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Extrusion as an energy-efficient manufacturing process for thermoplastic organosheets

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Abstract. Organosheets combine the advantages of reinforcement fibers and thermoplastic polymers. By pairing these two materials, composites with outstanding mechanical properties and low densities can be produced. These semi-finished products can be further processed into complex and functionalized components by thermoforming or injection molding. There are a number of different manufacturing processes for continuous fiber reinforced thermoplastics (CFRT), however, most of them require long production times and recurrent melting of the polymer resulting in high energy and manufacturing costs. This study presents a novel extrusion process, that enables a continuous production of reinforced thermoplastic sheets with only one melting step. Due to the high energy efficiency and wide range of processible materials, this process shows a high potential for an economical production of CFRT. To investigate the extrusion process in more detail, the influence of the processing and the flow behavior of the polymer on the impregnation quality and the mechanical properties of the composites were studied. The results showed increasing fiber volume contents with lower polymer viscosities. Furthermore, higher die temperatures and pressures resulted in higher fiber volume contents and thus in higher mechanical properties. The experiments also revealed that a complete impregnation can currently not be achieved without an additional small double belt press due to the line load of the calender, the high viscosity of the melt and the short impregnation time.

Introduction

Organosheets – continuous fiber reinforced thermoplastics (CFRT) – are semi-finished flat sheets that consist of continuous reinforcement fibers and a thermoplastic matrix polymer. The promising material class provides excellent weight specific mechanical properties [1] and can be recycled in contrast to conventional thermoset composites [2]. Due to their suitability for circular economy and the applicability of renewable and biobased fibers and polymers, continuous fiber reinforced thermoplastics enable sustainable lightweight construction [3]. Organosheets can be melted repeatedly [4] and further processed in highly automated mass production processes such as injection molding, thermoforming or other joining technologies [4].

Despite their high lightweight potential [2] and the wide range of well-suited processing options [5], organosheets have not yet been established on the market [6]. The main reason is the high price of the reinforced sheets [6], which is caused by high costs for the textiles [4], limited throughputs of common manufacturing processes [7] and intense energy consumption during the production [8]. To increase the economic attractiveness of the continuous reinforced thermoplastic composites new energy- and cost-efficient manufacturing processes must be developed [9]. In recent years, there have already been increasing efforts to optimize the production speeds of the conventional processes [5]. However, until now, the industrial usage of organosheets remains hampered by their high prices.

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One approach to reduce the costs during the manufacturing of CFRT is the reduction of processing and melting steps. State of the art are continuously or semi-continuously working presses that process plastic films or powder pre-impregnated fabrics [4]. These films have to be extruded or granules must be milled into powder prior to the production of the reinforced sheets. In a subsequent process step the semi-finished plastic product is molten again in a press and the impregnation occurs. First, macro-impregnation takes place and the voids between the fiber bundles are filled with melt. Due to the small gaps between the individual fibers, the wetting of the individual filaments – the micro-impregnation – occurs after the completion of the macro-impregnation. Finally, the thermoplastic composite is cooled and solidifies.

In contrast to the state of the art, the extrusion process enables a continuous impregnation of the reinforcement textile with only one melting step. The thermoplastic granules are molten by an extruder and are directly applied onto the dry fabric. Additionally, due to the direct processing of the granules, the process is suited for a wide variety of materials.

Several publications already discussed the influence of the process parameters such as pressure or temperature on the impregnation quality and the mechanical properties of organosheets. However, in these studies, only conventionally used presses were considered. Due to the line load between the calender rolls the extrusion process cannot be directly compared to these manufacturing processes and the existing models cannot be applied to describe the matrix flow into the fiber bundles.

Materials

Three different polypropylenes (PP) were investigated. The three polymers are typically used for injection molding and had comparable densities, melting and crystallization temperatures (Tab. 1). Differential scanning calorimetry (DSC 204 F1 Phoenix, NETZSCH-Gerätebau GmbH, Germany) with a heating rate of 20 K/min was used to determine the melting and crystallization temperatures.

