MANUFACTURING OF GLASS FIBRE/PBT-SPINCOM YARNS FOR INNOVATIVE COMPOSITE PROCESSING

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Abstract

The aim of this work was the development of GF (glass fibre)/ PBT (polybutylene therephthalate) SpinCOM yarns that are commingled online during the spinning process using sizing formulations that enable textile processability as well as high fibre-matrix interaction. Epoxy- and polyurethane-based sizings have been applied and were evaluated by micromechanical (single fibre pull-out test) and macromechanical (transverse tensile test of unidirectional plates) testing. Finally, a material combination was selected to be processed by Tailored Fiber Placement (TFP) to textile preforms with a variable-axial, load adapted fiber design. These preforms were consolidated in a low energy and resource consuming process using novel light and low cost forming tools produced by incremental sheet metal forming (ISF) technology. Finally, a low cost solution for thermal processing even for complex shaped TPC parts is demonstrated by the geometry of a bicycle saddle.

1. Introduction

The development of fibre-reinforced thermoplastics was strongly proceeding during the last years because they offer great advantages regarding short cycle capabilities, impact resistance, toughness and recyclability. However, fibre impregnation is challenging due to the high melt viscosity of the thermoplastic matrix. Commingled yarns provide a promising routine to introduce the polymer in forms of filaments into the composite structure resulting in very short impregnation paths. In our previous work we showed the manufacturing of glass fibre/SpinCOM yarns which are based on the principle of homogeneously distribution of continuous matrix filaments and reinforcing glass filaments during melt spinning [1]. Recently, thermoplastics with a wide application field such as polypropylene (PP), polyamide 6.6 (PA 6.6) and polylactic acid (PLA) have been processed to SpinCOM yarns using different sizings in the spinning process that determine the mechanical performance of the thermoplastic composites [2]. In the current work we present the processing of SpinCOM yarns using polybutylene terephthalate (PBT) as thermoplastic polymer in combination with E-glass fibres (GF) and different sizing formulations with the aim to achieve high mechanical performance by an appropriate fibre-matrix-interaction.

2. Experimental
2.1 Processing of SpinCom yarns

In contrast to other commingling techniques the SpinCOM yarns are produced by parallel spinning of glass and polymer filaments and commingling while passing the sizing applicator roll (Figure 1). Advantageously, the filament distribution homogeneity is reasonable high since commingling is done at a state where both, matrix and glass fibre yarns do not posses pronounced fibre integrity. The SpinCOM yarn integrity is later on achieved by applying a sizing. The mechanical load on the yarn during commingling is negligible low as compared to air jet texturing. Neither glass fibres are broken nor polymer fibres are stretched which results in high yarn strength and avoids thermal shrinkage during consolidation. Different sizings were applied during the fibre spinning as shown in Table 1, containing a silane and a film former that are compatible to PBT.

Two kinds of PBT filaments are produced with different melt volume rates (MVR) out of raw materials from two commercial suppliers: PBT1 with MVR = 46 cm^3/10 min and a molecular weight of M_w = 55 700 g/mol as well as PBT2 with MVR = 19 cm^3/10 min (for both: 250 °C; 2.16 kg) and M_w = 75 600 g/mol. The optimized processing conditions for a homogeneous mix of the GF and PBT filament arrays has been established after comparing different technological routes including the adaption of the spinning velocity and the use of applicable nozzles for the PBT spinning. The melt temperatures of GF and PBT are 1200 °C and 260 °C, respectively. The cooling behavior of the filaments controlled by the processing speed is crucial to determine the fiber diameter as well as the commingling and the winding process. PBT and glass filaments were spun at a speed of approximately 700 m/min with fiber volume fraction of 50 % and a fineness of about 160 tex. The cross sections of GF filaments show diameters of 16 µm and PBT filaments of 25 µm according to these processing conditions.

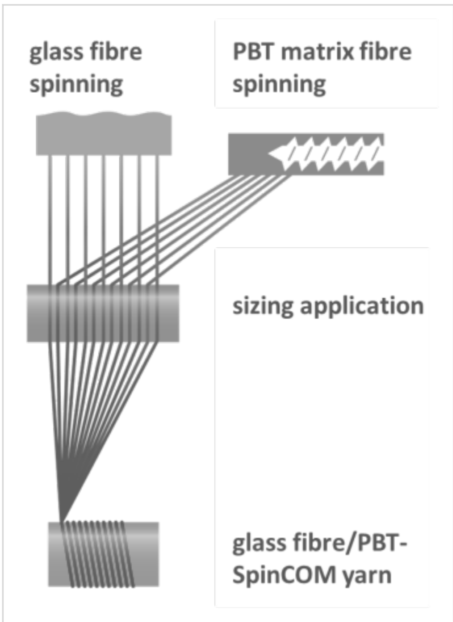


Figure 1. Schematic drawing of the manufacturing of glass fibre/PBT-SpinCOM yarns.

