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# Advanced molds and methods for the fundamental analysis of process induced interface bonding properties of hybrid, thermoplastic composites

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**Abstract**

Hybrid thermoplastic composites play an increasingly important role in lightweight applications. One key challenge in the intrinsic hybridization is to achieve an adequate interface bonding strength. In order to optimize the interface strength specific methods for characterization and process monitoring need to be developed. Thus, within this paper two advanced mold concepts are presented allowing online monitoring of the welding conditions at the spot and time of interest. The resulting part geometries are optimized for characterization of interface properties in a defined and reliable manner. Regarding this, adopted characterization setups are presented and validated allowing the characterization of three stress states.



*Keywords:* hybrid thermoplastic composites, intrinsic hybridization, injection molding, overmolding, online process monitoring, interface bonding strength, warpage

## 1. Introduction

Forming and overmolding of continuous fiber reinforced semi-finished products with technical thermoplastic matrices like PP and PA6 currently evolves from niche products to serial automotive applications [1]. The main reason for this is the possibility to take advantage of material and structure related weight savings, while these hybrid parts can be manufactured in highly automated and integrated processes leading to low cycle times and economic feasibility. Due to these advantages, increasing lot sizes and the need for simplified integration as well as recyclability, these production processes will become also of higher interest in aerospace applications in the near future [2,3]. Here, especially flame retardant, high temperature thermoplastics like PPS, PEI or PEEK will be used. Depending on the type of material and the processing sequence it might be challenging to achieve an adequate interface bonding strength. Thereby, the temperatures within the welding zone [3-5] as well as in-mold pressures are most determining for the interface bonding properties. Additionally, the cooling behavior leads to the development of mechanical properties during solidification and thus influences dimensional part stability [6]. However, the determination of the product temperature at the time and spot of interest is quite challenging in closed-mold processes. Thus, in this study two advanced mold concepts are presented in section 2 allowing precise online monitoring of the welding conditions at the spot and time of interest. Along the flow path contact-free temperature sensors as well as contact-based mold temperature and cavity pressure sensors are utilized. Both molds are designed for processing a variety of thermoplastics like PP, PA6, PA66 as well as for high-temperature matrices like PPS, PEI or PEEK. The resulting part geometries are optimized for characterization of interface properties in a defined and reliable manner. Regarding this, detailed information is given in section 3 about the developed characterization methods allowing the characterization of the failure modes: pure shear, pure tension and mixed mode (peel). The presented methods are compared and validated using samples of PEI/CF organo sheets overmolded with


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PEI/GF short fiber reinforced granulate. The developed molds and methods can be additionally used for the validation of advanced process simulation models predicting welding temperatures and pressures which lead to specific interface properties and final dimensions.

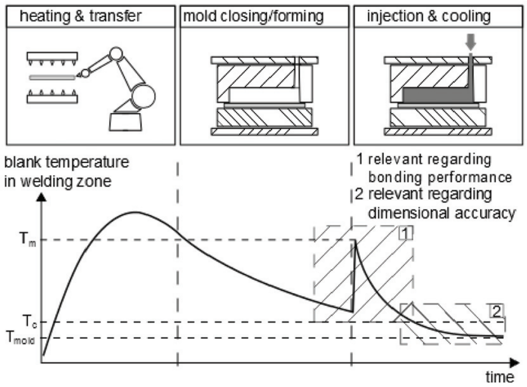


Fig. 1. General process chain illustration for the manufacturing of hybrid thermoplastic composites with exemplary temperature profile of the overmolded blank ($T_m$: Melting temperature; $T_c$: crystallization temperature; $T_{mold}$: mold temperature)

To validate the developed molds and corresponding part geometries as well as the testing procedures and setups, the results of online monitoring and testing are discussed in section 4.

## 2. Mold Concepts & Part Geometries

Hybrid thermoplastic components usually contain areas where the substrate is planar overmolded as well as areas were the substrate is locally overmolded (e.g. ribs). To accomplish for both cases two molds were developed.

