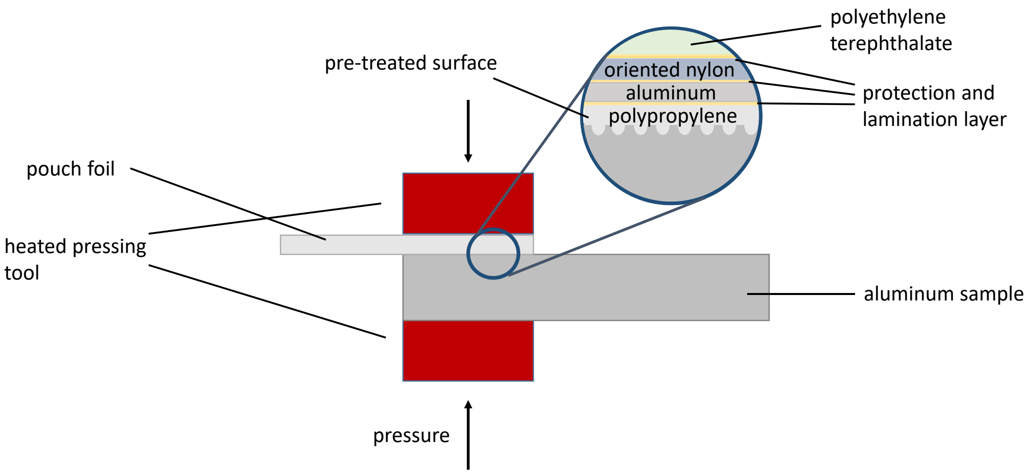

Fig. 3. Schematic illustration of the thermal joining process

4.3. Analysis methods

A scanning electron microscope (VK-X1000, Keyence Corporation) was used to analyze the topography of the samples and to determine the arithmetic surface roughness based on DIN EN ISO 4287.

A tensile testing machine (Z050 TE, ZwickRoell GmbH & Co. KG) was used to evaluate the mechanical strength. The hybrid specimens were tested based on DIN EN 1465 at a speed of 200 mm/min and no pre-load was applied. The maximum tensile strength was measured and evaluated.

5. Results and discussion

The laser power was adjusted to a maximum value of 30 W as a higher laser power led to an increased ablation volume and, thus, to a reduced structuring time. The pulse duration was set to 0.15 ns because the pulse peak intensity and at the same time the material removal increased with shorter pulse durations. Two different patterns, which are displayed in Fig 4, were evaluated. On the one hand, circular patterns, which showed advantageous properties in previous studies (Wunderling et al., 2020), e.g. increased productivity and adaptability to complex geometries for the enhancement of polymer-metal joints, were created. On the other hand, micro-drillings with a diameter of approximately 30 µm were introduced into the aluminum in a squared pattern (compare Fig 4, right). The pattern width was set to 10 mm for both cases.

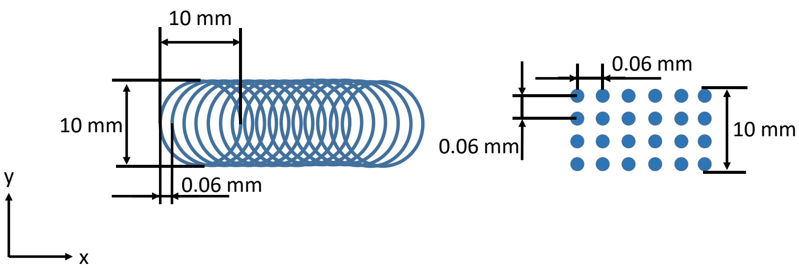

Fig. 4. Structuring patterns

5.1. Topographical surface analysis after laser structuring with circular beam oscillation

Initially, the influence of the pulse repetition rate (f_P), the scanning velocity (v_L) and the number of exposures (n_L) on the arithmetic surface roughness (*Sa)* was evaluated. To determine the influence of the frequency, pulse repetition rates of 60 kHz and 1200 kHz were chosen. Experiments with forward feed velocities of 200 and 400 mm/s were performed because preliminary tests have shown that these represent a good compromise between process time and the measured surface roughness. The pulse overlap therefore was between 99.4 % (f_p = 1200 kHz, v_L = 200 mm/s) and 77.7 % (f_p = 60 kHz, v_L = 400 mm/s). Additionally, the structuring process was repeated four times for two samples to evaluate the influence of additional passes on the surface roughness.

