INFLUENCE OF THE SHEET FORMING TECHNIQUE ON THE IMPREGNATION QUALITY OF TEXTILE REINFORCED THERMOPLASTIC COMPOSITES

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SUMMARY: As a thermoplastic matrix is more suitable for short cycle production, low cost weft inserted warp knitting is applied to produce a cost-effective thermoplastic textile, containing both non-crimp reinforcing glass fibres and polypropylene matrix ribbons. The composite structures can be obtained by simple pressing the textile preform without any further addition of the matrix. A full consolidation at high pressure and temperature or a pre-compaction at low pressure and intermediate temperature to eliminate the uncontrolled shrinkage of the stretched thermoplastic ribbons are used for the laminate production. Moreover, the influence of a cold pressing after pre-compaction on the impregnation quality has been examined in more detail. A special definition has been set up to quantify the degree of impregnation, which includes the degree of wetting out of the individual fibres (void content) and the fibre distribution within the bundle. A correlation between the degree of impregnation, the void content and the flexural performance is shown.

KEYWORDS: Textile preforms, thermoplastics, glass fibres, fast processing, impregnation quality, flow behaviour, automotive applications

INTRODUCTION

Fibre reinforced polymer composites offer, besides Al and Mg, a significant light weight potential in structural body parts of vehicles [1]. Much potential and interest therefore exists for the fast processing of lightweight, inexpensive composite preforms in automotive and other transport applications. Due to some severe disadvantages of thermoset composites, like long production times, limited recyclability, constrained storage time as well as storage requirements, research activities in thermoplastic composites have developed recently [2]. Thermoplastic polymers indeed offer a better fracture toughness, better impact tolerance, unlimited shelf life of raw material, recycling potential and a cleaner working environment [2][4][5]. However, the most important advantage of thermoplastics as a matrix in composites lies in their potential for rapid, low-cost mass production [3]. Thermoplastic composites using continuous textile reinforcements moreover constitute a new class of tailored intermediates to meet a variety in property and performance requirements. A growth in the use of thermoplastic composites currently depends on the further reduction of the manufacturing cost and the optimisation of the part performance. A decrease in the fabrication cost of thermoplastic composite parts can be obtained if the separate impregnation step is eliminated, either by combining impregnation and forming in one process or by means of a 'dry impregnation', implying intimate mixing of the thermoplastic and reinforcement [1].

Techniques for improving the *impregnation* of reinforcing fibres with thermoplastic polymers are basically divided into three categories.
First, those that aim to reduce the viscosity in order to achieve rapid impregnation in *liquid* form, the ***pre-impregnation processes***. Most important techniques are solvent and melt impregnation.
Second, the difficult impregnation of the reinforcing fibre bundles due to the high matrix viscosity and low shear flow rates can be overcome by the use of several types of "combined yarns", where the reinforcing fibres are intensely mixed with the matrix prior to composite processing [4]. These ***mingling processes*** attempt to combine the matrix and the reinforcement in such a way that it minimises the subsequent flow length required to fulfil impregnation, which is generally the rate determining step in composites processing. Amongst other techniques such as powder impregnation and film stacking, fibre co-mingling [1][2][3] has recently been studied. A high degree of intimacy can indeed be obtained by providing the matrix in fibre form and intermingling polymer fibres with reinforcing fibres. They should ideally be combined in the same strand, as Vetrotex has achieved in the commercialised co-mingled yarn Twintex, where glass fibres are mingled with PP or PET polymer fibres.
Third, to further optimise the impregnation during processing, the use of ***textile preforms*** is of recent interest, where new structures of yarns are studied.

SPLIT-WARPKNIT THERMOPLASTIC TEXTILE

Low cost weft-inserted warp knitting is chosen to produce a novel thermoplastic textile preform, containing both non-crimp reinforcing fibres and thermoplastic matrix ribbons (figure 1). The knitting equipment and the knitting process have been adapted for an efficient and economically feasible textile production of glass rovings with PET tapes, on Rashel machines (RS3 MSU-V with gauge E12 and a width of 79") with weft insertion system. Since all structures contain split-film and reinforcement rovings of rather high linear density, special guide elements are used to prevent frequent yarn breakage during production. A typical tube guide finger is applied which can guide reinforcement roving, split-film or a combination of the two. Moreover, the split-films have been fibrillated (figure 2) and varied in profile to enhance the melt distribution during the impregnation process [7][8][9].