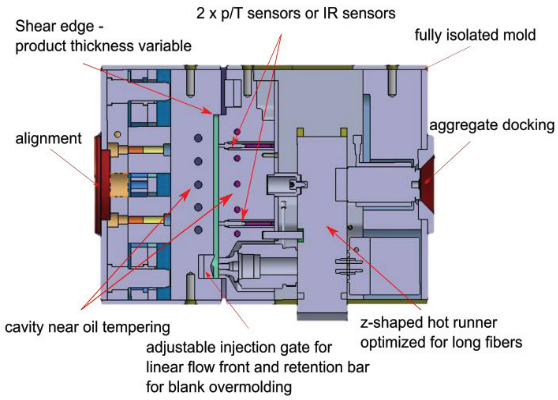


Fig. 2. Mold 1 for hybrid plate manufacturing – Visualized is a section profile of the mold

Figure 2 illustrates the mold for hybrid plate manufacturing with a size of 180 mm x 180 mm. Due to the shearing edge design, plate thickness can be adjusted as needed up to 8 mm in total. By utilizing the retention bar in the gate zone frontal injection to the substrate is prevented which could lead to substrate damage. Substrates with a thickness of up to 2 mm can be inserted. For homogeneous tempering of the mold, cavity near oil cooling as well as water heating of the mold platens are integrated. The z-shaped hot-runner is optimized for long fiber thermoplastic materials and includes an 8 mm needle valve nozzle.

Mold 2 for local rib overmolding of substrates is illustrated in Fig. 3 In a vertical closing injection molding machine the substrate can be placed on the lower part of the mold. The cavity needs to be sealed by the substrate. Thus the clamping force is applied onto the substrate. Due to a small projected area of the rib ground, necessary closing forces are rather small and no substrate damages takes place. Additionally, spacer blocks can be used for sensitive or softened substrates. The hydraulically activated sliders are necessary to allow product release before mold opening, since the rib is designed with an undercut (cf. Fig. 4). The rib thickness is 3 mm and is reduced to 2 mm in the rib ground. During testing this design leads to a defined failure zone within the welding zone of the hybrid structure. It is not intended to test an optimized rib design, but rather the bonding interface strength in a defined and reliable manner. For an optimized gripping in the testing procedure no draft angle is introduced to the rib geometry.

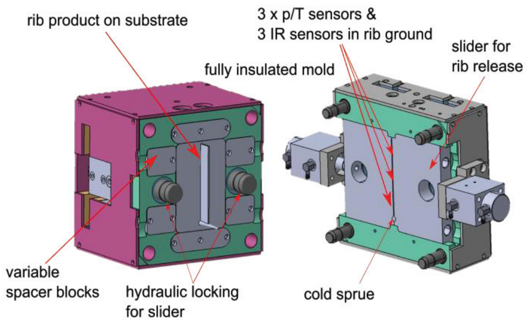


Fig. 3. Mold 2 for hybrid rib-substrate manufacturing – left: lower part of the mold, right: upper part of the mold

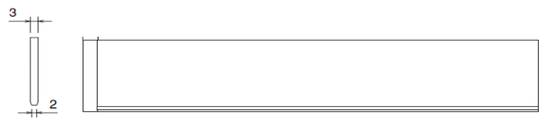


Fig. 4. Rib geometry illustrating the undercut – rib height is 30 mm and length approx. 160 mm – optimal substrate length is 180 mm

### 2.1. Online Process Monitoring

In mold cavity sensors have been applied in order to determine the process conditions during overmolding. The integration of IR sensors enables to measure the substrate surface temperature at the time and spot of overmolding. Within the rib mold additionally thermocouples can be integrated into the blank substrates, since no shearing edge is present. Cavity pressure sensors and mold temperature sensors are additionally integrated along the flow path.

### 2.2. Specimen Production

For the sample specimen production TenCate PEI/CF organo sheets and Sabic Ultem 2310 PEI/GF granulate was used. Materials were dried at 150 °C for at least 24 hours to 0.04 - 0.05 wt% moisture content. The organo sheet substrates were cleaned with isopropanol before processing and preheated using a convective oven. Transfer to the mold was manually at a reproducible transfer time of 6 ± 1 s. The materials are processed using an Arburg Allrounder 320C-600-250 injection molding machine. Due to a vertical closing of the machine, substrate fixation in the molds is not needed. If not indicated differently, the process parameters according to Table 1 were used.

