EFFECTS OF DIFFERENTLY IMPREGNATED CARBON FIBER REINFORCED TAPES WITH POLYAMIDE 6 ON FURTHER PROCESSING

V. Radlmaier[1], H. Baumann[1], P.-V. Brudzinski[2], A. Erber[2], H. Koerber[1], K. Drechsler[1]

[1]Institute for Carbon Composites, Faculty of Mechanical Engineering, Technische Universität München, Boltzmannstrasse 15, D-85748 Garching b. München, Germany
Email: radlmaier@lcc.mw.tum.de, Web Page: http://www.lcc.mw.tum.de
[2]SGL Group, Werner-von Siemens-Strasse 18, D-86405 Meitingen, Germany
Web Page: http://www.sglgroup.com

Keywords: Degree of impregnation, tapes, polyamide 6, flexural properties

Abstract

In this study, the impregnation progress of unidirectional carbon fiber reinforced tapes (UD tapes) with different initial degrees of impregnation (DOI_i) is followed throughout a production process chain for carbon fiber reinforced thermoplastics (CFRTP). Partially impregnated tapes can be manufactured with reduced cycle times along with cost savings. They are assumed to be completely impregnated throughout the repeated heating processes during CFRTP manufacture. UD tapes with polyamide 6 matrix were produced in a double-belt press yielding a DOI_i of 80 %, 90 % and 100 %. The tapes were stacked and consolidated in a press with different dwell times and some laminates were additionally thermoformed. Longitudinal and transverse flexural tests were conducted to evaluate the effect of the DOI_i on composite properties. Micrographs of tapes and test panels were post-processed to evaluate the DOI_i and the final degree of impregnation (DOI_f).

Impregnation progress throughout further processing was observed for all test panels. Complete impregnation was reached for 90 % impregnated tapes after a dwell time of 300 s and after thermoforming. After pressing tapes with a DOI_i of 80 % for 300 s the final DOI_f of 95 % indicated that longer dwell times are required to complete impregnation. The longitudinal flexural strength of completely and 90 % impregnated tapes is comparable after pressing for 300 s and after thermoforming. The transverse flexural strength is affected by the DOI_i to a stronger extent indicating that dwell times need to be increased to enable fiber bed relaxation and consolidation of tape plies with an DOI_i of less than 100 %.

Excerpt from ISBN 978-3-00-053387-7

1. Introduction

Enabling short forming and consolidation times, the fusibility of thermoplastics is one of the major advantages that lead to their increasing use as matrix materials in continuous fiber reinforced polymers (CFRP). Reactive thermoset matrix systems show typically viscosities that are lower by several magnitudes than most thermoplastics so that impregnation of fibrous components is facilitated [1–3]. In the last decades, several solutions were found to overcome the difficulties during impregnation of fibers with thermoplastics. These can be divided into completely impregnated intermediate products such as tapes or consolidated laminates and partially impregnated intermediate products such as commingled yarns [2] or powder-prepregs [4–6] with reduced flow path. In addition, there are reactive thermoplastic systems with low-molecular components that ease impregnation and react during processing known from thermoset matrix processing [7, 8].

Manufacture of completely impregnated prepreg materials is time- and energy-consuming leading to high material cost. In a typical production process chain to manufacture thermoplastic composites, several heating processes follow to consolidate tapes to multi-ply laminates and to thermoform them to the final component geometry. All process steps are associated with heating the polymer above glass transition or melting temperature enabling the thermoplastic matrix to further flow. This study aims to investigate if partially impregnated tapes can be completely impregnated throughout a typical production process for thermoplastic composite components. This principle is also known for Out-of-Autoclave (OoA) prepregs [9, 10]. Partial impregnation of tapes e.g. by increasing velocity in a double-belt press can significantly decrease the cost of intermediate products due to increased output. By using less expensive matrix systems such as Polyamide 6 (PA6), Co-Polyamides (PA6T, PA10T) or Polyetherimide (PEI) and Polyether sulfone (PES) [11] along with cost-efficient intermediate product manufacture and time-efficient processing, the overall component cost can be reduced.