Tab. 1 Overview of the polymers used in this work

	Borealis® BC612WG	Ducor DuPure® SR76	Ineos Rigidex® 450-HP90
Melt flow rate (230 °C / 2.16 kg, ISO 1133)	5 g/10 min	15 g/10 min	90 g/10 min
Melting temperature	175.4 °C	176.4 °C	175.4 °C
Crystallization temperature	106.1 °C	110.3 °C	119.3 °C

As can be seen in Tab. 1, the major difference between the polymers is the melt flow rate. To assess the different flow behavior in more detail, a rotational rheometer (Modular Compact Rheometer MCR 302, Anton Paar GmbH, Germany) with a cone-plate setup was used. The viscosity data for the different temperatures, shear-rates and polymers are shown in Fig. 1. All investigated polymers show a pronounced newtonian plateau and a shear thinning behavior at higher shear rates. Since shear rates below 100 1/s usually occur during calendering, it can be expected that the different viscosities at low shear rates influence the impregnation [9].

A glass fiber textile (style number 92130), supplied by Porcher Industries Germany GmbH, with plain weave was used in all experiments. The warp consisted of five 68 tex rovings and the weft of one 272 tex roving. The rovings consisted of e-glass filaments with a 9 µm diameter and had an areal weight of 390 g/m². The fabric was coated with a volan chromium complex finish (FK144) und had a finish content between 0.08 and 0.28%.

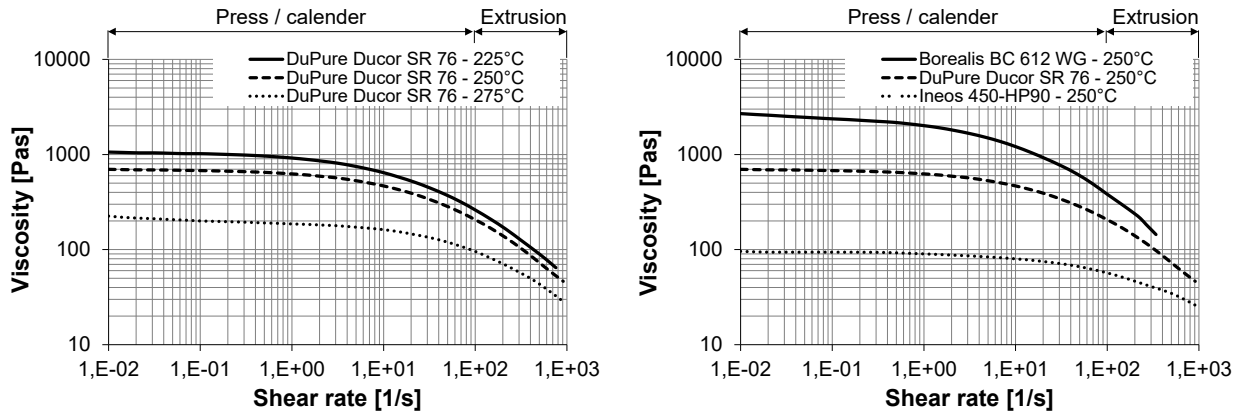

Fig. 1 Viscosity data for different temperatures (a) and polymers (b)

Experimental

The aim of this work was to investigate the potential of the extrusion process for the manufacturing of CFRT and to determine the influences of the flow behavior of the matrix polymer and the processing on the quality of the reinforced sheets. With the extrusion process, shown in Fig. 2, the polymer is applied directly onto the dry fabric and converted into a reinforced sheet. The granules are molten by a co-rotating twin-screw extruder. A melt pump separates the melt stream into two streams and ensures a consistent output. In the extrusion dies the melt is evenly spread and applied to both sides of the reinforcement textile to minimize the impregnation length. Due to the pressure of the calender rolls, the melt infiltrates the fabric and the macro impregnation between the rovings takes place. Because of the short impregnation time and the line load between the calender rolls, a complete micro-impregnation does not occur at this stage. An additional small double belt press can be used to ensure a complete wetting of all filaments. In contrast to the conventionally used double belt presses, this press only requires a reduced heating power, as the pre-impregnated organosheets enters the machine at an elevated temperature.

