HYBRID-MATRIX PROCESSING: HOW TO CO-INJECT MULTIPLE RESIN SYSTEMS INTO ONE COMPOSITE PART?

J. Krollmann [1], C. S. Alvarado [1], P. Carqueville [1], R. Snajdr [1], S. Zaremba [1], K. Drechsler [1]

[1]Institute for Carbon Composites, Faculty of Mechanical Engineering, Technische Universität München, Boltzmannstraße 15, D-85748 Garching b. München, Germany
Email: krollmann@lcc.mw.tum.de, www.lcc.mw.tum.de
Email: patrick.carqueville@tum.de
Email: c.sancheza@outlook.es
Email: roman@snajdr.de
Email: zaremba@lcc.mw.tum.de
Email: drechsler@lcc.mw.tum.de

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Abstract
Conventional composite structures consist of fiber reinforcement embedded in a single matrix system. Hence, the matrix dominated composite properties are defined for the entire part and the matrix selection represents a compromise between the dominant demands. The Hybrid-Matrix approach aims at the defined integration of multiple matrix materials in one composite structure with an out-of-plane matrix transition. The composite properties can be adapted to locally changing requirements and functionalities. This study presents the development of a closed mold injection process in which different resin systems are co-injected to form defined transition areas and a transition line where resin system merge and co-cure. This Hybrid-Matrix-RTM (HyMa-RTM) process concept is based on the control of the shapes and the reduction of velocities of the flow fronts by local and reversible compaction of the preform. Injection experiments, using a transparent modified RTM tool for flow front analysis are conduct to evaluate the concept of reducing the flow front velocity. The results show that the flow front velocity is reduced by approximately 95% - 98% at compacted areas. Based on the proof-of-concept to control the flow front experimental investigations are conducted to define an injection strategy and process parameters for a co-injection process. The results of the co-injection, using colorized resin systems, show the potential of the HyMa-RTM process. A test plate is manufactured fulfilling the requirements regarding tolerances of the transition area and the course of the transition line.

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1. Introduction

At fiber reinforced plastics (FRP) the two constitutive components e.g. the fibers and the polymer matrix system fulfill different functions and possess different influences on the composite properties (see Figure 1). The main function of the matrix is the load introduction as well as the load transfer from fiber to fiber. The rigid matrix system defines the geometry and the positioning of the fiber reinforcement in the composite part. The anisotropic fibers are used to carry the loads. Dependent on the conventional design parameters like type of fiber, type of reinforcement (unidirectional, textiles), ply number, ply orientation and fiber volume content mechanical properties like stiffness and strength can be influenced. Respectively, the matrix material dominantly predicts composite properties like bending stiffness, strain rate dependency, damage tolerance, thermo-mechanics, chemical resistance as well as the processing.

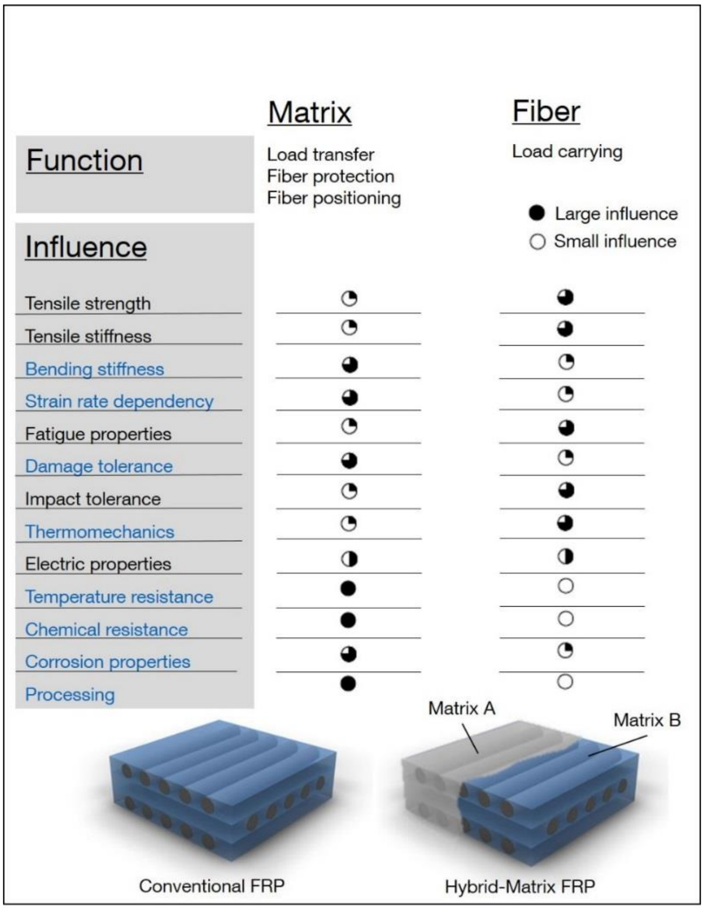

Figure 1 Functions and influence of matrix and fibers at fiber reinforced plastics and the concept of Hybrid-Matrix [1] [2]