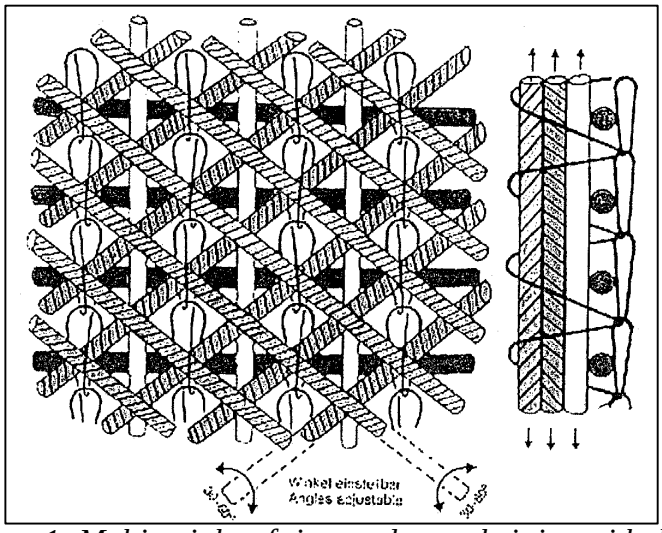


Figure 1: Multi-axial weft-inserted warp knitting with thermoplastic split-films

The split-films are inserted in the straigth yarns, along with the reinforcement rovings. The loops are hence exclusively consisting of a binding yarn (figure 2). The hybrid split-warpknit textile allows for a more even fibre distribution because it is possible to bring the inlays closer together, as the binding yarn has a considerable lower count than the split-films and the volume between the rovings is kept low. This type of textile structure evenso permits for the reinforcement yarns to be placed straighter.

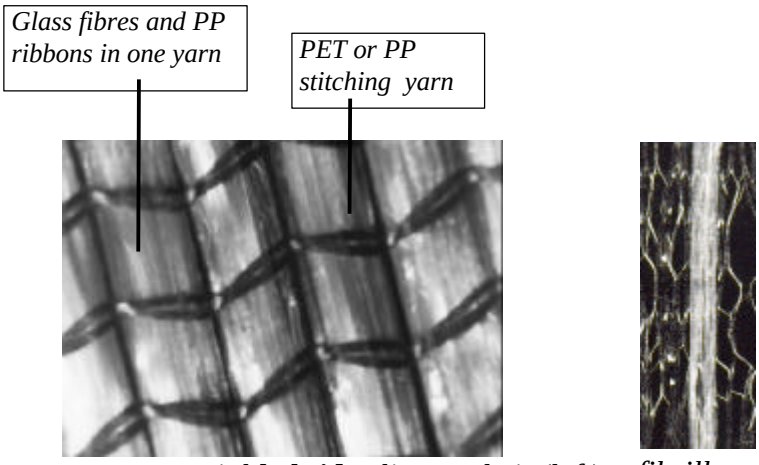


*Figure 2: Mono axial **hybrid** split-warpknit (left), fibrillated, split film (right)*

The composite structures are obtained by simple hot pressing the textile preform without any further addition of matrix. Special glass sizings and low viscosity matrix materials are developed to ensure a good wetting out of the reinforcement and an easy, thus fast impregnation.

SHORT CYCLE PROCESSING CONCEPTS

SHRINKAGE OF THE SPLIT-FILM RIBBONS

The thermoplastic split film ribbons are severely stretched to guarantee sufficient tenacity during knitting. During the stretch operation, the polymer chains get a high degree of orientation. Free heating without the application of external pressure, increases the possibility for the molecular chains to return to their more natural state of random coil orientation. This will cause the film to shrink and may result in a deformation (in-plane contraction or curling) of the split-warpknit structure (figure 3). Due to the shrinkage of the ribbons upon heating, it is mandatory to support the textile throughout the heating phase, either by sideways clamping and stretching, or by applying a flat-wise pressure on the preform.

