A CONSOLIDATION PROCESS MODEL FOR FILM-STACKING GLASS/PPS LAMINATES

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SUMMARY

The applied pressure, processing temperature and holding time influence the consolidation of thermoplastic laminates. A model to optimise the processing parameters is proposed. The influence of heating rate, processing temperature and pressure is investigated. Short textile impregnation times, in the order of seconds, are predicted. The model is validated in an experimental programme.

Keywords: thermoplastic composites, film stacking, process modelling

INTRODUCTION

The application of thermoplastic polymers as a matrix material in fibre reinforced composites has grown steadily over the last decades [1]. Thermoplastic composites offer clear advantages over their thermoset counterparts in terms of improved fracture toughness, potential for recycling and, most notably, the possibility to reshape the product at higher temperatures [1,2]. The latter allows rapid processing techniques, such as stamp forming or rubber pressing, with short cycle times and potentially low costs.

The production of fibre reinforced composites requires the impregnation of a fibrous reinforcement with a resin. A major disadvantage of using thermoplastic polymers is their high viscosity, in the range of 200-600 Pa.s for the polyphenylenesulfide (PPS) at processing temperature used in this study, which makes textile impregnation more difficult. The impregnation can be eased by minimising the distance required for the matrix to flow. This can be achieved by mixing the two constituents before processing [3], for which several approaches have been developed; examples are film-stacking [4], solvent impregnation [5] or the use of commingled yarns [6].

The present study focuses on the production of flat textile reinforced thermoplastic laminates using a film stacking procedure. Ten Cate Advanced Composites produces the specific laminates under the trade name Cetex®. The laminates serve as a semi-finished product and obtain their final shape using stamp forming or rubber pressing. Figure 1 schematically shows the film-stacking procedure. A stack, comprising alternate plies of reinforcing textile and thermoplastic film, separated by steel or aluminium tinplates or TPFE foil, is placed between two press platens. The production process consists of three steps: 1. heat the press to lower the matrix viscosity, 2. increase the pressure to force the thermoplastic matrix material to impregnate the textile, and 3. cool the press to solidify the laminates.

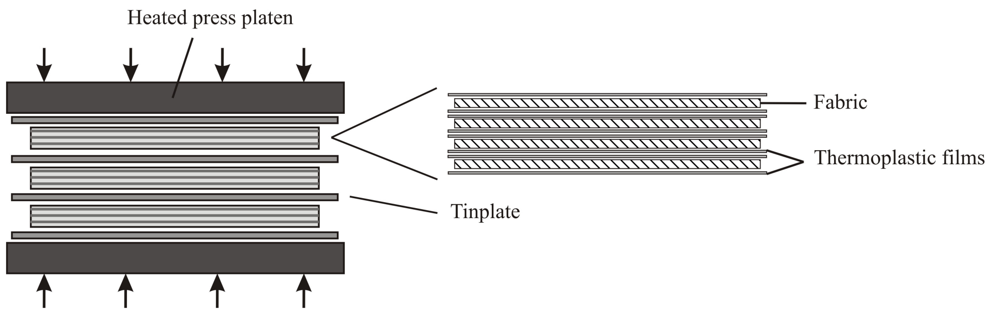


Figure 1. Film-stacking production process for three thermoplastic composite laminates

Currently, the processing parameters such as heating rate, processing temperature and pressure are determined using a trial and error procedure with extensive material inspection. The development of a predictive process model could provide an inexpensive and fast alternative to optimise the processing parameters.

The development of such a process model is subject of the present paper. The first section presents a short experimental study to qualify the different stages in which consolidation takes place. The second section introduces a modelling approach treating the thermal heating part of the process and the impregnation part separately. A transient thermal model predicts the through-thickness laminate temperature as a function of time during the heating step of the process, while a simple Darcy based model is used to account for the impregnation kinetics. An experimental programme to validate the thermal and impregnation model is presented in the third section. The final conclusions are presented in the last section.

