CONSOLIDATION MODEL FOR OPTIMIZATION OF THERMOPLASTIC COMPOSITE LAMINATION PROCESSES

Huajie Shi[1], Jeffrey Lugo[2], Bert Rietman[1] and Nikhil Verghese[1]

[1] SABIC, P.O. Box 319, 6160AH, Geleen, The Netherlands.
[2] SABIC, 475 Creamery Way, Exton, PA 19341-2537, US.
Huajie.shi@SABIC.com
Jeffrey.Lugo@SABIC.com
Bert.Rietman@SABIC.com
Nikhil.Verghese@SABIC.com

Keywords: Consolidation model, Thermoplastic composites, Lamination, Unidirectional tape

ABSTRACT

Lamination by co-consolidation of fibre reinforced thermoplastic prepreg is an efficient way to make thermoplastic composite laminates. The quality of the laminate is to a large extent defined by the lamination process and is dependent on the applied temperature and pressure profiles, but also on the quality of the incoming prepreg used. The objective of this study was to identify the primary influencing parameters and develop a consolidation model that allows one to optimize the lamination process with respect to quality and throughput efficiency.

A consolidation model was developed to analyse the underpinning mechanisms during lamination, namely intimate contact and autohesion (or molecular inter-diffusion). Unidirectional glass fibre reinforced polypropylene (GF/PP) tapes were laminated using a static hot press and sensitivity studies were performed to assess the key material properties and processing parameters. A reasonable agreement was achieved between the consolidation quality predicted by the model and the interlaminar shear strength measured from the experiments. The work and the results of this study provide good guidance to optimize the lamination process towards cost-effectiveness and mass productivity.

1 INTRODUCTION

The application of continuous fibre reinforced composites has developed from their origins in the aircraft industry to other industries such as automotive, oil & gas, and consumer electronics due to the potential to achieve cost effectiveness at large build volumes (i.e. mass production) and still deliver light weighting. Thermoplastic composites provide additional advantages such as high toughness, good damage tolerance, shapeability and recyclability. Thermoplastic composites can also be easily integrated with injection molding grades to extend their shape complexity and achieve a balance between performance and cost. Lamination by co-consolidation of fibre reinforced thermoplastic prepreg/semi-preg has been found to be an efficient way to make thermoplastic composite laminates, for which high temperatures and pressures are usually involved. The quality of the thermoplastic laminates as well as the conversion cost are therefore largely dependent on the lamination process, in particular the temperature and pressure profiles.

To date, the optimization of lamination processes is based largely on previous experience and trial and error. The aim of this study is therefore to identify the main influencing parameters on lamination and develop a model to help optimize the lamination process and identify a feasible processing window. Better understanding of lamination of thermoplastic prepreg-based composites will be gained by analysis of the underpinning mechanisms in the lamination process, and by sensitivity studies of the key processing parameters. The validity of the consolidation model will be verified on a selected material system for a selected lamination process, and the consolidation quality of the laminates will be measured by short beam shear tests and compared to the model predictions of interlaminar strength development. The results of this study can be used as a guide to optimize the lamination process towards cost-effective mass production. The framework of such a model can also be used in other classes of problems such as welding, joining and sealing of thermoplastic polymers.

2 MATERIALS AND EXPERIMENTAL SET-UP

SABIC FRT's UDMAX™ GPP 45-70 tape (unidirectional fully impregnated GF/PP tape) [1], with nominal thickness of 0.25 mm and fibre weight fraction of 70%, was used in this study. The laminates were made of 16 layers of tapes in a stacking sequence of $[0]_{16}$, yielding a final laminate thickness of around 4 mm. Consolidation was performed in a WABASH MPI 100 ton hydraulic vacuum compression press, model# S100H-24-BCX. The tape preform was laid-up in a custom-designed 350x350 mm^2 picture frame steel tooling, as shown in Figure 1. Time, compaction pressure, as well as platen and external mold temperature were directly measured through the controller from Yokogawa. Thermocouple wires were embedded into the laminate preform for precise recording of the spatial temperature profile.

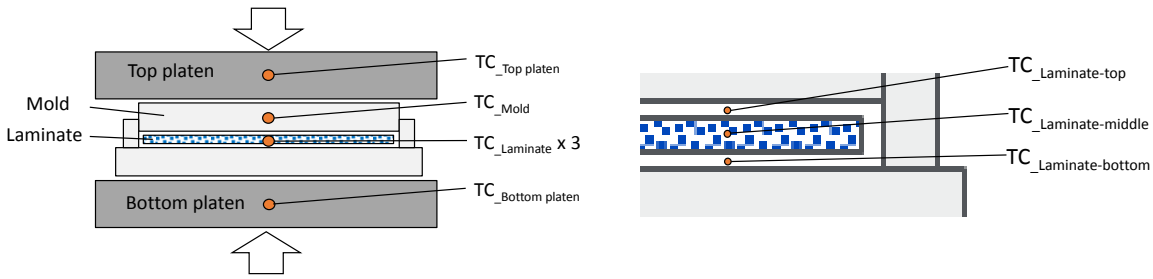


Figure 1: Steel tooling used to house tape preform and the positions of the thermocouples used during lamination.

