



Bonding Strength of FRP-Metal Hybrids

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Abstract

The lightweight credo “the right material in the right place” raises an interesting concern once different materials are meant to provide a watertight bond. Therefore, we investigate the bonding behavior of metals with Fiber-Reinforced-Plastic (FRP) materials. In order to optimize the bond, the major influencing factors and their interactions are studied.

In order to identify the above interactions, FRP-metal hybrid specimens were investigated with regard to peel forces and shear strengths. During manufacturing the influencing factors such as sheet metal and FRP type, surface treatments, and bonding processes were varied.

Considering the peel force, a thermoset plastic matrix adhesively bonded to steel provided the best results, along with the use of a novel surface etching method by Kobelco. The latter yielded the highest shear strengths within this investigation. No bond could be obtained applying thermoset plastic matrices for in-operandi connections.

Using adhesives or surface treatments introduced additional production costs. Hence, in-operandi bonding would be a favorable option, however, one requiring further research. Compared to the material costs, the additional production costs could prove to be insignificant once the bonding process has been properly robustified and automated.

1 Introduction

Today, mechanical fastening and adhesive bonding are generally used to connect composite structures with metals. On the one hand, mechanical fastenings like bolts or rivets provide a reliable joining strength; on the other hand, they lead to a weight increase and poor sealing properties. Furthermore, the deepening of holes damages the structure and results in stress increase. In comparison to that, adhesive bonding offers additional sealing and a lower stress level in the joining area. However, the degeneration of the adhesive can reduce the bonding strength with time, emphasizing the importance of correct adhesive selection. In hybrid bonding, mechanical fastening on top of adhesive bonding is added to improve the overall joining strength [1].

Simultaneous joining during curing by co-curing (e.g. by using excess resin) is one option to reduce process time and manufacturing effort. However, the process time is reduced at the expense of bonding strength. Intensive surface preparation is necessary to improve bonding between adherents [2, 3]. For this purpose, Matsuzaki et al. introduce additional means of reinforcement such as bolts [2] or inter-adherent fibers [3].

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Monden et al. evaluated the influence of different surface modification techniques on the bond strength between metal and carbon fiber reinforced polymers (CFRP). Short-beam shear tests were used to determine the apparent interlaminar shear strength (ILSS). "Grit blasted surfaces increased ILSS values, but interfacial failure was mainly adhesive. TiO₂ coating and silane treatment showed lower ILSS values, but higher degree [sic] of cohesive damage. Combinations of TiO₂ coating and silane treatment showed ILSS values almost identical to those of pure CFRP" [4].

Schimanski et al. investigated the "[...] usage of wires (titanium) as transition elements between CFRP and aluminum. As a possible alternative to the liquid phase processes, the solid state process diffusion bonding has been applied. The experimental results showed high application potential of this process with respect to the transferable loads for integral transition structures" [5].

Saleema et al. employed a technique immersing aluminum substrates in a solution of sodium hydroxide (NaOH). They studied the effect of varying the treatment period on the adhesive bonding characteristics with an epoxy adhesive via single lap shear (SLS). Excellent adhesion characteristics with complete cohesive failure of the adhesive were encountered on the NaOH treated surfaces that are comparable to the benchmark treatments such as anodization, which involve use of strong acids and multiple steps of treatment procedures [6].

At Kobe Steel Ltd., Sugimoto developed several composite structures of aluminum and GFRP for weight and noise reduction, which were applied to actual vehicles. In addition to reducing weight by using aluminum, compounding with GFRP is a promising means for providing other functions such as vibration control and high rigidity [7].

A commercial method for enhancing the bonding strength is provided by the company plasmatreat. As the name implies, "plasmatreat"[ment] relies on applying a plasma coating to the bonding zone. Doing so, the organosilicon-compound of the coating is meant to provide an improved adhesion to metal or oxide surfaces. In addition, the organic components increase the binding force to plastic matrices [8].

In this study, a new surface etching method for aluminum alloys developed by Kobe Steel Ltd. is investigated regarding the influence of the joining force with CFRP compounds. The aluminum specimens are prepared with different surface treatments. The bonding between aluminum and CFRP is realized in two different ways: on the one hand using adhesive and on the other hand by welding thermoplastic CFRP in an autoclave process. Shear strength and peel resistance of coated samples are compared with uncoated samples using an SLS-test and peel-test.