PROCESS DESCRIPTION

A fully consolidated laminate requires the thermoplastic resin to replace all the air in the textile. This is achieved at high temperature by increasing the applied pressure and force the thermoplastic resin to impregnate into the reinforcing textile. The architecture of a woven textile dictates that the impregnation takes place on two scales. The meso-scale concerns the space between the bundles, while the micro-scale concerns the space between the individual filaments. Based on the difference in characteristic pore dimension of the two scales it is assumed that meso-scale impregnation will precede micro-scale impregnation. A glass-PPS laminate is processed in an experimental press mounted in an *Instron* tensile tester to validate this assumption. The tensile tester measures the press force and platen displacement, while a thermocouple placed in the centre of the laminate records the temperature during the process. Figure 2 shows the applied pressure, platen displacement and laminate temperature during the process. The measured displacement suggests the consolidation takes place in three stages (labelled A to C).

Two additional laminates are processed to quantify the impregnation at stage A and B. The first stage starts when the glass transition temperature (90 °C) of the thermoplastic film is reached. The micrograph in figure 3, labelled A, shows that the thermoplastic film plastically deforms and fills open meso-scale pores of the textile. The second impregnation stage starts at the melt temperature (280 °C) of the PPS thermoplastic film. The second micrograph in figure 3 shows that meso-impregnation is complete; the matrix material now surrounds the individual bundles. However, impregnation of the bundles themselves is not achieved yet. An increase of the applied pressure causes the last stage of impregnation: the applied pressure forces the thermoplastic matrix to impregnate the individual bundles. The last micrograph in figure 3 clearly shows that all the air inside bundles is now replaced by matrix material.

The processing graph in figure 2 in combination with the micrographs in figure 3 shows that meso-impregnation is achieved during the heating step of the process, while bundle impregnation requires an increased pressure. Therefore, bundle impregnation is regarded as the rate-determining stage during the impregnation step.

FILM-STACKING PROCESS MODEL

This section proposes a modelling approach to optimise the heating and micro-impregnation step of the film-stacking production process. Firstly, a one dimensional transient thermal model is proposed to predict the through thickness temperature of the laminate during the heating step of the process. The model provides the possibility to investigate the influence of laminate and tinplate material properties on the time required to fully heat the stack.