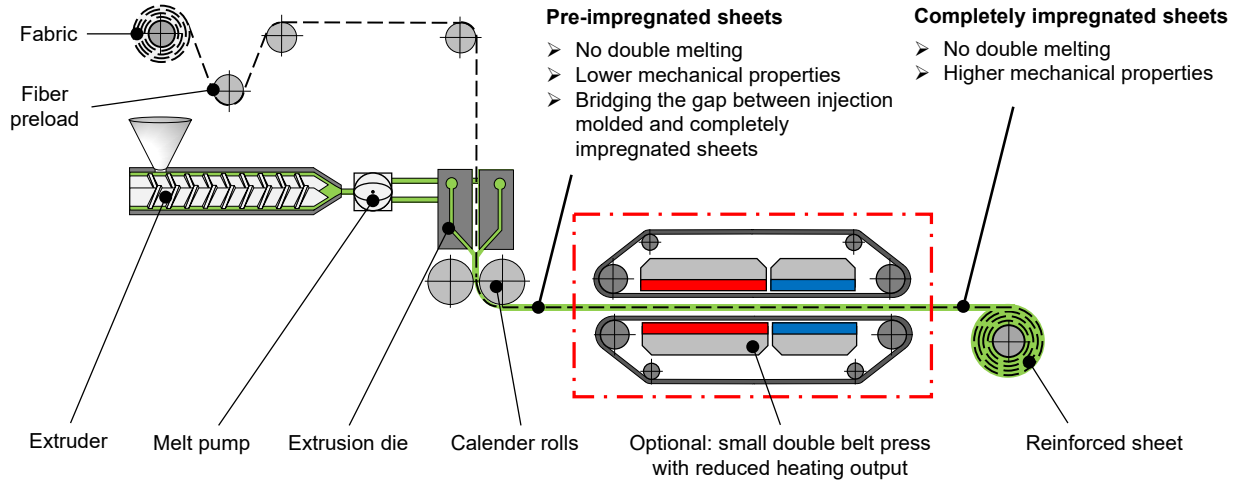

Fig. 2 Schematic of the developed extrusion line [10]

The focus of this study is the influence of the process and material parameters on the quality of the CFRT after the impregnation between the calender rolls. The key components of the extrusion line and the design of the extrusion dies are illustrated in Fig. 3. The dies are positioned directly in front of the calender rolls to minimize the temperature loss until the impregnation of the fabric. Furthermore, the position above the calender prevents an uneven influence of gravity on the melt.

The two dies have an identical, but mirrored design and redirect the melt before the stream exits the die to minimize the distance to the fabric.

