Article

Mechanical Properties of Thermoplastic-Based Hybrid Laminates with Regard to Layer Structure and Metal Volume Content

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Abstract: Multi layered lightweight material compounds such as hybrid laminates are composed of different layers of materials like metals and unidirectional fibre-reinforced plastics and offer high specific strength. They can be individually tailored for applications like outer cover panels for aircraft and vehicles. Many characteristics especially layer structure, volume contents of the embedded materials as well as layer surface adhesion determine the performance of a hybrid laminate. In this study, the influence of layer structure and metal volume content are evaluated with regard to the mechanical properties of the recyclable hybrid laminate CAPAAL (carbon fibre-reinforced plastics/aluminium foil laminate), which consists of the aluminium alloy AA6082 and a graded structure of glass and carbon fibre-reinforced polyamide 6. Hybrid laminates with different ratios of the fibre-reinforced plastic and numbers of aluminium layers were manufactured by thermal pressing. The consolidation quality of in total four laminate structure variations, including 2/1 and 3/2 metal-to-fibre-reinforced plastic layer structures with fibre orientation variation, were investigated by light microscopy through cross-sections and further on computed tomography. For determination and evaluation of the mechanical properties metrologically instrumented quasi-static tensile and three-point bending tests, as well as tension-tension fatigue tests for the establishment of S-N curves were performed. The results were correlated to the microstructural observations, revealing significant influence by the consolidation quality. The layer structure proved to have a proportional impact on the increase of quasi-static tensile and flexure strength with a decrease in metal volume content. Orienting some of the fibre-reinforced plastic layers in ±45° leads to a more evenly distributed fibre alignment, which results in a higher consolidation quality and less anisotropic bending properties. Fatigue results showed a more complex behaviour where not only the metal volume content seems to determine the fatigue loading capability, but also the number of metal-fibre-reinforced plastic interfaces, hinting at the importance of stress distribution between layers and its longevity over fatigue life.

Keywords: fibre-metal laminate; thermoplastic; polyamide 6; CFRP; AA6082; metal volume content; layer structure; surface energy; tension/tension fatigue loading; three-point bending

1. Introduction

The hybrid laminate CAPAAL (carbon fibre-reinforced polyamide/aluminium foil laminate) [1] is a build-up of metals and thermoplastic-based carbon fibre-reinforced plastics (FRPs), which is an attractive alternative to those based on thermosetting matrix systems like CARALL [2] (carbon-reinforced aluminium laminate), because they can easily be shaped by thermoforming [3] and consolidated in
shorter cycle times. A significant advantage of the layered design is the demonstrably better fatigue behaviour than pure aluminium through the fibre bridging effect [2] and increased damage resistance compared to FRP. The composition of a hybrid laminate is described by the number of metal (N\text{Metal}) and FRP layers (N\text{FRP}) as “N\text{Metal}/N\text{FRP}”. Typically, hybrid laminates are constructed in a multiple changing arrangement of metal and FRP layers with the layer structure description of 2/1, 3/2, or 4/3, etc.

An essential aspect of CAPAAL and other fibre-metal laminates (FML) is the surface treatment of the metal component since it impacts the interface strength between the metal component and FRP. Regarding the criteria, such as suitability for large series production [4] and interlaminar shear strength [5], mechanical blasting has proven to be appropriate in previous work. It has been found that mechanical adhesion is not alone responsible for high shear strength due to increased roughness. In [5] different strong chemical bonds of the surface states are applied. Narbon et al. show that roughness differences caused by mechanical treatment (grinding) or chemical etching (HCl) do not have a strong influence on the surface energy of AA6082, but they always measured an increase in comparison to the untreated sheet [6]. The aluminium surface treatment possesses a clear influence on the quasi-static tensile properties of the laminate, but significantly more in fatigue tests where anodization and phosphating of the aluminium surface led to the highest fatigue lifetimes [7].

Reyes et al. were able to determine a linear increase in strength with increasing glass fibre-reinforced plastic (GFRP) content in tensile tests independent of the layer structure 2/1, 3/2, or 4/3. The Young’s modulus, on the other hand, decreased linearly [8]. The relationships are following the rule of mixture and their values depend on the properties of the base materials, which was also shown by Sun et al. [9] for titanium/carbon-fibre epoxy laminates. It can be expected that the bending behaviour of the laminates will behave significantly different since the distance of the FRP from the neutral fibre has an influence. Bellin et al. [10,11] showed that for inverted layer structures 1/2 and 2/3 of CARALL the flexural strength, strain, and modulus are decreased for latter, if the fractions of laminate partners and overall thickness of the laminate are held constant. All investigated specimens showed composite tensile failure with delamination occurring in-between the fibre-metal interface in specimens without additional adhesive layers. The changes in mechanical properties were confirmed by Cortés et al. [12], which investigated a 2/1 layer structure made of magnesium alloy and thermoplastic GFRP in terms of varying FRP content, and also by Wu et al. [13] in investigations from 2/1 with up to an 8/7 layer structures of laminates containing magnesium alloys and carbon fibre-reinforced plastic (CFRP).