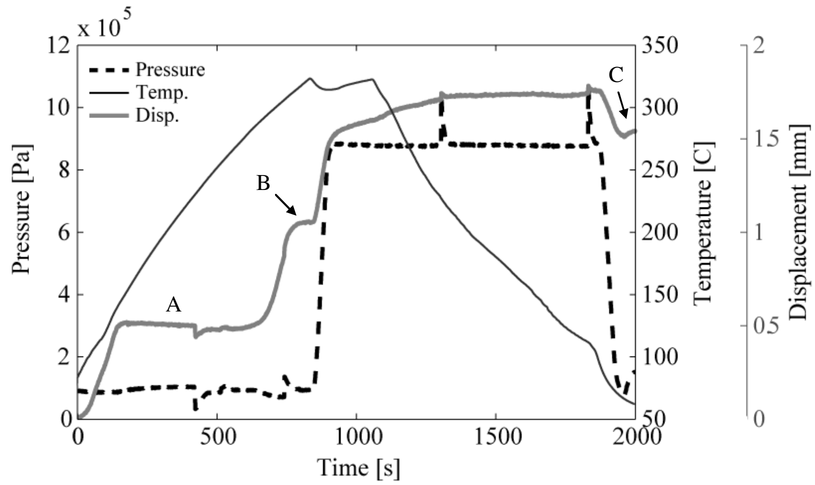


Figure 2. Typical processing graph for film-stacked glass-PPS

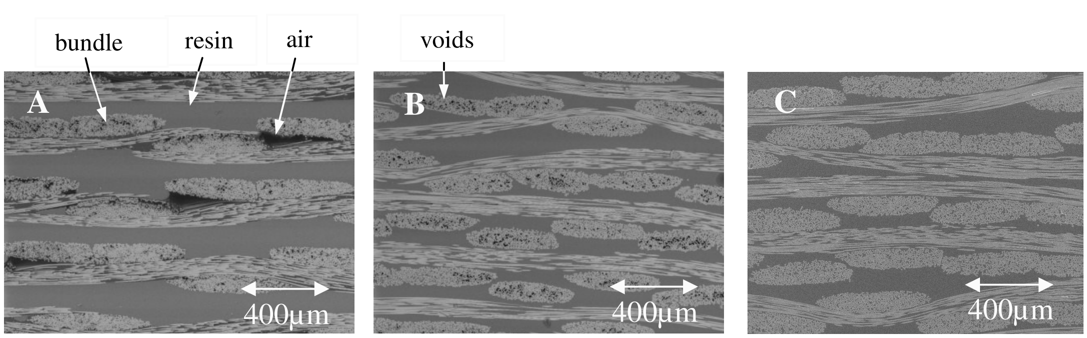
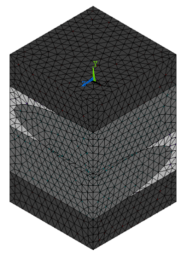
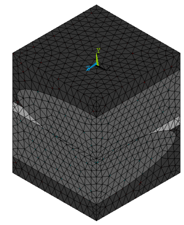
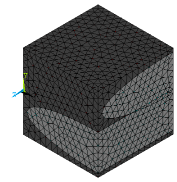


Figure 3. Micrographs showing the stages of impregnation for film-stacked glass/PPS

Table 1. Thermal properties of the different unit cells for glass-PPS

Unit Cell			
Temp range [°C]	$T < T_g$ (90°C)	$T_g < T < T_m$	T_m (280°C) < T
Conductivity [$W.m^{-1}.K^{-1}$]	0.15	0.23	0.29
Heat Capacity [$kJ.kg^{-1}.K^{-1}$]	0.82	0.82	0.82
Density [$kg.m^{-3}$]	1125	1405	1590

Secondly, a Darcy impregnation model is proposed to study the influence of applied pressure and processing temperature on the bundle impregnation time. The combination of both models allows optimisation of the film-stacking process in terms of time and costs.

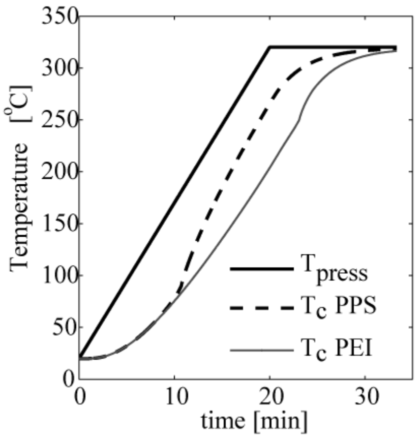
Step I: Transient thermal model

A simple transient one dimensional thermal model is applied to predict the thickness-wise temperature distribution in the laminate as a function of time and applied platen temperature. The one dimensional conduction equation reads:

$$\frac{1}{\alpha(T,y)}\frac{\partial T}{\partial t}=\frac{\partial^2 T}{\partial y^2} \qquad (1)$$

in which $\alpha(T,y)$ represents the thermal diffusivity, which varies over the thickness of the stack. The structure and subsequently the thermal properties of the composite laminate change while heating the stack. The previous section showed that the film is forced in the open meso-scale pores at the glass transition temperature and meso-impregnation is achieved above the melt temperature. The thermal diffusivity of the composite is therefore also function of the local temperature.

A three dimensional finite element analysis of representative unit cells is performed in *Ansys* to estimate the thermal conductivities of the glass-PPS composite laminate during the three stages. The thermal conductivities of PPS and air equal 0.28 $W.m^{-1}.K^{-1}$ and 0.026 $W.m^{-1}.K^{-1}$ respectively. The bundle has a fibre volume fraction of 0.7 and is assumed to have an isotropic thermal conductivity of 0.2 $W.m^{-1}.K^{-1}$. Table 1 shows the unit cells and the resulting homogenised thermal properties. The resulting steady state through-the-thickness thermal conductivity is seen to increase with the degree of consolidation.