Table 1. Processing parameters

| Processing parameter | value | unit |
|---|---|---|
| Mold temperature | 165 | °C |
| Injection mass temperature | 370 | °C |
| Blank preheating temperature | 200 | °C |
| Injection velocity | 15 | cm³/s |
| Back pressure | 25 | bar |
| Holding pressure | 450 | bar |

## 3. Interface testing methods and developed setups

For a complete mechanical characterization of interface properties it is convenient to consider the three load cases pure shear, pure tension and peel stress. Pure shear and pure tension are distinct stress states and the test is giving a specific failure stress as output parameter. In real application often complex stress states occur. Thus, a peel test represents a more realistic stress state, allowing the determination of a specific energy which is necessary to detach the two components related to its bonding surface. The mentioned failure modes and the testing realizations are shown in Fig 5. The pure tension and peel apparatus are specifically developed setups, while for the pure shear characterization a device according to [7] was used. All Setups were installed in a standard Hegewald & Peschke universal tensile testing machine. For the comparison of the presented testing setups and the output reliability a set of five specimens is tested for each method. All specimens are produced using the same set of processing parameters.

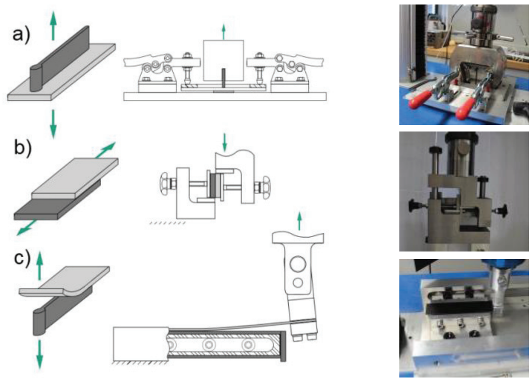


Fig. 5. Failure modes (left) and testing setup realization (right) – a) pure tension, b) pure shear, c) peel / mixed mode

### 3.1. Pure Tension

For the tensile test the rib sample geometry is used (mold 2). From the 160 mm rib, sections with 30 mm length are cut using a water cooled circular saw resulting in a joining interface of 60 mm² per sample. On the one hand this specimen size allows differentiation between probes near and far to the sprue giving a deeper insight into flow path depending properties. On the other hand, mechanical machining might cause micro cracks near to the bonding interface. The organo sheet substrates are clamped using a blank holder and four quick release clamps (cf. Fig. 5a). To minimize the risk of substrate bending during measurement, the blank holder reproduces the undercut of the rib geometry leaving only 0.1 mm tolerance between blank holder and rib. The injected rib is clamped using a standard mechanical clamping jaw. The testing sequence is performed according to DIN EN ISO 527-1 resulting in a traverse speed of 2 mm/min. A 10kN force transmitter is used. The rib detachment tensile strength is calculated according to eq. 1.

$$\sigma_t = \frac{F_{t,\max}}{A} \quad (1)$$

$\sigma_t$: tensile strength, $F_{t,max}$: maximum tensile force, $A$: interface area

### 3.2. Pure Shear

The shear test is realized as a compression shear test according to [7], having quite a few advantages compared to common tensile shear tests. Especially the risk of torsional moments during the measurement is significantly reduced allowing a distinct stress state and an improved reproducibility. Specimen sample probes have the dimensions of 10 mm x 8 mm and are waterjet cut from the middle of the overmolded plates (mold 1). The testing direction is along the 0° direction of the PEI organo sheets. The testing is performed using a 10 kN force transmitter at a traverse velocity of 1 mm/min.

$$\sigma_s = \frac{F_{s,\max}}{A} \quad (2)$$

$\sigma_s$: shear strength, $F_{s,max}$: maximum shear force, $A$: interface area