In conventional FRP parts only one matrix material is applied. Hence, the matrix-dominated composite properties are defined for the entire part. Dependent on the key requirements at the different fields of industry for example costs (automotive), curing time (automotive), toughness (aviation) or temperature resistance (aviation) the material selection takes place. Since a large number of composite properties are dependent on the matrix selection conflicts of objectives exist and compromises at the material selection are made lowering the potential of overall performance of the composite structure. Furthermore, at integral composite structures requirements regarding mechanical properties or functionality which are influenced by the matrix materials can vary locally. To fulfill the different requirements commonly cost intensive matrix systems or differential design are applied. This introduces drawbacks regarding weight, cost and manufacturing.

The conventional use of single matrix material prevents the adaptation of matrix dominated composites properties to locally changing requirements and herewith an optimized part design.

The Hybrid-Matrix approach offers the possibility to overcome the dogma of single matrix application and its disadvantages by the integration of multiple resin systems into one composite structure. The intention is to realize out-of-plane transition lines (see Figure 1) of different resin systems at continuous fiber reinforcement. Figure 2 gives an example of the possibilities of the Hybrid-Matrix approach. At the displayed bicycle rim different functional areas exist. An optimal and cost efficient fulfillment of these local demands can only be realized by the application of different matrix materials with suitable properties regarding damage tolerance, temperature resistance and costs.

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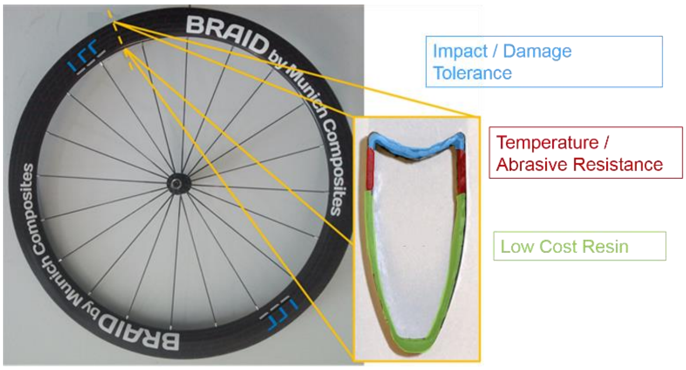

Figure 2 Braided bicycle rim with functional areas dominantly influenced by the matrix material [3]

In literature manufacturing processes and design approaches can be found trying to enhance structural performance and functionality by integration multiple resin systems. Gillio et al. and Fink [4–6] introduced a single step co-injection RTM process (CIRTM) using a separation film between different layers of fiber reinforcement to separate the matrix material during injection. Based on diffusion enhanced adhesion the injected matrix materials bond to the film. With this process and suitable matrix materials impact tolerance at ballistic structures was enhanced. 2010 Kaps [7] presented a combined prepreg infusion technology for integral fiber reinforced plastic structures. Here, pre-impregnated layers of fiber reinforcement are combined with dry layers which are impregnated via resin infusion with flexible tooling. To enhance the bonding properties between the different matrix systems polymer films were integrated. Characteristic for both approaches is the in-plane separation of matrix material (see Figure 3). The composite properties are influenced by both matrix systems. A local adaptation of matrix dominated composite properties via different resin systems cannot be realized.