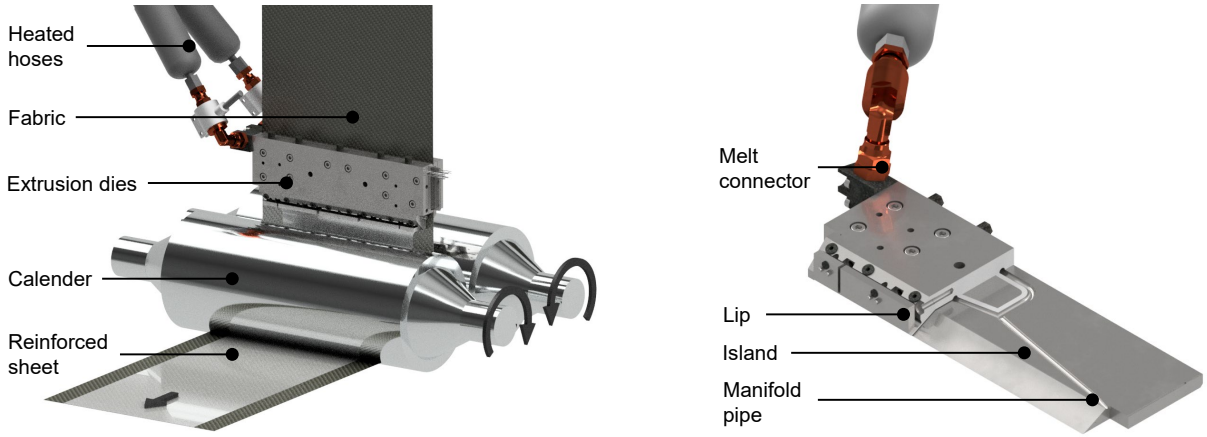

Fig. 3 Impregnation of the fabric (a) and design of the extrusion dies (b) [10]

In the conducted experiments a full factorial experimental plan was used and the die temperature and the pressure of the hydraulic cylinders driving the calender rolls were varied in two stages. The temperature was varied between 225 and 275 °C, while a hydraulic pressure between 5 and 9 MPa was used during the experiments. Furthermore, the center point of the experimental plan (250 °C, 7 MPa) was investigated to identify non-linear relationships. The material used was Ducor DuPure SR 76.

The influence of the flow behavior on the quality of the CFRT was investigated using three different matrix polymers (Ducor DuPure SR76, Borealis BC612WG and Ineos 450-HP90) at the same operating point. For the comparison of the materials, the center point of the full factorial test plan was selected with a hydraulic pressure of 7 MPa and a die temperature of 250 °C.

The haul-off speed and the extruder throughput were kept constant at 1.2 m/min and 3.1 kg/h in all experiments of this study. Previous studies showed that these two parameters have the greatest influence on the fiber and void volume content, as they directly affect the matrix-fiber-ratio [10]. However, it has been found that the impregnation quality is not influenced by these parameters [10]. The extruder temperature and the temperatures of the heated hoses were kept constant in all experiments, to prevent a thermal degradation of the matrix polymer.

Characterization

The impregnation quality of the obtained composites was evaluated by determining the fiber and void volume content. The combination of fiber and void volume content essentially defines the resulting mechanical properties of fiber reinforced composites for a given fiber orientation [11]. For each experiment ten rectangular specimens with the dimensions of 40 mm x 20 mm were cut out from the center of the composite and evaluated. Samples were taken every 500 mm along the haul-off direction. The weight and the density of the samples were measured with a laboratory balance (Sartorius MSU224S, Sartorius AG Germany, Germany). The burn-out test was carried out in a muffle furnace at 550 °C for a duration of 45 minutes. After the complete thermal decomposition of the resin, the weight of the fibers m_f was determined and the fiber volume content v_f was calculated according to equation 1.

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$$v_f = \frac{\frac{w_f}{\rho_f}}{\frac{w_f}{\rho_f} + \frac{1-w_f}{\rho_m}} = \frac{\frac{m_f/m_c}{\rho_f}}{\frac{m_f/m_c}{\rho_f} + \frac{1-m_f/m_c}{\rho_m}}. \quad (1)$$

Where w_f is the fiber mass content, ρ_f and ρ_m are the densities of the fibers and the matrix and m_c is the mass of the composite. With knowledge of the density of the composite ρ_c the void volume content v_v was calculated according to DIN EN 2564.

$$v_v = 1 - \left[w_f \cdot \frac{\rho_c}{\rho_f} + \left(1 - w_f\right) \cdot \frac{\rho_c}{\rho_m}\right] = 1 - \left[\frac{m_f}{m_c} \cdot \frac{\rho_c}{\rho_f} + \left(1 - \frac{m_f}{m_c}\right) \cdot \frac{\rho_c}{\rho_m}\right]. \quad (2)$$

Furthermore, tensile tests were conducted based on DIN EN ISO 527-4 (test specimen geometry type 2b). Ten rectangular specimens with the dimensions 250 mm x 25 mm were again cut out from the center of the composites every 500 mm for each experiment. All mechanical tests were performed in warp direction using a universal testing machine (Shimadzu SFL-50KNAG, Shimadzu Corp., Japan). All tests were performed at room temperature and at a constant testing speed of 5 mm/min.

