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Basic material and technology investigations for material bonded hybrids by continuous hybrid profile fabrication

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Abstract. The development of multi-material hybrids by injection molding has been studied very intensively at the IPF in the past. For that, a material bonding between the different substrates was achieved by using a newly developed two-step curing powder coating material as latent reactive adhesive. The aim of the project “Hybrid Pultrusion” was to perform a novel approach for the fabrication of material bonded metal-plastic joints (profiles) in a modified pultrusion process. Therefore, powder pre-coated steel coil is combined with a glass-fiber reinforced epoxy resin matrix. For initial basic studies, the impregnated fiber material has been applied on the pre-coated steel sheets using the Resin Transfer Molding process (RTM-process). It was proved via lap shear tests, that this procedure resulted in very high adhesive strengths up to 35 MPa resulting from the formation of a covalent matrix-steel bonding as well. In addition, the failure mechanism was subsequently studied. Furthermore, by adapting the successful material combination to the pultrusion process it was demonstrated that material bonded hybrids can be achieved even under these continuous processing conditions.

1. Introduction

In many areas, weight reduction plays an important role in saving energy, especially in future electric vehicles. For that reason, multi-material combinations became more and more important during the last years. According to the state of the art, there are several ways to produce multi-material structures [1]. In the past those joints were achieved mostly mechanically, e.g. by form fitting or clamping. Now, material bonding plays an outstanding role for the production of lightweight structures, especially in the automotive and transportation industry, but also for machines and household appliances. In practice, material bonding is usually achieved by applying an adhesive or primer. However, high adhesive strengths in combination with a high durability under application conditions, e.g. for metal-plastic joints, can only be reached by additional cost and time-consuming pre-treatment steps. A direct adhesion works only for less material combinations.

During the last few years a two-step curable powder coating system based on commercially available uretdione (internally blocked isocyanates) cross linkers, OH-functionalized polyester resins and a specific catalyst system was developed at the IPF (Figure 1) [2]. The curing mechanism runs via two crosslinking steps that can be separated by curing time and temperature selectively. At temperatures lower than 160 °C a polyallophanate structure is obtained that shows a high level of post-formability.

At temperatures higher than 180 °C the polyallophanate structure can be transferred to polyurethane within a short time period. Additionally, the post formability of the coating system as well as the surface appearance can be maintained at the high level. For this reason, the second process step can subsequently be used for interlayer reactions and a covalent linkage of different materials. Based on that, the two-step crosslinking for the fabrication of Al-powder coating-plastic hybrids by injection molding was studied very intensively at the IPF. In that manner, 3- or 4-layer metal-plastic hybrids with a very high bond strength were generated [3].

The subject of the current investigation was the adaptation of this concept to the manufacture of metal-plastic joints consisting of powder-coated steel and a glass-fiber-reinforced epoxy matrix. At IPF, powder coating and material development was performed using the discontinuous RTM process. Subsequently, preferred material combinations were transferred to the pultrusion process and a proof of the technological feasibility of such material joints was provided under continuous conditions at IWU.

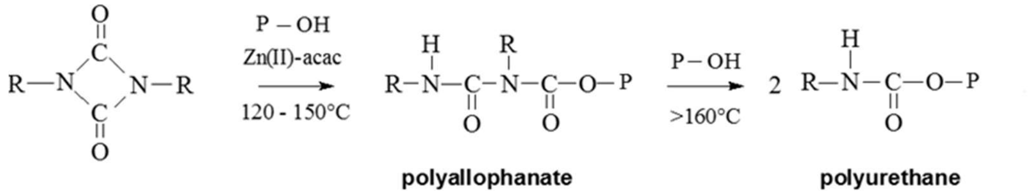

Figure 1: Curing reaction of uretdione-based powder coatings in the presence of a special catalyst [2]

2. Hybrid-materials by using the RTM process

Test plates with a total thickness of 4 mm were fabricated via RTM process in extensive laboratory tests in order to investigate the best process conditions and material combinations.

2.1. *Manufacturing of the hybrids*

First, the steel sheet substrate (DC01, 2 mm thick) was sandblasted with high-grade corundum and pre-treated with zinc phosphate in an industrial facility, resulting in a fine crystalline layer. In this manner, a good corrosion protection was received for the metal. In addition, previous studies have shown that such treatments are necessary to achieve a high adhesion of the powder coating film in the subsequent hybrid structure [4]. Then, the pretreated steel sheets were coated with various latent reactive adhesives in the form of powder coatings by using corona discharge. The electrostatically applied powder particles on the steel sheet were then cured (150 °C / 15 min). During the crosslinking step, the powder coating, while strongly bonding to the metal, was transformed into a polyallophanate network.