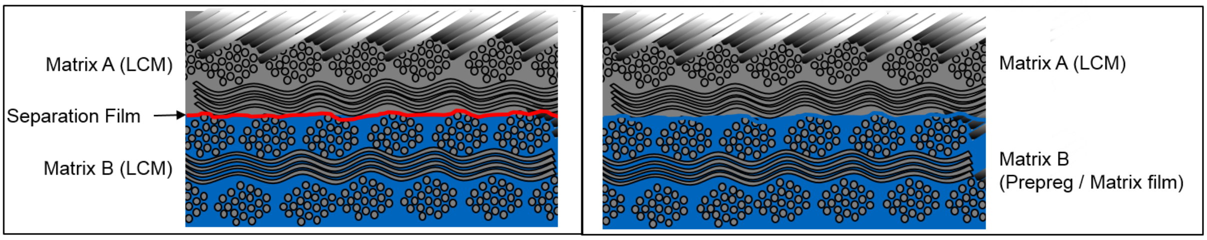

Figure 3 In-plane separation of multiple matrix materials

2012 Plenk [8] introduced the integration of thick film elastomer material into the RTM process between different layers or on top of the fiber reinforcement. During injection and curing the elastomer (EPDM) is vulcanized leading to a strong bonding to the injected resin. The integration can be realized locally in the composite part. Composite properties like impact tolerance, fracture process as well as vibration damping behavior was enhanced. 2008 Todoroki [9] presented a fiber reinforced reinforcement to realize a functional composite structure. Manufacturing process is based on ply wise wet lamination of the silicon matrix material and subsequent infusion of the epoxy matrix material. Compared to Gillio et al. and Fink [4–6] and Kaps [7] an out-of-plane transition line can be realized within the composite. Both approaches focus on the combination of elastomer and thermoset matrix materials and do not offer a material independent realization of hybrid-matrix composites.

The presented paper focuses on the introduction, development and validation of an innovative closed mold manufacturing process which is capable of the simultaneous injection of two different matrix

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materials in one composite structure forming an out-of plane transition line based on industry relevant process requirements regarding serialization, reproducibility, robustness and quality.

2. Hybrid-Matrix-RTM process concept

At the HyMa-RTM processes multiple resin systems are co-injected into on preform. Suitable process concepts and modifications are necessary to enable a defined matrix transition. In the following basic process objectives are presented describing an aspired filling process. Based on theoretical assumptions a promising concept is presented and realized using local reversible compaction of the preform within the tooling.

2.1. Basic process objectives

The intention of the hybrid-matrix process is to realize complex part and complex matrix transition geometries based on the RTM process to enable a broad application at relevant composite structure. As it can be seen in Figure 4 co-injected matrix materials are supposed to merge at a pre-defined transition area. Dependent on inlet locations and transition area geometries, flow fronts can converge head on or with various contact angle. It is believed that due to the complex filling process and the geometry of the transition area flow fronts arrive at the pre-defined transition area at different point of times. To realize reproducible transition areas within the composite part it must be guaranteed that flow front velocity is reduced in these areas to allow the continuous approach of the second flow front.

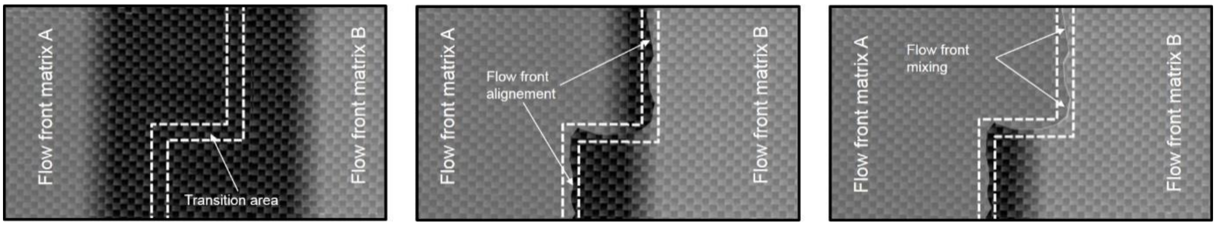

Figure 4 Hybrid-Matrix process concept with complex pre-defined transition area [11]

Flow behavior at closed mold processes can analytically described by Darcy´s Law [12]. According to this equation (1) the flow front velocity is a function of injection pressure (p), viscosity (η), permeability (K) and flow front distance to the inlet (x).

$$v(x) = (K * \Delta p)/(\eta * x) \quad (1)$$

It is the intention of the HyMa-RTM approach to enable a broad use with a minimum of process limitations regarding suitable materials or process design. Hence, it is found that the realization of the Hybrid-Matrix process needs to be as independent as possible from these process parameters.