2 Materials and methods

2.1 Overview and experimental design

Within the initial screening, a set of four major factors was set up and investigated using a full factorial experimental design. The factor levels chosen are shown in table 1.

Table 1: Experimental design for screening

Factor	Factor Level		
	Low	Medium	High
Sheet Metal	Mild Steel	-	Aluminum
Interface Surface	Polished	Cleaned	Roughened
Composite Matrix	Thermoplastic (Organo Sheet)	-	Thermoset (Prepreg)
Hybrid Material Bonding Method	In-Operando	-	Adhesive

For each factor combination, material for a set of five specimens was provided. However, due to losses during specimen preparation, the number of repetitions was reduced to three.

Based on the result of the above screening, a second experimental batch on most promising factors was conducted. For this purpose, the thermoplastic matrix with in-operando bonding procedure was omitted, while a surface coating was investigated instead of a mechanical preparation. Furthermore, by focusing only on aluminum, a simplified experimental design could be used as shown in table 2. Similar to the earlier testing set-up, a set of five specimens was prepared and experimentally investigated.

Table 2: Experimental design using chemically treated surfaces

Metal sheet	Coating / interface treatment	Composite matrix	Hybrid material bonding
Aluminum	Kobelco etching	Thermoset	In-Operando
Aluminum	Kobelco etching	Thermoset	Adhesive
Aluminum	PST coating	Thermoset	In-Operando
Aluminum	PST coating	Thermoplastic	In-Operando

2.2 Materials

Considering their application in the automotive field, two common sheet metal materials – steel and aluminum - were chosen. For steel, the commonly used DC01 (1.0330), as described in [9], was selected. As the lowest grade of high-quality forming steel materials, DC01 offers the advantage of resulting in comparable quality to higher grades, if processed within 6 months, while offering the most economical choice in sheet forming.

For the second factor level in metals, the wrought aluminum-magnesium alloy EN AW-5754 H22 (3.3535) was used. This material combines good forming properties with a high resistance to corrosion – even in seawater. In the given condition of H22, the material is work hardened by rolling and then annealed to quarter hard. Thus the tensile strength of 220 – 270 MPa in H22 condition is smaller, but close compared to DC01, ranging from 270–400 MPa.

The next material factor is the FRP material used for creating the hybrid structure. Here, prepreg sheets were tested against organo sheets. With respect to prepreg sheets - referred to as thermoset in the experimental design – the material “TenCate E644” was used. The denoted material is an epoxy component prepreg which toughens during curing. It consists of a weave of T300 carbon fibers in 0°/90° orientation, resulting in a layer thickness of 0.5 mm. It requires curing at 80°C for 105 min where post curing for higher temperatures is possible. Furthermore, tempering (post curing) at 140°C for 4h at a glass transition temperature of 124°C is possible.

Organo sheets – referred to as thermoplastic in the experimental design due to their PA 6.6 matrix – resemble the second factor level in FRP materials. In contrast to the prepreg material, depending on the required test specimen (see next section), two different initial sheet thicknesses were used. Here the organo sheets 201-C200(2) – featuring a layer thickness of 0.5 mm – and 201-C200(4) – with a thickness of 1.0 mm – were used. Both FRP sheets feature carbon fiber with a fiber volume content of 45%.

As the investigation compares the bonding strength of in-operando bondings versus adhesive-bonded hybrids, suitable adhesives must be chosen for the corresponding FRP materials. Thus, the choice of the adhesive is a vital, albeit neither a varied nor an investigated factor for the bonding strength. In this study, two different adhesives were used, based on the FRP to be included in the joint. For prepreg sheets, the two-component epoxy adhesive Scotch-Weld DP 490 by 3M was selected. On the other hand, organo sheets were joined using the adhesive L-F503 by L&L Products. Aiming to achieve a high level of industrial applicability, the adhesives were chosen based on the recommendations of our suppliers.

2.3 Bonding methods and specimen preparation

Depending on the test to be performed, two different specimen geometries are created. For the SLS tests, a sheet metal strip of 1.5 mm thickness was joined to an FRP sheet of a resulting similar thickness. The bond was created over a length of 12.5 mm. For the peel-test, a sheet metal strip of a total length

of 250 mm was bonded over a bonding length of 145 mm to an FRP sheet of 195 mm length. As a result, the layers of the FRP sheet added up to a total thickness of 2.5 mm, whereas the sheet metal has a thickness of 0.5 mm. All specimens have a uniform width of 25 mm. The geometry is depicted in figure 1.