Fatigue bending of CAPAAL with layer structures 2/1 und 3/2 under elastic loading of the laminate, which is below the fatigue limit, withstands one million load cycles without any measurable damage in the individual subcomponents. Higher loads are leading to crack propagation inside the aluminium top sheet, whereas only the highest loads applied induced additional damage occurrence inside the thermoplastic matrix [14].

To extend previous research on the influence of the layer structure and metal volume content on the mechanical properties of hybrid laminates like CAPAAL more investigations, using expanded metrological instrumentation for a detailed understanding while not relying on just one specific loading case, need to be conducted. Thus, this study provides a comprehensive overview about the impact of the layer structure and metal volume content on the mechanical properties under bending and fatigue tension/tension loading for a hybrid laminate based on aluminium and carbon fibre-reinforced polyamide 6, containing a glass fibre-reinforced graded layer. The aluminium surface treatment and thereby implied interlaminar interface changes between the aluminium surface conditions and the FRP are considered as important factors as well. The aim of this study is gathering a wide impression about the structural differences and its importance regarding load capacity and laminate integrity for high and lasting mechanical performance, to establish fundamental knowledge, which is needed for, e.g., design of components.
2. Materials and Methods

2.1. Design and Manufacturing

The hybrid laminate based on CAPAAL [1] is composed of a surface-treated aluminium AA6082-T4 (0.5 mm thickness) and a graded structure of unidirectional glass- and carbon fibre-reinforced polyamide 6. The aluminium alloy AA682 was mechanically blasted as described [15]. Additionally, wetting tests were performed on other surface treatments such as untreated, anodized, phosphate, adhesion promoted, and etched as also described in [5,15]. The FRP layer of the investigated CAPAAL is a build-up of multiple layers of CePreg (Cetex Institut gGmbH, Chemnitz, Germany), a preconsolidated thermoplastic prepreg with a calculated fibre volume fraction of 0.5. The carbon fibre-reinforced polyamide 6 (CFR-PA6) consists of two 40 µm thick PA6 foils (mf-Folien GmbH, Kempten, Germany) and the carbon fibres ZOLTEK PX35 50K (ZOLTEK Corporation, Bridgeton, MO, USA), which are continuously processed into 3000 mm wide tapes as described in [3]. The glass fibre-reinforced polyamide 6 (GFR-PA6) contains the fibre TURov 4588 (Nippon Electric Glass Co. Ltd., Tokyo, Japan).

Between the interface of aluminium and the high strength fibre-reinforced CFR-PA6 an additional foil of polyamide 6 (PA6) and a glass fibre-reinforced PA6 are added to compensate the mismatch in the coefficient of thermal expansion. The individual layers were combined for a 2/1 and 3/2 layer structure as well as with 4 and 8 layers of FRP, Figure 1.

![Material composition of the hybrid laminates](image)

**Figure 1.** Material composition of the hybrid laminates: (a) 2/1 (GC) with four fibre-reinforced plastic (FRP) layers, (b,c) 2/1 (GGCC) with eight FRP layers but different glass FRP (GFR-PA6 orientations (±45° for 2/1 (G’G’CC)), and (d) 3/2 with 2× four FRP layers between aluminium sheets.

The orientation of the unidirectional FRP semi-finished products is specified according to their fibre orientation to the rolling direction of the aluminium alloy. Often this is done parallel to each other. In the case of one specific investigated type 2/1, the GFR-PA6 tapes are oriented in ±45° (Figure 1c). Additionally to a short notation, a designation of the laminates in the style of the classic laminate inscription is recommended, Table 1. This requires a suitable description according to multidirectional compositions. Moreover, a description of the laminates based on the classic laminate designation after [16] is proposed, which takes into account the kind, number, and orientation of the used FRP as well as the included metal and symmetry of the hybrid laminate.