As a suitable concept to reduce the flow front velocity at designated areas the local modification of preform permeability is identified. According to experimental investigation of Walbran and Körber [13] preform compaction leads to an increase in fiber volume content and a decrease in permeability. The basic process idea of the HyMa-RTM process is to enable reversible, local compaction of the preform to reduce the permeability at transition areas during the co-injection process. The reversible character is pursued to reduce the risk of thickness variation and modification of the initial fiber course at the finished part.

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2.2. Concept realization

Local reversible compaction is realized by a flexible compaction device which is integrated into one half of the RTM tooling. It features a solid core embedded in elastomer material. It can be inflated and deflated (see Figure 5). During injection the inner pressure of the compaction device is higher than the inner mold pressure. After the flow fronts merged in the compacted area the pressure of the compaction device is removed. The inner pressure of the mold as well as the elastic material properties of the compaction device lead to a reversible deformation.

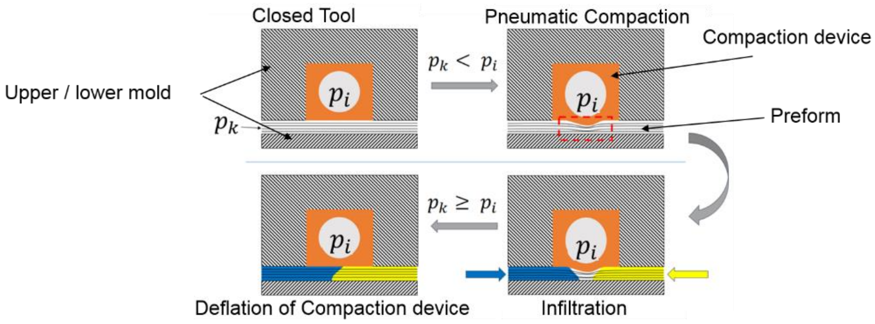

Figure 5 HyMa-RTM process and mold design with compaction device

It is believed that dependent on the compaction pressure the flow front velocities can be reduced and aligned along the compaction area resulting in reproducible out-of-plane transition lines within the composite. It is assumed that due to elastic effects preform deformation possesses a reversible character. This reduces the risk of permanent deformation and fiber angle deviations caused by the compaction.

3. Experimental investigation of the Hybrid-Matrix Process

The experimental validation of the process concept is proceeded in two steps: validation of reduction of flow front velocity, validation of HyMa-RTM process by co-injection of two resin systems. The basic principle is the local reduction of flow front velocity enabling the modification of flow front geometry. Single matrix injection experiments are conducted to investigate the potential reduction of flow front velocity. The proof of the HyMa-RTM concept is realized by co-injection experiments where colorized resins systems are simultaneously injected into a modified RTM tool.

3.1. Influence of local preform compaction on flow front velocity

Based on a transparent tooling a compaction device is realized in one half of the RTM test tooling. A solid core is placed into the 23,5 x 20 mm (width x depth) grove and connected to a pressure vent. The grove is filled with silicone casting resin and cured (see Figure 6). The silicone elastomer does not bond to the solid core. The preform facing side of the silicone material separates and deform outwards when pressurized.

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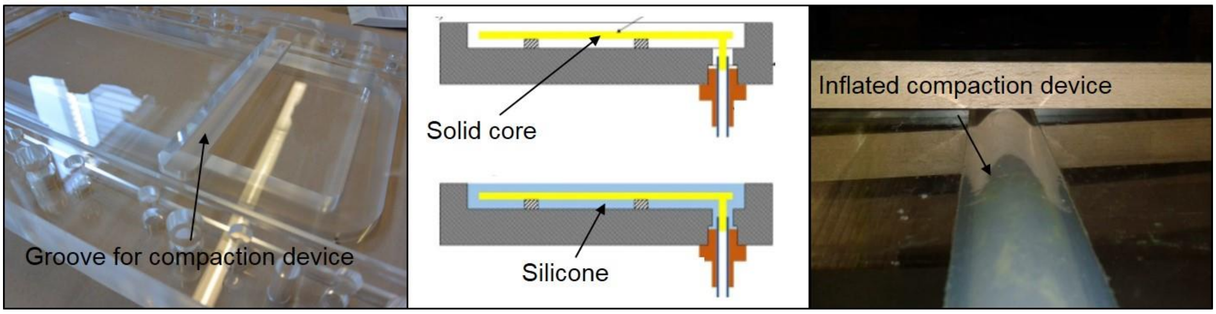


Figure 6 Modified RTM Tooling with groove for compaction device, inflated compaction device