2. Experimental

2.1. Manufacture of tapes with different Initial Degrees of Impregnation (DOI_i)

Three different initial degrees of impregnation (DOI_i) of unidirectional carbon fiber reinforced tapes (UD tape) with PA6 as matrix were produced by using varying velocities and pressures in a double-belt press. The matrix material was supplied by BASF (tradename: Ultramid B3Ssw), grinded to powder and deposited from both sides on nine SIGRAFIL C T50-4.0/240-T140 roving spread to a nominal width of 200 mm (powder-prepregs). Subsequently, the powder-prepregs were consolidated in a double-belt press. By varying velocity and pressure of the double-belt press different DOI_i of the UD tapes were yielded. For each process parameter set given in Table 1 the temperature of the heating zones were adapted to maintain similar conditions for all produced tapes. Since the duration of impregnation is governed by the transverse impregnation of fiber bundles [12], the DOI_i of all produced tapes is determined for several single fiber bundles instead of entire tape cross-sections. The DOI_i was evaluated by post-processing of micrographs using Matlab R2014b. In general, subtracting the non-impregnated area ($A_{porosity\,within\,fiber\,bundle}$) from the entire fiber bundle area ($A_{fiber\,bundle}$) divided by the entire fiber bundle area is considered [13, 14] to yield the DOI as presented in Equation 1.

$$Degree\ of\ Impregnation\ (DOI) = \frac{A_{fiber\,bundle} - A_{porosity\,within\,fiber\,bundle}}{A_{fiber\,bundle}} \quad (1)$$

After preparing micrographs of the manufactured tapes the DOI_i was determined for five fiber bundles each and the results are presented in Table 1 along with the normalized process settings in the double-belt press.

Table 1. Double-belt press settings and DOI_i of differently processed tapes

Designation of tapes	DBP Settings Velocity [m/min]	Pressure [bar]	DOI_i determined from micrographs with Matlab [%]
Tape_100 %	v_0	p_0	99.98±0.00
Tape_90 %	$2v_0$	$0.125p_0$	86.43±0.10
Tape_80 %	$3v_0$	$0.125p_0$	81.94±0.20

Excerpt from ISBN 978-3-00-053387-7

2.2. Laminate Production

The tapes with different DOI_i were stacked to obtain laminates with the layup $[0_{12}]$ yielding a nominal thickness of 2 mm. After drying of all stacks in a vacuum oven at 80 °C for at least four hours, they were transferred to a press heated to 260 °C. As soon as the stacks were heated uniformly a pressure of 10 bar was applied. Two different dwell times (90 s and 300 s) were applied while holding the pressure constant before cooling the laminates to 80 °C with a cooling rate of 20 °C/min.
Some of the laminates pressed for 90 s were additionally “thermoformed”. They were positioned in a flat-shaped tooling and pre-heated in an infrared field. As soon as 270 °C were reached, controlled by thermocouples, the laminates were rapidly transferred to non-heated press plates and pressed with 10 bar. Thus, the potential impregnation progress due to the thermal history during typical thermoforming processes is received while maintaining the flat shape of the test panels.
The designation of the test panels follows the scheme “DOI_i of tapes in % _ Dwell time in seconds in a static press _ Dwell time in seconds during thermoforming (TF)”. Here, the dwell time describes the time of the polymers heated above melting temperature (225 °C) while pressure is applied. Before pressure application, the test panels dwell above melting temperature for 250-291 s during heating while thermoforming and approximately 180 s during heating in the static press .
After test panel manufacture, micrographs were prepared and the final Degree of Impregnation (DOI_f) of all test panels was determined by Equation 1 in an analogous manner to DOI_i of the produced tapes.

