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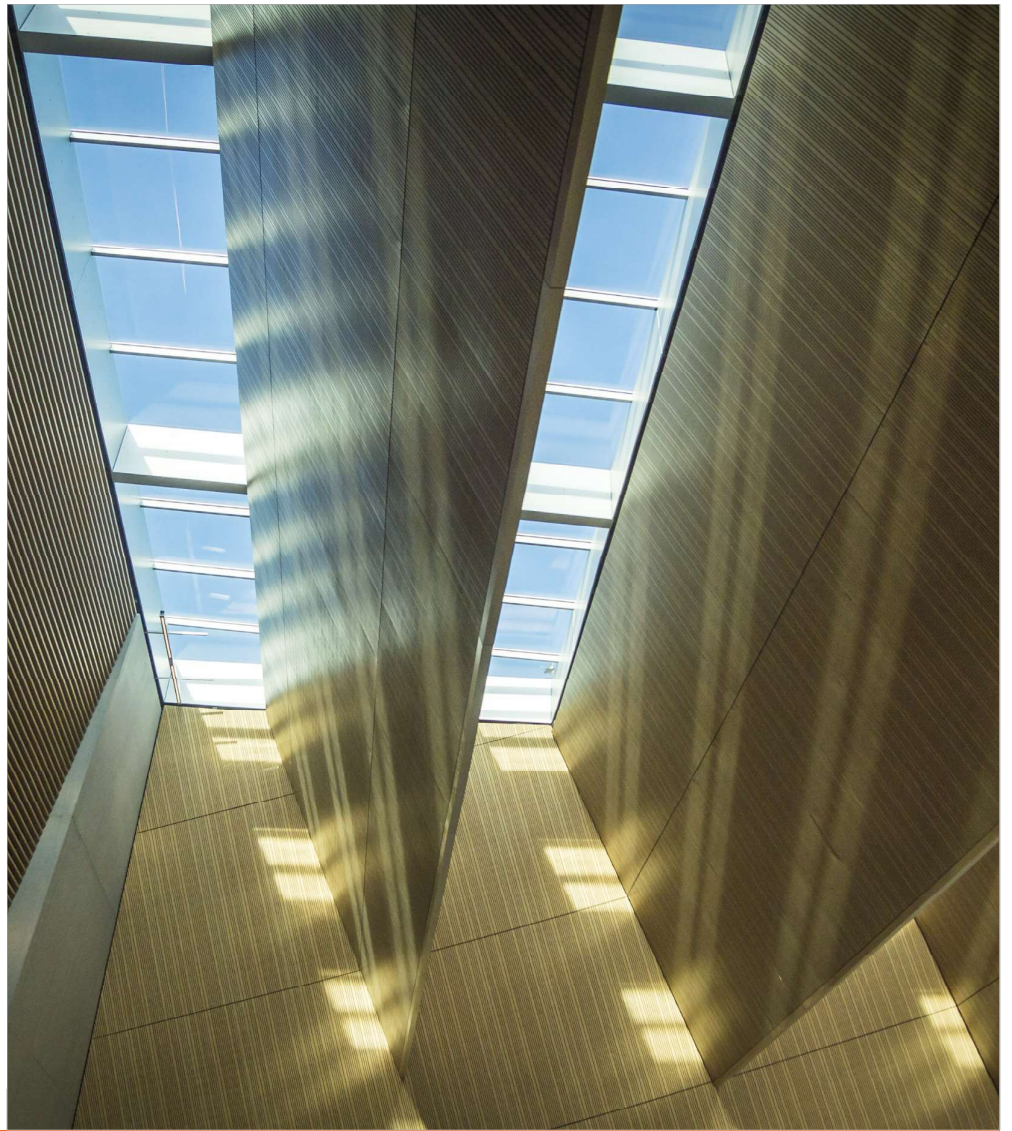
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CONSOLIDATION OF HYBRID TEXTILES FOR AEROSPACE APPLICATIONS

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Abstract: *Side-by-side hybrid textiles consist of layers of woven reinforcing fibres and flexible thermoplastic layers alternatively stacked on each other. They possess an elevated design freedom as many properties can be easily and locally changed. Side-by-side hybrid textiles can be consolidated with press molding to produce near-net shape final parts. Appropriate process parameters and local tailoring of the composite properties would enable to fully exploit their potential and create very high-performance, defect-free parts. However, the prerequisite for such an optimization approach is a three-dimensional consolidation model, which is not available yet. As a first step, we propose an impregnation model for hybrid textiles which considers air entrapment and dissolution and demonstrate that these two effects greatly influence impregnation. The model captures all relevant phenomena and is shown to be able to properly predict impregnation, and thus porosity.*

