Influence of Microstructure in Near-Surface Areas of Feedstocks on the Bond Strength of Roll Bonded Aluminum Clads

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This work determines the impact of the different grain sizes and oxide layers in near-surface areas of aluminum semi-finished products on the bond strength after warm roll bonding. To adjust the grain microstructures in the initially twin-roll cast feedstocks, they are differently treated to generate different grain sizes in the near-surface areas. Semi-finished products of the same near-surface microstructure are then joined by warm roll-bonding. In industrial applications, it is common to influence the bond strength of clads by applying a post heat treatment. Therefore, a part of the samples was annealed after warm roll bonding. Post-treated samples with coarser grain size exhibit up to two times higher peel strength than untreated samples. Higher peel strengths of the post-treated samples can be explained both by improved diffusion bonding due to heat exposure, and by recrystallization and grain coarsening of microstructure in the interface of sheets. Increase in peel strength of the composites made of feedstocks with coarser grains can be the result of stronger grain rotation, which damages oxide layer on the interface. To summarize, twin-roll cast and soaked material with the coarsest grains in the near-surface areas is the best feedstock for roll-bonding, if an additional heat treatment is applied.

1. Introduction

Aluminum has established itself as a lightweight building material, particularly in the transport sector, and for years has also been replacing structural components that were previously only made of steel. In the automotive industry, aluminum is used, for example, for engine components as a heat shield, in the chassis and in the body as a carrier and outer skin. However, the full potential of the lightweight construction concept was only made possible by the development of new composite materials, which combine the positive properties of different materials. One option for the manufacture of metallic composite materials is roll bonding, whose core competences lay in the creation of new material combinations, the targeted influencing of the microstructure during the roll-bonding process, and the optimization of roll bonding with regard to an improved control of 3D structural elements.

Due to the high pressure and friction forces during rolling, the brittle hydroxide and oxide layers break on the metal surfaces (Figure 1a) and the underlying pure metal extrudes through the cracks. Here, the purely metallic surfaces along the separation plane are in a closed environment. Therefore no oxidation, which would have prevented bonding, occurs. The connection in the layered composite is then made by mechanical cladding in the cracks and by atomic bonding. The interaction mainly occurs through grain boundary diffusion. As a result of forming the connection via roll bonding, the broken oxides in the interfacial plane are island-shaped, which impacts the bond strength negatively. To minimize this effect, chemical and mechanical surface pretreatments are recommended.[1–3]

A strong fusion of roll-bonded sheets depends mainly on the microstructures in their near-surface areas before roll bonding and the chosen process parameters. Important influencing factors are the initial sheet thickness,[4] the surface roughness due to pretreatments,[5–7] the reduction degree,[8,9] the roll-bonding temperature,[10] and the rolling speed.[11,12] Especially heat treatments[5] before and after the roll-bonding process have a significant effect on the peel strength (Figure 1b). A preliminary heat treatment reduces the hardness of the semi-finished sheets, which improves both the material flow and the bond strength.[12] The highest values of peel strength are characteristic for preliminary heat-treated roll-bonded sheets, but the reason of this effect remains unclear. The aim of this study is to determine the influence of the grain size in near-surface areas of feedstocks on the
bond quality during roll bonding with the same hardness of the initial material.

2. Experimental Section

Twin-roll cast 4.9 mm thick sheets of the aluminum alloy EN AW-1050A were used for the study. With its purity of 99.5 wt%, the alloy is classified as technically pure aluminum. The chemical composition of the alloy was specified in DIN EN 573-3:2019 and is shown in Table 1.

2.1. Preliminary Treatment of Aluminum Sheets

As the onset of recrystallization and new grain formation depends on the dislocation density in the lattice, it was necessary to perform cold forming with a reduction above the critical strain. Therefore, some of the aluminum sheets were cold rolled with a reduction of 40% to a thickness of 2.91 mm. At least one surface of the other part of the sheets was machined until the same thickness of 2.91 mm was achieved to allow different near-surface grain structures with the same reduction during the roll bonding.