The metal volume content (MVC) additionally describes the volumetric amount of the introduced metal component using the number of layers (N) and layer thickness of the metal \( t_{Metal} \) and hybrid laminate \( t_{HL} \). The following equation can be used for the calculation:

\[
MVC = \frac{N \cdot t_{Metal}}{t_{HL}}
\]
The FRP semi-finished products have a preconsolidated character (Figure 2) and are fully consolidated by the pressing process. As PA6 tends to absorb water, the tapes are dried in an oven at 80 °C for one hour before processing.

**Table 1.** Hybrid laminates of AA6082 and glass/carbon fibre-reinforced polyamide 6 with different layer structures and metal volume contents.

| Short Notation | Composition [Orientation FRP] | Description                  | Thickness mm | MVC % |
|----------------|-------------------------------|------------------------------|--------------|-------|
| 2/1 (GC)       | Al/GCCG/Al                    | [Al/GCCG/Al]                 | 1.64         | 61    |
| 2/1 (GGCC)     | Al/GGCCG/Al                   | [Al/GGCCG/Al]                | 2.28         | 44    |
| 2/1 (G’G’CC)   | Al/GGCCG/Al                   | [Al/GGCCG/Al]                | 2.28         | 44    |
| 3/2 (GC)       | Al/GCC/Al                     | [Al/GCC/Al]                  | 2.78         | 54    |

**Figure 2.** Cross section of the semi-finished FRP products: (a) glass and (b) carbon fibre-reinforced polyamide 6.

The consolidation of the materials with a size of 260 × 260 mm² takes place in dipping edged tool, Figure 3a, with a laboratory platen press Collin PM300 (COLLIN Lab & Pilot Solutions GmbH, Maintenbeth, Germany). A maximum temperature of 295 °C is held for 24 min with a pressure of 1.0 MPa in the first 10 min and 1.5 MPa during the last consolidation process, Figure 3b. After the pressing they were cut in specimens by a water jet.

**Figure 3.** (a) Dipping edge tool and (b) cycle of thermal pressing for the hybrid laminates.
2.2. Characterization Methods

As shown in [5], a high value in interlaminar shear strength can be achieved for better mechanical properties despite low surface roughness, which can be attributed to chemical adhesion. For this purpose, the surface energy and wetting behaviour was investigated to establish a basis for optimal laminate properties for the investigations regarding layer structure and metal volume content. The determination of the surface energy was performed by contact angle measurements of three different test liquids (thiodiglycol, ethylene glycol, and diiodomethane) on the contact angle meter OCA20 Dataphysics OCA20 (Dataphysics Instruments GmbH, Filderstadt, Germany). Five drops per surface were applied and optically analysed in the lying position. The calculation of free surface energy was performed according to Owens, Wendt, Rabel, and Kälble (OWRK) according DIN 55660-1/2:2011-12. To reproduce the wetting behaviour with the actual matrix material, PA6 granules with a quantity of 10 mg were melted onto the treated surfaces in a laboratory furnace at 240 °C and then cooled. The resulting droplets were recorded with a stereo microscope.

For microstructural analysis cross-sections of the different structured hybrid laminates according to consolidation quality were observed by light microscopy with a Keyence VK X200 (Keyence Deutschland GmbH, Neu-Isenburg, Germany) laser scanning microscope. Also, small specimen (3 mm width, 5 mm length) were examined via computed tomography (CT) with a Nikon XT H 160 (Nikon Metrology GmbH, Alzenau, Germany) to evaluate the volume distribution. The Nikon XT H 160 contains an X-ray tube with a maximum voltage of 160 kV, maximum power of 60 W, and a 1008² pixel detector. The CT acquisition parameters were optimized with regard to a grey value distribution suitable for multi-material analysis and high image quality, resulting in the following parameters: tube voltage 60 kV, tube current 117 A, exposure time 1000 ms, 1583 projections, 8-fold superimposition, and 6 μm voxel size. For the image analysis, the software VG Studio Max 2.2 from Volume Graphics (Volume Graphics GmbH, Heidelberg, Germany) was used.