Keywords: Hybrid textiles; Impregnation; Press molding; Modeling; Thermoplastic Composites

1. Introduction

Fibre-reinforced thermoplastics (FRTP) display several advantages over the more widespread thermoset-based composites such as a potential for high volume production, better impact resistance and an ability to be recycled [1]. To produce FRTP, the base constituents are usually first architected to an intermediate material which is then consolidated in the final part in a subsequent step. The intermediate material can be classified according to its degree of mingling, for which film stacking, co-woven fabrics, commingled yarns and organosheets can be cited as examples with increasing degree of mingling.

As a rule of thumb, a higher mingling reduces the flow length and the cycle time but increases the costs. In addition, pre-consolidated materials and film stacking do not possess the drapeability inherent to textiles, which significantly decreases the geometrical complexity that can be achieved. Furthermore, the limited range of organosheets available on the market currently reduces the design freedom.

Hybrid textiles are an alternative intermediate material in which layers of woven reinforcing fibres and a flexible thermoplastic layer are alternatively stacked on each other as represented in Figure 1. The thermoplastic layer can take the form of a veil or woven thermoplastic fibres to conform to the drapeability of the reinforcing fibres. Recently, Reynolds investigated stamp forming with hybrid textiles based on veils and produced structural parts within a cycle time of 330 s. [2], proving the relevance of such a process for the industry.



Figure 1: Picture of a partial cutaway of quasi-UD carbon / PEI hybrid textile manufactured at the Faserinstitut Bremen.

An advantage of hybrid textiles is their vast design freedom, as many parameters such as the fabric architecture, orientation or fibre volume fraction could be easily and even locally changed. An adequate selection of the parameters would therefore allow to locally tailor the composite properties and fully exploit their potential. We therefore propose press moulding of hybrid textiles to produce semi-complex shapes towards high-end applications.

This approach requires a consolidation model to avoid the formation of unwanted defects such as porosity or fibre disorientation, which is, however, not available yet. As a first step towards the creation of a complete consolidation model, an impregnation model must be selected and validated. Thereby, not only the overall impregnation behavior should be well described but also the remaining porosity as it has a detrimental impact on mechanical properties [3]. Several authors addressed the impregnation of fabrics, such as Kobayashi [4] and Groupe [5]. They both considered the impregnation of elliptical tows and based their approach on the model of Van West [6], thereby the air in the tows was assumed free to escape. Other authors proposed other similar approaches [7, 8], yet no one validated their model from the beginning to the very end of impregnation. This is unfortunate, as entrapped air mostly influences the end of impregnation and the residual void content as demonstrated by Rozant [9]. However, as the diffusion of air into molten thermoplastic was disregarded his model predicts a constant residual porosity while measurements showed a decay instead.

In this study, we propose to extend existing impregnation models with air entrapment and dissolution. We demonstrate that entrapped air dissolution needs to be considered to properly model the impregnation behavior. Glass quasi-unidirectional (UD) fabric and polypropylene (PP) or high-density polyethylene (HDPE) are chosen as model materials for the investigation in the form of film stacking, since once molten barely any difference is thought to remain when compared to other flexible thermoplastic plies. A validation of the model with high-temperature polymers and veils is planned as further step.

2. Materials and methods

A woven glass fibre fabric with a quasi-UD fabric architecture and an areal weight of 931 g m² provided by Tissa Glasweberei AG, as described and characterized in [10], was used in this study. The tows in the weave direction have 2400 tex with and a density of 3.5 tows cm⁻¹. Micrograph analysis was performed; the tow fibre volume fraction was measured to be 0.77 and the fibre diameter to be 9 µm with a standard deviation of 0.8 µm. The packing of the fibres can be overall well described with a hexagonal arrangement.

Two thermoplastics were investigated in this study, PP BJ100HP from Borealis and HDPE Lupolen 5031L from LyondellBasell. The technical datasheets specify that the PP has a melting temperature of 165 °C and a density of 906 kg m⁻³ in solid state while these values are respectively 131 °C and 952 kg m⁻³ for the HDPE. The polymers were obtained in the form of granulates and have been processed in 0.15 mm thick foils with a Collin FT-E20T-MP extruder.