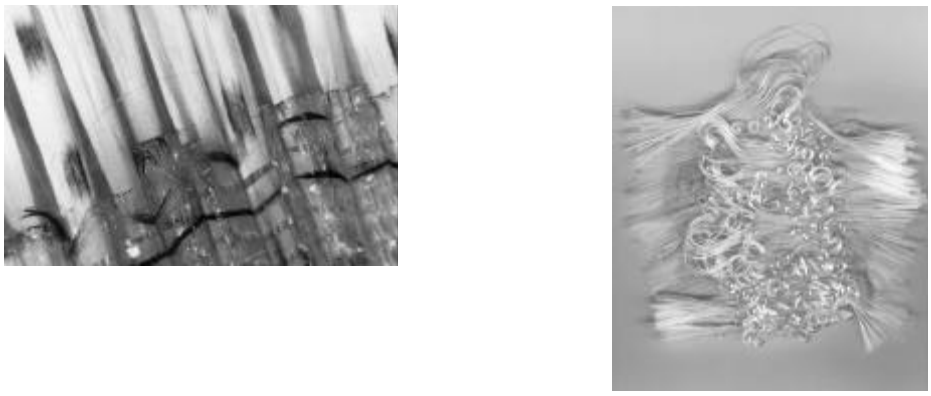


Figure 3: Shrinkage of the thermoplastic film: waviness and curling of the fabric

SHORT CYCLE PROCESSING OF FLAT LAMINATES

There are mainly two different ways of processing this new split-warpknit textile into a flat stable laminate *prior to deep drawing*.

First, a ***full pre-consolidation*** of the textile into an organic sheet under high temperature and pressure (T=240°C, p=25 bar). Consequently, the pre-consolidated sheet is re-heated and formed during deep drawing. A classical *in-mould heating and cooling* method, where a loose stack of textile layers is placed in a cold mould, heated, pressed and cooled down could be used to produce a pre-consolidated sheet. It is however very energy and time consuming and hence far too expensive for industrial use. However, a more economical feasible pre-consolidation method is provided by a double belt press, in which continuous manufacturing of fully consolidated sheets, to be formed afterwards, is performed.

Second, i***mpregnation and full consolidation*** press forming are combined. A ***pre-compaction*** at lower pressure and intermediate temperature (T=210°C, p=5.5 bar and t=5.5 min) is necessary for the relaxation of internal stresses and hence the elimination of the shrinkage of the thermoplastic ribbons during convection or infrared heating. During pre-compaction, a limited matrix infiltration of the fibre bundles is already achieved. Lab-scale tests with a hot press are performed (batch wise) to evaluate the influence of the parameter settings on the impregnation quality (infiltration depth of the fibre bundles).

A comparison between two possible ways of processing flat split-warpknit laminates – ***pre-consolidation and pre-compaction*** – is carried out. Both processes mainly differ in four ways:

- A *higher temperature and pressure* are applied for pre-consolidation
- There is a substantial difference in the *heating and cooling rate*. During pre-consolidation, a slow heating and cooling of 3°C/min is achieved, while during pre-compaction a heating and cooling rate of about 40°C/min is reached. This difference greatly affects the degree of crystallisation of the polypropylene matrix and hence the mechanical performance of the composite part.
- Secondly, the *processing equipment differs*. During pre-consolidation, the textile layers are wrapped in aluminium foil and sealed inside a mould, while during pre-compaction no mould is used. Hence, matrix flow-out and fibre misalignment are avoided during pre-consolidation and a higher temperature, pressure and time level can be applied. However, during pre-compaction, trapped air can be squeezed out, reducing the void content. The trapped air can not escape during pre-consolidation and is dissolved in the thermoplastic matrix due to the high pressure.
- The pre-consolidated laminates can be applied as such, while pre-compaction is only a necessary process prior to compression moulding or deep drawing to avoid the shrinkage of the split film ribbons.

A ***GMT based cold pressing method***, i.e. an external preheating and cold pressing, is applied on the pre-compacted laminates. The GMT parameter settings are as follows: preheating at 240°C for 4 min, pressing at 125°C for 1.4 min at 60 bar.

EVALUATION OF THE IMPREGNATION QUALITY

WHAT IS IMPREGNATION QUALITY ?