In the next step, the powder coated steel sheets was combined with glass fiber reinforced epoxy resin via RTM process. For that, an epoxy resin system was used (Huntsman, Table 1).

Table 1. Epoxy matrix formulation.

Component	Component name	Supplier
Resin	Araldite LY 3585 CH	Huntsman
Curing agent	Aradur 917-1 CH	Huntsman
Catalyst	Accelerator DY080	Huntsman
Internal Mold Release	IC25	ChemTrend
Filler	ASP 600 (0.6 µm)	BASF

The steel sheet and the fiberglass mats was placed in the RTM plate tool (Figure 2) and heated up to 90 °C under vacuum after closing. Afterwards, the previously mixed epoxy resin system was injected by using a pressure pot. After a curing cycle of 90 °C / 90 min the cross-linked hybrid structure was demolded. Half of hybrid panels (Figure 3) were then thermally treated at 160 °C for 3 h or at 200 °C

for 1 h, respectively, to investigate the effect of post-curing on the resulting bond strength between the powder coating layer and the fiber reinforced epoxy matrix.
It was expected that during the thermal treatment the 2nd reaction step from polyallophanate to polyurethane would occur. As a result, more covalent bonds should be generated in the interface between the powder coating layer and the epoxy resin, leading to higher bond strengths. When investigating the influence of the powder coating formulation, the main focus was on varying the OH number of the polyester resins contained and on adding OH-functionalized dendrimers at 1 or 3 wt% to study their influence on the adhesion strength to the epoxy matrix.

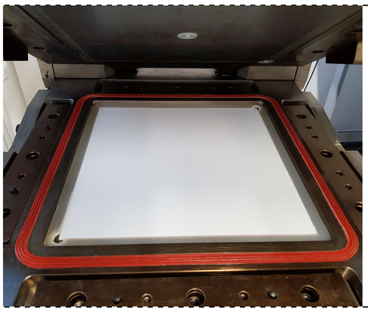

Figure 2. Opened RTM-tool for hybrid preparation.

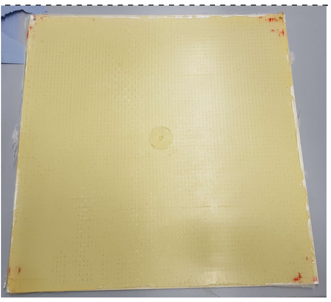

Figure 3. Material bonded steel - thermoset (glass fiber reinforced epoxy) hybrid material plate by RTM.

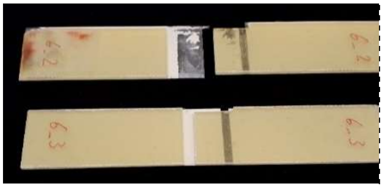

Figure 4. Standardized tensile test specimen according to DIN 65148 before and after the lap shear test.

2.2. *Lap shear tests*

To determine the bond strength, a lap shear test was carried out in accordance to DIN 65148 [5]. For this purpose, the hybrid parts were cut to standardized tensile specimen (dimension 250 mm x 25 mm x 4 mm) on a water-cooled separating device. In addition, two notches are milled into the specimen, resulting in a shear area of 25 mm x 12.5 mm, as recommended in DIN 65148 (Figure 4).
After testing, the occurred fractures were analysed by the possible failure patterns sketched in Figure 5 in detail. The percentage was determined from every possible type of breakage.

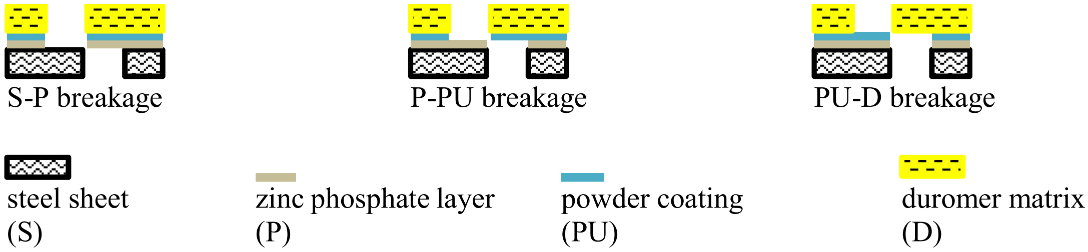

Figure 5: Main failure patterns/failures occurring during the strength test.