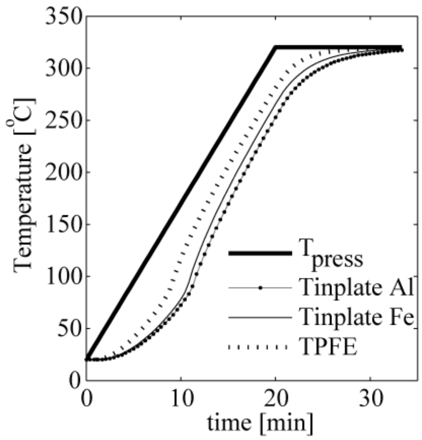


Figure 4. Temperature in the centre of a stack glass-PPS and glass-PEI laminates

Figure 5. Temperature in the centre of a stack for different separator materials

The conduction equation (1) is then solved for the homogenised laminate, subject to the following initial and boundary conditions:

$$T(y,0) = T_{start}$$
$$T(0,t) = T(h,t) = T_{platen}(t) \tag{2}$$

in which h corresponds to the thickness of the laminate. Equation 1 is solved numerically by using an implicit finite difference approach on a fixed grid. The thermal diffusivities are compensated to account for the thickness change during the process.

The dashed line in figure 4 shows the temperature in the centre of a stack comprising 10 laminates with a $[PPS/glass/PPS]_4$ layup separated by steel tinplates. The PPS has a glass transition temperature of 90 °C. The solid line in the graph shows the temperature in case the PPS is replaced with polyetherimide (PEI) with a glass transition temperature of 250 °C. A lower glass transition temperature decreases the time required for the through thickness temperature differences to fade away.

Figure 5 shows the influence of the tinplate material on the temperature in the centre of the laminate during heating. Three different materials are used to separate the laminates during production: tinplates of either aluminium or steel with a thickness of 1.1 mm or 0.25 mm thin TPFE foil. The figure shows that the application of TPFE foil between the laminates promotes the through thickness heat conduction in the laminate; the temperature difference diminishes faster compared to the aluminium or steel tinplates.

Step II: Darcy micro-impregnation model

The micrographs in the previous section show that the meso-scale impregnation precedes the micro-scale impregnation and is achieved while heating to the processing temperature. Therefore, it is assumed that the micro-impregnation governs the total impregnation time. Bundle impregnation with a thermoplastic resin has been subject of research in a number of comparable studies: for instance in commingled yarn composites [6,7] and film-stacking of knitted textiles [8]. The majority of the research assumes that all fibre bundles undergo impregnation simultaneously and all of the bundles have an identical geometry. This assumption allows the textile to be divided into repetitive elements or unit cells, which can have the geometry of a rectangle [5], a circle [9] or an ellipse [10]. The inward impregnation of the bundle unit cell is then described using Darcy's law:

$$u = -\frac{K}{\mu}\frac{dp}{dr} \tag{3}$$

A similar approach is adopted in this work to model the impregnation of the film-stacked glass-PPS laminates. The textile used in this investigation comprises bundles with an elliptic cross-section. The investigations of Van West et al. [11] showed that an equivalent one dimensional radial analysis can be used to model the impregnation kinetics. The equivalent bundle radius yields:

$$r_{eq} = \sqrt{2}\frac{a_0 b_0}{\sqrt{a_0^2 + b_0^2}} \tag{4}$$

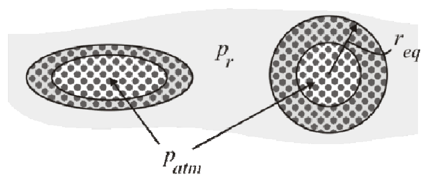
with a_0 and b_0 the major and minor axis of the bundle. The equivalent bundle radius for the glass S303 textile used in this investigation equals 70 µm.