For the first validation step a single matrix injection is performed with following process parameter:

- Injection pressure: 1 bar
- Injected media: Oil (constant viscosity)
- Compaction pressure: 4-8 bar
- Preform: 4 and 5 layers, Hexcel G0926 weave [14] (FVC: 46,5% - 58%)
- Cavity height: 1,8 mm

Flow front velocity is optically recorded analyzed qualitatively and quantitatively. In Figure 7 the injection process at four different injection times is displayed. At 10 seconds injection time the flow front runs through the preform forming a curved shape. It is believed that this shape is caused by friction effects at the side walls as well as tool deformation due to the local compaction and pressurization. After 19 seconds injection time the flow front reaches the compaction area. At 25 seconds it can be observed that the curved flow front geometry is aligned at the compaction device before the flow front propagates constantly at the compaction area. After 35 seconds the flow front propagated only a short distance within the preform.

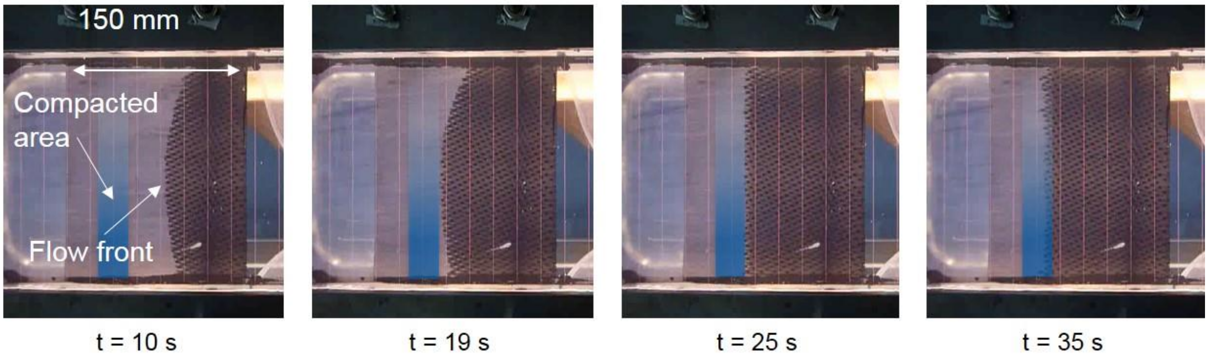


Figure 7 Injection process with local preform compaction at RTM process

For a quantitative evaluation of the flow front velocity is determined and analyzed. As it can be seen in Figure 8 flow front velocity is reduced significantly due to the local compaction of the preform. The velocity is reduced between 95% - 98% dependent on the initial FVC of the preform.

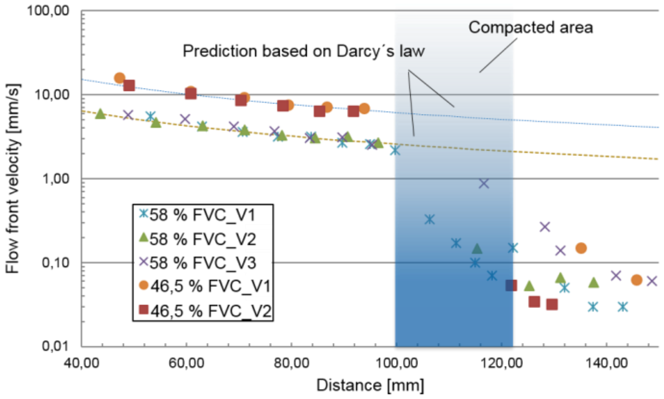
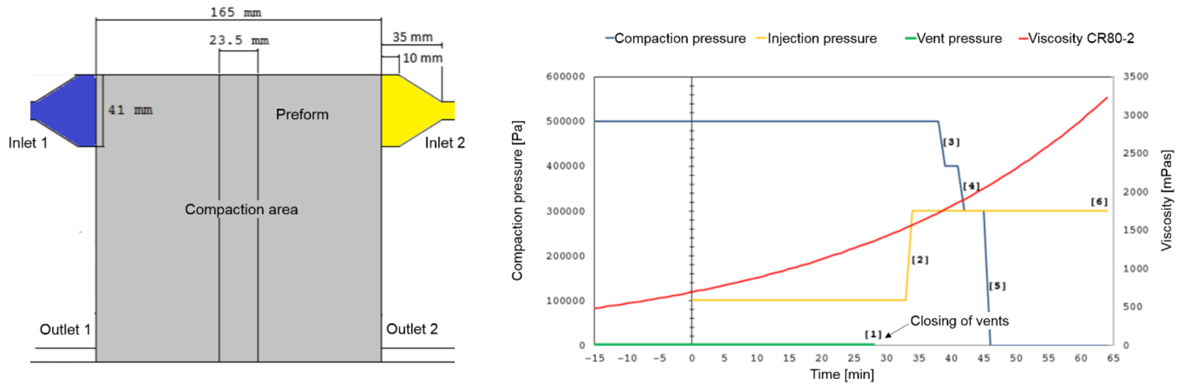