After consolidation, test specimens were cut from the laminates using a diamond-blade wet saw with the fibre orientation aligned along the length of the specimens. Short beam shear tests were performed according to the ASTM D2344 standard [2], with the test fixture and sample setup shown in Figure 2. A span-to-thickness ratio of 4:1 was used, and the cross-head speed was set constant to 1.27 mm/min. An Instron® testing machine was used for the study, and at least six test samples were obtained per consolidation setting. The consolidation degree, or degree of bonding, of each laminate was obtained by comparing the interlaminar shear strength (ILSS) of the laminates with the ILSS of the benchmark samples (consolidated under a high enough temperature, i.e. 194°C, for long enough time, i.e. 20 min, which gives the highest ILSS) and calculating the ratio. Optical microscopy images were made on cross-sections of a selected number of interesting samples.

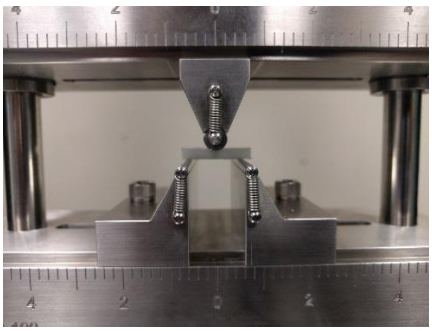


Figure 2: Fixture and test set-up for short beam shear test of GF/PP laminates.

3 MODELLING

Theories of consolidation model and characterization of the material properties required in the model are discussed in this Chapter.

3.1 Consolidation model

Several mechanisms and steps are involved during the lamination process of tape/prepreg-based thermoplastic composites. These include: intimate contact of layers, molecular autohesion (or healing), void dynamics, fiber impregnation, squeeze flow, and crystallization (in case semi-crystalline

thermoplastics are used). However, not all of these mechanisms contribute to the consolidation quality. Intimate contact and healing have been regarded as the two most important steps that govern the consolidation quality if fully impregnated tapes/prepregs are used [3-7], and therefore are the main focus of this study.

Intimate contact is defined as the relative amount of surface area that is physically in contact at the interface between two plies and is directly related to the applied temperature, pressure and residence time at a location given a certain surface roughness. The model employed for this work [8] describes intimate contact by considering that the surfaces are an approximate distribution of identical rectangular elements, as shown in Figure 3

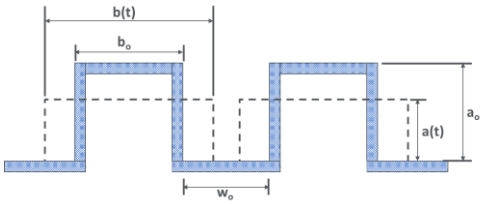


Figure 3: Idealization of surface roughness as array of identical rectangular elements [8].

Given the geometric relations and conservation of mass for a given control volume, the degree of intimate contact, D_{IC}, can be described [8]:

$$D_{IC} = R_C \left[\int_0^{t_p} \frac{P}{\mu} dt \right]^{\frac{1}{5}} \tag{1}$$

$$R_C = D_{ICO} C_1^{0.2} \tag{2}$$

$$D_{ICO} = \frac{1}{1+\frac{w_o}{b_o}}, \; C_1 = 5\left(1+\frac{w_o}{b_o}\right)\left(\frac{a_o}{b_o}\right)^2 \tag{3}$$

in which a_o, b_o and w_o refer to the initial geometry as shown in Figure 3. D_{ICO} refers to the initial intimate contact, and R_C is the lumped surface roughness constant, which allows for the experimental characterization of the initial status of intimate contact. Fitting the surface of a material based on its asperities allows us to determine the intimate contact at a discrete time.

Autohesion or the degree of polymer healing, describes the migration of polymer chains across the interface. It determines the development of interfacial bonding strength which is critical for laminate quality. The reptation theory for polymer chain diffusion [9] is used to describe the motion of molecular chains within the bulk material to the motion across an interface [10, 11]. The model employed in this work [12, 13] describes the degree of healing, D_{AU}, with the bond strength σ:

$$D_{AU} = \frac{\sigma}{\sigma_\infty} \propto \left(\frac{t}{t_r}\right)^{\frac{1}{4}} \tag{4}$$

where σ_∞ is the strength of a polymer in the bulk material, and t is the time for healing.