2.3. Test Setup and Procedure

Four-point flexure tests (DIN EN ISO 14125 B) were conducted to determine the effect of the DOI_i of UD tapes on the longitudinal and transverse flexural moduli (E_{1b} and E_{2b}) and longitudinal as well as transverse flexural strengths (σ_{1b} and σ_{2b}). Prior to testing, all test specimens were dried in a vacuum oven at 80 °C for at least 60 hours to ensure complete water removal. Longitudinal tests were conducted with a support span of 45 mm and a load span of 15 mm at a crosshead speed of 5.00 mm/min. Transverse tests were carried out with a support span of 81 mm and a load span of 27 mm at a crosshead speed of 2.00 mm/min. All tests were carried out in a displacement-controlled mode on a Hegewald & Peschke universal testing machine at room temperature. The mid-point deflection was determined by means of a video-extensometer with telecentric lenses. The outer fiber strain was derived from the deflection in accordance with DIN EN ISO 14125 B - section 10.2.3. For each case seven specimens were tested. Altogether, 126 specimens were tested in this study. The fiber volume content (FVC) of each test panel was determined by acid digestion according to DIN EN 2564 B.

3. Results and Discussion

3.1. Determination of DOI_i and DOI_f

Micrographs of tapes with different DOI_i and of laminates manufactured in a press for 90 s, 300 s and after thermoforming are presented in Figure 1. The micrographs served to determine the DOI_i of the produced tapes and the DOI_f of the laminates. As presented in Figure 1, completely impregnated tapes (100 % DOI_i) exhibit fiber bundles that are entirely infused by the matrix. Independent of dwell time or additional thermoforming, all laminates pressed from completely impregnated tapes show uniformly spread consolidated plies without porosity. Tapes with a DOI_i of 90 % show some dry areas within a fiber bundle as presented in Figure 1. After subjection to a dwell time of 90 s non-impregnated areas are detected to a very limited extent. An extension of the dwell time to 300 s yields completely impregnated tapes without entrapped air. The same observation is made after thermoforming tapes with a DOI_i of 90 %. In contrast, tapes with a DOI_i of 80 % show large non-impregnated areas within the fiber bundle.

Excerpt from ISBN 978-3-00-053387-7

Progress of impregnation can be detected after pressing these tapes for 90 s but non-impregnated areas are clearly visible for large fiber bundles. Non-impregnated areas are still existent but further shrink when the laminate is pressed for 300 s. After thermoforming, the laminate produced from tapes with a DOI_i of 80 % exhibit completely impregnated fiber bundles.

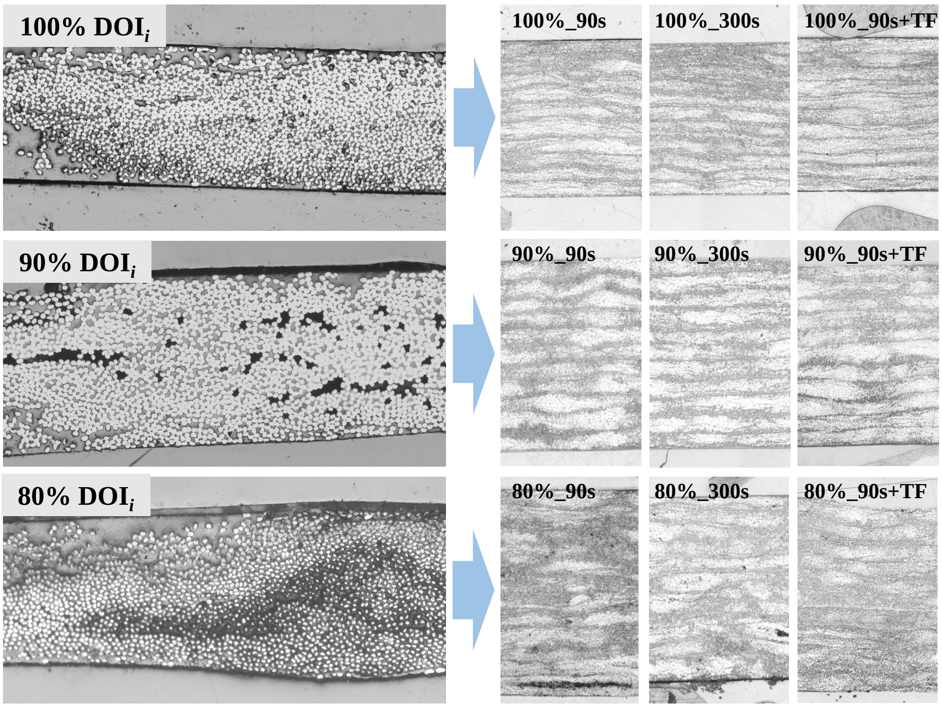

Figure 1. Micrographs of tapes with different DOI_i and of test panels pressed for 90 s, 300 s and after thermoforming

The results for DOI_f of all test panels determined from micrographs are presented in Table 2 along with the used press settings.