Results and discussions

Fig. 4 shows the influence of the matrix polymer on the resulting fiber and void volume content. The comparatively high viscosity of the Borealis BC612WG results in a lower fiber volume content of 36%. The two polymers with a lower viscosity and therefore better flow show a higher fiber volume content around 40%, a further improvement with decreasing viscosity could not be determined. When examining the void volume content, no significant influence could be observed due to standard deviations around 1%. Although a higher fiber volume content can be achieved due to the better flow, the lower viscosity does not lead to a significant improvement of the void volume content or a better micro-impregnation. This observation may be attributed to the short impregnation duration while calendering. Due to the cooled calender rolls, the matrix polymer solidifies within a short time.

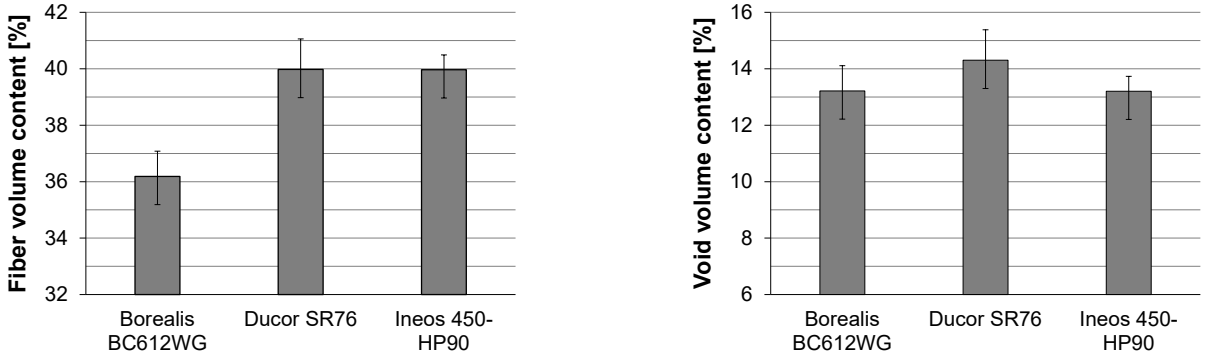

Fig. 4 Fiber (a) and void volume content (b) for different matrix polymers

The influence of the process parameters pressure and die temperature on fiber and void volume content are illustrated in Fig. 5 for Ducor SR76. Both parameters show a significant influence on the fiber volume content, while only the die temperature has a significant influence on the void volume content. The interactions between the two parameters were not significant. The statistical investigation was carried out with an error probability of 1%.