The quasi-static properties were investigated by tensile tests and bending tests. The tensile behaviour was investigated in accordance to DIN EN 527-5 with water jet cut specimens (type A with length of 160 mm) by a servo-hydraulic testing system EHF-EV50 (Shimadzu Europe GmbH, Duisburg, Germany) with a maximum force \( F_{\text{max}} = \pm 50 \text{kN} \), Figure 4a. The gauge length is 80 mm to reduce the technologically monitored gauge length. The specimen edges were polished to reduce stress concentrations due to notches. In the clamping area, on both sides additional AA6082 aluminium tabs were adhered with epoxy glue to reduce stress concentrations at the transitional area of clamping and testing area. The three-point bending tests were performed after DIN EN ISO 14125 on a universal material testing machine Zwick 100 kN (Zwick/Roell GmbH & Co. KG, Ulm, Germany), Figure 4b. The size of the water jet cut specimens related to class III with a dimension of \( 80 \times 15 \text{ mm}^2 \) and a thickness that depends on the composition of the laminate (Table 1). The distance between the supporting pins is 40 mm. The bending speed is 2 mm/min. A minimum of three specimens for each laminate was tested.

For fatigue behaviour investigations constant amplitude tests (CAT) were conducted using the same specimen geometry and preparation and testing system as for quasi-static tensile tests. Sinusoidal load-time functions at a stress ratio \( R = 0.1 \) (tension/tension loading) and testing frequency \( f = 10 \text{ Hz} \) were applied under ambient temperature according to ISO 13003. Investigations focused on low (LCF) and high cycle fatigue (HCF), for which the maximum stress levels were determined with the help of the quasi-static results.

The technological instrumentation setup successfully used to investigate the influence of aluminium surface treatment on the mechanical properties of thermoplastic-based hybrid laminates in [15] was applied partly to detect material changes in order to generate comparable results and complement the results. The setup contains a Limess Q400 digital image correlation (DIC) system (Limess Messtechnik und Software GmbH, Krefeld, Germany), which was used as a video extensometer for strain analysis of the front aluminium sheet (precision lens, 28 mm focal length) during tensile testing. The specimens were spray-painted with a speckle pattern for DIC analysis. A Shimadzu TCK 1 LH \( (\Delta l = 25 \text{ mm}, \Delta l = \pm 1 \text{ mm}) \) extensometer recorded the strain during fatigue tests. Due to occurring
development of aluminium cracks and aluminium delamination from the FRP during fatigue testing, which led to extensometer slippage resulting in recorded data deviations and less reliability, additionally the machine piston displacement \( s \) was recorded by a linear variable differential transformer (LVDT) for stress-displacement hysteresis values.

![Image](https://via.placeholder.com/150)

**Figure 4.** (a) Instrumented tensile and fatigue testing setup containing the servo-hydraulic testing system, digital image correlation camera perpendicular to the specimen (face view), and tactile extensometer; (b) Three-point bending testing setup according DIN EN ISO 14125.

3. Results

3.1. Surface

The results of the surface energy of the different aluminium surface treatments are displayed in Figure 5, where for the PA6 foil the used values for dispersive and polar surface energy were extracted from [17]. The phosphated aluminium alloy reaches the highest value of 46.4 mN/m. This is an increase of 42.4% compared to the untreated sheet metal (26.7 mN/m). The lowest value was measured on the mechanical blasted surface (21.3 mN/m) despite the highest interlaminar shear strength determined in double notch shear tests as published in previous work [5,15]. This implies that the main adhesion mechanism of mechanical blasting is the mechanical interlocking of the rough surface and the PA6 matrix. The other investigated surface treatments contributed to an increase in surface energy, with different values of the polar component. The highest value of 3.2 mN/m was measured on the anodized surface. The oxide layer increases the polar character, which is negative for the wetting behaviour, because the PA6 has a high fraction of the polar component with 11.5 mN/m. This can be recognised by the largest wetting angle, Figure 6c. The lowest polar fraction was determined on a sodium hydroxide etched surface (0.2 mN/m). This treatment leads to a reduction of the natural oxide layer compared to the untreated surface. All other surfaces have a higher polar component than the untreated surface. Remarkable is the low polar fraction of the surface with the applied adhesion promoter, which is based on a copolyamide. A significantly higher fraction comparable with the PA6 film was expected.

From the surface energy values alone, no conclusion can be made about the direct wetting behaviour of the PA6. However, the wetting test tends to show a worse wettabillity on the mechanically blasted (Figure 6b) surface as well as on the anodized (Figure 6c) surfaces. Phosphating (Figure 6d) and etching (Figure 6f) seems to produce a slight improvement compared to the untreated surface (Figure 6a). In this simple wetting test, the molten PA6 has the lowest wetting angle on the adhesion promoted copolyamide surface compared to all the other surfaces (Figure 6e).
Interestingly, mechanically blasting does not exhibit the best surface energy conditions or wettability for PA6 but leads to the highest interlaminar shear strength of all for the interface between the aluminium sheet and the copolyamide. A significantly higher fraction comparable with the PA6 film was expected.