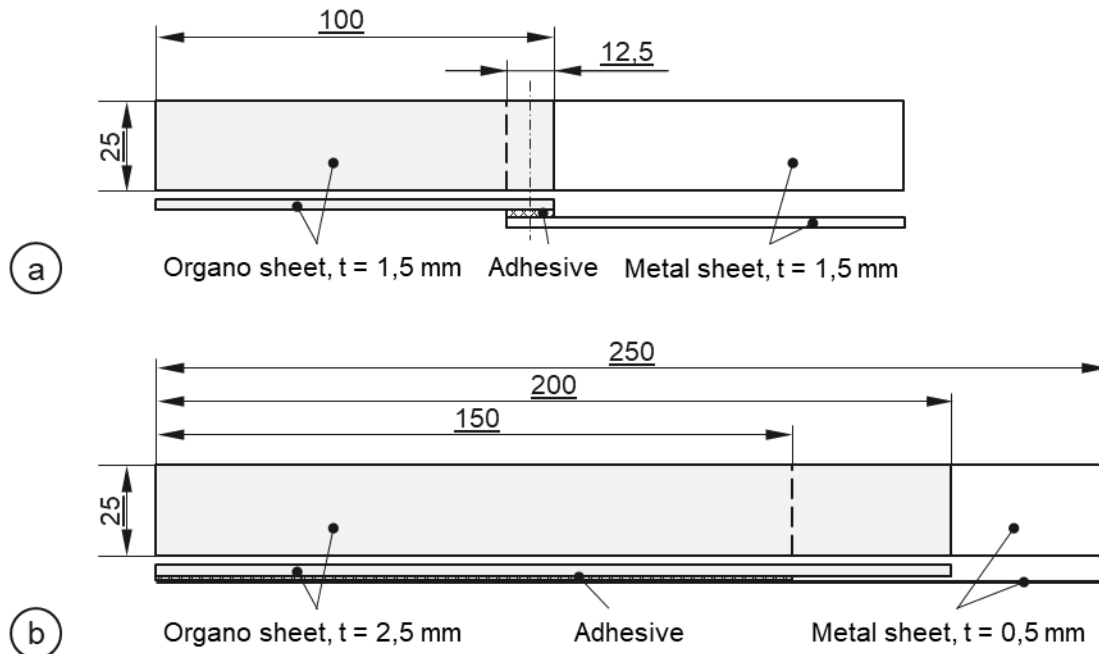


Figure 1: Specimen geometries (*not to scale*); a: SLS-test specimen; b: Peel-test specimen

The sheet metal parts of the specimen are cleaned using acetone prior to the surface preparation (cleaning, roughing or polishing) of the bonding zone. Next, the prepared sheet metal parts enter either a prepreg autoclave process for creating the thermoset specimens or a press forming process using preheated organo sheets.

In the autoclave prepreg process, a typical series of manufacturing processes is used. For both bonding methods, metal as well as prepreg sheets are positioned in the preheated autoclave at 80 °C to be held at a pressure of 4 bar for 105 minutes. In the subsequent joining process, the adhesive is applied to the positioned metal sheets. Next, the thermoset sheets from the autoclave are placed on the adhesive, as shown in figure 1. In order to create the hybrid-adhesive-prepreg-metal bond, the hybrid structure is heated to 65 °C under a pressure of 0.5 bar for 120 minutes.

Similar to the above autoclave process, the thermoplastic pressing process is considered a typical series manufacturing process. It basically involves the press-forming of preheated organo sheets. Using this approach, the organo sheets are first heated to 320 °C for 15 seconds. Meanwhile, the metal sheets are either subject to adhesive application at room temperature or heated up to 280 °C and cooled down in the mold to 200 °C for in-operando bonding. Next, the heated organo sheet is cooled down in the press to approximately 300 °C. Finally, the press forming commences at a mold temperature of 110 °C and 100 kN for the SLS or 200 kN for the peel-test specimen.

Regarding the surface interface, treatments were considered. First, a mechanical treatment of the bonding zone was conducted. Here, we aimed to increase the surface roughness, ranging from polishing to roughening. Based on the results of the screening, the second experimental design (see table 2) introduced chemical treatments. In doing so we investigated the effect of chemical etching versus plasma coating on the bonding strength. Using chemical etching, a fine unevenness was formed on the aluminum surface in the micron order in anticipation of the anchor effect.