Figure 8 Reduction of flow front velocity

The experimental results reveal that the flow front velocity can be significantly reduced due to the local compaction and that the flow front geometry can be modified and aligned along the compaction.

3.2. Co-injection experiments

Based on the initial validation a co-injection process is designed. A crucial process parameter is the inlet location. Hereby, the merging angle of the flow fronts is predicted. According to Pearce et al. [15] convergent flow fronts at RTM processes influences the void content of the composite parts in this area. The more frontal the flow front meet the higher the void content. It is the intention to manufacture high quality test plates with low void content at the transition area. Therefore, flow fronts displays the inlet, outlet design and the location of the compaction device. Based on filling simulation process parameters are defined regarding injection pressure for the two inlets, the compaction pressure and the deflation of the compaction device.



(a) (b)

Figure 9 Inlet and outlet design (a), representative process design (b)

For the experimental evaluation of the HyMa-RTM process an epoxy resin systems (SIKA Biresin CR80-2 [16]) is colorized (blue, yellow), representing two different resin systems, and simultaneously injected. The yellow colored resin system is additionally moderately enriched by soluble fluorescence powder (2 % proportion of weight). Both modifications have no noticeable influence on viscosity and cure kinetics. Tooling cavity is 3,8 mm. The preform consists of 8 layers Hexcel G0926 [14] woven textile leading to a theoretical fiber volume content of approximately 45 %. Test plate dimensions are 165 mm by 165 mm.

The resins systems are separately mixed and degassed for 15 min. During this time the mold is evacuated and 5 bar absolute compaction pressure is applied in the middle of the preform. According to Walbran and Körber [13] the presented material shows time dependencies in compaction stress when the textile is compacted. It is believed that this stress relaxation is based on setting effects and nesting. Therefore, preform compaction is established prior to injection to guarantee constant permeability properties at the compaction area.

The two matrix materials are simultaneously injected with 1 bar injection pressure. After 27 min both flow fronts reach the respective outlets. Both outlet vents are closed. After 5 minutes injection pressure at both inlets is increased to 3 bar. Hereby, final filling of unfilled areas at the compaction area is enabled. After 4 minutes the pressure of the compaction device is stepwise reduced and vacuum is applied to guarantee full removal of the deformation. Resin viscosity is approximately 1700 mPa*s at this point of time. Reversible deformation of the compaction area needs to take place at a viscosity range where matrix materials can still flow since a cavity is created by the deflation which needs to be filled. Contra wise elevated viscosities are aspired to reduce the risk of uncontrolled flow of the two matrix systems.

4. HyMa-RTM Results

In
Figure 10 (a) an example of a HyMa-RTM injection can be seen. This test plate features a distinct transition line between the two matrix materials. Its course is centric to the compaction area. Obviously, no significant material transfer between the two matrix systems exists.

For the quantitative evaluation of the transition area fluorescence photographs of both sides of the plate are analyzed using ImageJ software. UV-light is used to illuminate the test plate. Only fluorescence enriched material emits the UV-light which is captured by the camera. Based on a RGB value analysis of the fluorescence photos the transition line can be geometrically determined.
Figure 10 (b) displays the analysis methodology. At 29 sections along the y-axis the green value is determined for each pixel. Critical green values between 0 and 255 can statistically be determined for both areas (yellow, blue) where neat resin appearance in the composite is assumed. According to the pixel wise analysis x-coordinates can be allocated where the green values undergoes the critical green value of matrix A resulting in a high limit (HL). Respectively, an x-coordinate can be defined representing the low limit (LL) where the critical green value of matrix material B is exceeded. Between these limits material transition takes place. Plotted along the plate surface the transition area can be determined and visualized (see Figure 10 (c), (d)).

