

Development of PLA hybrid yarns for biobased self-reinforced polymer composites

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Abstract. Lightweight materials are a necessity in various industries. Lightweight design is in the key interest of the mobility sector, e.g. the automotive and aerospace industry. This trend applies also for the consumer industries, e.g. sporting goods. In addition, the worldwide demand for replacing fossil-based materials has led to a significant growth of bioplastics. Due to their low mechanical performance and durability, their use is still limited. Therefore, it is necessary to develop biobased, sustainable polymeric materials with high stiffness, high impact and high durability without impairing recyclability at a similar price level of non-biobased solutions. Biobased self-reinforced polymer composites offer these unique properties.

1. Introduction

In general, fibre reinforced composites consist of at least two different materials. A reinforcing material is embedded in a polymeric matrix material. Usually glass or carbon fibres are being used as the reinforcement material. These fibres carry the loads applied to the composites. The matrix mainly holds the reinforcing fibres in place and protects them from environmental conditions. Adding reinforcing fibres cause an increase in mechanical properties of the matrix phase. As a result, fibre reinforced composites combine the advantage of high mechanical properties (strength and stiffness) with low weight compared to metals such as steel or aluminium. [1]

Self-reinforced polymer composites (SRPCs) are a special type of composite. In SRPCs the same polymer is used for the reinforcing and the matrix phase [2]. Capiati and Porter introduced the concept already in 1975 [3]. Due to the use of a thermoplastic reinforcing fibre, SRPCs combine high impact and high durability. The density of SRPCs is lower compared to for example glass fibre reinforced composites. The recyclability of such composites is also increased by using the same polymer type as reinforcing and matrix material [4].

Self-reinforcing a polymer with fibres of the same polymer type results in an increase of the following material properties:

- strength,
- stiffness,
- durability,
- and impact behaviour [5].



The most significant improvement is observed for the impact behaviour [5]. SRPCs also offer superior fibre-matrix-adhesion compared to traditionally fibre filled polymers as fibre and matrix are made of the same material. The high chemical similarity leads to a strong composite as well as to a high nucleation density for the transcrystallization of the fibres into the matrix. The mechanical properties are strongly affected by the fibre-matrix-adhesion. Poor fibre matrix adhesion leads to peeling, cracking and reinforcement fibre pull-out. [5]

Using thermoplastic reinforcing fibres for self-reinforced composites also introduces some disadvantages. The main problem is the dependency of the reinforcing fibre's mechanical properties on temperature. Therefore, the processing window for consolidation is limited. It has to be precisely controlled in order to receive the best mechanical properties. The small processing window (consolidation temperature and cycle time) is the major challenge for producing SRPCs.

In order to reduce the cycle time and therefore heat exposure, hybrid yarns with short melt flow paths can be used. The reinforcing fibres as well as the matrix fibres are mixed on a filament level during the Commingling process. The use of commingled yarns allows the combination of a large variety of fibres and therefore a wide range of material properties. The melt flow paths of the matrix material are considerably lower compared to the Film-Stacking process [6]. COMFIL ApS, Gjern, Denmark currently produces SRPCs in form of high-tenacity PET mixed with low melting PET in form of yarn, fabrics, consolidated plates and rods/tapes. Self-reinforced Polypropylene (PP) is also available [7]. The production process of SRPCs using commingled yarns is illustrated in Figure 1.

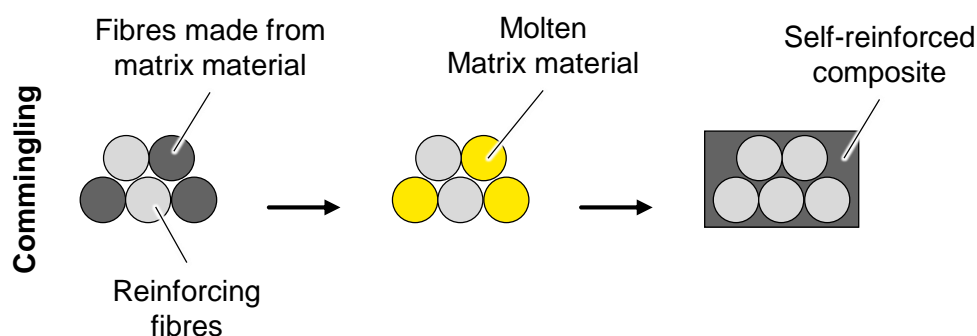


Figure 1. Consolidation process of commingled yarn based production process for SRPCs

2. Experimental

The material provided for the development of self-reinforced PLA is procured directly in filament yarn form as part of the EU project BIO4SELF [8]. Two project partners are developing the fibres in their research facilities. The reinforcing fibres (High T_m) are provided by Centexbel, the Belgian research centre for textiles and plastics, Gent, Belgium. The matrix filaments (Low T_m) have been acquired from the AMIBM Aachen-Maastricht Institute for Biobased Materials, Maastricht, Netherlands.