By dividing the thermal history into sufficiently small intervals ($\tau_{i+1} - \tau_i = \Delta\tau$), a non-isothermal history can be treated as the sum of those isothermal steps:

$$D_{AU} = \frac{\sigma}{\sigma_\infty} = \sum_{i=1}^{\tau_H/\Delta\tau} \left[\frac{(\tau_i)^{\frac{1}{4}} - (\tau_{i-1})^{\frac{1}{4}}}{t_r(T)^{\frac{1}{4}}} \right] \tag{5}$$

where $t_r(T)$ is the reptation time at temperature T, and $\tau_H$is the elapsed time of the healing process. Thus, the degree of healing is found at each time step and summed to give a final degree of healing.

As intimate contact is a prerequisite for healing at the surface to occur, a coupled bonding model is used to capture the development of consolidation, or the degree of bonding D_B [14, 15]:

$$D_B(n) = min\left(\sum_{i=1}^{n-1}\left[\Delta D_{IC}(i)\cdot min\left(\sum_{j=i+1}^{n} D_{AU}(T_j,t_j),1\right)\right],1\right) \quad (6)$$

where $D_B(n)$ stands for the local development of bonding at step n, $\Delta D_{IC}(i)$ is the increase of intimate contact at step i, $D_{AU}(T_j,t_j)$ is the extent of autohesion occurring during step j, T_j and t_j are the average temperature and time elapsed during step j. The degree of bonding is limited to a maximum value of 1.

3.2 Material characterization

Accurate model inputs are of great importance to ensure good model predictions, and therefore, the GF/PP tape and the thermoplastic resin used in the tape were carefully characterized.

The initial surface roughness of the tape was measured using a Keyence® VK-X100 3D laser microscope. To obtain a sufficiently adequate representation of the tape surface, average values were obtained from horizontal scans of the tape from 10 different locations. Surface fitting was used to calculate the surface roughness constant - R_C, and an average value of 0.4 was found.

Due to the presence of a resin-rich layer on the surface of the GF/PP tape, surface deformation will mainly happen in the resin area, and therefore, the viscosity of PP, instead of the viscosity of GF/PP composite, was used for the simulation. The viscosity of PP was measured by parallel-plate rheometer and dynamic mechanical analysis (DMA) for the low temperature range. The plateau modulus, G_N^n, and the zero shear rate viscosity, η_0, the two important parameters for estimating the reptation time [8], were also calculated from the rheology and DMA measurements.

Reptation theory states that the reptation time has the same value as the terminal relaxation time [12]. By performing steady-state shear experiments with a parallel plate rheometer, the terminal relaxation time can be determined by the critical angular frequency (onset of nonlinear viscosity behavior). However, this method is only applicable for material in molten state. For the lower temperature range, reptation time was calculated from the reptation theory [16-21]:

$$\tau_d = \frac{45}{\pi^2}\left(\frac{G_N^0}{\rho R_G T}\right)^2 \frac{M^3\eta_0(M=M_c)}{M_c G_N^0} \quad (12)$$

where ρ is density, R_G is gas constant, T is temperature, M is molecular weight, M_c is critical molecular weight, and $\eta_0(M=M_c)$ is the viscosity at the critical molecular weight. The full range reptation time calculated for the PP used in this study is plotted in Figure 4.

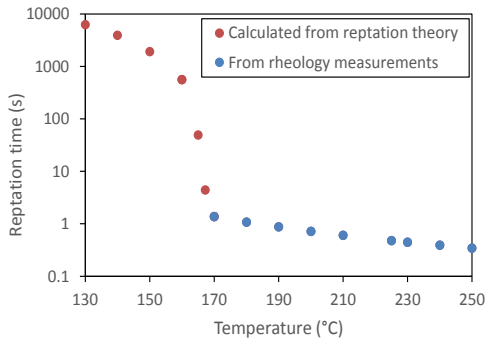


Figure 4: Reptation time of the PP polymer used in the GF/PP tape

The value of reptation time is dependent on the nature of the polymer, such as the molecular structures, weight and distribution. Therefore, considering the distribution of molecular weight, the reptation time of macromolecule is usually not a single value but a wide range at a given temperature. Due to the lack of a well-established relationship between the polydispersity and reptation time, a measurement value was simply used instead of a wide range for each temperature.

4 RESULTS AND DISCUSSION

In this Chapter, a sensitivity study was performed to assess the influence of key input parameters

on consolidation quality, and validation of the present model framework is given.

4.1 Sensitivity study of input parameters

With the aid of the presented model framework, the development of consolidation quality or the degree of bonding - D_B can be predicted for a given temperature and pressure profiles, as shown in Figure 5. Besides the degree of bonding degree, the degree of intimate contact - D_{IC} and the degree of autohesion - D_{AU} were also shown in the figure to provide detail information during the development of consolidation.