Subsequently, some of the cold-rolled and all-machined sheets were soaked at 400 °C for 45 min to create different grain sizes in the near-surface area and to set a common hardness value for the material. The hardness values were determined using the Brinell method (DIN EN ISO 6506) with the Frank Frankoskop [13] device. In this manner, three states of the feedstocks—machined on both sides and annealed at 400 °C (M2A400), machined on one side and annealed at 400 °C (M1A400) as well as cold rolled and annealed—were prepared for the roll bonding. One part of the cold-rolled sheets was annealed at 400 °C (CRA400 state), whereas the other part was soaked at 520 °C for 45 min to influence the oxide layer on the feedstocks’ surfaces. The latter samples were labeled as CRA520.

To identify the microstructure in the cross section of the samples, they were prepared by grinding, polishing, and electrochemical etching for an examination in the Zeiss Axiophot light microscope [14]. The grain sizes in the near-surface areas were determined with the line analysis technique from a distance from the surface of 30, 60, and 90 μm with a 1000 μm long analysis line. The three resulting values were then used to determine the average near-surface grain size of each specimen. In addition, the surfaces of two samples (CRA400 and CRA520), which had been cold rolled and annealed at different temperatures, were cut with focused ion beam (FIB). The cut sections were analyzed in the scanning electron microscope (SEM) for the oxygen distribution in the near-surface areas using energy-dispersive X-ray spectroscopy (EDS). The cuts were performed and characterized in an FIB–SEM Zeiss NEON40 [15]. The acceleration voltage of the gallium ions was 30 kV. The SEM images of the cuts were taken with an inlens detector at a magnitude of 20 000 and an acceleration voltage of 2 kV. The EDS measurements were performed at the same magnitude at an acceleration voltage of 5 kV.

2.2. Roll Bonding of Preliminary Heat-Treated Aluminum Sheets

Before the roll bonding, the surfaces of the sheets were cleaned with ethanol. Then, identically pretreated sheets were put together in two-layer sandwiches so that the identical near-surface microstructures faced each other. In the case of sheets that were machined on one side (M1A400), these surfaces touched.

Table 1. Chemical composition of EN AW-1050A according to DIN EN 573-3:2019 in wt%.

| Alloy   | Al | Si  | Fe  | Cu  | Mn  | Mg  | Cr  | Zn  | Ti  | V  | Other |
|---------|----|-----|-----|-----|-----|-----|-----|-----|-----|----|-------|
| EN AW-1050 | ≥99.50 | 0.25 | 0.40 | 0.05 | 0.05 | 0.05 | 0.05 | 0.7 | 0.05 | 0.03 | 0.07 |

a) Limit of impurity

Figure 1. a) Scheme of the bonding mechanism during roll bonding. Reproduced with permission [3] © 2008, National Institute for Materials Science. b) Effect of preliminary and post heat treatment on the peel strength. Reproduced with permission [4] © 2009, Elsevier B.V.
Afterward, the two-layer sandwiches were preheated to a temperature of 350 °C, which was below the recrystallization temperature of the alloy, and subsequently warm rolled. The forming process was realized by means of the duo rolling stand of the Paderborn University. The preload forces of the rolls amounted to 10 kN, the rolling speed was set to 0.15 m s⁻¹, the roll diameter was 210 mm with a barrel length of 300 mm, and the composites were produced with a thickness reduction of 70%. After warm roll bonding, every composite was divided into two samples. Considering the influence of grain size on the bond quality of semi-finished sheets after post heat treatment, one half of the clads was annealed at 520 °C with a soaking duration of 45 min before being analyzed regarding the bond properties (labeled with ending “-W”), whereas the other half was investigated untreated (labeled with ending “-O”). The experimental procedure is shown in Figure 2, and the final labeling of the samples is given in Table 2.

### 2.3. Characterization of Bond Strength

First, seven rectangular specimens from each warm roll-bonded composite were sampled in the rolling direction (RD). Each specimen was 10 mm wide and 35 mm long, with the bonding area extending over the whole width and a length of 20 mm. The bond strength was then investigated by means of the quasi-static peel testing according to ASTM D 1876:2008 with the servo-hydraulic test machine MTS 858 Table Top System. To perform the peel tests quasi-statically, the peeling rate for determining the maximum peeling force was set to 0.015 mm s⁻¹ in the first minute of loading. After that, the pulling rate was increased to 0.3 mm s⁻¹ until the total fracture of the composite. In addition to the peel strength, the specific delamination energy of the composites was calculated as well, using Equation (1), which represents the energy in relation to the unit width used for a full composite delamination.