Apart from the chemical surface treatments, which were applied to the sheet metal either by Kobelco (etching) or PST (coating), all specimen manufacturing was carried out as subcontracted work at our partner facility, the Leichtbau-Zentrum Sachsen (LZS) GmbH.

2.4 Testing methods

For the assessment of the bonding, the Single-Lap-Shear-Test (SLS-Test), as described in the standards [10,11], as well as the peel-test with respect to [11] were considered. The individual test parameters are listed below in table 3.

Table 3: Testing Parameters

Parameter	Single-Lap-Shear-Test (SLS-Test)	Peel-Test
Testing Machine	Zwick UMP 1475 (LZS ID: LIZ PR 01 11)	
Fixture/Clamping	Mechanical clamping jaws	Peel-testing device
Deformation Measurement	Machine Cross Head	
Force Measurement	10 kN load cell (LZS ID: LIZ PR 01 170)	
Test Climate	Controlled: 23 °C, 50 % RH	
Pre-Force	10 N	20 N
Testing Velocity	3 kN/min	100 mm/min

Like the specimen preparation, the experiments were conducted as subcontracted work at our partner facility, the Leichtbau-Zentrum Sachsen GmbH.

3 Results

Prior to the actual testing, it was found that bonding failed when combining any organo sheets with metal parts using the in-operando method. Consequently, a further discussion of the above results is omitted.

3.1 Single-Lap-Shear Test

The results of the Single-Lap-Shear-Test are shown in figure 2.

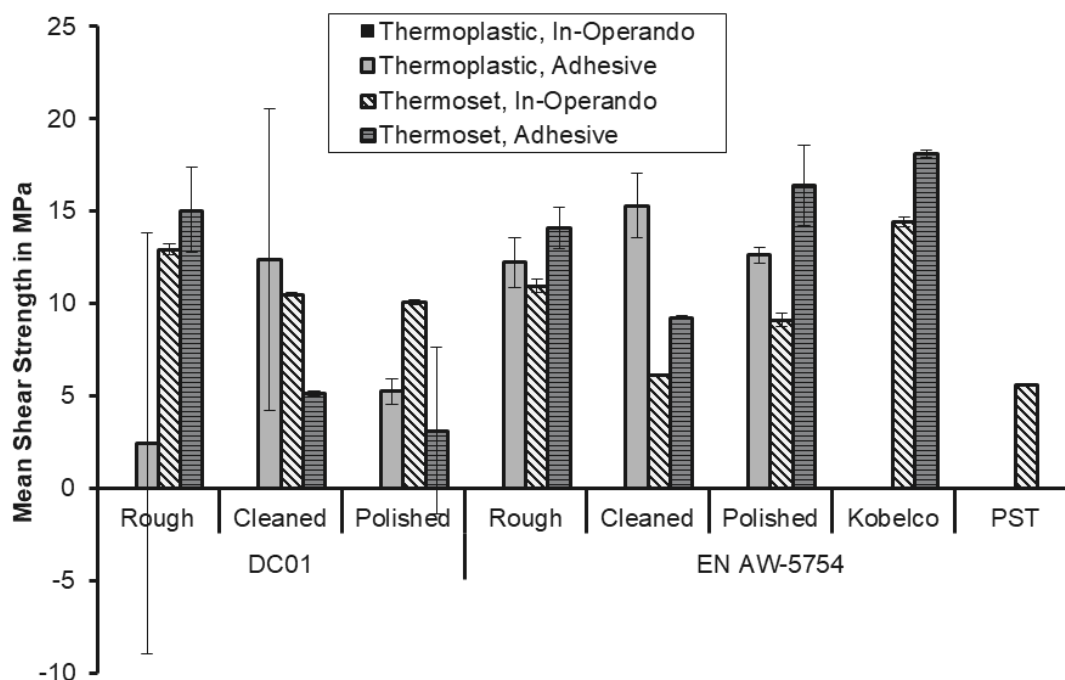


Figure 2: Resulting mean bonding strengths in a 95 % confidence interval (error bars)

As described in DIN EN 2243-1 [11], we calculated the mode II bonding strength σ_a using equation 1,

$$\sigma_a = \frac{F}{lw} \quad (1)$$

where F is the breaking load in N, l the overlapping length in mm, and w the width of the overlapping area in mm. For each set of factor combinations, we evaluated the resulting mean value by calculating a 95 % confidence interval using the student distribution.