Figure 6 shows the influence of the different OH numbers of the polyester resin in the powder coating formulation on the resulting shear strength. Shear strength was expected to increase when larger hydroxyl numbers were used. However, it was found, that the shear strength decreases with the increasing hydroxyl number. It can be assumed that other parameters affect the tensile testing results as well, for example the glass transition temperatures of the powder coatings and resulting thermal stresses during the cooling process after the manufacturing in the RTM plate tool. It was found that the HP-01 powder coating (OH number: 51 mg/g) should be preferred for further testing. Additionally, it was observed, that the failure mainly happens between the zinc phosphate layer and the powder coating (P-

PU) and not the real interesting PU-D interlayer. So only can be concluded that the adhesion between powder coating and the epoxy resin (PU-D) is higher than the determined shear strength values.

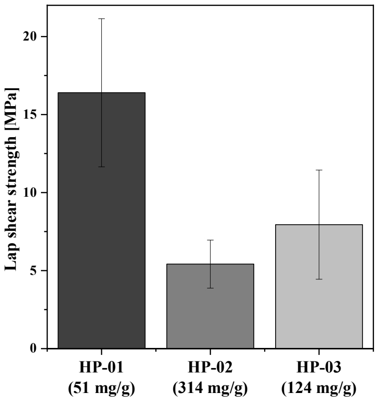

Figure 6: Lap shear strength by varying the OH-group number of the containing polyester resins in the powder coating formulation.

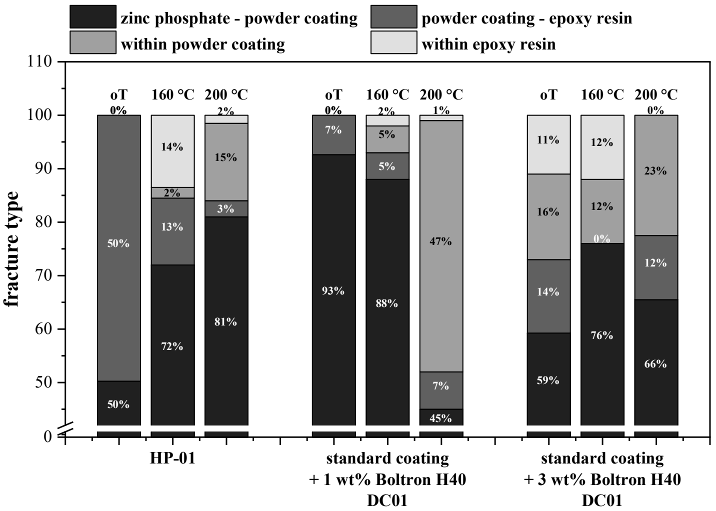

Figure 7: Fracture percentage of the failure modes occurring in the lap shear test using different dendrimer amounts compared to the standard powder coating and influence of post-curing (3 h / 160 °C or 1 h / 200 °C) before the lap shear test.

When considering the influences of the use of dendrimers in the preferred powder coating HP-01 have on the interlayer strength, almost no effect on the resulting shear strength was found in comparison with the standard formulation. Thus, the addition of the expensive dendrimer to the coating is not effective in this case. In contrast, annealed test specimens showed a change of the occurring failure types in the shear test (Figure 7). In the absence of dendrimers in most cases the failure happened between P-PU and PU-D. By using dendrimers the failure percentage between PU-D decreased and therefore the amount of the interfacial failure between P-PU increased. However, also other failure types occurred beside those. The influence of the dendrimers has to be studied more intensively later on.

2.3. *Hybrid pultrusion*

The preferred results from the RTM process then were adapted to the continuous pultrusion process at IWU (Figure 8). The individual fabrication steps can be described as follows: Semi-finished fiber products (1) are pulled from bobbins by alternately moving pulling devices (4) and pass through a resin bath (2). Afterwards, the impregnated fibers are pulled through a heated die (3), in which the liquid thermoset plastic cures completely within seconds. A saw (5) cuts the profiles to the desired length [6].