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\text{Specific delamination energy} \left( \frac{J}{\text{mm}} \right) = \sum_{i=1}^{n} \frac{\text{peeling force} \times \text{tensile path}}{\text{strip width} \times 1000}
\]  

(1)

With its help, a qualitative statement about the evolution of the peel strength over the entire composite surface could be made. However, the tensile path did not correspond to the peeling path over the clad length, as material deformation and crack tip propagation influenced the incremental peeling path along the interface. Based on the experimental results, statistical analyses were performed with the confidence interval of 95% = 1.96 σ and the degree of freedom n = 6. The standard deviation was corrected by the t-factor of 2.447.

After the peeling tests, the fracture surfaces were examined with the Zeiss Ultra Plus SEM for a more precise analysis of the amount of metallic bonds. The images were taken with a secondary electron detector at an acceleration voltage of 10 kV.

### 3. Results

The grain microstructure produced by soaking the unilaterally machined twin-roll cast strip (state M1A400) at 400 °C is shown in Figure 3a. The heat treatment led to a visible microstructure evolution in the near-surface area (marked by black squares), but not in the central area, which is typical for twin-roll cast materials. As in twin-roll casting the center of the strip has a higher temperature than the areas near the surface, the central zone is subjected to more intensive staining. It subsequently recrystallized, whereas the near-surface areas were work-hardened with a relatively low strain, which is indicated by a higher hardness in these areas. As a result of work hardening with a low strain, coarse grains were formed near the surface during the subsequent annealing.[20,21] If the work-hardened layers were milled from both sides prior to the heat

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**Table 2. Labeling of composite samples for the investigations of bond strength.**

| Samples without post heat treatment | Samples with post heat treatment at 520 °C for 45 min |
|------------------------------------|---------------------------------------------|
| M1A400-O                           | M1A400-W                                   |
| M2A400-O                           | M2A400-W                                   |
| CRA400-O                           | CRA400-W                                   |
| CRA520-O                           | CRA520-W                                   |

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**Figure 2. Scheme of the experimental procedure.**
Treatment, a fine-grained near-surface zone in the M2A400 state was formed (Figure 3b). Cold-rolling combined with a 45-min soaking at 400 °C resulted in the recrystallization of the entire sheet cross-section in the CRA400 state (Figure 3c). The inhomogeneous distribution of the initial dislocation density, which was additionally increased by cold rolling, led to the formation of differently sized grains. Much finer grains were visible in the near-surface areas, as the higher dislocation density therein produced many nuclei for new grains during recrystallization. These areas had already been work hardened during twin-roll casting. This observation coincides with previous studies.[23]

The higher temperature also increased the average grain size in the cold-rolled samples, with the strongest grain growth occurring in the sheet core. In contrast, the microstructure in the near-surface areas was almost unaffected by the temperature increase from 400 to 520 °C (Figure 3d). The quantitative values of the average grain sizes in the near-surface areas of all samples are given in Table 3.

The hardness and near-surface grain size values for the CRA400 and CRA520 states were almost identical; the difference being comparable with the accuracy of the experimental methods used. For this reason, an additional fused CRA400/520 state with an average grain size of 44 μm in the near-surface areas was separated. Furthermore, Figure 4 shows how the peel strength of warm roll-bonded aluminum sheets changed when the grain size in the near-surface areas of the raw material increased. At a grain size of 26 μm the peel strength was 3.7 N mm⁻¹ (M2A400-O) and 5.3 N mm⁻¹ (M2A400-W) for the states without and with a post heat treatment. By increasing the grain size to 44 μm, the peel strength of the cladding improved to 8.7 N mm⁻¹ (CRA400/520-O) and 12.6 N mm⁻¹ (CRA400/520-W), respectively. A further grain coarsening to 184 μm resulted in a decrease in peel strength to 6.9 N mm⁻¹ without a post heat treatment (M1A400-O) and an increase to 14.4 N mm⁻¹ with one (M1A400-W).