3.2 Peel-Test

We assessed the peel resistance τ according to the standards DIN EN 1464 and DIN EN 2243-2. By omitting the first and last 25 mm of the total peeling length, an average peel force F_{avg} was calculated for each measurement. Taking w , the width of the overlapping area into account, the peel resistance can be calculated according to equation 2.

$$\tau = \frac{F_{avg}}{w} \quad (2)$$

Applying the same statistical approach used for the evaluation of the shear strength, the results shown in figure 3 are obtained.

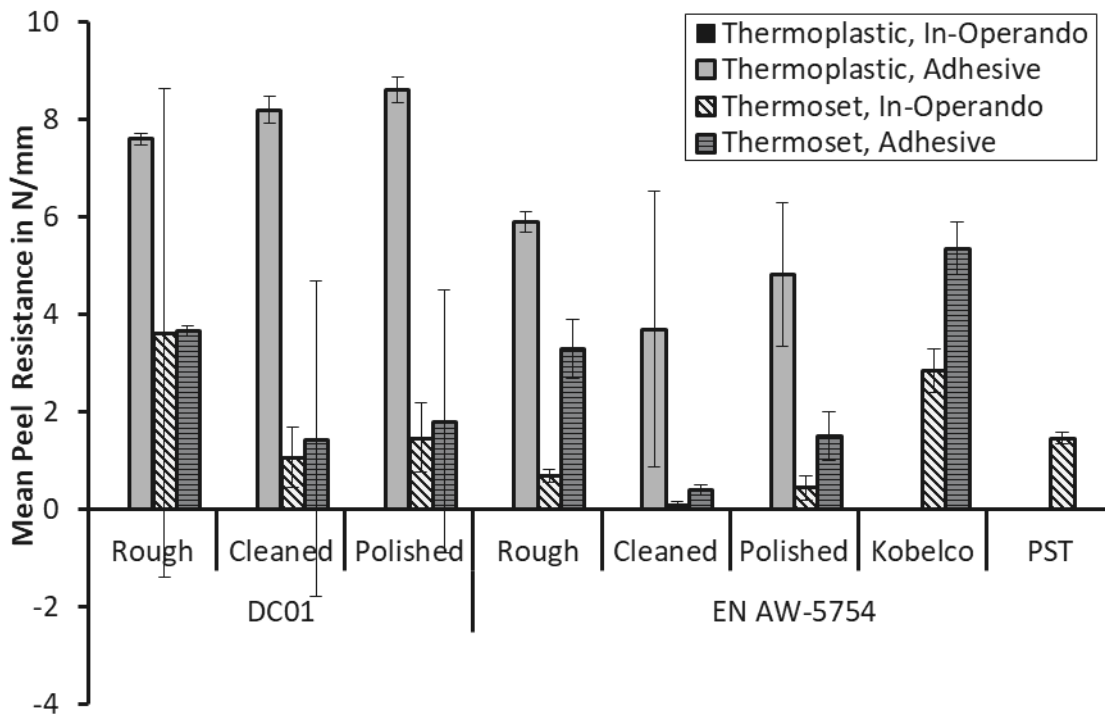


Figure 3: Resulting Mean Peel Resistance in a 95 % confidence interval (error bars)

3.3 Influencing factors and main effects

While the above results depict only the raw data, a concise conclusion about the main influencing parameters is not apparent. Thus, we performed a main effect analysis by calculating the mean values of all high settings of a given factor and all low values respectively. The difference between the mean effects at all high and all low states yields the main effect of a factor. Those results are shown in figure 4.

Figure 4 uses the same scaling for both the SLS and the peel-test results. Therefore, a common ordinate is used. Since all results contribute to the statistical calculation of the main effects, the confidence intervals are very small. As such, apart from the impact of the choice of plastics on the SLS-Test, all effects are within the range of 99.9% confidence and can thus be considered significant.

The main effects show a strong positive effect of the coating on both the peel and shear strength. The factors bonding method and metal sheet result in a medium effect in both tests. On the other hand, the factors treatment and plastic yield a negative effect (i.e. the assumed “low” factor results in a high output and vice versa) for the peel-test.