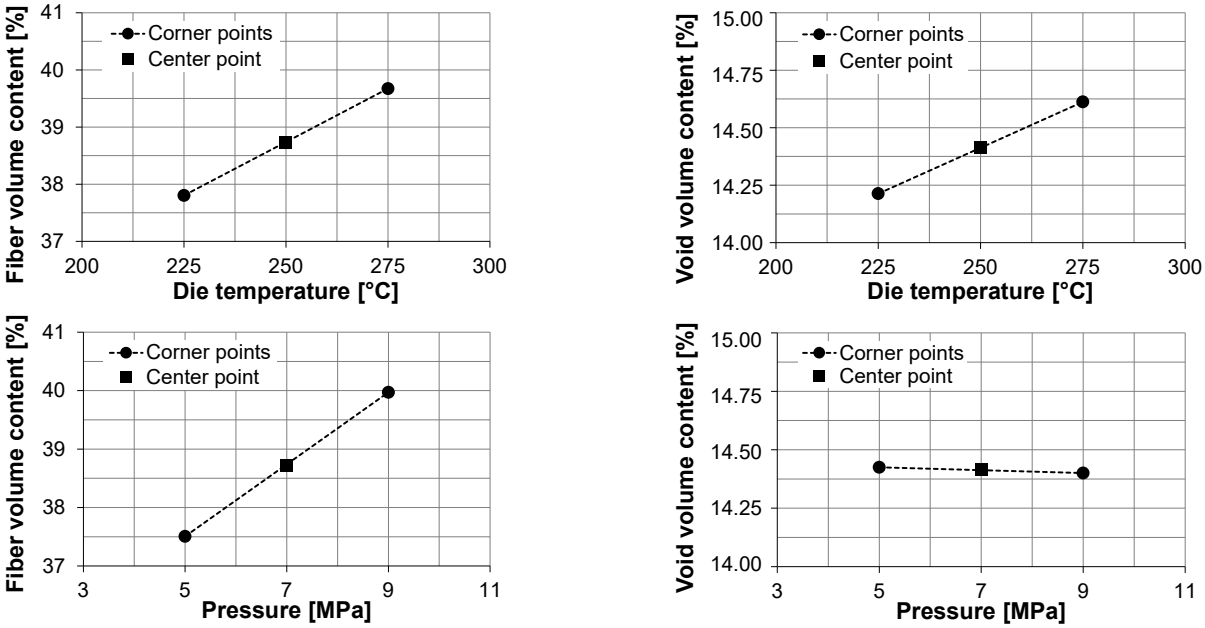

Fig. 5. Effects of die temperature and pressure on fiber and void volume content

A closer look at Fig. 5 shows that the change in pressure has the most distinct effect on the resulting fiber volume content, while a higher pressure does not significantly affect the void volume content. An increase in pressure from 5 to 9 MPa leads to a linear increase of 2.5% in fiber volume content. Contrary to the findings in the literature, a higher die temperature results not in a better impregnation [12], but in both a higher fiber and void volume content. A linear relationship can be observed for fiber and void volume content. At first impression, the higher melt temperature and consequently lower viscosity, as can be seen in Fig. 1, should also lead to a better impregnation and a lower void volume content. However, it turns out that during extrusion, the viscosity also affects the extrusion width. Due to the increased flow and the later solidification of the polymer, a wider melt film is formed in front of the calender rolls, which leads to a higher extrusion width. This reduces the matrix volume in the center of the composite, which results in an increase in the void volume content for identically sized pores. The change in fiber and void volume content is therefore primarily attributed to the changed dimensions and not to better impregnation. Since the width of the extruded sheets also increases at higher pressure, the effects of better impregnation and higher width seem to compensate each other.

The fact that changes in fiber and void volume content primarily results from the changed dimensions and not from changed impregnation can be confirmed when analyzing the relationship between extruded width and fiber volume content. Fig. 6 shows the fiber volume content in context to the normalized width, which corresponds to the ratio between extruded width and die width. Regardless of the material investigated, process and material parameters, which result in a larger extruded width due to better flow behavior and later solidification, also lead to a higher fiber volume content. A linear correlation between the width and fiber volume content can be identified.

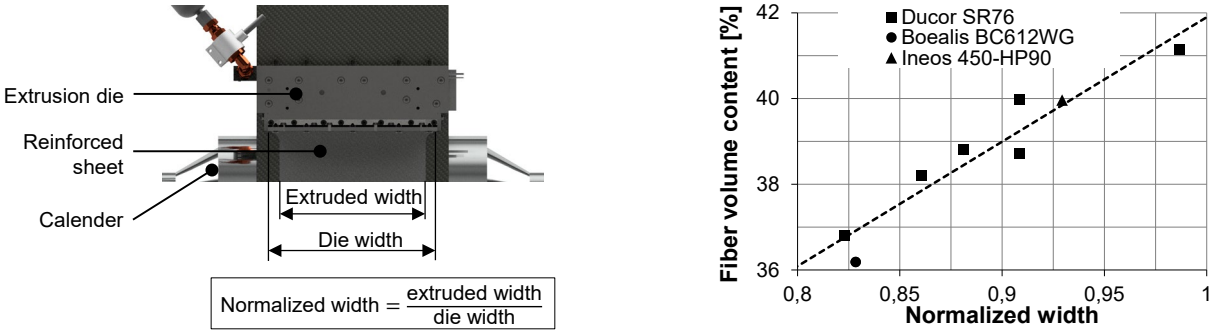

Fig. 6. Definition of the normalized width (a) and correlation between normalized width and fiber volume content (b)

The results of the mechanical characterization show a dependency of the mechanical properties on fiber volume content (fig. 7). Deviations can be attributed to the different mechanical properties of the polymers and slightly varying void contents and fiber waviness of the specimens.