5 plies of textiles were film stacked with two polymer foils between each layer and one on the top and bottom to target a final fibre volume fraction of 0.45. They were then pressed into 170 x 85 mm plates with a 200 kN Vogt hydraulic press. The mould was placed in the heated press with spacers to avoid pressure on the composite during heating up. Upon reaching the desired cavity temperature the spacers were removed and pressure was applied for different durations before cooling down under the same pressure. For each thermoplastic the experiment was repeated at different pressures as summarized in Table 1. The plates were then cut with a Compcut 200 circular saw and finally photographed to analyze the impregnation behavior over the whole surface. A precision ruler was photographed along each picture to determine the pixel size. The software ImageJ was used to manually select the dry areas and the whole tow and to measure properties such as the tow width, height, area, and impregnation degree. At least 30 measurements were performed for each plate.

Table 1: Summary of the test matrix

Thermoplastic	Cavity temperature [°C]	Cavity pressure [bar]	Press time [mn.]
PP	185	10.4 / 17.3	0/2/5/15/30/60
HDPE	155	35 / 69	0/2/5/15/30/60

3. Consolidation model

An analytical approach based on the model of Van West [6] is adopted, which propose to solve the impregnation for a cylinder equivalent to the ellipse as shown in Figure 2. The equivalent radius is given as:

$$R_{eq} = \sqrt{2} \frac{l_w l_h}{\sqrt{l_w^2 + l_h^2}} \quad (1)$$

Where l_w and l_h are the semi major and minor axis of the ellipse. The impregnation can be solved analytically for cylinders as follows:

$$r \ln \left(\frac{r}{R_{eq}} \right) \dot{r} = \frac{K_{tow}}{\eta (1-\nu_f)} (P_m - P_g) \quad (2)$$

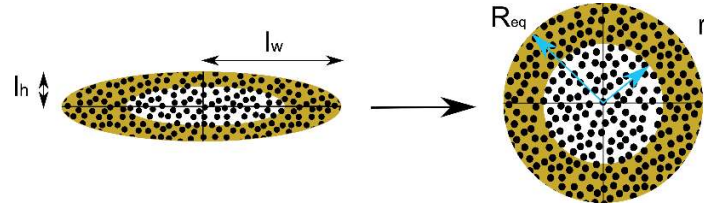


Figure 2: Representation of a partially impregnated tow and its equivalent cylinder according to Van West [6].

Where r is the radial distance to the impregnation front, R_{eq} the equivalent tow radius, K_{tow} the tow permeability, η the viscosity, ν_f the tow fibre volume fraction, P_m the matrix pressure boundary on the periphery of the tow and P_g the gas pressure. The reader is redirected to [11] for more information. The tow permeability is predicted with the equation of Gebart for hexagonal packing [14] and the impregnation degree ξ is defined as:

$$\xi = \frac{R_{eq}^2 - r^2}{R_{eq}^2} \quad (3)$$

Assuming mechanical equilibrium between the applied pressure and full saturation of the inter-tow space, the fibre bed stress response and the matrix pressure yields:

$$P_{mat} = P_{applied} - A e^{\nu_f B} \quad (4)$$

Where P_{mat} is the matrix pressure, $P_{applied}$ the applied one and the term $A e^{\nu_f B}$ relates to the quasi-static stress response of the fabric, which was characterized in [11]. Following the ideal gas law, the air pressure is given as:

$$P_g = \frac{n R T}{V} = \frac{n R T}{A_{tow}(1-\nu_f)(1-\xi)} \quad (5)$$

Where A_{tow} is the tow surface and n the amount of gas in moles trapped in the tow per unit depth. Assuming that all the air in the hybrid textile is pushed inside the tows in the first moments of consolidation when the meso structure collapses as shown in Figure 3, the initial amount of air trapped in each tow reads:

$$n_0 = \frac{P_{atm} A_{tow} (1-\nu_0) \nu_f}{R T_0 \nu_0} \quad (6)$$

Where it is assumed that the amount of air initially present in the hybrid textile and in a textile stack with the same number of layers of the reinforcing ply at rest are equal. In Equation (6) P_{atm} is the atmospheric pressure and ν_0 is the fibre volume content of the textile stack at rest.