In Fig 5, the analysis of the surface roughness for different samples is shown. At a frequency of 60 kHz, the surface roughness was around 0.35 µm, independent of the scanning velocity and the number of exposures. By increasing the pulse repetition rate to 1200 kHz, the surface roughness rose by 100 % and 66 % at scanning velocities of 200 mm/s and 400 mm/s, respectively. A reason for that could be that the number of laser pulses reaching the surface at a repetition rate of 1200 kHz is much higher, which results in an increased heat accumulation at the surface. This leads to higher temperatures and increased material removal. Increasing the number of repetitions from 1 to 5 led to 80 % (v_L = 200 mm/s) or 35 % (v_L = 400 mm/s) enhancement of the surface roughness, which can be explained by the increased energy that was introduced into the surface. It can be assumed that the surface roughness did not increase by a factor of 5 because the laser beam was gradually defocused while the kerf depth increased.

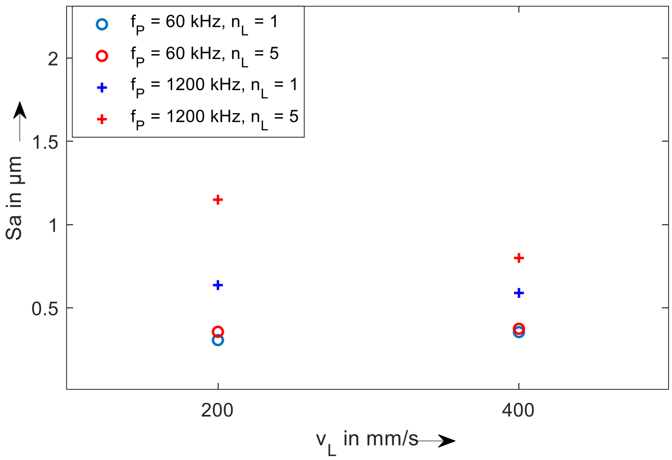

Fig.5. Analysis of the surface structure of samples pre-treated with circular structuring patterns

5.2. Shear strength analysis of samples with circular patterns

The structured aluminum samples were thermally joined with the pouch foil and tensile shear tests were conducted. In Fig 6, the tensile strengths of three samples with different arithmetic surface roughnesses are shown. A higher surface roughness led to a higher tensile strength and is therefore favorable. Increasing the surface roughness by a factor of 2 led to a 40 % increased tensile shear strength (blue line compared to yellow line). The significantly higher strain of the samples with a surface roughness of 0.78 µm and 1.15 µm can be explained by the high elongation of the pouch foil. Until a shear strength of approximately 0.5 MPa was reached, the strain of the pouch foil was relatively small (ca. 1 mm). While the shear strength was further increased, the foil was plastically deformed. The foil elongated nearly linearly with 2.5 mm as the shear strength was increased by 0.1 MPa.

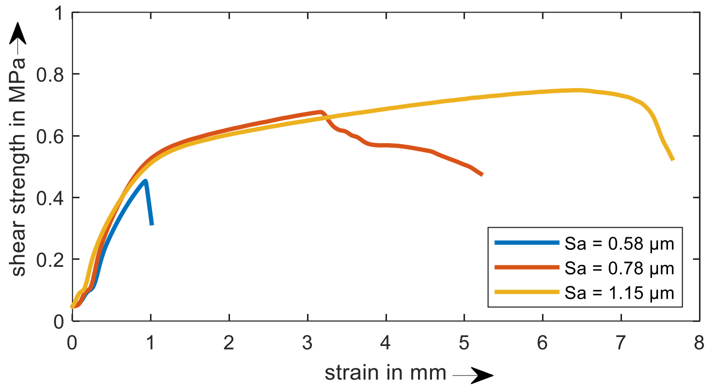

Fig. 6. Tensile shear strengths of thermally joined samples, which were laser structured with circular patterns

5.3. Topographical surface analysis after laser structuring with micro-drillings

For a comparison of the circular with the micro-drillings pattern, identical measurements were performed. In this case, no continuous velocity was applied, but a laser processing time per drilling (t_D) was defined. The processing time per drilling is the period of time in which one spot of the surface was structured by the laser beam. To improve the comparability, the period of time in which the laser structured the surface was kept constant for the circular pattern and the pattern with micro-drillings. In analogy to the circular pattern, the surface roughnesses of the samples resulting from a pulse repetition rate of 60 kHz were nearly the same (independent of the laser processing time per drilling or the number of repetitions). The surface roughness resulting from a repetition rate of 1200 kHz increased by 40 % (t_D = 0.5 µs) or 85 % (t_D = 1 µs) compared to the