Both multifilament yarns (reinforcing and matrix fibres) are mixed using a modular commingling machine at ITA. A mixing ratio of 50:50 is chosen. The filament yarns are delivered separately and commingled inside a mixing box using an air jet nozzle. Compressed air is used to open and to intermingle the fibres. This results in the production of a hybrid yarn. The turbulences causing the mixing are controlled by increasing or decreasing the air pressure. The amount of filament breaks as well as production costs rise with increasing air pressures. Both multifilament yarns can be overfed. This influences the quality of the yarn (fibre distribution and orientation). The overfeeding is caused by the relative speed between the delivery godets and the take-off godet. In addition, the production

speed constitutes a relevant process parameter for the hybrid yarn quality as well as production costs. The production process is shown in Figure 2. The production speed, pressure and overfeeding rate are varied during the commingling trials to identify a parameter set that offers best properties for the commingled yarn.

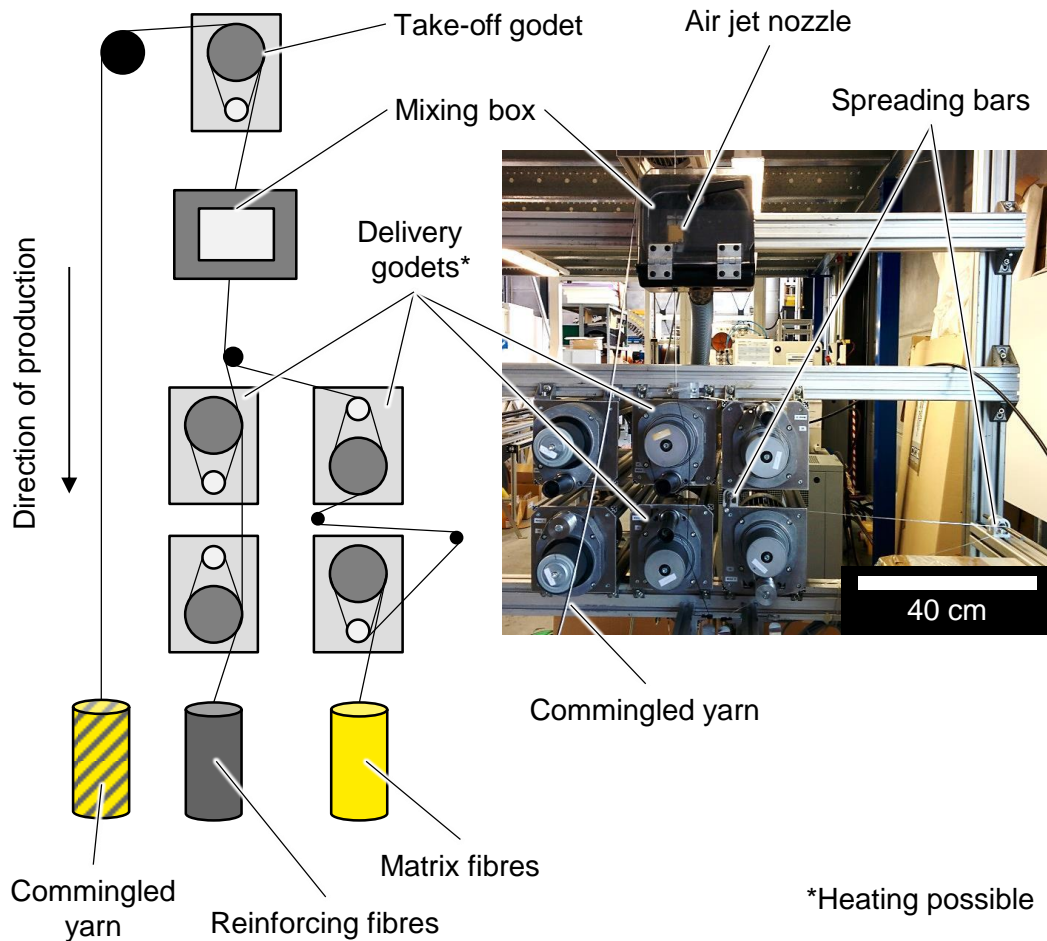


Figure 2. The commingling process

3. Results

The reinforcing fibres and matrix fibres used in this paper are characterised regarding their thermal and mechanical properties. Differential Scanning Calorimetry (DSC) from Mettler-Toledo GmbH, Greifensee, Switzerland has been used to analyse the melting temperature of the fibres. It was not possible to determine the melting temperature of the low melting PLA. Therefore, the melting temperature from the data sheet is given. The yarn count of the fibres is tested according to DIN EN ISO 2060. The tensile strength is analysed according to DIN EN ISO 2062 using a STATIMAT 4U from Textechno, Herbert Stein GmbH & Co. KG Textile Mess- und Prüftechnik, Mönchengladbach, Germany. The relevant fibre characteristics are presented in Table 1.

Table 1. Materials used for the experiments

	Low T_m PLA	High T_m PLA
SRPC phase	Matrix phase	Reinforcing phase
PLA grade	Ingeo TM Biopolymer 6302D	PURAPOL L130
PLA manufacturer	Nature Works	Corbion
Yarn count T_t [dtex]	$211,87 \pm 0,64$	$248,52 \pm 3,17$
Tensile strength σ_t [cN/tex]	$10,22 \pm 2,15$	$30,09 \pm 2,84$
Melting temperature T_m [°C]	125 - 135	176,99
Filaments produced by	AMIBM	Centexbel

The development of the hybrid yarns is successful. Reinforcing fibres and matrix fibres are doubled prior to the Commingling process in order to increase the yarn count of the hybrid yarn. In total 54 different parameter combinations are investigated. The first analysis of the hybrid yarns is a visual examination. The hybrid yarns are graded regarding the intermingling as well as the yarn structure (ondulation). The matrix fibres are coloured to ease the evaluation. An exemplary image of the hybrid yarns is displayed in Figure 3.