As the grain size increased, the specific delamination energy of samples manufactured without the post heat treatment rose up to a grain size of 44 μm, after which it decreased (Figure 5). In this case, however, the specific delamination energy was very low. Especially with a grain size of 26 μm, the composite delaminated after a short period of loading. The bond strength was significantly improved due to coarser grains in the interface zone of the clads. At a grain size change from 26 to 44 μm, for example, the specific delamination energy increased from 0.012 to 0.109 J mm⁻¹ without post heat treatment. In contrast, the specific delamination energy for samples with an additional heat treatment went up from 0.0825 to 0.214 J mm⁻¹. At a grain size of 184 μm, the specific delamination energy was reduced to 0.031 J mm⁻¹ (M1A400-O) without a post heat treatment and increased to 0.281 J mm⁻¹ (M1A400-W) with one.

The near-surface grain enlargement from 26 to 44 μm also increased the necessary maximum peeling forces, whereas further grain coarsening to 184 μm reduced it (Figure 6). They rose from 13.8 (M2A400–O) to 35.4 N mm⁻¹ (CRA400/520-O) and then fell to 26.6 N mm⁻¹ (M1A400-O), whereas the post heat treatment reduced the required maximum peeling force values to 11.7 N mm⁻¹ (M2A400-W), 27.1 N mm⁻¹ (CRA400/520-W), and 24.9 N mm⁻¹ (M1A400-W), respectively. All values and deviations of the charts are listed in Table 4.

The following is a comparison of the states of CRA420 and CRA520. Although the characterization of the initial state of the feedstocks showed strong similarities (Table 3), the analysis

![Figure 3. Light microscope images of cross-sections of sheets after electrochemical etching: a) M1A400 state, b) M2A400 state, c) CRA400 state, d) CRA520 state. Black squares mark the positions of bonding surfaces.](Image 48x743 to 145x765)
of the warm roll-bonded clads revealed a clear difference in their bond strength (Figure 7). As result of the various preliminary heat treatments, the peel strength values increased at higher soaking temperatures of the CRA520 state, whereby the near-surface grain size varied only slightly. The peel strength was 9.9 N mm$^{-1}$ in the CRA520-O state compared to 7.4 N mm$^{-1}$ for CRA400-O. The specific delamination energy also increased significantly, as did the maximum peeling force.
Figure 8 shows the corresponding FIB-section images of the sample’s near-surface areas in both the CRA400 and the CRA520 state. The focus is on the formation of the aluminum oxide layers in the areas marked by white circles. It should be noted that the oxide layers were of different thicknesses. Due to a higher thermal load on the material surface, a thicker and partially porous oxide layer formed in the CRA520 state (Figure 8b) than in the CRA400 state (Figure 8a). The findings were confirmed by EDS of the near-surface area, as seen in Figure 8c,d. Between the cut area with a higher aluminum concentration (red) and the surface area with a higher oxygen content (blue), an oxide layer with a mixture of both elements (encircled) was visible. It was thicker and contained more pores in the feedstock in the CRA520 state than in the CRA400 state. An additional quantitative EDS measurement along the white line supported this observation, with the oxygen concentration in the layer of the CRA520 sample being 29.41%, compared to 11.37% for the CRA400 state (Table 5 and 6). The sharper layers shown in the lower part of the images in Figure 8a,b are to be understood as deposits during FIB process.

3.1. SEM Fractography

The peeling tests were conducted transverse to the RD (TD: Transversal direction). All SEM shoots of the delaminated surfaces showed elongated asperities and dimples in the peeling direction in addition to the flat areas characteristic for roll bonding. The asperities and dimples represented the areas of former local metallic bonds between the cladded layers. The peeling led to the numerous cohesive ductile fractures of the aluminum clad, which were visible as bright elevations on SEM images (marking A in Figure 9b). In flat areas adjacent to the ductile fractures, no metallic bonding between the aluminum layers occurred (marking B in Figure 9b). Figure 9a,b show the fracture surface of the M2A400-O state, whose grain size was smallest before roll bonding, at 26 μm. The images revealed larger areas with former metallic bonds (marking C in Figure 9a) as well as areas with few and weakly formed connections (marking D in Figure 9b). In contrast, the CRA400-O state (Figure 9c,d) with a near-surface grain size of 43 μm had an evenly distributed pattern characteristic for cohesive ductile fractures over the entire separation plane. At the former interface of the slightly larger grain size of 45 μm in feedstocks of the CRA520-O state (Figure 9e,f ), a magnification between the large plastic deformation areas (marking E in Figure 9f ) and other smaller deformation areas was identified (marking F in Figure 9f ). As a result of the grain enlargement from 26 to 45 μm, the density of the metallic bonds on the surface increased during the roll-bonding process until a peak was reached. Subsequently, further grain coarsening to 184 μm led to a weaker bond, as the M1A400-O state showed (Figure 9g,h).