Table 1. Sizing formulations applied during GF/PBT-SpinCOM yarn processing.

Sizing	Sizing formulation	Matrix Polymer	Mechanical properties determined by	
			UD-composite	Single Fibre Pull-Out
V1	3-glycidoxypropyltrimethoxysilane, phenoxy film former	PBT1	X	
V3	3-aminopropyltriethoxysilane, polyurethane film former	PBT2		X
V4	3-aminopropyltriethoxysilane, epoxy film former 2	PBT1/PBT2	X	X
V6-3	3-glycidoxypropyltrimethoxysilane, epoxy film former 3 (low content)	PBT2	X	
V6-5	3-glycidoxypropyltrimethoxysilane, epoxy film former 3 (high content)	PBT1/PBT2	X	
unsized1	none	PBT1	X	
unsized2	none	PBT2	X	

2.2 Composite Manufacturing and Transverse Tensile Test

Filament winding of the SpinCOM yarns followed by isothermal compression moulding was used to manufacture unidirectional (UD) composites. The fiber volume fraction of the composites ranged from 49.7 % (V6-3/PBT2) to 56.8 % (V6-5/PBT2). The processing parameters were varied in terms of time, temperature and pressure to optimize the consolidation and achieve low void contents. Samples for the transverse tensile test were cut out of the composites and tested in accordance to DIN EN ISO 527-5.

2.1. Preparation and Testing of Single Fibre Model Composites

The interfacial adhesion strength was evaluated by single fiber pull out (SFPO) test [3][4]. Using an embedding equipment designed and constructed at IPF Dresden, the model micro-composites were prepared by accurately embedding one end of the single fiber in the matrix perpendicularly with a pre-selected embedding length l_e (target l_e = 150 µm) and an embedding temperature of 265 °C at controlled atmosphere. The pull out test was carried out on a self-made pull-out apparatus with force accuracy of 1 mN, displacement accuracy of 0.07 µm and a loading rate of 0.01 µm/s at ambient conditions. The force- displacement curves were detected and the resulting embedding length l_e was determined. After testing, the fiber diameter, d_f was measured by optical microscopy and the fractured surfaces was investigated by scanning electron microscopy (SEM). The interfacial parameters (apparent interfacial shear strength (IFSS) τ_{app}, local interfacial shear strength τ_d, frictional shear strength τ_f, were determined based on maximum force F_{max} and debonding force F_d according to our previous work [5]. Each fiber/matrix combination (GF with sizing V3 and V4 in PBT2) was evaluated in about 15-20 single tests.

3. Results and Discussion

After spinning the SpinCOM yarn bobbins the behavior during the textile processing was evaluated by the extent of fuzz formation during winding, which is highly depending on the sizing formulation and is of great importance for further processing in TFP. Figure 2 shows that sizing V1 leads to damage and yarn splicing during winding. In contrast, yarns with sizing V4/PBT2 reveal good textile processability.

Besides the bobbin quality, PBT2 was also found to lead to a higher level of the mechanical properties in the transverse tensile test (Figure 3). Also in the results of the composites made of SpinCOM yarns without sizing a strongly reduced standard deviation was found. For both PBT polymers, composites with the sizing formulations based on epoxy film formers achieved the highest transverse tensile strength indicating a high fiber-matrix-interaction.