A special definition has been developed to quantify the degree of impregnation within the fibre bundle. In a perfectly impregnated part, there are no voids. Every fibre is surrounded by matrix material and the matrix is evenly distributed over the composite (no matrix-rich zones). As it was not yet feasible to achieve a uniform distribution of the fibres in the

composite, the term impregnation should only address the local situation in a given area with a high fibre concentration. The *degree of impregnation* ***inside*** *a fibre bundle* is defined as:

$$DOI = \left(\frac{A_{matrix}}{A_{in-between-fibres}} \right) \left(1 - \frac{k\bar{N}}{6} \right) = \left(\frac{A_{matrix}}{A_{matrix} + A_{voids} + \ldots} \right) \left(1 - \frac{k\bar{N}}{6} \right) \qquad (1)$$

A: cross-section area of the different phases (matrix-fibres-voids) (figure 4)
N: number of fibres, touching a given fibre (N= 6 for the most dense hexagonal packing)
k: an empirical factor (≤ 1) accounting for the fact that, due to the existence of matrix material in between the fibres, even in an array with hexagonal packing, better impregnation is obtained and hence increased mechanical properties than the ones of a loose, dry fibre bundle. Set k=1.

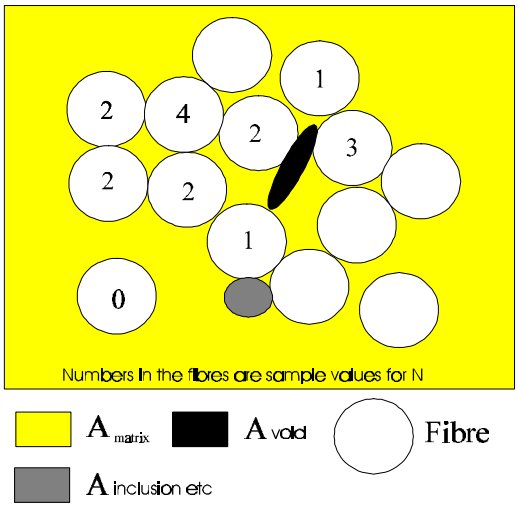


Figure 4: Cross-section of the different phases, meaning of the N-factor

The first term in formula 1 is called the **wetting term** and measures to what extent the area between the fibres is filled with matrix material. This term is influenced by the viscosity of the matrix and the surface properties of the fibres and the thermoplastic melt.
The second term, **the fibre spacing term,** accounts for the local fibre arrangement and is influenced by the fibre geometry, the distribution of the fibres and matrix phase in the virgin rovings and the overall processing conditions, determining the viscosity of the thermoplastic matrix and hence the ease of wetting out the fibres and flowing within the closely packed fibre bundles.

The first term in the former equation is difficult to evaluate because voids can only be detected under Reflection Optical Microscopy (less contrast than Transmission Light Microscopy). Therefore it is more accurate to evaluate the first term by determining the fibre volume fractions with the ignition loss test.

$$DOI = \left(\frac{V_{matrix}}{V_{matrix} + V_{voids} + \ldots} \right) \left(1 - \frac{k\bar{N}}{6} \right) \qquad (2)$$

The area fraction of fibres is often assumed identical to the volume fraction, which is a good assumption only if the fibres are continuous, parallel and perpendicular to the polished surface (as for the mono axial structure).

For the evaluation of the infiltration quality of the polypropylene into the fibre bundle, a microscopic study is carried out. The samples are embedded in an Epofix-red dye mixture. The fibre contact distribution is only calculated in the fibre rich areas, thus within the fibre bundle. The "*Random points*" method is used where a grid of points is placed on the picture and the number of fibres in contact with a certain fibre is recorded [5].

The impregnation phase in non-twisted split-film glass fibre yarns consists mainly of two steps [2][6]:
First, the initially separated fibre bundles are flattened out and move towards each other when pressure is applied. This phase starts already during conductive preheating where at least a low pressure is needed to avoid the uncontrolled shrinkage of the PP ribbons. If the temperature reaches the melt temperature of the thermoplastic matrix, the polymer melts and firstly fills the spaces in between the fibre bundles (inter-bundle impregnation due to the larger porosity and hence better permeability). The border of the reinforcing bundle is still visible (figure 5).