Figure 7. Influences of different annealing temperatures of the cold rolled material on a) peel strength; b) specific delamination energy; c) maximum peeling force of the aluminum clads after warm roll bonding.

| Sample | Grain size [μm] | Peel strength [N mm⁻¹] | Max. peeling force [N mm⁻¹] | Specific delamination energy [J mm⁻¹] |
|--------|----------------|------------------------|-----------------------------|----------------------------------|
| M2A400-O | 26 | 3.7 ± 0.6 | 13.8 ± 4.4 | 0.012 ± 0.01 |
| CRA400-O | 43 | 7.4 ± 0.3 | 30.1 ± 4.1 | 0.091 ± 0.03 |
| CRA520-O | 45 | 9.9 ± 4.6 | 40.6 ± 15.8 | 0.127 ± 0.08 |
| CRA400/520-O | 44 | 8.7 ± 2.0 | 35.4 ± 7.1 | 0.109 ± 0.04 |
| M1A400-O | 184 | 6.9 ± 0.8 | 26.6 ± 4.7 | 0.031 ± 0.01 |

With post heat treatment 520 °C / 45 min.

| Sample | Grain size [μm] | Peel strength [N mm⁻¹] | Max. peeling force [N mm⁻¹] | Specific delamination energy [J mm⁻¹] |
|--------|----------------|------------------------|-----------------------------|----------------------------------|
| M2A400-W | 26 | 5.3 ± 0.7 | 11.7 ± 2.1 | 0.082 ± 0.01 |
| CRA400-W | 43 | 11.5 ± 1.1 | 25.8 ± 2.1 | 0.190 ± 0.01 |
| CRA520-W | 45 | 13.7 ± 1.4 | 28.5 ± 1.5 | 0.237 ± 0.03 |
| CRA400/520-W | 44 | 12.6 ± 1.0 | 27.1 ± 1.3 | 0.214 ± 0.02 |
| M1A400-W | 184 | 14.4 ± 1.2 | 24.9 ± 2.3 | 0.281 ± 0.03 |
previous delaminated specimens in Figure 9a–h, the post-annealed aluminum clads in Figure 10a–h showed more areas of cohesive ductile fractures for the same near-surface grain sizes. Furthermore, the surfaces demonstrated a higher frequency of metallic bonds between the cladding layers with larger grains (mark G Figure 10d,f,h). Even at a grain size of 26 μm (Figure 10a,b), larger areas of microscopically small metallic bonds were visible. The highest number of cohesive ductile fracture areas were found both at grain sizes of 43 and 45 μm (Figure 10c–f) and also at 184 μm (Figure 10g,h).

4. Discussion
The evaluation of the peeling tests showed a strong increase in the peeling force at the beginning of the pulling (Figure 11), which led to a first microscopic delamination of the clad, when the maximum peeling force was reached. The measured maximum of the peeling force always exceeded the calculated peel strength in the tests by a factor of about 2.

The reason for the force peak is a stress state at the crack tip. In the beginning, a normal stress was induced in the separation plane of the clad, which deformed the notch and caused a plastic area to grow in front of the crack tip. Cracking followed, and the peeling force decreased. As a result, its peak was a brief moment, just before the first microscopic failure of the bond. The peak value was thus dependent on component geometry, notch geometry, stress state, and microstructure. The peel strength, in contrast, was the mean of a peeling force function over the entire sample length. During the continuous crack growth, i.e., the motion of the plastic zone along the interface until the bonding between the clad layers fails, the peeling force was assumedly constant.