**Figure 3.** PLA-PLA hybrid yarns

The yarn count of the fibres is characterised according to DIN EN ISO 2060. The tensile strength is analysed according to DIN EN ISO 2062 using a STATIMAT 4U from Textechno, Herbert Stein GmbH & Co. KG Textile Mess- und Prüftechnik, Mönchengladbach, Germany. Three well performing hybrid yarns are chosen for UD-composite production. The production of UD-composites is possible. The manufacturing parameters and material properties of these hybrid yarns are displayed in Table 2.

Table 2. Manufacturing parameters and material properties of hybrid yarns

	A	B	C
Pressure p [bar]	6	6	6
Manufacturing speed v [m/min]	150	200	200
Difference of overfeeding rate between fibres $\Delta OR_{rf/mf}$ [%]	0,5	0,5	2
Yarn count T_t [dtex]	$2070,71 \pm 13,25$	$2070,10 \pm 8,52$	$2079,81 \pm 11,74$
Tensile strength σ_t [cN/tex]	$12,83 \pm 0,97$	$12,33 \pm 1,30$	$11,92 \pm 1,38$

4. Discussion

High production speeds during the commingling process can be used to produce hybrid yarns. The best mechanical properties of the three processing parameter sets displayed in Tab. 3 are received at low overfeeding rates (0.5 %) at a processing speed of 150 m/min. However, higher production speeds (200 m/min) do not lead to a significant loss in tensile strength. The impact of an overfeeding ratio of 2 %, between reinforcing and matrix fibre, also does not reduce the tensile strength of the hybrid yarn significantly.

A differentiation between the commingled yarns is difficult. Therefore, the production of UD composites from the hybrid yarns is required. The production of such composites is possible. The SRPCs are currently being tested according to DIN EN ISO 527-5. Previous results indicate high quality PLA SRPCs [9].

5. Conclusion and Outlook

The research conducted demonstrates the possibility to produce hybrid yarns for biobased SRPCs. Current research is conducted on the production of UD-composites to further investigate the quality of hybrid yarns. The next research step includes the evaluation of different mixing ratios of reinforcing and matrix fibres. In addition, the production of fabrics is investigated. The fabrics will also be consolidated and tested as well. The effect of the processing window of the consolidation process on the mechanical properties of the SRPC is going to be analysed. Further improvements on the material side may include adding nanofillers in order to increase mechanical properties. Additional research has to be conducted regarding the environmental impact of the biobased SRPCs.

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