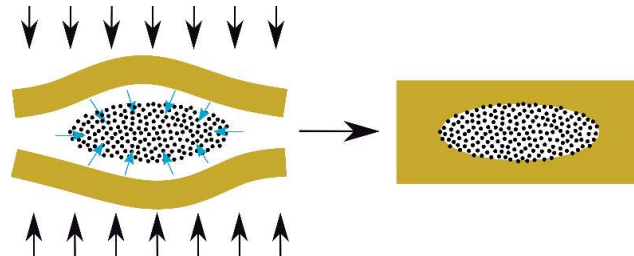


Figure 3: Schematical representation how additional air gets entrapped in the tow. The initial situation is represented in the left, as pressure is applied the molten thermoplastic fills the inter-tow space and pushes air into the tow.

The dissolution rate \dot{n} is obtained by modifying the equation proposed by Epstein and Plesset [12] for bubbles in pure molten polymers by adding a correction factor G to the diffusivity to consider that the fibres are impermeable and disturb dissolution, as schematically represented in Figure 4. Thus:

$$\dot{n} = 2 \pi r J = - 2 \pi r G D (C_s - C_\infty) \left(1 + \frac{r_i}{\sqrt{\pi G D t}}\right) \quad (7)$$

Thereby, J is the flux of species, D the diffusivity, t the time elapsed since the start of consolidation and C_s the saturation concentration at the bubble interface and C_∞ the initial concentration of the molten polymer which is assumed to be the saturation concentration at atmospheric pressure. The diffusivity of PP and PE, respectively $5e-9$ and $6e-9$ $m^2 s^{-1}$, and Henry's constant, respectively $3.61e-5$ and $4.28e-5$ $mol Pa^{-1} m^{-3}$, were taken from [13].

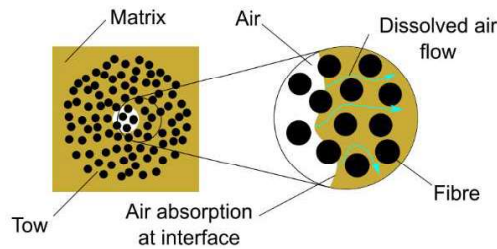


Figure 4: Schematical representation of air entrapment and dissolution. The air dissolves at the flow front interface and diffuses outwards, thereby the fibres obstruct the species flow.

4. Results

Figure 5 shows a photograph of partially impregnated plate cross-section, displaying regularly arranged tows surrounded in polymer without apparent porosity. One can distinguish between the impregnated and dry region of the tow, which has a darker shade. The impregnation front shape is overall centered in the tow with an elliptical shape, which corresponds quite well to the analytical predictions.

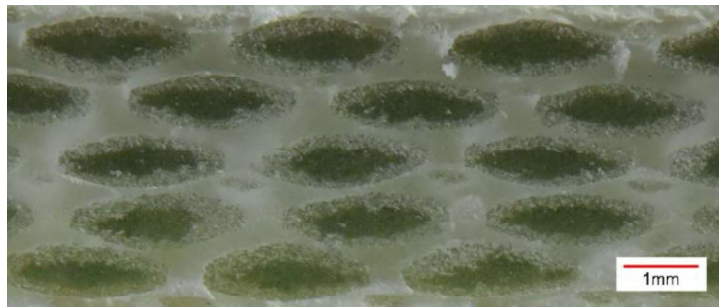


Figure 5: Picture of the cross-section of a PP plate pressed at 10.4 bar for 5 minutes.

It was observed that that the impregnation degree raises sharply at the beginning, then flattens significantly. This can be observed in Figure 6 where the measurements for PP pressed at 10.4 bar are displayed along with different predictions. Thereby, as the cooling time required for polymer solidification was measured to be 2 minutes, the total consolidation time is approximated as the press time plus one minute. The proposed model is found to satisfactorily predict the impregnation degree, thereby the parameter G was fitted using a simplex search method. Using the same model and neglecting air entrapment and dissolution, as many of the models available in literature do, results in a complete failure to predict impregnation as soon as flattening starts to occur. If diffusion is neglected and only the air originally in the tows is

entrapped, meaning that no air migrates from the inter-tow space into the tow, the predictions significantly overshoot. This tends to indicate that more air is trapped inside the tow to counteract impregnation and taking into account air migration is necessary.