Different grain sizes of the semi-finished sheets had a significant influence on the peeling tests. The peel strength increased analogously with the grain size up to a point where it slowly decreased. By increasing the grain size from 26 μm (M2A400-O state) to 44 μm (CRA400/520–O state) before warm roll bonding, the peel strength increased (Table 4), which corresponds to an increase of approximately +135%. The composite
Figure 9. SEM of fracture surfaces after peeling test of not post heat treated samples with the highest peel strength: a,b) M2A400-O; c,d) CRA400-O; e,f) CRA520-O; g,h) M1A400-O.
Figure 10. SEM of fracture surfaces after peeling test of post heat treated samples with the highest peel strength: a,b) M2A400-W; c,d) CRA400-W; e,f) CRAS20-W; g,h) M1A400-W.
with the largest grain size of 184 μm exhibited a sloping peel strength. These results included samples that were not post heat treated. The fracture surface analyses of the corresponding samples lead to the conclusion that the samples with the near-surface grain size of 26 μm (M2A400-O state) had fewer microscopic metallic bonds than the ones with coarser grains (Figure 9a,b). In addition, there were large areas without microconnections. These facts suggest that for the aluminum feedstocks with 26 μm grains, less cracks in the interfacial oxide layers occurred during warm roll bonding, which would have been necessary for the generation of juvenile surfaces. Thus, there were fewer metallic bonds, which resulted in a low peel strength of the M2A400-O state. At a grain size of 44 μm (CRA400/520-O state), in contrast, more cohesive ductile fracture areas were visible on the delaminated surfaces, which were much larger in their width, branching and length (Figure 9c,f). The higher peel strength of the CRA400/520-O state could be explained by the increased number of metallic bonds. As a result, the maximum peeling force increased in line with the grain size, increasing the crack resistance of the composite. This is also shown in Figure 5, which showed the specific delamination energy growing analogous to the more frequently detected areas of microscopic bonds on the fracture surface (CRA400/520-O state), caused by the grain coarsening. Nevertheless, the investigations also showed a peak in the near-surface grain size in terms of the specific delamination energy, as it dropped in the very coarse 184 μm grains of the M1A400-O state.

A comparison of the states CRA400 and CRA520 in Figure 7 demonstrated very different bond strengths, with the difference in their grain sizes of 43 and 45 μm varying only slightly. The FIB investigation (Figure 8) of the near-surface areas showed that the CRA520 state, which exhibited a significantly higher peel strength than the CRA400 state, also had a larger aluminum oxide layer before warm roll bonding. Literature on the forming of aluminum describes a higher susceptibility to cracking of thicker oxide layers as a result of rolling force.\(^{[24]}\) Thus, there were favorable conditions for the generation of juvenile surfaces and the formation of atomic bonds in the interface area. However, the thicker oxide layer on the interface before the roll bonding resulted in a higher peel strength deviation of the composite, as it increased from CRA400-O state to CRA520-O state. The additional heat treatment caused a reduction in the maximum deviation (CRA520-W). The fractography of the separation planes after the peeling tests confirmed that the asperities and dimples were less homogenously distributed on the fracture surface of the CRA520-O sample compared with the CRA520-W state (Figure 9f and 10f). This correlates with the higher peel strength deviation.

In general, the additional post heat treatment increased the peel strength of every sample (Figure 4). For the near-surface grain size of 26 μm for example, the peel strength increased from M2A400-O state to M2A400-W state. The specific delamination energy (Figure 5) also increased from M2A400-O state to M2A400-W state. This significant increase in bond strength over the entire length of the sample was also confirmed by the analysis of the fracture surfaces. In the M2A400-O state, only fine and diffusely distributed cohesive ductile fracture areas were found, indicating a weak bonding. The post-treated sample M2A400-W, on the other hand, showed a higher density of cohesive fracture areas, which corresponds to the higher specific delamination energy and peel strength. This suggests a stronger bonding. Enlarging the grains of the feedstocks to 44 μm made the increase in peel strength between not annealed and post heat treated samples even greater, as it increased from CRA400/520-O state to CRA400/520-W state. This was due to the improved diffusion conditions on the interface with more juvenile surfaces. However, the highest increase in peel strength and specific delamination energy was registered for the samples with the initially coarsest near-surface grains of 184 μm. The former increased by +109% from M1A400-O up to M1A400-W. Thus, the warm rolled bonds with the initially coarsest near-surface microstructure had the best peel strength and specific delamination energy after the post heat treatment.