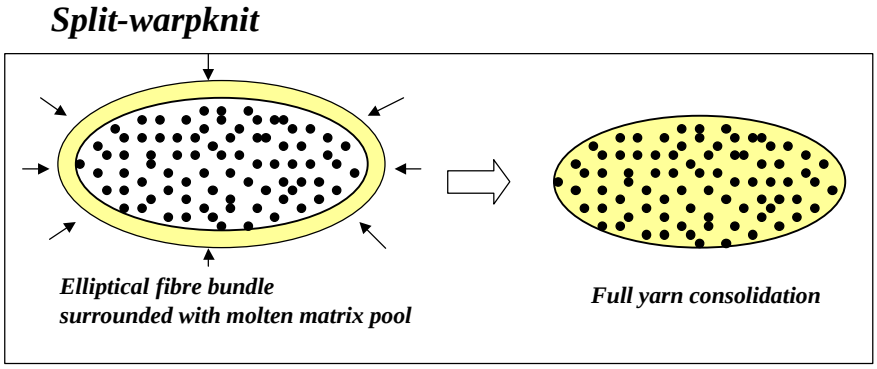


Figure 5: Starting point for the melt-impregnation of the split-warpknit textile: every dry fibre bundle is surrounded by a molten matrix pool

Consequently, the molten matrix melt-impregnate the individual fibres under the applied pressure

Although with ***pre-consolidation*** a degree of impregnation of 100% can be reached, the individual fibre bundles remain still visible despite of the high temperature and pressure (figure 6). The higher the impregnation pressure and the longer the time, the more homogeneous the fibre distribution *within* the fibre bundle will be.

Hence, with ***pre-compaction*** at low pressure and at low temperature, it is impossible to obtain full melt impregnation of the individual fibres. The border of the fibre bundles remains clear, resulting in a less homogeneous fibre distribution where matrix rich zones separate not fully-wetted fibre bundles (figure 7). Moreover, the non-meltable stitches act as a periodic screen to the bulk flow of the thermoplastic matrix between the fibre bundles. Presence of air bubbles or voids in the fibre bundles also results in poor fibre wetting and consequently poor fibre bonding yielding composite parts with non-uniform mechanical strength.

Because a ***cold pressing*** is applied during the GMT technique, the viscosity of the thermoplastic matrix decreases very rapidly and only a little improvement of the inter-bundle impregnation of the pre-compacted sheets is achieved (figure 8).

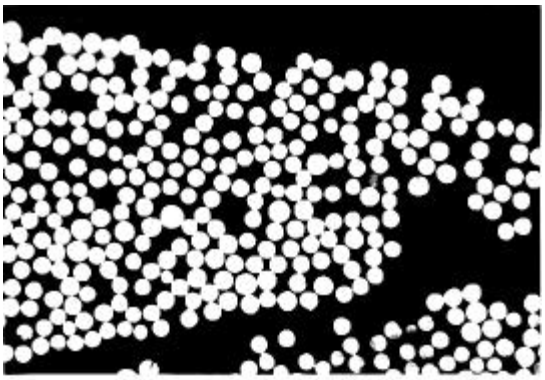


Figure 6: Pre-consolidated laminate (DOI= 100%)

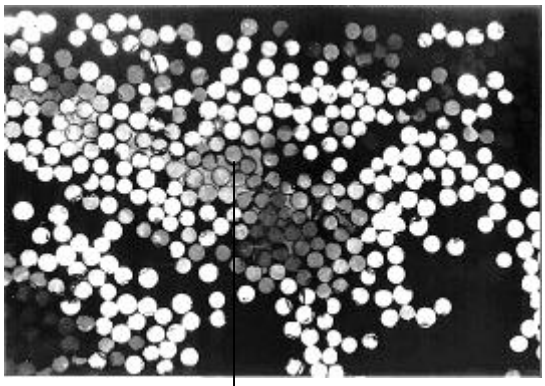
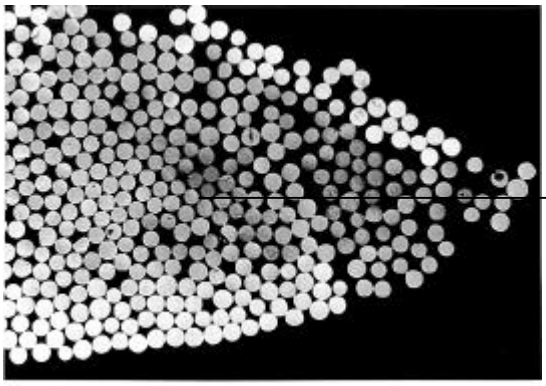