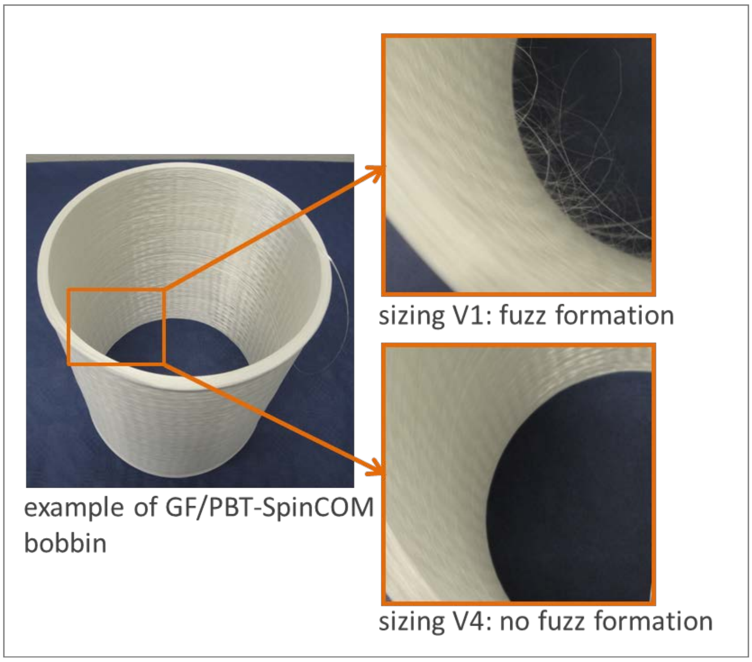


Figure 2. Comparison of SpinCOM yarn fuzz/friction properties depending on the sizing formulation (matrix filaments: PBT2)

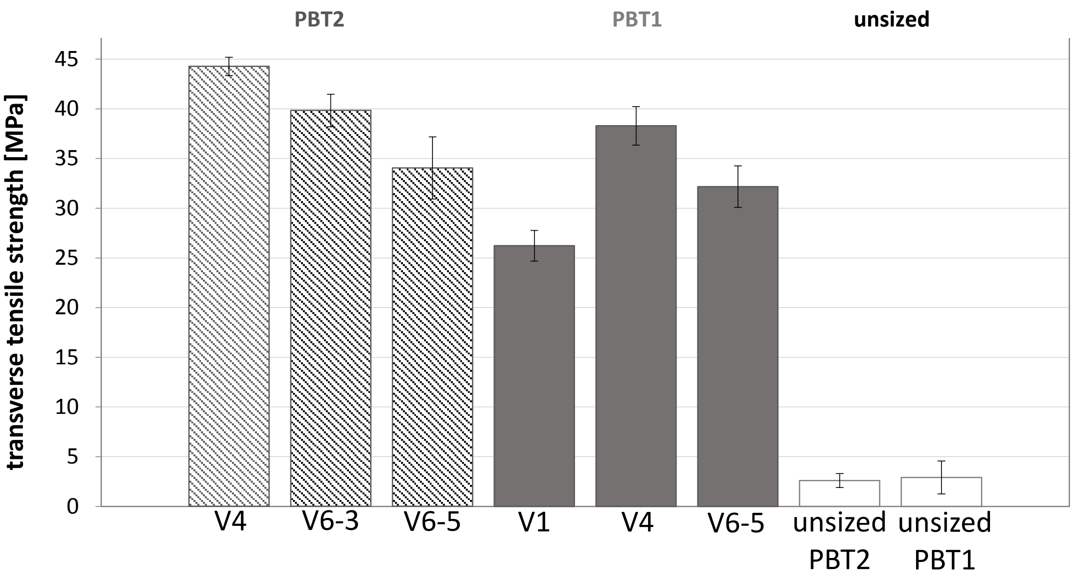
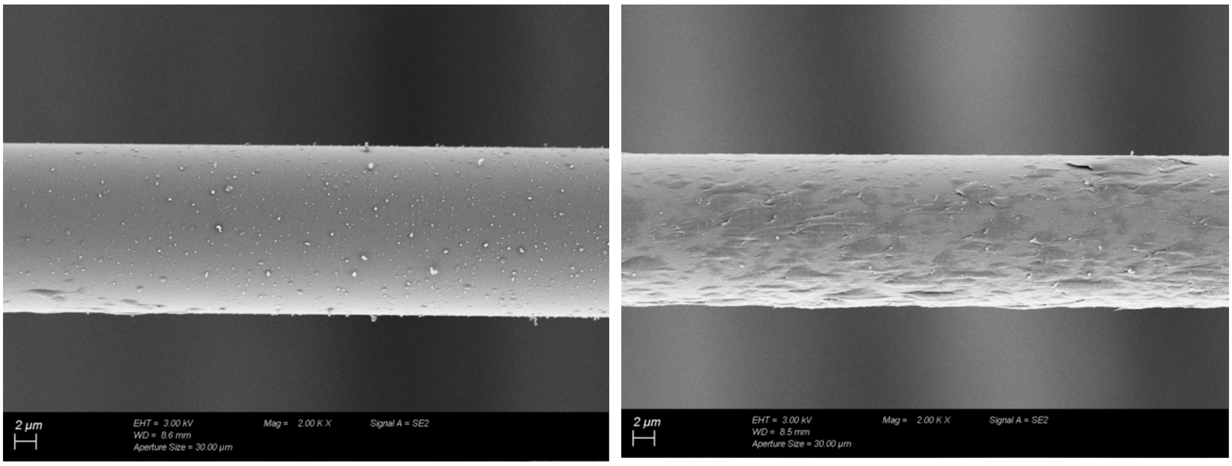


Figure 3. Transverse tensile strength of unidirectional GF/PBT composites depending on sizing formulation and PBT polymer.