A slight improvement compared to the untreated surface (Figure 6a). In this simple wetting test, the molten PA6 has the lowest wetting angle on the adhesion promoted copolyamide surface compared to all the other surfaces. The low est polar fraction was determined on a sodium hydroxide etched surface (0.2 mN/m). This treatment leads to a reduction of the natural oxide layer compared to the untreated sheet metal (26.7 mN/m). The lowest value was measured on the mechanical blasted surface (21.3 mN/m) despite the highest interlaminar shear strength determined in double notch shear tests as published in 2020.

Various surface treatments were investigated in order to improve the wettability of the PA6. These treatments are the mechanical blasting, the anodizing, phosphating, etching, the application of an adhesion promoter and the etching with a sodium hydroxide solution. The surface energy of the different treatments are shown in Figure 5.

Figure 5. Surface energy of treated aluminium alloy AA6082 and PA6 foil with dispersive and polar component. Absolute value of the polar component of surface energy is highlighted through digits.

Figure 6. Wetting behaviour between (a) untreated, (b) mechanical blasted, (c) anodized, (d) phosphated, (e) adhesion promoted, and (f) etched AA6082 and melted PA6.

These results give an impression about how the PA6 wettability can be influenced by surface treatments. Interestingly, mechanically blasting does not exhibit the best surface energy conditions or wettability for PA6 but leads to the highest interlaminar shear strength of all for the interface between the aluminium sheet and the PA6. Moreover, it leads to the highest tensile strength when applied to the hybrid laminate with 2/1 (GC) layer structure [5,15]. Mechanical interlocking therefore is assumed to be the most important adhesion mechanism since superior wetting and high surface energy seem to have only a supporting effect. With regard to the investigations in this study, containing tensile tests and bending tests, as well as fatigue tests with tension/tension loading, mechanically blasted aluminium surface treatment was chosen for the investigations of the influence of layer structure and metal volume content, extending on previous findings [5,7,15] and securing comparability between the results.

3.2. Consolidation

The cross sections of the consolidated hybrid laminates show a good quality of the consolidation without pores or accumulation of matrix areas, Figure 7. The fibres have a dense and homogeneous packing. For unidirectional orientation of all fibres in Figure 7a,b,d a wavy orientation of the CFR-PA6 layer can be observed. For the ±45° orientation of the grading GFR-PA6 layer, this effect can be prevented and contributes to stabilization of the CFR-PA6 layer, Figure 7c. Uneven distribution of the fibres in the cross section, especially for the 3/2 (GC) layer structure observed, leads to a partially uneven thickness of the FRP component. The reason for this effect is believed to be the waviness caused by the mechanical blasting, which in the molten state causes local displacement processes in the consolidation process.
While for latter deviating FRP content between the upper and lower layer visible. These are attributed to the smoother flow properties of the molten CFR-PA6 due to the straight layer separation from the aluminium layers and the FRP layer (Figure 9b). These seem to be caused by the roughness of the mechanically blasted aluminium surface and the GFR-PA6 layer in hybrid laminates, (Figure 9a). A possible reason could be superior flow properties of the larger amount of molten FRP, leading to a floating of the interlaminar aluminium layer in-between the molten PA6 during the consolidation process as well as the waviness of the mechanically blasted aluminium sheets. 2/1 (G′G′CC) with its ±45° oriented glass fibres achieved not just areal, but along the investigated volume the most homogeneous fibre distribution. But for this layer structure also a higher metal volume content is attributed to the waviness caused by the mechanical blasting, which in the molten state causes the exceeding of the aluminium yield stress, as mentioned in [15]. Whilst the structures 2/1 (GC) with 61% MVC exceed the aluminium yield stress, as mentioned in [15]. Whilst the structures 2/1 (GC) with 61% MVC; 2/1 (GGCC) with 44% MVC; 3/2 (GC) with 54% MVC.