It is the target of the HyMa-RTM process to realize defined and finite transition areas within the composite structure. Process quality and reproducibility can be characterized by the width of the transition area as well as the course of the transition area and its deviation from the pre-defined course.

The average width of the transition area at side 1 (facing the compaction device) of the test plate is 3,7 mm with a standard deviation of 1,3 mm. The mean value of transition width for side 2 is 6,7 mm with a standard deviation of 3,1 mm. A deviation between both sides of the plate exists. It is believed that on the one hand local pressure deviation as well as local, ply-wise permeability variation (gaps) leads to a wider transition are. On the other hand the contact properties between preform and tooling is influenced by the compaction device. During pressurization the flexible material of the compaction device allows the adaption of the textile structure and strong nesting. Gaps between tows and crossover points are closed. This effect might lead to a modification of permeability of the outer layer in this area. It is believed that matrix mixing particularly takes place in between rovings, gaps and crossover points. Reduction of mixing cavities within the preform might lead to smaller transition areas.

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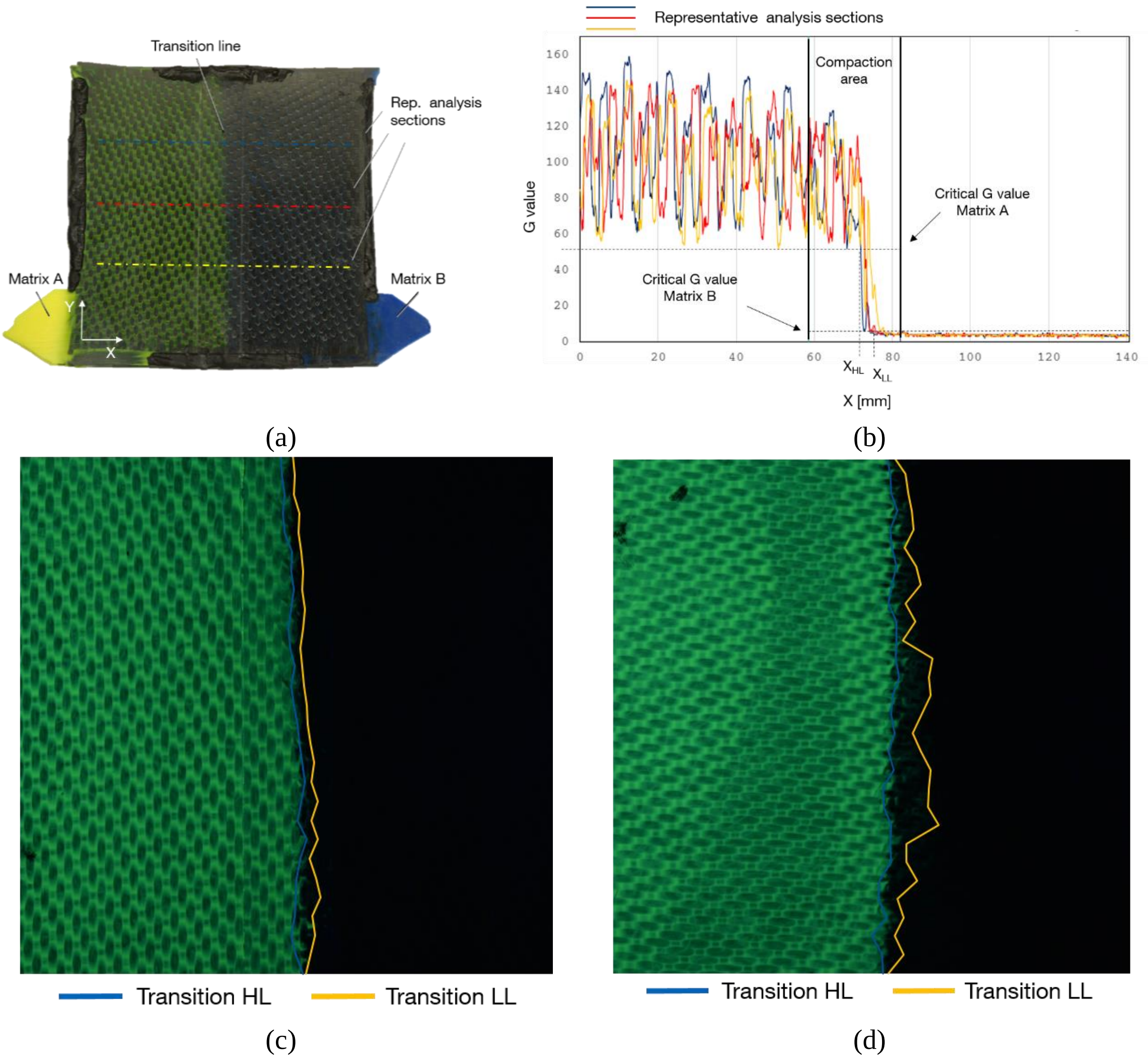