### *3.3. Mixed Mode (Peel)*

For the peel test a new clamping device was developed for testing the rib geometry. The rib is clamped heading downwards in a roll guided sliding rack. The whole rack is fixed on the bottom of the testing machine. The substrate is clamped near to the sprue using a tilting clamp. The testing and assessment is performed in accordance to DIN EN 6033, which is normally used for the determination of mode 1 interlaminar fracture toughness energy of laminated composites. Thus, testing is performed at 10 mm/min constant traverse speed. The initial crack length is marked at the sample probe after approx. 20 mm to 30 mm crack length and the corresponding displacement $D_1$ is quoted. The measurement is performed until a total crack length of about 100 mm has been reached. Therefore, testing is stopped at approx. 120 mm to 130 mm crack length. The final crack length is marked at the sample probe and the corresponding displacement $D_2$ is quoted. The total propagated crack length $a$ is measured at the sample probe. For the presented tests a 500 N force transmitter was used. The peel toughness in J/m² is given by eq. 3.

$$G_I = \frac{A}{a \cdot w} \quad (3)$$

$G_I$: peel toughness / fracture toughness energy, $A$: energy to achieve a total propagated crack length, $a$: total propagated crack length (final crack length minus initial crack length), $w$: width of bonding interface

The energy A is calculated from the measured force-displacement curve via integrating over the cross head displacement between initial and final crack length (cf. Fig. 6).

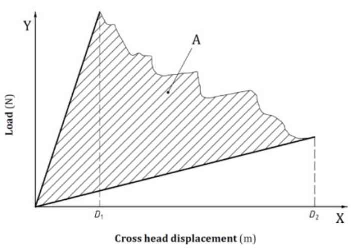


Fig. 6. Exemplary graph visualizing how the crack propagation energy is calculated - $A$: energy to achieve a total propagated crack length, $D_1$: displacement at initial crack length, $D_2$: displacement at final crack length

## 4. Results & Discussion

### *4.1. Online temperature & cavity pressure measurement*

Fig. 7 visualizes recorded IR data during molding for two different blank preheating temperatures and three different injection mass temperatures using the introduced PEI materials. Since the sensors are calibrated and focused to a closed mold distance, real temperatures can be detected as soon as the mold is fully closed and until injected material reaches the sensor. Beforehand temperatures are underestimated (cf. Fig. 7). Afterwards the injection mass temperature is measured, which will be overestimated due to the short distance between sensor and material. For measuring the cooling of the product the sensors need to be recalibrated. Nevertheless, it is also possible to detect the real product cooling behavior which is determining for material solidification and part warpage phenomena. Interestingly, in this example regardless for the preheating temperature of the blank, the blank reaches the mold temperature until the injected mass is in contact with the blank substrate. This indicates the importance of mold temperatures in over-molding processes.

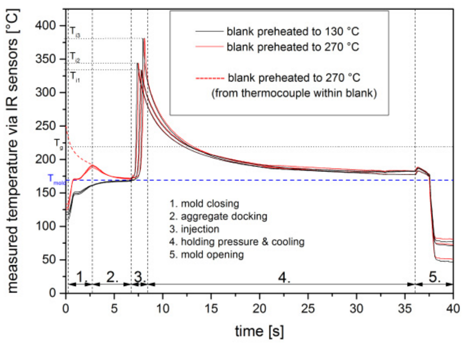


Fig. 7. Time-temperature chart for experimental data using cavity IR sensors – Shown are the curves for two different blank preheating temperatures and three different injection mass temperatures

Using a linear mixing rule to calculate the contact welding temperature for the prepared samples - considering the substrate surface temperature from the IR sensors (~165 °C) and the injection mass temperature of 370°C - leads to approx. 267 °C welding temperature. This is quite above the glass transition temperature of PEI and should therefore lead to a sufficient bonding for validation of the testing setups.

Figure 8 shows the recorded pressure data from the cavity sensors. Near to the sprue, highest cavity injection and holding pressures are found.