Additionally, to the 2D-limited light microscope images 3D CT images were acquired to check the consolidation quality and FRP distribution along a volume. Figure 8 shows CT images of partial volumes of the investigated layer structures as 3D volumes and cross-sectional images to visualize the structure and fibre distribution. The corresponding specimens were taken from the middle area of the thermally pressed laminates. Based on the light microscopy images comparable laminate structures can be achieved between the individual joining partners in the layer structures 2/1 (GC) and 3/2 (GC), while for latter deviating FRP content between the upper and lower layer visible. These are attributed to the floating of the interlaminar aluminium layer in-between the molten PA6 during the consolidation process as well as the waviness of the mechanically blasted aluminium sheets. 2/1 (G′G′CC) with its ±45° oriented glass fibres achieved not just areal, but along the investigated volume the most homogeneous fibre distribution. But for this layer structure also a higher metal volume content is visible than in 2/1 (GGCC), despite having the same number of FRP and metal layers. This is attributed to the smoother flow properties of the molten CFR-PA6 due to the straight layer separation from the GFR-PA6, resulting in the molten CFR-PA6 flowing out of the laminate due to the pressure during consolidation. Since this reduces the amount of carbon fibres in the FRP, this results in a significant change in mechanical properties and needs to be considered for result interpretation.

Figure 7. Cross section of hybrid laminates: (a) 2/1 (GC), (b) 2/1 (GGCC), (c) 2/1 (G′G′CC), and (d) 3/2 (GC).

Figure 8. 3D computed tomography images including cross-sectional images of the fibre distribution of hybrid laminates with mechanically blasted aluminium surface treatment and different layer structures/metal volume content (MVC). 2/1 (GC) with 61% MVC; 2/1 (GGCC) with 44% MVC; 2/1 (G′G′CC) with 44% MVC; 3/2 (GC) with 54% MVC.
Furthermore, CT cross sections of the interface between aluminium and GFR-PA6 (red plane in Figure 9a) revealed that 2/1 (GC) and 3/2 (GC) have locally isolated pores as well as pore accumulations between the aluminium layers and the FRP layer (Figure 9b). These seem to be caused by the roughness of the mechanically blasted aluminium surface, leading to trapping of air. The same pores were observed by Park et al. [18] for a similar structured GLARE (glass laminate aluminium reinforced epoxy) laminate, which was consolidated within an autoclave cure cycle. In view of the pore-free appearance of 2/1 (GGCC) and 2/1 (G′G′CC), the FRP volume also seems to be of importance.

**Figure 9.** Computed tomography cross-section images showing pore occurrence between the mechanically blasted aluminium surface and the GFR-PA6 layer in hybrid laminates, (a) red plane visualizes displayed cross-section plane; (b) cross-section image for the layer structure/metal volume content (MVC) 2/1 (GC) with 61% and 3/2 (GC) with 54%.

A possible reason could be superior flow properties of the larger amount of molten FRP, leading to a flushing out of pores during consolidation. Either way, the effect of such pores needs to be considered when evaluating the mechanical properties, especially regarding fatigue loads, where interface health is a key property for stress distribution between the layers and therefore impacts lifetime. Since these CT images were acquired from specimens taken out of the middle sections of the laminates the pore distribution results are considered representative for the laminate.

### 3.3. Quasistatic Properties

To evaluate the influence of the laminate structure and MVC on the mechanical properties under quasi-static load tensile tests were executed, which stress-displacement and stress-strain (measured via the DIC virtual gauge length) curves are visible in Figure 10. All curves for each investigated laminate structure show an overall similar development until initial failure. The kink visible at each start of the curves is assumed to be representing the exceeding of the aluminium yield stress, as mentioned in [15]. Whilst the structures 2/1 (GC) with 61% MVC and 3/2 (GC) with 54% MVC show immediately total failure, the structures 2/1 (GGCC) and 2/1 (G′G′CC) with 44% MVC show all a stepwise failure, with an initial drop to around 1/3 of the ultimate tensile strength ($\sigma_{\text{UTS}}$). After this drop the aluminium sheets and majority of fibres failed. This is attributed to the higher effective stress on the aluminium at lower MVC. Due to the lower MVC, the fibre structure carried more load and gradually failed after the aluminium, which in this case was noticeable by continuous acoustic perception of breaking fibres. With a high MVC, on the other hand, there seemed to be a more homogeneous distribution of the effective stress, which led to a failure of the laminate components that was close in time and resulted in direct total failure.