Table 2. Press settings and final DOI_f of test panels

Designation of test panels	Press Settings: Dwell time static press [s]	Press Settings: Dwell time thermoforming [s]	DOI_f of test panels analyzed with Matlab from micrographs [%]
100 % 90s	90	-	100.00±0.00
90 % 90s	90	-	97.29±0.01
80 % 90s	90	-	94.51±0.01
100 % 300s	300	-	100.00±0.00
90 % 300s	300	-	100.00±0.00
80 % 300s	300	-	95.66±0.01
100 % 90s TF	90	8	99.81±0.00
90 % 90s TF	90	9	99.40±0.01
80 % 90s TF	90	8	100.00±0.00

The FVC is depicted in Figure 2 along with the mean average thickness of all test panels. The scattering

Excerpt from ISBN 978-3-00-053387-7

of the FVC is found to be high except for test panels pressed for 90 s and made from completely impregnated tapes and laminates produced from tapes with a DOI$_i$ of 80 % that were thermoformed. There is no correlation between measured FVC and panel thickness. The significantly increased panel thickness of laminates produced from tapes with a DOI$_i$ of 80 % and pressed for 90 s is not accompanied by a decrease in FVC. FVC being independent of the test panel thickness is explained by dimensional changes without mass loss during processing in closed tooling. A large width and low consolidated ply thickness was detected (Figure 1) for completely impregnated tapes. In contrast, partially impregnated tapes show increased consolidated ply thickness and reduced width. In general, the test panel thickness appears to be increasing with decreasing level of DOI$_i$ for all press settings.