The polyurethane based sizing V3 did not allow the manufacturing of UD-composites because of insufficient quality during winding. However, to enable the comparision of the interfacial shear strenght of polyurethane- and epoxy-based film formers micromechanical characterization by SFPO was applied. As already observed during UD-composite testing sizing V4 leads to good interfacial adhesion. Compared to model composites with sizing V3 the interfacial parameters of specimens with sizing V4 also increased (Table 2), whereby also the frictional shear strength τ_f was found to be enhanced. As shown by SEM-images (Figure 4) the crack path during pull-out was close to the fibre surface for specimens with sizing V3. In the case of sizing V4 a deformation of matrix-residuals on the fibre surface was observed indicating good fibre-matrix interaction, but also a more tough failure behavior. Furthermore, the remaining matrix on the surface leads to mechanical interlocking after debonding explaining the increase of τ_f.

Table 2. Interfacial parameters determined by SFPO test

Specimen	τ_{app} (MPa)	τ_d (MPa)	τ_f (MPa)
V3 in PBT2	27.6 ± 11.1	52.4 ± 19.4	4.9 ± 2.4
V4 in PBT2	34.7 ± 13.2	66.5 ± 24.8	6.3 ± 3.2



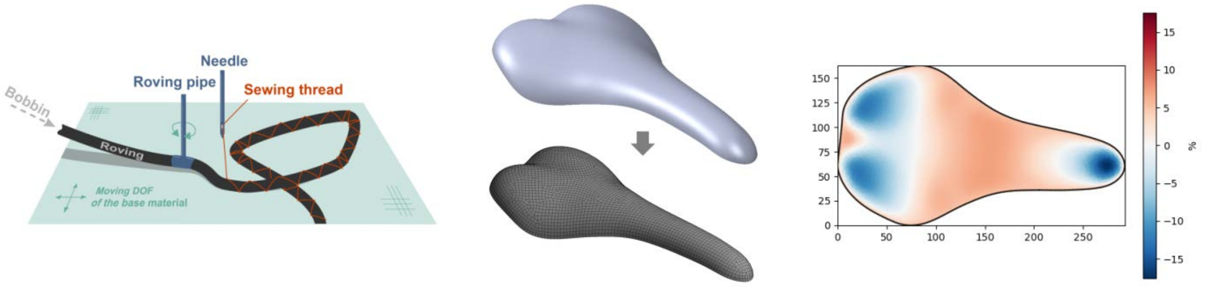
a) V3 b) V4

Figure 4. Comparison of fractured surfaces of glass fibres with a) polyurethane-based sizing V3 and b) epoxy-based sizing V4 after being pulled out of matrix PBT2 during SFPO test.

3. Design and processing of 3D thermoplastic composites

3.1. Optimization of 3D shaped variable-axial composites made by TFP

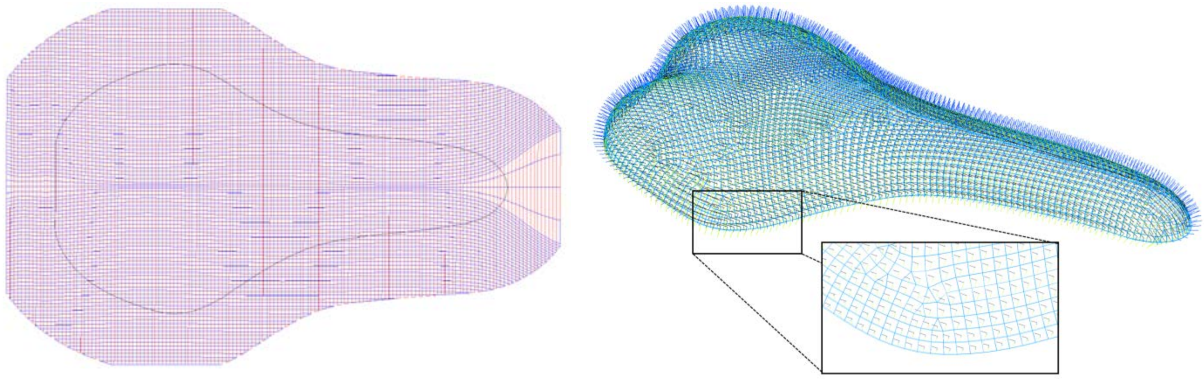
Do validate the processing of the SpinCOM yarn for textile preforms the Tailored Fiber Placement (TFP) technology was applied [7]. The principle of placing and fixing fibers with help of the TFP process is shown in Figure 5 a). As demonstration part a saddle geometry was chosen. The 3D model and the thereof derived finite element base mesh of the saddle geometry are shown in Figure 5 b). With help of the software tool *AOPS*, which was recently developed at IPF Dresden for the design of variable-axial composites, the 3D finite element mesh was flattened into a 2D domain (Figure 5 c)).