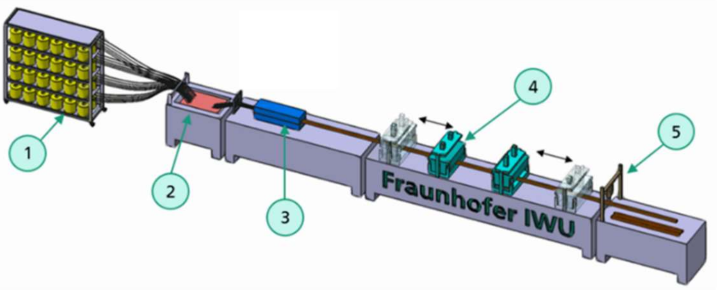

Figure 8: Pultrusion line

With a special manufactured tool and steel sheets, that were powder coated with HP-01, it was possible to produce a side sill demonstrator (Figure 10) via the continuous pultrusion process successfully. The dimension of the 3 chamber-hollow hybrid-demonstrator has a wall thickness of 4 mm, a cross-section area of 3.567 mm^2 and two stands with a wall thickness of 6 mm in the inner for the necessary stiffness (Figure 9 (a)). To further improve the crash properties, a powder-coated steel sheet/coil of 90 mm length was integrated on top of the demonstrator. The pultrusion process was performed with various temperature zones from 150 to 190 °C and a processing speed of 100 mm/min. A simulation (finite element method – FEM) with the program "ANSYS Workbench 2019 R2" and the special program add-on "ANSYS Composites Cure Simulation" of the pultrusion process was realized to identify the best process parameters. The starting point for a valid calculation was formed by a large number of basic trials and corresponding measurements of the process parameters (e.g. temperature, production speed). These pultrusion trials were carried out on strip samples with a profile cross-section of 50x4 mm². In addition to different sheet metal thicknesses (0.5; 1.0; 1.5 and 2.0 mm), the influence of the sheet position in the thickness direction was also investigated. The results obtained in this way were incorporated in the construction of a simulation model. Thus, it was possible to calculate not only the different temperatures of the materials but also the degree of cure of the epoxy resin. Coupled with a structural mechanical simulation, it was also possible to determine the distortion due to the different material properties (Figure 9 (b)). This knowledge gained was used for the design of the demonstrator component, so that, for example, the layer structure of the fiber composite profile including the sheet metal insert could be optimized to minimize the resulting distortion. However, the focus of the simulation was on determining relevant process parameters in order to minimize time-consuming and cost-intensive trials. Thus, after only three trials with minor adjustments to the process parameters, a qualitatively appealing demonstrator could be realized.

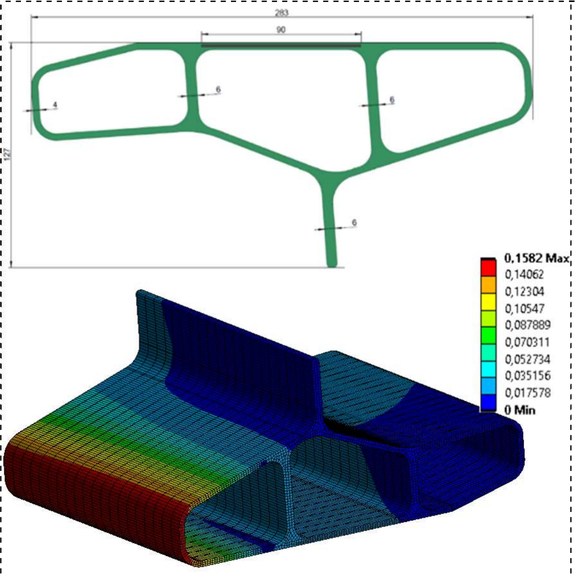

Figure 9:
(a) Cross-section geometry of the side sill demonstrator with a powder coated steel sheet on top.
(b) Simulation result on distortion [mm] due to different shrinkage behavior for the demonstrator

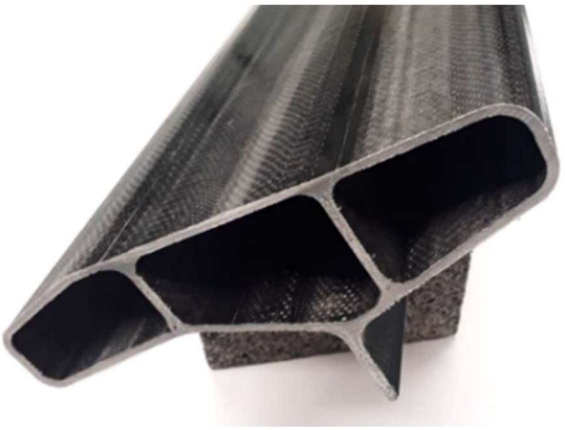

Figure 10: Finished side sill demonstrator manufactured via pultrusion process**.**

3. Summary

Studies have shown that it is possible to generate material bonded hybrids based on powder coated steel and glass fiber reinforced epoxy resin with a very high bond strength via the RTM process as well as continuous pultrusion by manufacturing a demonstrator for a side sill. The variation of the OH number of the containing polyester resins in the powder coating showed an influence on the resulting adhesive strength. In contrast, the addition of dendrimers to the powder coating formulation showed no clear effect on the bond strength of the manufactured hybrid materials so far.

4. Acknowledgement

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