Figure 7: Pre-compacted laminate (DOI<<<100% due to void areas and closely packed fibre bundle)

Voids between individual fibres



Closely packed fibres. Non-homogeneous fibre distribution

Figure 8: Cold pressed laminate (after pre-compaction)(DOI <<<100%)

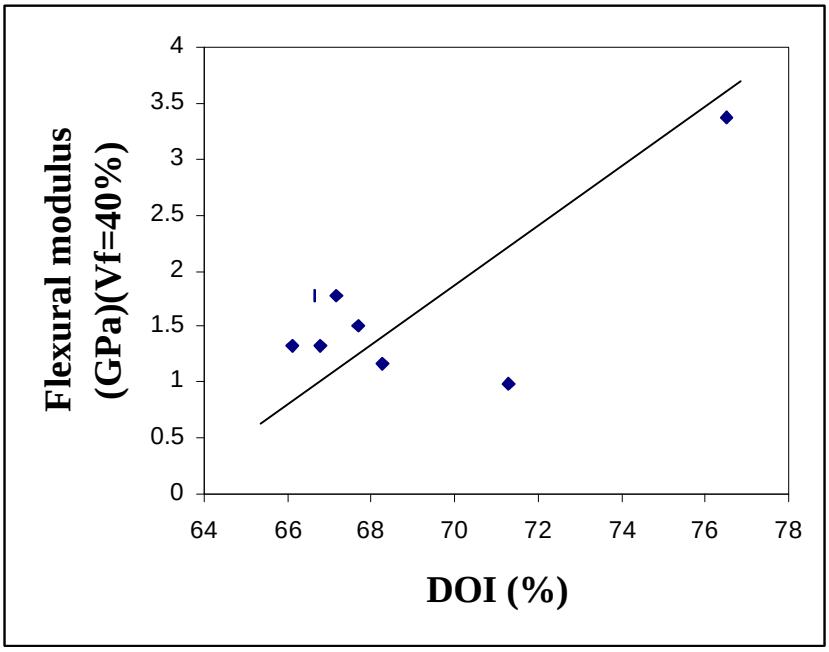
The pre-consolidated samples clearly show a more homogeneous fibre distribution where almost no fibres are in contact. This is primarily due to the good penetration of the less viscous PP (at 240°C) in the fibre bundles under high pressure (25 bar). The less homogeneous distribution of the fibres in the pre-compacted samples is obvious because the matrix flow-out limits the parameter settings (temperature and time) and reduces the amount of matrix present in the laminate. Hence, more fibres are still in contact. The highest degree of impregnation is reached for average pre-compaction parameter settings (T=210°C, p=5.5 bar, t=5.5 min) (table 1). For the GMT pressed samples, the resulting infiltration quality depends on the infiltration quality of the sample after pre-compaction (table 1).

	DOI (%)
Pre-consolidation T240p25t5	76.5
Pre-compaction T192p2.8t2.8	68.3
Pre-compaction T210p5.5t5.5	66.1
Pre-compaction T210p5.5t10	71.3
Pre-compaction T192p2.8t2.8 +GMT	66.6
Pre-compaction T210p5.5t5.5 + GMT	67.7
Pre-compaction T210p5.5t10 + GMT	66.8
GMT direct T240t4T125p60t1.4	67.2

Table 1: Influence of the processing technique on the impregnation homogeneity

If the infiltration of the polypropylene into the fibre bundles is poor during the pre-compaction (due to matrix flow out or too viscous matrix), the high pressure of the GMT cold pressing will move the fibres towards each other and the number of dry, not-wetted fibres in contact increases. The re-melting of the PP during the GMT process does however lead to an equalisation or evening of the polypropylene distribution in the infiltrated areas of the bundle and hence an increase in the mechanical performance.