a) b) c)

Figure 5. a) Principle of the Tailored Fiber Placement process, b) 3D geometry and derived plane finite element mesh of chose saddle part and c) geometrical flattened shape

The resulting flat geometry was exported into the 2D-CAD format "DXF" and used as base shape to generate the TFP fiber pattern. This pattern, as shown in Figure 6 a), consists of two layers (red and blue). In a final step the initial generated base mesh was overlaid with this fiber path pattern. With help of *AOPS* tool a reverse transformation of the 2D pattern with locally derived element thicknesses and element orientations to a 3D finite element model was carried out [6]. With help of the so created finite element model with its highly complex anisotropic characteristics a numerical analysis can be carried out and a purposeful adaptation of the TFP pattern design can be realized.



a) b)

Figure 6. a) TFP path pattern and b) derived 3D finite element model with adapted element thicknesses and element coordinate systems according to the two layers of the TFP pattern

3.2. TPC consolidation molds made by incremental sheet metal forming

The aim of the incremental sheet forming (ISF) process is to generate a lightweight consolidation mold. The shape of the intended mold is formed by a sequence of kinematic movements of a trivial forming element (Figure 7). Thus, the geometry is not defined by the shape of the tool itself but moreover by the kinematic sequence of the tooling along the forming path.
An initial CAD data set was achieved by an optic 3D scan of a commercial saddle. Therefore, the functional surface is convex and the corresponding mold face needs to be concave. To avoid surface defects on the contact side of the mold, the shell was formed from the backside, which is convex again. This layout implies a partial support die. To ensure a minimum resource impact, this die was milled from a wooden block. On the final component, nearly perpendicular wall angles could be achieved. Different flange lengths were generated by a trimming process. This should enable a complete vacuum bagging solution (short flange) or a one-sided approach by application of a sealing on the elongated flange as an alternative. During the trimming operation, no distortion or twist of the

shell could be observed. Due to the double-curved surface, the shell does provide a good stiffness, stability and shape integrity.

Figure 7. Process chain of 3D composite manufacturing using SpinCOM yarns.

3.3. Vacuum bag based consolidation of hybrid yarn based preforms

The use of technical polymers such as PBT, PC, PPS or even PEEK require high processing temperatures that could be provided by an autoclave available at SIRRIS allowing processing temperatures up to 400 °C. Concerning the TFP technology, it has to be assumed that it is starting from a 2D architecture of a unique yarn to be developed during the consolidation phase in a 3D structure. So the use of an extra support is required for the handling and the cohesion of the yarn into the layer definition. Because of the support is closely linked to the reinforcement structure, it will remain present into the final part. Then it is needed to use a substrate material that is strong enough to support the TFP preform during its creation, to be manipulated between the different processing units. It also needs to be flexible enough to follow the 3D shape without causing extra complications in terms of wrinkle creation and finally that is compatible with the matrix of the TP composite. Currently proposed solutions are a glass woven fabric, a glass fleece and a PA based film. The first results of the via autoclave technology consolidated TPC saddle made of TFP preforms (stitched on PA based films) and GF/PBT2 SpinCom yarn is shown in Figure 7 (right) together with its corresponding ISF made mold. For the consolidation process a temperature of 235 °C and an additional pressure of 2 bar were applied. The red net-shaped surface results from a red colored commercial PBT stitching yarn, which was applied at the TFP process for roving fixation.

3. Conclusions

Summarizing the results of UD-composite and SFPO testing it can be stated that epoxy-based sizings have been shown to be an appropriate basis for further development of GF/PBT SpinCOM yarns. The combination of sizing-adapted SpinCOM yarn with the TFP technology allows high quality thermoplastic composites with precision placement and efficient material usage for preform production. Incremental sheet metal forming was applied to generate complex 3D shaped tools for thermoplastic composite consolidation; the consolidation process was carried out with help of a high temperature autoclave. Finally, the TPC consolidation process will be developed towards an out-of-autoclave manufacturing. Therefor vacuum bag technology with heated and easy to handle molds will be investigated in order to increase further cost-efficiency and energy saving as a novel strategy to design and produce composite structures with superior performance for lightweight applications.

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