Also, the metal volume content is decisive regarding laminate stiffness and ultimate tensile strength, but also the orientation of the FRP. 2/1 (GGCC) with the highest MVC shows the highest stiffness and ultimate tensile strength out of all tested layer structures, while the same number of layers with GFR-PA6 oriented in $\pm45^\circ$ led to an immense reduction in both. The reason is that the $\pm45^\circ$ oriented GFR-PA6 layers load bearing is ensured through the transfer of stress over the adjacent PA6 interface only and results in a worse overall stress distribution in the whole laminate.
For a direct comparison of the quasi-static tensile properties Figure 11 shows the arithmetic mean values for $\sigma_{UTS}$, the corresponding displacement of the machine piston ($s$) and total strain measured via the DIC virtual gauge length ($\varepsilon_{LVGL}$) on the aluminium surface. 2/1 (GGCC) with 44% MVC shows an around 40% higher $\sigma_{UTS}$ (860 MPa against 620 MPa) compared to the layer structure 2/1 (GC) with 61% MVC, while having a 28% lower MVC seen relative to 2/1 (GC). 2/1 (G’G’CC) (GC), because of the aforementioned ±45° oriented GFR-PA6 layers. 3/2 (GC) with 54% MVC ranks with 660 MPa 6% above 2/1 GC, despite the relatively seen 11% lower MVC, supporting the earlier statement that not just MVC determines the resulting $\sigma_{UTS}$, but also the FRP orientation and aluminium layer number. The partially large scattering of the $\sigma_{UTS}$ results is due to the inhomogeneity of the fibre distribution inside the FRP component, which occurs during the laminate consolidation. The displacement on the other hand shows an around 30% reduced $\sigma_{UTS}$ (428 MPa) compared to 2/1 of the machine piston shows deviating results, which are possibly due to different interactions between the epoxy applied aluminium tabs and strain deformation behaviour based on the MVC. However, the values of the recorded, overall achievable total strain $\varepsilon_{LVGL}$ before failure show a plausible ranking, where with an increase in metal volume content the achievable total strain increases. Only the 2/1 (G’G’CC) ±45° oriented GFR-PA6 layers shows lower strain values, which again is due to the higher effective stress on the CFR-PA6 layer resulting in an overall earlier failure and therefore the lower total strain.

Figure 11. Arithmetic tensile strength and strain values of hybrid laminates with aluminium alloy AA6082-T4 and a graded structure of glass and carbon fibre-reinforced PA6 for different layer structures and metal volume contents.
Besides the tensile tests also three-point bending tests were performed (see Figure 12a). With the FRP oriented along the specimen length (0°) the layer structure 2/1 (GC) reaches a flexural strength (σ_{flex}) of 420 MPa. The highest flexural strength (σ_{flex}) of 787 MPa can be achieved by the layer structure 2/1 (GGCC). By positioning the GFR-PA6 in ±45° orientation (2/1 (G′G′CC)) σ_{flex} is reduced to 428 MPa similar to the results from the tensile tests. Therefore, the orientation of the GFR-PA6 layers contributes significantly to the flexural strength of the laminate with comparable laminate thickness and MVC. While the layer structure 3/2 (GC) does not improve σ_{UTS} significantly, σ_{flex} is near the one of 2/1 (GGCC). Since the FRP volume is the same, this was expected.

The flexural strength with the FRP oriented perpendicular to the specimen length (90°) is significantly lower. The layer structures with unidirectional orientation of all fibres have a comparable value of σ_{flex} between 225 and to 260 MPa. Only the layer structure 2/1 (G′G′CC) reached a higher value of σ_{flex} with 300 MPa due to the GFR-PA6 oriented in ±45°, illustrating the better applicability for bending in terms of more quasi isotropic behaviour. Further it is the only laminate showing intact aluminium failure (see Figure 12b). The MVC did not seem to impact the σ_{flex} when the FRP orientation is 90°, since they all show comparable results.

The flexural strain (ε_{flex}) of the laminates with the FRP oriented along the specimen length (0°) was measured for the layer structure 2/1 (GGCC) as 5.8%. The change of the GFR-PA6 orientation to ±45° (2/1 (G′G′CC)) has only a minor influence. However, an increase in MVC as for 3/2 (GC) results in a significantly lower ε_{flex} value of 3.1%. For the specimens tested in 90° FRP orientation ε_{flex} decreases for the 2/1 (GGCC) layer structure and increases for 3/2 (GC). While the first exhibited an earlier metal failure due to the lower stiffness, the latter seems to extend its strain ability due to the fibres not limiting the deformation of the overall laminate. 2/1 (G′G′CC) reaches the highest value of flexural strain 9.1% for this case, which specimens show no macroscopic failure in aluminium and FRP (see Figure 12b). The specimens of the other layer structures and metal volume contents show cracks in the aluminium and particularly fractures in the FRP layer on the lower side where tensile stress is present.