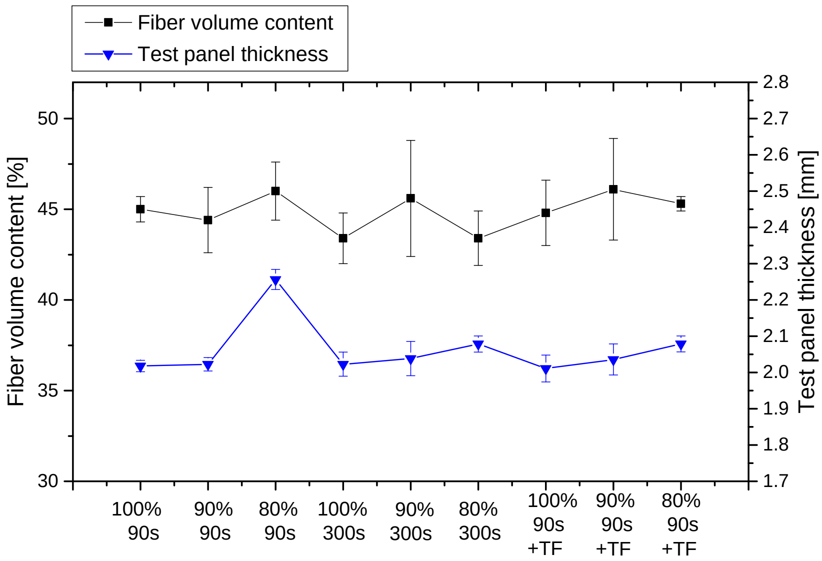

Figure 2. FVC and mean average thickness of all test panels used in this study

3.2. Longitudinal flexural properties

Figure 3 presents the longitudinal flexural strength σ_{1b} along with the flexural modulus E_{1b} in fiber direction for test panels after pressing with short and long dwell times by using tapes with varying DOI$_i$. Referring to short dwell times of 90 s, the longitudinal flexural strength σ_{1b} slightly decreases with decreasing DOI$_i$. This goes along with an increase in mean average thickness of test panels produced from 90 % and 80 % impregnated tapes as shown in Figure 2. In general, the level of σ_{1b} for all test panels pressed for 90 s is below the test panels that were pressed for 300 s or underwent thermoforming. These results indicate that a dwell time of 90 s is not sufficient to achieve full consolidation between the stacked tape plies. Extending the dwell time to 300 s or after thermoforming gives similar σ_{1b} for completely impregnated tapes and tapes with a DOI$_i$ of 90 %. For all test series, the tapes with a DOI$_i$ of 80 % yielded the lowest σ_{1b}.
The flexural modulus E_{1b} shows similar results for the tested specimen within standard deviation. Only 80 % impregnated tapes that were pressed for 90 s reveal a flexural modulus reduced by 32 % in comparison to the mean average of all test series (89.14 GPa). This is assumed to arise from incomplete impregnation (DOI$_f$ of 94.51 %) and significantly increased panel thickness by 12 %.
In general, the longitudinal strength correlates well with the obtained DOI$_f$ and is found to be a suitable method for evaluation of the effects of DOI$_i$ on the composite properties.

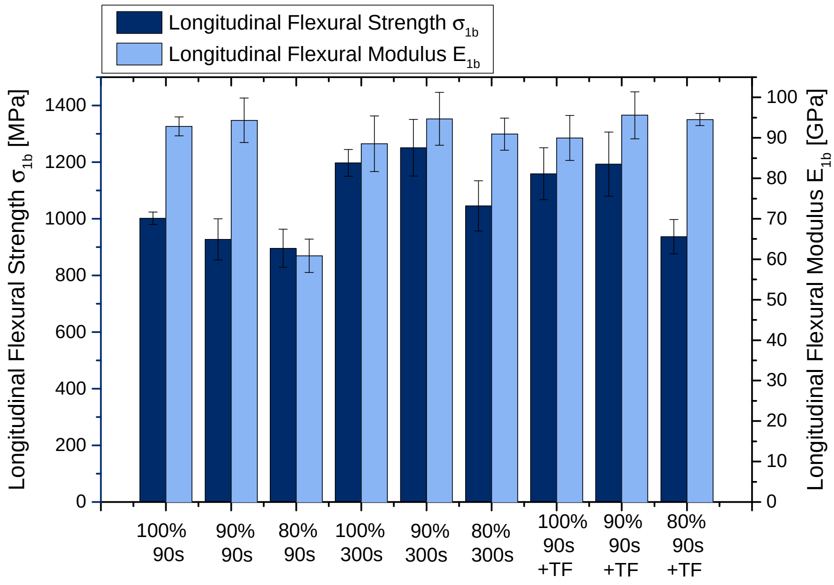

Figure 3. Longitudinal flexural properties of test panels made of tapes with different DOI$_i$ and pressed with varying dwell times and after thermoforming

3.3. Transverse flexural properties

The transverse flexural properties for test panels pressed from tapes with different DOI$_i$ are reported in Figure 4. Short dwell times of 90 s yielded comparable results for the transverse flexural strength σ_{2b}