The void fraction and the degree of impregnation, both a measure of the laminate quality, have been correlated with the "transversal" flexural modulus (figure 9) and strength of cross ply $[0,90]_s$ laminates. The better the impregnation homogeneity (higher DOI) and the lower the overall void content, the better the flexural performance. A clear effect of the degree of impregnation on the flexural strength could not be noticed. It should however be notified that the pre-consolidated laminate exhibits superior properties, not to be correlated anymore.



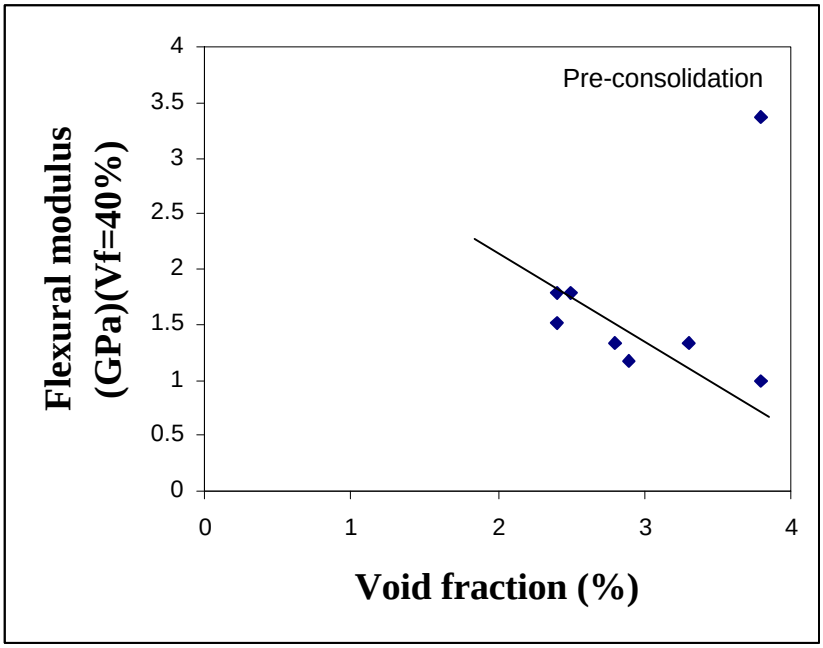


Figure 9: Correlation between the flexural modulus and the impregnation quality, i.e. the degree of impregnation DOI and the void content

CONCLUSIONS

The aim of this paper is mainly the evaluation of the feasibility of the pre-compaction technique at low pressure and intermediate temperature to eliminate the shrinkage of the split-film ribbons upon subsequent heating. The influence of other short cycle laminate production techniques – a pre-consolidation at high temperature and pressure and a cold pressing - has been evaluated as well: The process quality has been studied only based on impregnation homogeneity and flexural performance.

A new definition has been developed to quantify the impregnation quality *within the fibre bundle*. The degree of impregnation DOI both includes the degree of wetting out of the individual fibres (i.e. void content) and the fibre distribution within the fibre bundle. The impregnation homogeneity has been visualised by optical microscopical analysis.

The pre-consolidated samples clearly show a more homogeneous fibre distribution where almost no fibres are in contact. This is primarily due to the good penetration of the less viscous PP (at 240°C) in the fibre bundles under high pressure (25 bar). The less homogeneous distribution of the fibres in the pre-compacted samples is obvious because the matrix flow-out limits the parameter settings (temperature and time) and reduces the amount of matrix present in the laminate. Hence, more fibres are still in contact. The highest degree of impregnation is reached for average pre-compaction parameter settings (T=210°C, p=5.5 bar, t=5.5 min).

During GMT cold pressing, voids still present after pre-compaction are removed due to the very high pressure (60 bar). The re-melting of the polypropylene during heating improves the distribution in the areas of the fibre bundle, already infiltrated during the former pre-compaction phase. The degree of infiltration is not further improved. The sample quality after GMT processing is hence strongly influenced by the infiltration quality after pre-compaction.

A correlation between the impregnation homogeneity, i.e. the void content and the degree of impregnation, and the transversal flexural modulus is noticed. The better the impregnation homogeneity (higher DOI) and the lower the overall void content, the better the flexural performance. A clear effect of the degree of impregnation on the flexural strength could however not be detected.

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