3.4. Cyclic Properties

The quasi-static tensile properties show that the layer structure and MVC have a significant influence on the performance. When comparing 2/1 (GC) and 2/1 (GGCC see Figure 11) it can be derived that a larger proportion of higher-strength CFR-PA6 contributes significantly to increasing σ_{UTS}, which can partially be transferred to the fatigue behaviour, visualized through the S-N relationship.
in Figure 13. In general, the results show that with decreasing MVC the fatigue life achieved along the investigated maximum stress levels shows a flatter course. This is attributed to the more FRP dominated load absorption of the laminate. A 28% lower MVC from 2/1 (GGCC), relatively compared to 2/1 (GC), leads to a more FRP dominated load absorption and at least 25% higher fatigue strength over all investigated maximum stress levels. The layer structures 2/1 (GGCC) and 3/2 (GC) show, despite the relatively seen 23% higher MVC in latter, similar fatigue strengths, with 3/2 (GC) having an approximately 5–10% lower fatigue strength over all investigated maximum stress levels. This was not observed in the quasi-static tensile properties, where it achieved 23% lower σ<sub>UTS</sub>. Therefore, the number of aluminium layers influences the fatigue life, which is assumed to be due to the higher number of FRP metal interfaces, enabling a superior stress distribution over all laminate partners. The layer structure 2/1 (G′G′CC) on the other hand confirms that at tensile load the consequence of GFR-PA6 layer orientation change to ±45° is degradation of mechanical properties. Nevertheless, it becomes apparent that the around 30% reduction in σ<sub>UTS</sub> compared to 2/1 (GC) is not visible for fatigue load. Instead it performs in a way nearly identical to the layer structure 2/1 (GC) regarding fatigue life. This indicates that the laminate undergoes a change in aluminium stiffness and interfacial residual stress behaviour under fatigue load as observed in similar manner in [15], which in contrast to the quasi-static behaviour may prolong fatigue life.

![Figure 13. S-N relationship for hybrid laminates with different layer structures and metal volume contents.](image)

4. Conclusions and Outlook

For thermoplastic-based hybrid laminates made of aluminium alloy AA6082-T4 and unidirectional GFR-PA6 and CFR-PA6 the following conclusion can be drawn regarding the influence of layer structure and metal volume content on the mechanical properties:

The preliminary investigations regarding surface and wetting behaviour of the aluminium sheet for identification of an appropriate treatment for different layer structures show a contradictory result. Increasing the surface energy of the aluminium surface does not necessarily improve the wettability at the same time. Despite the mechanically blasted surface having the lowest free surface energy of all the surface treatments and a worse wetting behaviour than the untreated sheet, it offers the best mechanical properties regarding quasi-static load, which makes it the most suitable for the investigations in this study.

With the thermal pressing consolidation process used in this study dense FRP layers in 2/1 and 3/2 layer structure are successfully produced. CT images showed that the investigated layer structures 2/1 and 3/2 with lower metal volume content appears to have small pores in the interface area, assumed to be due to the mechanically blasted aluminium surface. The wavy and uneven distribution of the FRP layers can be improved by a ±45° orientation of the grading GFR-PA6 layer, resulting in a smooth, uniform distribution with a more continuous laminate thickness.
The influence of layer structure and metal volume content on the mechanical properties depends on the loading case. For quasi-static tensile load, a lower metal volume content with unidirectional fibre alignment in load direction is favourable. A ±45° orientation of the grading GFR-PA6 layer results in the worst performance of the tested variants, since tensile load is less bearable due to the higher effective stress on the, in load direction aligned, CFR-PA6 fibres and the aluminium sheets. However, the ±45°-oriented GFR-PA6 layer results in a more quasi isotropic behaviour in case of quasi-static three-point bending. Decreasing the metal volume content leads also for three-point-bending to higher strength, but only as long as the fibres are aligned to act against the bending force. Fatigue results showed that not just a decrease in metal volume content can achieve higher fatigue strengths, but also a suitable increase of metal volume content through an added aluminium layer, which enables a superior stress distribution over all laminate partners.

In future investigations the influence of existing pores in the interface between the mechanically blasted aluminium sheet and the PA6 layer on the mechanical properties needs a thorough analysis, which can be carried out with in situ CT analysis of specimens alongside mechanical testing. With this method the damage initiation, starting usually at the aluminium sheet and metal-FRP interface, can be analysed intralaminar and interlaminar with high resolution. Looking at the promising consolidation results when GFR-PA6 layers are oriented in ±45° direction regarding FRP distribution and laminate thickness, but worse mechanical properties, more layer structures with emphasis on the fibre orientation need to be investigated. Further the damage mechanisms and development during fatigue load is a key aspect regarding knowledge for applicability of such a hybrid laminate in the industry, which needs to be considered for future studies.

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