In contrast to the peel strength and the specific delamination energy, the maximum peeling force was decreased by the post heat treatment (Figure 6), which also reduced the material strength. This caused a lower threshold value of the stress intensity factor, which indicates the crack initiation.

The assumed reason that the peel strength of the coarser near-surface bonds increased up to a certain point was an enhanced grain rotation. It is known that the ductility of materials depends on their grain size. Those with coarser grains usually have a lower ductility. This is because materials with coarser grains are less likely to have grains with dislocation slip planes oriented advantageously to the shear stress than materials with smaller grain sizes.\(^{[25,26]}\) Therefore, fewer slip planes can be activated during the loading, causing larger grains to rotate until they are favorably oriented for the dislocation sliding. This suggested that the coarser grains damaged the oxide layer at the interface of the aluminum feedstocks more during roll bonding, as the
probability of grain rotation was higher. Subsequently, more juvenile surfaces are generated and the aluminum could flow into the cracks, which increased the number of metallic bonds between the cladded layers. Nevertheless, a grain size peak must be considered, above whose value a further grain coarsening leads to a weakening of the bond strength. Furthermore, it is known that diffusion along grain boundaries proceeds much faster than through the grains.\textsuperscript{[13, 27]} Thus, with further grain coarsening, the number of grain boundaries along the interface became smaller and diffusion less effective. This led to the determined drop in peel strength above a threshold value of the near-surface grain size. To achieve the optimal bond strength between the roll-bonded layers, the interaction between the two effects that facilitate the breaking of the oxide layer and increase the number of diffusion paths must be considered. The latter was additionally favored by an additional heat treatment, which resulted in both a higher peel strength and specific delamination energy for the twin-roll cast feedstocks with the coarsest near-surface grains.

5. Conclusions

As the study has shown, different grain sizes and oxide layers in near-surface areas of aluminum semi-finished products have a noticeable impact on the bond quality of roll-bonded composites. With a grain size increase in the near-surface areas of the feedstocks from 26 to 44 μm, the peel strength increased from 3.7 to 8.7 N mm\(^{-1}\) (±135\%) when no post heat treatment was applied. This marked the peak of the peel strength, as it decreased to 6.9 N mm\(^{-1}\) at a grain size of 184 μm. However, when a post heat treatment was conducted, the peel strength continuously improved up to a grain size of 184 μm due to an enhanced diffusion. For the smallest grain size of 26 μm, the peel strength increased to 5.3 N mm\(^{-1}\) (±43\%) compared with the same sample without a subsequent heat treatment. At 184 μm, it increased even further to 14.4 N mm\(^{-1}\) (±109\%). Grain coarsening also influenced the specific delamination energy. Without a post heat treatment, it reached its highest value of 0.109 J mm\(^{-1}\) at a grain size of 44 μm. Similar to the peel strength, the specific delamination energy was highest in the post heat-treated samples with a near-surface grain size of 184 μm and amounted to 0.281 J mm\(^{-1}\). The better bond quality of the post heat-treated samples could be explained both by the improved diffusion bonding due to the heat exposure, and by the recrystallization and grain coarsening of the microstructure in the interface of the sheets. The increase in peel strength of the composites made of feedstocks with coarser grains could be the result of a stronger grain rotation. This damages the oxide layer on the interface during roll bonding and leads to an enhanced metallic bonding.

In addition, a closer examination of two differently preliminary heat-treated samples with near-surface grain sizes of 43 and 45 μm, which were not subjected to a post annealing, showed the influence of the oxide layer thickness on the bond strength. FIB images confirmed a stronger oxidation for the sample annealed at 520 °C, although the near-surface grain sizes and the hardness of both samples were almost equal. The more pronounced oxide layer on the surface of these feedstocks led to an increase in peel strength of the warm roll-bonded aluminum composites from 7.4 to 9.9 N mm\(^{-1}\).

In summary, this study revealed that the near-surface microstructure of feedstocks significantly influences their bond strength after roll bonding, and established relationships between the post heat treatment and the initial grain sizes. It was found that the twin-roll cast and isochronally soaked material with the coarsest grains in the near-surface areas is the best feedstock for roll bonding, if an additional post heat treatment is applied.

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Conflict of Interest

The authors declare no conflict of interest.