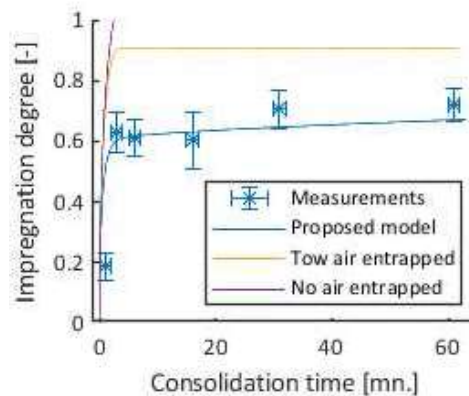


Figure 6: Measured impregnation degree in function of the time along with the predictions of the proposed model, the predictions of the same model if only the air originally in the tows is trapped without diffusion and the predictions if air entrapment is neglected.

The proposed model with appropriate parameter value describes the impregnation kinetics in a satisfactory manner. The value of G was found to be $8.1 \cdot 10^{-4}$, meaning that the fibres do very significantly hinder diffusion or that the diffusion coefficient taken from the literature is overestimated. Figure 7 displays the results for the experiments with HDPE with the same value for G .

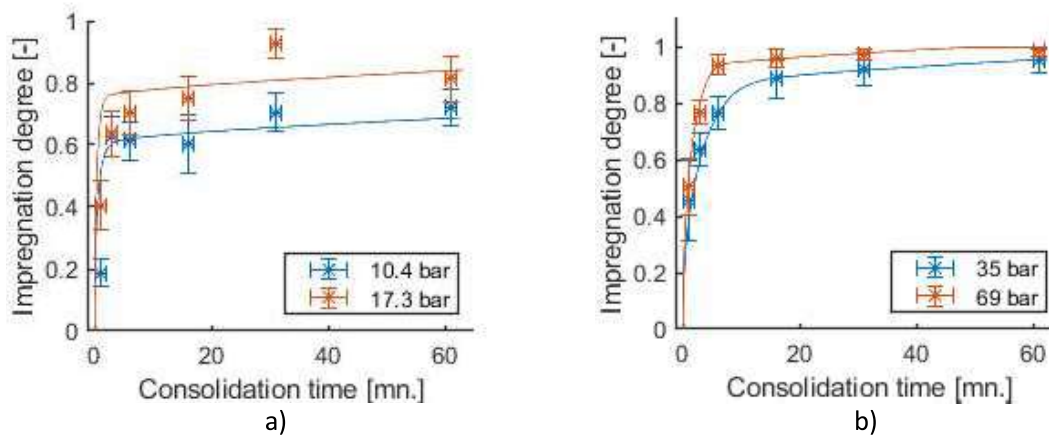


Figure 7: Measured impregnation degree in function of the time along with the predictions of the proposed model with PP as matrix in Figure 7a and HDPE as matrix in Figure 7b.

5. Discussion

The proposed model was found to be able to correctly describe the impregnation behaviour given appropriate parameter selection. The tow permeability predicted by the model of Gebart [14] was found to be well-suited to the measurements and the assumption that the whole air originally present in the textile remains trapped explains well the onset of the measured flattening of the impregnation degree. This effect coupled with diffusion can explain the large differences between the predicted time for full consolidation and those obtained in practice [5]. In this study the correction factor G for the reduced diffusion in porous structures was measured

empirically, however in future research the validity of existing approach for similar problems should be investigated [15].

This research sheds a new light on the underlying processes taking place during impregnation and allow a comprehensive understanding on how the different parameters affect it, which should be examined in detail. For instance, while previous models held that doubling the tow radius quadruples the impregnation time, it should be investigated this relation changes when air entrapment and dissolution are considered. Many of the parameters in this model depend on either pressure or temperature, not only the viscosity but also the saturation concentration and the diffusivity, which exponentially depends on temperature [13]. The optimal set of process parameters for impregnation is still unclear and should be investigated. Since entrapped air has such a predominant effect on residual porosity and on the impregnation time, air removal strategies and the possibility to press under vacuum should be evaluated.

In further work, the model will be validated with high-performance hybrid textiles and integrated into a three-dimensional consolidation model. Characterization of the solubility and diffusivity of gas in molten high-performance thermoplastics is completely lacking and should be performed as these values are required in the impregnation model.

6. Conclusion

This study investigated the impregnation behaviour of hybrid textiles made with a glass fibre quasi-UD textile and either PP or HDPE foils. We propose a novel impregnation model taking into account air entrapment and dissolution. We show that this is necessary for the correct prediction of the late-stage impregnation behaviour, which current impregnation models fail to predict. The proposed model was found to be able to correctly describe the impregnation behaviour with appropriate parameter selection of the diffusion reduction factor.

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