As a first approach the bundles are assumed to be rigid entities comprising ideally stacked filaments. The bundle permeability K is therefore assumed constant and depends on fibre volume fraction and the fibre packing. The formulation developed by Gebart [12] is used to determine the bundle permeability in this work. The fibre volume fraction and packing type of a bundle is estimated from micrographs of fully consolidated laminates: a hexagonal packing with a fibre volume fraction of 0.7 seemed most appropriate. Furthermore, it is assumed that the impregnation takes place at isothermal conditions. The viscosity μ of the PPS film as a function of temperature is obtained from data specified by the supplier [13].

The applied press force F_A causes a pressure gradient dp/dr over the bundle which will drive the bundle impregnation. During the process the press force F_A, is distributed over the resin and textile simultaneously:

$$F_A = \int_{A_r} p(x,y,t)dA + \int_{A_f} \sigma_f(x,y,t)dA \tag{5}$$

in which σ_f is the textile compaction stress and p is the resin pressure. Initially, when there is negligible deformation of the textile, the applied force is carried solely by the resin pressure, causing bundle impregnation. As the impregnation proceeds, the textile is compressed and thereby carrying an increasing part of the applied load. The textile compaction behaviour, therefore, needs to be taken into account to obtain the resin pressure. The stress-strain compaction curve of the dry textile is acquired experimentally and fitted with a power law function.



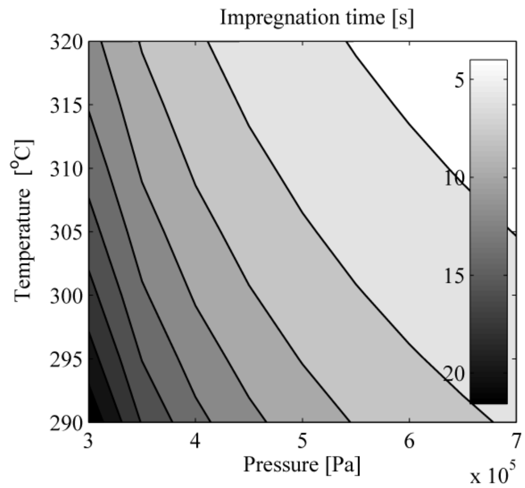


Figure 6. Bundle unit cell (left) and its circular representation

Figure 7. Bundle impregnation time for the glass-PPS system

The time required for impregnation is determined using a simple numerical solution scheme. Darcy's law is applied to calculate the impregnation rate using the effective resin pressure as an input. For a small time increment this results in a volume flow, which can be used to update the textile compaction strain. Subsequently, the effective resin pressure follows from equation 5, which can then again be used to determine the bundle impregnation rate.

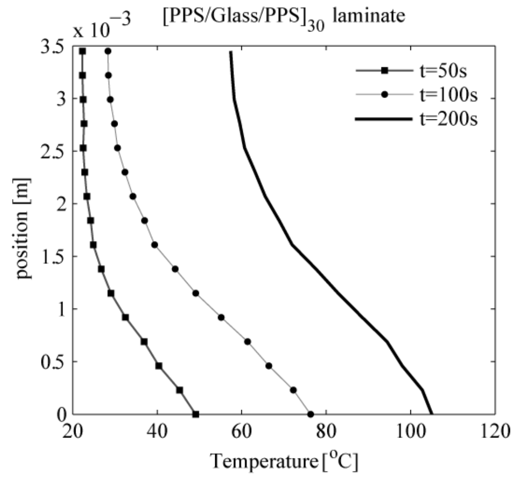
Figure 7 shows the bundle impregnation time as function of processing pressure and temperature for glass-PPS thermoplastic laminates. The figure shows that the bundle impregnation for the glass-PPS laminates is achieved in less than 25 seconds. An experimental programme is designed to validate these results.