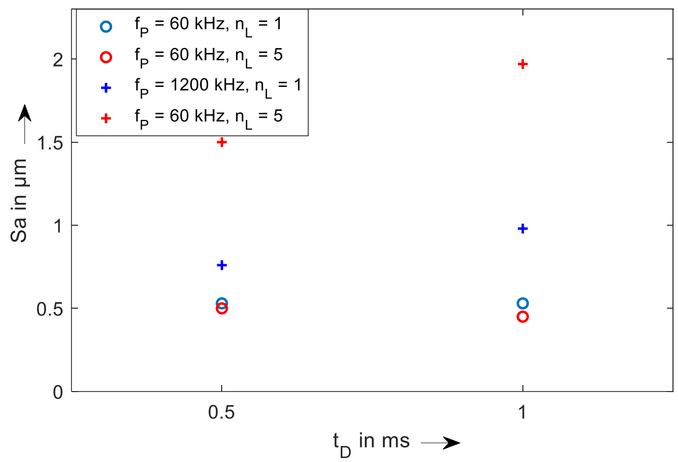

Fig. 7. Analysis of the surface structure of samples with micro-drilling patterns

samples resulting from 60 kHz. One possible explanation was already stated in section 5.1. The increased number of repetitions led to a by 97 % or 100 % increased surface roughness (t_D = 0.5 µs and respectively t_D = 1 µs). The higher surface roughness which was seen in the case of the increased number of repetitions can be explained by the higher energy input for each drilling hole. From Fig 7, it becomes clear that the surface roughness of the pattern with the micro-drillings was higher in comparison to the circular pattern. Since the micro-drillings were not generated using a continuous forward feed, the individual points were exposed to laser radiation for a longer period of time resulting in deeper indentations.

5.4. Shear strength analysis of samples with micro-drillings

The shear strength analysis of the structured samples with micro-drillings is presented in **Fig** 8. The blue line shows the results for a micro-drilling pattern with a surface roughness of 0.53 µm. For probes with surface roughnesses of 0.76 µm and 1.52 µm, the maximum shear strength (0.83 MPa) was similar. For both cases, the pouch foil elongated and therefore the shear strength could not be increased any further. When comparing the results to the tensile shear strength of the samples with the circular patterns (compare **Fig** 6) it can be concluded that the samples with micro-holes and a similar surface roughness performed better within the shear tests. It is assumed that the polypropylene interlocks better within the holes in comparison to the continuous kerfs of the circular pattern.

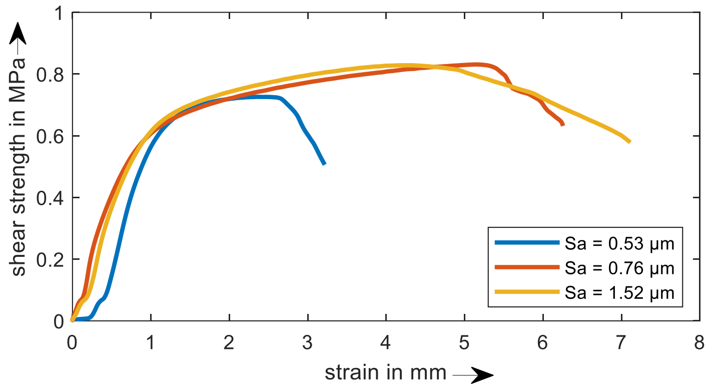

Fig. 8. Tensile shear strength of thermally-joined samples pre-treated with micro-drilling patterns

6. Conclusion

In this paper, strategies for the laser pre-treatment of surfaces to improve the adhesion between aluminum and aluminum-polymer-composite foils due to thermal joining were investigated. An increase of the pulse repetition rate and the number of passes as well as a reduction of the forward feed resulted in a higher surface roughness. Additionally, tensile shear tests were performed to investigate the strength of the thermally joined samples. It was observed that the micro-drillings pattern showed a higher shear strength in the tests in comparison to the circular pattern. For samples with surface roughnesses of 1.15 µm (circular pattern) or 0.78 µm (micro-drillings pattern), it was identified that the strain of the pouch foil was the limiting factor. For lithium-ion batteries, the gas- and liquid tightness has to be ensured since the battery components are sensitive to contamination and also partly toxic. Therefore, in further investigations the gas- and liquid tightness of the joint will be examined to guarantee a safe operation of the hybrid batteries.

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