Keywords

aluminum alloys, bond strengths, grain sizes, near-surface areas, warm roll bonding

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[1] C. Schlögl, C. Planitzer, O. Harrer, B. Buchmayr, Berg-Huettenmaenn. Monatsh. 2011, 156, 249–254.
[2] L. Long, K. Nagai, F. Yin, Sci. Technol. Adv. Mater. 2008, 9, 23001.
[3] I. Kaur, Y. Mishin, W. Gust, Fundamentals of Grain and Interphase Boundary Diffusion, Vol. 3, Wiley, Chichester 1995, pp. 32–34.
[4] R. Jamaati, M. R. Toroghinejad, Mater. Sci. Eng. A 2009, 527, 2320.
[5] R. Jamaati, M. R. Toroghinejad, J. Mater. Eng. Perform. 2011, 20, 191.
[6] C. Tang, Z. Liu, D. Zhou, S. Wu, Strength Mater. 2015, 47, 150.
[7] K.-I. Mori, N. Bay, L. Fratini, F. Micari, A. E. Tekkaya, CIRP Ann. 2013, 62, 673.
[8] M. Heydari Vini, M. Sedighi, M. Mondali, Trans. Indian Inst. Met. 2017, 53, 609.
[9] M. Naseri, M. Reihanian, E. Borhani, J. Manuf. Process. 2016, 24, 125.
[10] M. Eizadjou, H. Danesh Manesh, K. Janghorban, Mater. Des. 2008, 29, 909.
[11] H. Yan, J. C. Lenard, Mater. Sci. Eng. A 2004, 385, 419.
[12] R. Jamaati, M. R. Toroghinejad, Mater. Des. 2010, 31, 4508.
[13] A. Puda, Makrohärteprüfer, https://mb.uni-paderborn.de/en/labstaff/laboratory-equipment/makroharterprüf (accessed: March 2020).
[14] A. Puda, Lichtmikroskop/Stereomikroskop, https://mb.uni-paderborn.de/labstaff/laboratory-equipment/lichtmikroskop-stereomikroskop (accessed: March 2020).
[15] M. Traut, Focused Ion Beam (FIB) mit dem Zeiss Neon 40, https://chemie.uni-paderborn.de/arbeitskreise/technische-chemie/cmp/labstaff/ausstattung/fib (accessed: March 2020).
[16] O. Grydin, Walzanlage, https://mb.uni-paderborn.de/labstaff/laboratory-equipment/walzanlage (accessed: March 2020).
[17] G. Chen, J. T. Li, H. L. Yu, L. H. Su, G. M. Xu, J. S. Pan, T. You, G. Zhang, K. M. Sun, L. Z. He, Mater. Des. 2016, 112, 263.
[18] L. d. Silva, M. El-Sharif, C. Chisholm, L. Stuart, Adv. Manuf. Technol. 2016, 3, 161.
[19] L. Tasche, Zeiss Ultra Plus, https://mb.uni-paderborn.de/ werkstoffkunde/laborausstattung/rasterelektronenmikroskopie-rem (accessed: March 2020).
[20] M. Cieslar, J. Bajer, M. Šlapáková, B. Klivská, M. Zimina, M. Stolbchenko, O. Grydin, Acta Phys. Pol. A 2018, 134, 145.
[21] Y. Birol, J. Alloys Compd. 2009, 471, 122.
[22] O. Grydin, M. Stolbchenko, M. Schaper, Int. Mater. Rev. 2017, 69, 2648.
[23] O. Grydin, M. Schaper, V. Danchenko, Metall. Min. Ind. 2011, 3, 7.
[24] E. Doege, B.-A. Behrens, Handbuch Umformtechnik. Grundlagen, Technologien, Maschinen, Springer-Verlag, Berlin, Heidelberg 2007, p. 446.
[25] G. Gottstein, Materialwissenschaft und Werkstofftechnik. Physikalische Grundlagen, Springer Vieweg, Berlin, 2014 pp. 265–272.
[26] N. Tsuji, Y. Ito, Y. Saito, Y. Minamino, Scr. Mater. 2002, 47, 893.
[27] G. Stechauner, E. Kozeschnik, J. Mater. Eng. Perform. 2014, 23, 1576.