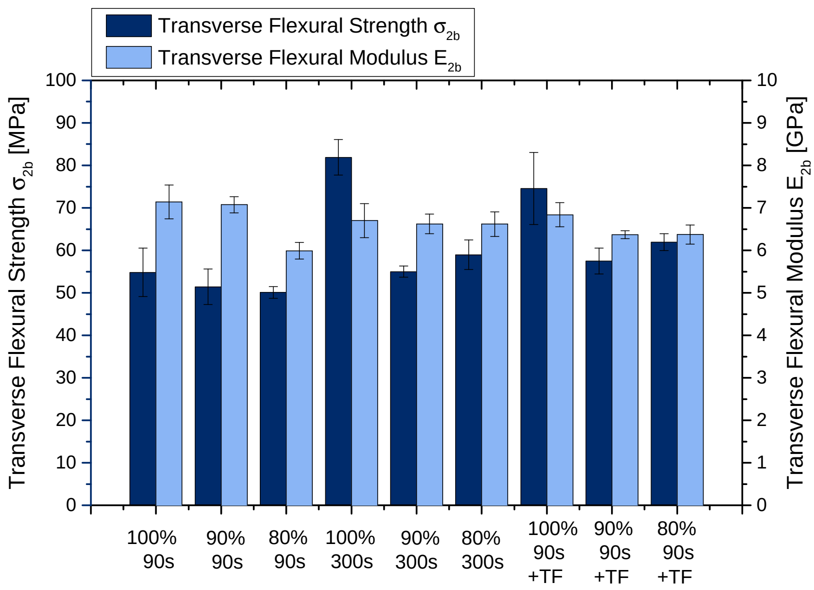

Figure 4. Transverse flexural properties of test panels made of tapes with different DOI$_i$s and pressed with varying dwell times and after thermoforming

within standard deviation independent of the DOI$_i$. As observed for longitudinal strength, σ_{2b} of all test panels pressed for 90 s remains at a lower level than laminates that were pressed for 300 s or were thermoformed. Comparing to short dwell times of 90 s, σ_{2b} of laminates made of tapes with DOI$_i$ of

80 % and 90 % improves when the dwell time is extended or a thermoforming step is added. However, the level of σ_{2b} obtained from laminates with completely impregnated tapes is not reached when tapes with DOI_i of 80 % and 90 % are used. σ_{2b} of test panels pressed for 300 s and thermoformed from tapes with a DOI_i of 90 % is found to be slightly lower than for laminates made from tapes with a DOI_i of 80 %. Slight increases in FVC for the correspondent laminates made from 90 % impregnated tapes are assumed to cause lower values for σ_{2b}.
Even if the DOI_f of test panels made of tapes with a DOI_i of 80 % and 90 % yield nearly 100 % after subjection to 90 s in a press with subsequent thermoforming, the transverse flexural strength σ_{2b} shows lower values than for test panels produced from completely impregnated tapes. Insufficient air evacuation and less time for relaxation of the fiber bed during manufacture is assumed to cause the deterioration in transverse flexural strength. Large fiber bundles in the test panel produced from 80 % impregnated tapes are believed to cause incomplete impregnation throughout a dwell time of 300 s.
The transverse flexural modulus E_{2b} of completely and 90 % impregnated tapes is comparable for short and long dwell times. Only 80 % impregnated tapes that were pressed for 90 s resulted in a reduction in E_{2b} by 16 % compared to completely impregnated tapes. This is assumed to arise from incomplete impregnation and a lack of consolidation between tape plies.

4. Conclusion

UD tapes with PA6 were produced in a double-belt press yielding three different DOI_i of 80 %, 90 % and 100 % analyzed from micrographs. The tapes with different DOI_i were stacked and consolidated to laminates by using varying dwell times in a static press and an additional thermoforming step. The final DOI_f was analyzed from micrographs of all test panels. The micrographs document that a further progress of the impregnation is possible throughout a production process chain for CFRTP.
To enable the evaluation of effects of DOI_i on composite properties, flexural tests were conducted for all laminates. A dwell time of 300 s or additional thermoforming was sufficient to complete impregnation of tapes with a DOI_i 90 % confirmed by the results for the longitudinal flexural strength σ_{1b}. Manufacturing tapes with a DOI_i of 90 % goes along with doubling the velocity in a double-belt press. The increased throughput reduces the cost during tape manufacture and can yield potential for cost saving throughout the production of CFRTP. Tapes with a DOI_i of 80 % could not be completely impregnated after pressing for 300 s in a press resulting in DOI_f of 95 % that affected the longitudinal flexural strength. Both the longitudinal and transverse flexural strength yielded lower values independent of the DOI_i after processing the stacked tapes in a press for 90 s in comparison to long dwell times of 300 s indicating a lack of consolidation between the stacked tapes. Referring to the deviations in flexural strength when completely impregnated and tapes with a DOI_i of 80% are compared, the transverse flexural strength is more influenced by the DOI_i than the flexural strength in fiber direction. In general, both longitudinal E_{1b} and transverse flexural modulus E_{2b} appear to be barely affected by the DOI_i of the used tapes.
Altogether, the impregnation progress of fibers can further advance during typical production processes of CFRTP. The chosen dwell times of 300 s are considered to complete impregnation of initially 90 % impregnated tapes but may be insufficient for tapes with a DOI_i of 80 %. In addition, the increased panel thickness and low values for transverse flexural strength of tapes with a DOI_i of 80 % indicate that dwell times of more than 300 s are required to enable complete impregnation but also sufficient consolidation between the tape plies.