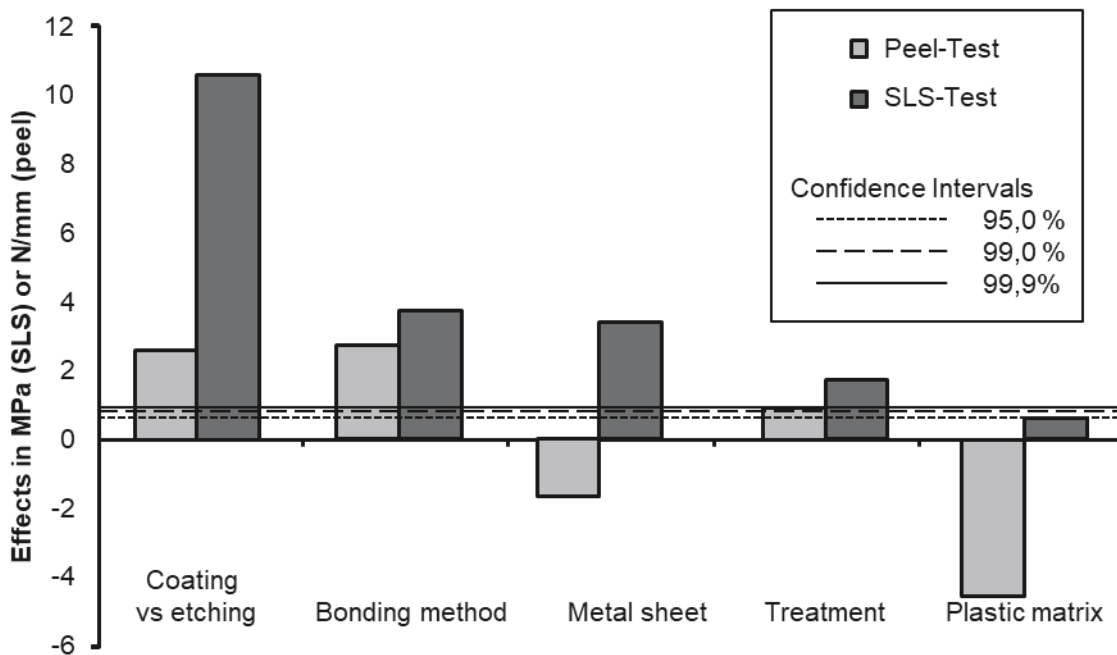


Figure 4: Influencing factors and main effects

4 Discussion and conclusion

The failure in bonding using the in-operando method in combination with organo sheets can be explained by the resulting thermal expansion (and shrinking) during the manufacturing process.

Considering the SLS-test shown in figure 2, the steel-thermoplastic specimens result in very large confidence intervals, thus leaving no statistically profound space for discussion. On the other hand, the aluminum-based specimens, yield significantly high values once adhesives are used. The best results can be obtained using the Kobelco coating. Doing so, even the in-operando procedure provides very good results, while the combination with adhesives exceeds in shear strength.

With respect to the peel resistance in figure 3, the highest values could be obtained by applying thermoplastic sheets using adhesives. Thermoset matrices, while allowing the use of in-operando bonding, show resulting force levels significantly smaller compared to thermoplastics. With respect to the material, steel yields better results compared to aluminum. However, the confidence interval when using aluminum is very large, thus calling for an extension of the experimental run. Similar to the failure in the in-operando joining of organo sheets, we assume the higher thermal stresses are the main cause for failure when using aluminum. The effect of an increased roughness is discernible, while its resulting total increase is rather small.

Considering the bonding strength using PST coating, our results of 5.6 MPa are fundamentally lower than the results of 14.35 MPa, reported by PST [8]. In the discussion with PST we have found that using an furnace for specimen preparation may result in contamination of the bonding area. In addition to the temperature of both the metal and the plastic matrix, this contamination results in a fundamental loss in bonding strength.

When evaluating the main impact on the resulting bonding strength, figure 4 clearly refers to the coating with a strong positive effect on shear strength and peel resistance. While the bonding method still has a relevant effect on both test results, the other factors result in a major difference between peel and SLS-test.

Concluding, we recommend using steel with an adhesively bonded thermoplastic if aiming for the best peel resistance. With respect to shear strength, the Kobelco treatment along with adhesives yields the best results. Considering an efficient in-operando process, the only statistically promising option is using the treatment by Kobelco.

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