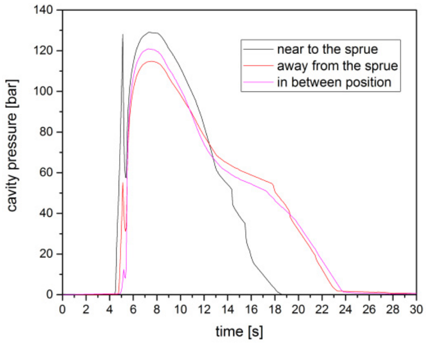


Fig. 8. Cavity pressure chart during molding

## 4.2. Interface testing

All tests resulted in a distinct and defined failure of the sample probes in the bonding zone. This allows for process dependent interface characterization. In this section the results from the different testing methods are compared and discussed.

### a) Pure tension

Fig. 9 depicts the testing results of the tensile rib detachment test.

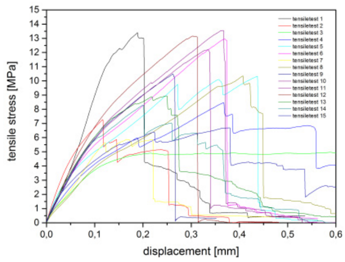


Fig. 9. Results from the tensile rib detachment test

In this case three specimens are cut from each of the five samples leading to 15 specimens in total. The rib detachment tensile strength is determined to $\sigma_t = 10.03 \pm 2.83$ MPa which is a quite high variance of 28.2 %. This variance is a bit smaller if only specimens from the same position are compared. However, for the selected material it was not possible to determine a distinct dependency on the probe position near or far away from the sprue. There are a few reasons for this high variance. Firstly, due the fact that a distinct maximum stress is determined the method is naturally subject to a high variance. Very small local differences can strongly influence the testing result. Secondly, due to the sample preparation the probes might be already weakened. Thirdly, due to the relatively small bonding interface area also local material differences related to the morphology of the organo sheet substrates might cause the a high variance.

### b) Pure shear

Fig. 10 depicts the results from the shear test.

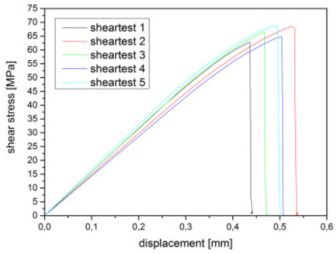


Fig. 10. Results from the shear test

The results reveal very high reproducibility with a variance of 3.5 % at a determined shear strength of $\sigma_s = 66.22 \pm 2.31$ MPa. Due to the increased interface area of the specimen local material difference probably play a minor role.

### c) Mixed mode (peel)

Fig. 11 depicts the peel force-displacement curves. Determination of the peel toughness according to equation 3 leads to 2.05 ± 0.22 kJ/m² which means a variance of approx. 10.6 %. This indicates a quite good reproducibility of the test. Due to the force-displacement curve integration local differences are averaged.

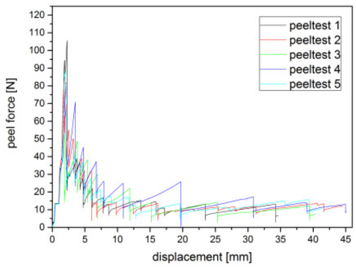


Fig. 11. Results from the peel test

## 5. Conclusion & Outlook

The presented molds allow the production of specimens for bonding interface characterization even for high temperature materials like PEI as presented in this study. The integrated sensors enable to measure the welding conditions at the time and spot of interest. The contact free IR sensors give detailed temperature information of the overmolded substrate shortly before overmolding and allow to track the product cooling. Additionally, three interface property testing devices are presented, validated and compared using PEI materials.

Depending on the failure mode of interest all methods are applicable. However, in comparison the tensile rib detachment is subject to the highest variance, while pure shear and the peel test show improved reproducibility. Therefore, especially the latter techniques are suitable for the determination of bonding properties depending on specific processing parameters. In focus of a follow up study a process parameter design of experiment is performed. By linking the obtained interface properties to the measured process conditions, optimized processing parameters for the intrinsic hybridization of PEI materials can be found.

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