Acknowledgements

The preparation of test specimens and micrographs by Adrián García López is gratefully acknowledged by the authors.

References

[1] J.A.H.M. Buijs and P. J. Nederveen. A study of consolidation in filament winding with thermoplastic prepregs. *Journal of Thermoplastic Composite Materials*, 5(4):276–286, 1992.

[2] N. Svensson, R. Shishoo, and M. Gilchrist. Manufacturing of thermoplastic composites from commingled yarns - a review. *Journal of Thermoplastic Composite Materials*, 11(1):22–56, 1998.

[3] P. McDonnell, K.P McGarvey, L. Rochford, and C.M Ó Brádaigh. Processing and mechanical properties evaluation of a commingled carbon-fibre/PA-12 composite. *Composites Part A: Applied Science and Manufacturing*, 32(7):925–932, 2001.

[4] M. Connor, S. Toll, Manson, J. -A. E., and A. G. Gibson. A model for the consolidation of aligned thermoplastic powder impregnated composites. *Journal of Thermoplastic Composite Materials*, 8(2):138–162, 1995.

[5] S. R. Iyer and L. T. Drzal. Manufacture of powder-impregnated thermoplastic composites. *Journal of Thermoplastic Composite Materials*, 3(4):325–355, 1990.

[6] C. Steggall-Murphy, P. Simacek, S. G. Advani, S. Yarlagadda, and S. Walsh. A model for thermoplastic melt impregnation of fiber bundles during consolidation of powder-impregnated continuous fiber composites. *Composites Part A: Applied Science and Manufacturing*, 41(1):93–100, 2010.

[7] H. Parton, J. Baets, P. Lipnik, B. Goderis, J. Devaux, and I. Verpoest. Properties of poly(butylene terephthatlate) polymerized from cyclic oligomers and its composites. *Polymer*, 46(23):9871–9880, 2005.

[8] J. Verrey, M. D. Wakeman, V. Michaud, and J.-A.E. Månson. Manufacturing cost comparison of thermoplastic and thermoset RTM for an automotive floor pan. *Composites Part A: Applied Science and Manufacturing*, 37(1):9–22, 2006.

[9] T. Centea and P. Hubert. Modelling the effect of material properties and process parameters on tow impregnation in out-of-autoclave prepregs. *Composites Part A: Applied Science and Manufacturing*, 43(9):1505–1513, 2012.

[10] T. A. Cender, P. Simacek, and S. G. Advani. Resin film impregnation in fabric prepregs with dual length scale permeability. *Composites Part A: Applied Science and Manufacturing*, 53:118–128, 2013.

[11] P. Mitschang, M. Blinzler, and A. Wöginger. Processing technologies for continuous fibre reinforced thermoplastics with novel polymer blends. *Composites Science and Technology*, 63(14):2099–2110, 2003.

[12] X. Wang, C. Mayer, and M. Neitzel. Some issues on impregnation in manufacturing of thermoplastic composites by using a double belt press. *Polymer Composites*, 18(6):701–710, 1997.

[13] B. Thorfinnson and T. F. Biermann. Degree of impregnation of prepregs - effects on porosity. *32nd International SAMPE Symposium, Anaheim, California*, April 6-9, 1987.

[14] P. Peltonen, K. Lahteenkorva, E. J. Paakkonen, P. K. Jarvela, and P. Tormala. The influence of melt impregnation parameters on the degree of impregnation of a polypropylene/glass fibre prepreg. *Journal of Thermoplastic Composite Materials*, 5(4):318–343, 1992.

Excerpt from ISBN 978-3-00-053387-7