Figure 10 HyMa-RTM process test plate (a), Green value profile at rep. analysis section (b), Matrix transition area side 1 (c), Matrix transition area side 2 (d)

Compliance of the actual transition line with the pre-defined course is established by a profile tolerance. A tolerance zone along the pre-defined transition line is determined in which the actual transition line lays. This approach allow the quantitative analysis of the quality criteria. At each side of the plate a centerline within the transition areas is used as a representative of the transition (see Figure 11).

At the initial experimental investigation the pre-defined transition line lays in the middle of the compaction area. At side 1 the actual transition line can be found within a tolerance zone of 11,56 mm width. The transition line of side 2 lays within a tolerance of 20,66 mm width. Both transition lines can be found on one side of the pre-defined converging line resulting in a general displacement from the pre-definition. It is believed that this displacement is caused by a temporal offset between the arrivals of the flow fronts at the compaction area. This offset seems to remains during the injection leading to the constant displacement.

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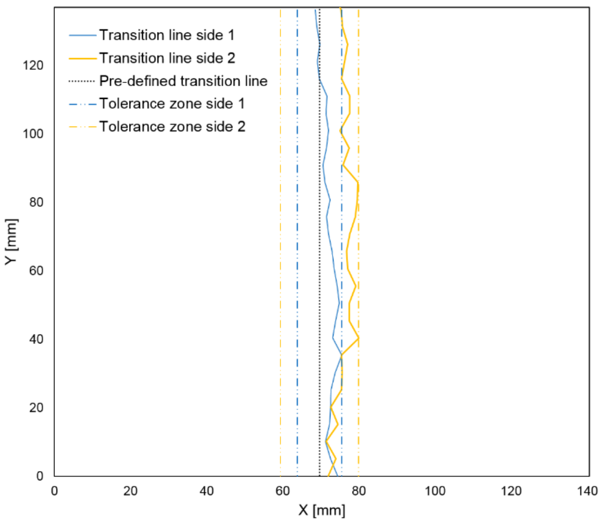

Figure 11 Tolerance zones of the transition lines

Filling processes based on textile reinforcements with local variation in compression stress are highly complex. Inhomogeneity of the preform have strong influences on the micro and macro flow affecting the transition properties in the composite. It is assumed that up to a certain degree, local inhomogeneity of the permeability and by this the formation and properties of transition areas can hardly be controlled. The results of the transition width and the transition tolerance are promising in relation to the size of the different matrix regions of the hybrid-matrix composite part as well as the part size itself. The presented results represents suitable values for industrial application.

5. Conclusion

Studies on the co-injection of two resins systems into one composites structure were undertaken, highlighting the potential of the HyMy-RTM process. Based on local, reversible compaction of the preform experimental investigation reveal that flow front velocity can be significantly reduced during the injection process within the preform. The local compaction leads to an alignment of the flow front along the compacted area. This supports the general process idea to modify the flow front geometry and propagation of different flow fronts to create a defined transition line within the composite part.
Experimental evaluation of a co-injection process using differently colorized and fluorescent matrix materials lead to promising results. A test plate was manufactured and analyzed. With the help of florescence photography transition areas at both sides of the plate were determined showing good results regarding transition width within the composite part. Process quality was assessed based on the deviation of the actual transition lines of both sides from the pre-defined transition line. The results show deviation in an acceptable range.

The experimental investigation show that co-injection of different matrix materials in one composite part with defined matrix transition is feasible. In this study only one possible injection strategy e.g. inlet-, outlet location and constant injection pressure was presented. With regards to the application at actual structural parts more complex transition geometries and injection strategies need to be investigate. Filling simulations need to be validated and integrated into the development process to allow the cost and time efficient evaluation of HyMa-RTM processes. It is believed that the HyMa-process quality can be enhanced by intelligent process control using in mold sensor technology for flow front and pressure detection in combination with mass flow controlled injection devices.

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