EXPERIMENTS AND DISCUSSION

An experimental programme was designed to validate the transient thermal model and bundle impregnation model. The present section compares obtained experimental data with the model predictions and discusses the obtained results. The Dutch-based company Ten Cate AC kindly provided glass S303 woven textile and PPS thermoplastic film. The textile has an areal density of 300 $g.m^{-2}$ and an initial thickness of roughly 0.3 mm, while the PPS film has a thickness of 60 μm. The thermoplastic PPS has a glass transition temperature of 90 °C and a melt temperature of 280 °C. The textile and film are stacked in a $[PPS/glass/PPS]_n$ lay-up. A fully consolidated laminate has a fibre volume fraction of approximately 50% and a thickness of 1.95 mm.

Step I: Thermal model - validation

A glass-PPS laminate with a $[PPS/glass/PPS]_{30}$ lay-up is processed in an experimental press. Sixteen thermocouples, placed in the bottom half of the laminate between every two PPS films, measured the through thickness temperature distribution during heating,



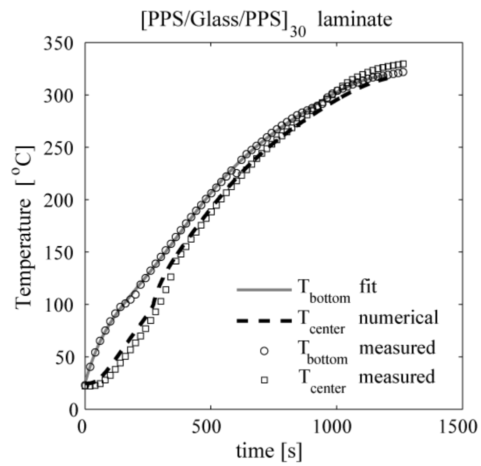


Figure 8. Temperature distribution over the bottom half of the laminate

Figure 9. Comparison between predicted and measured temperature in the centre of the laminate

as presented in figure 8. The centre of the laminate corresponds with the top of the figure. A large temperature difference of approximately 50 °C was found over the laminate thickness during the heating process. The transient thermal model is validated by fitting the measured temperature at the bottom of the laminate with a polynomial function and using this as a boundary condition in the model. The predicted and measured temperatures in the centre as well as the measured and fitted edge temperature are shown in figure 9. Despite the simplicity of the implemented model the predicted temperature corresponds well with the experimental results for the glass-PPS system used in this research. The applicability of the proposed model for other thermoplastic laminates is subject of future research.

Step II: Bundle impregnation - validation

The Darcy bundle impregnation model predicts for an impregnation time of less than 20 seconds for processing temperature above 290 °C and pressures above 0.3 MPa. A [PPS/glass/PPS]$_8$ laminate (#2) is processed to validate the predicted impregnation time. The stack is placed between the press platens and the press is heated to 300 °C. The pressure is increased to 0.4 MPa, after which the press is cooled down immediately to room temperature. As a reference a standard laminate (#1) is processed at 315 °C and 1.0 MPa, with a holding time of 10 minutes. The processing graphs for both laminates are shown in figure 10.

After processing, specimens were cut from the laminates and visually inspected using a scanning electron microscope. Figure 11 shows a micrograph taken from the standard laminate processed at 315 °C and 1.0 MPa. The thermoplastic resin impregnated all the individual bundles and replaced all the air in the laminate. Figure 12 and 13 show the micrographs obtained from the laminate processed at a temperature of 300 °C and a pressure of 0.4 MPa. The impregnation in terms of Darcy has been completed; no unimpregnated elliptical zone is present in the bundles. The short impregnation times predicted by the simple Darcy micro-impregnation model seem therefore realistic.

However, the micrograph in figure 13 also shows some micro-voids between the individual filaments, while the standard laminate shows negligible air being present between the filaments.

Based on the obtained results two important questions need to be answered. Firstly, the influence of the small micro-voids on the mechanical properties has to be investigated. Secondly, the disappearance of the small air pockets needs attention. The air apparently dissolves into the PPS matrix material. This assumption needs to be validated and modelling strategy has to be developed to predict the time required for this process.