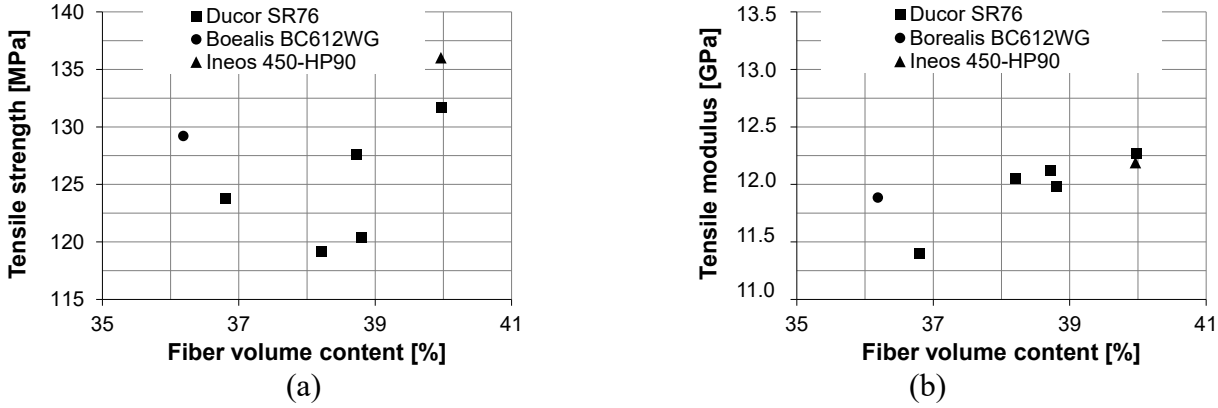

Fig. 7. Tensile strength (a) and tensile modulus (b) as functions of the fiber volume content

The tensile strength ranges from 120 MPa to 137 MPa, while the tensile modulus ranges from 11.4 to 12.3 GPa. A higher fiber volume content leads to both a higher tensile strength and modulus concurrent to the rule of mixtures. Compared to fully impregnated and consolidated organosheets [1], the extruded CFRT show around 33% lower mechanical properties. This can partially be attributed to the respective fiber volume contents. Additionally, the void volume contents of the extruded CFRT are higher and the impregnation is incomplete. Due to entrapped air in the fiber bundles, the force transmission between the fibers is disturbed, leading to premature failure. Further, the individual fibers are not perfectly aligned during the mechanical testing as a slight displacement of the fibers can occur during the extrusion. Previous investigations suggest that sizing and adhesion promoters also have a significant influence on the mechanical properties [2].

Conclusions

This study demonstrates the high potential of the extrusion process for the continuous production of CFRT. The extrusion enables an energy-efficient and economically reasonable manufacturing of pre-impregnated sheets by sparing one melting step. Due to the short line load between the calender rolls, fiber impregnation is still incomplete, even with low viscosity polymers. The investigations of process parameters showed that higher pressures and processing temperatures lead to increased fiber volume contents. Varying flow behavior and pressure, influences the melt film before the calender rolls and the width of the composites. Both tensile modulus and -strength tend to increase with higher fiber volume contents. Compared to fully impregnated organosheets, the extruded and not completely consolidated composites still have lower mechanical properties

even at a high fiber volume content of 40%. To achieve complete impregnation, an additional small double belt press can be used, which can be operated with reduced heating power. Due to their lower costs, the pre-impregnated sheets might be able to bridge the price and performance gap between short fiber reinforced injection molded parts and fully impregnated sheets.

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