CONCLUSIONS

The consolidation process of film-stacked glass-PPS laminates is investigated. A one dimensional transient thermal model is proposed to optimise the heating step of the process. The structural change of the material is taken into account by implementing a temperature dependent thermal diffusivity. Despite the simplicity of the model, good agreement with experimental data is observed.

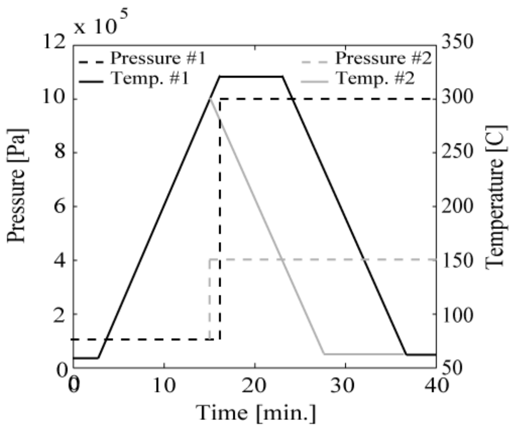


Figure 10. Processing graph for two laminates

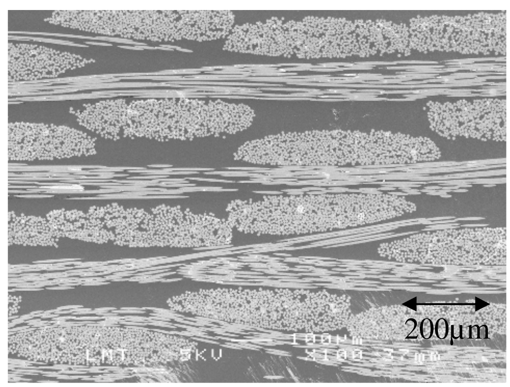


Figure 11. Micrograph of the glass-PPS laminate #1

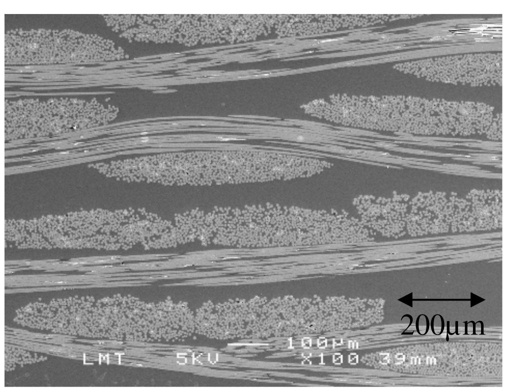


Figure 12. Micrograph of the glass-PPS laminate #2

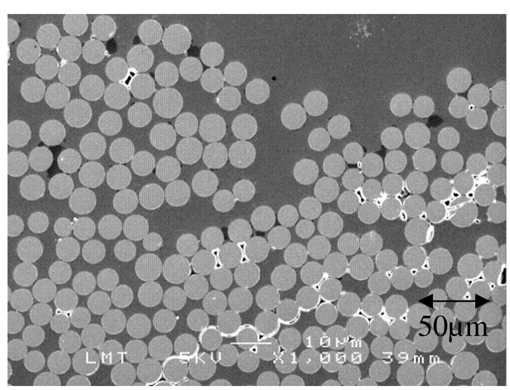


Figure 13. Micrograph of the glass-PPS laminate #2

The impregnation of the textile takes place on different scales. It is observed that bundle impregnation is the rate determining step. A simple Darcy based impregnation model is applied to calculate the impregnation time of the textile. The obtained results predict an impregnation time in the order of seconds. A laminate processed at low temperature, low pressure and short holding time was processed to validate the model. Micrographs from the laminate show that impregnation in terms of Darcy was achieved. However, small micro-voids were present between the individual filaments. Future work aims to investigate the influence of these voids on the mechanical properties of the laminate. Furthermore, the dissolution of the voids in the thermoplastic PPS material will be investigated.

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