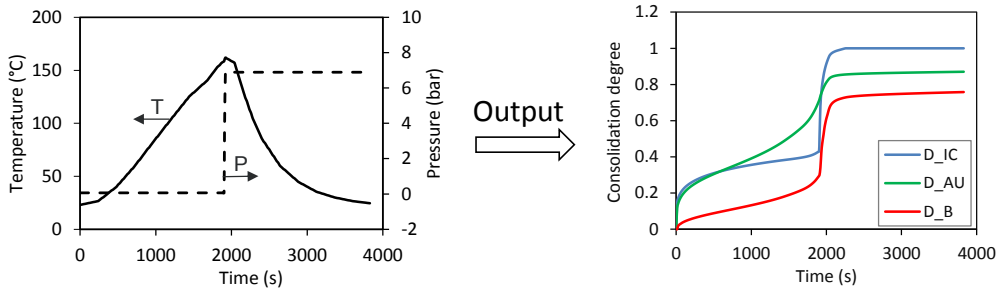
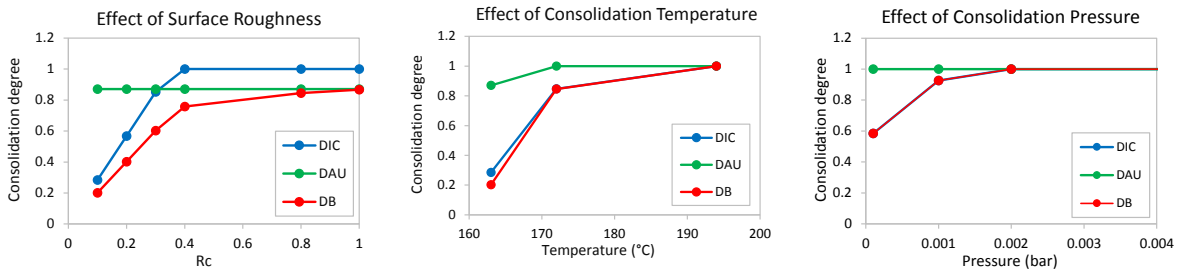


Figure 5: (a) An example of the input temperature and pressure profiles, and (b) the model predictions.

In order to understand the influence of the key input parameters on consolidation quality, a sensitivity study was performed to assess the influence of surface roughness, consolidation temperature and consolidation pressure. As shown in Figure 6 (a), surface roughness shows an influence on the development of intimate contact, while showing no influence on autohesion. For a system with increased R_C, or in other words a smoother tape, the development of intimate contact is faster, and as a consequence a higher bonding degree is obtained.



(a) (b) (c)

Figure 6: Sensitivity study of the key parameters: (a) surface roughness ($T_{con} = 163$ °C, $P_{cons} = 6.9\ bar, t_{dwell} = 0\ min$), (b) consolidation temperature ($P_{con} = 6.9\ bar, t_{dwell} = 0\ min, R_C = 0.4$) and (c) consolidation pressure ($T_{con} = 163$ °C, $R_C = 0.4, t_{dwell} = 0\ min$).

The matrix of the composite should be heated above the glass transition temperature for an amorphous polymer or melting temperature for a semi-crystalline polymer in order to promote molecular inter-diffusion at the ply interface. As shown in Figure 6 (b), both intimate contact and autohesion are found to increase with increasing temperature, and as a result of that, a higher bond quality is obtained for a higher consolidation temperature. A dramatic increase of bonding degree was found when the processing temperature increased from 163 °C to 172 °C, which falls into the melting range and correlates with the dramatic decrease of reptation time in this temperature range (see Figure 4).

During consolidation, a certain pressure is required for the development of intimate contact, which is clearly shown in Figure 6 (c). It has to be noted that once the consolidation pressure is higher than a critical value, further increase of consolidation pressure does not improve the bond quality.

4.2 Model validation

The validation of the presented model was performed by lamination of GF/PP tapes in a static press and subsequent assessment of the interlaminar shear properties. This was performed on the basis of a

number of experiments with varying dwell time, temperature and pressure. Furthermore a feasible process window for static press consolidation of the considered materials was constructed.

As the production rate and cost are closely related to the cycle time, the influence of dwell time on consolidation quality was investigated. Current study showed that a dwell time of 20 min gave full consolidation, such that this time is used as the reference. Therefore lamination tests with shorter dwell times of 10 min and 0 min were performed, and the temperature and pressure profiles are shown in Figure 7. The temperatures measured at the middle of the laminates ($TC_{\_laminate\_middle}$) were used for the simulation, although very little difference on temperature was found through the thickness. Figure 8 shows a comparison of degree of consolidation based on model predictions and experimental calculations from ILSS measurements. Full consolidation was predicted for all three laminates, which has also been confirmed by the experiments. The dwell time was found to be not important for the studied material with this particular temperature and pressure settings. As shown in Figure 9, the cross-section microscopy image shows no clear gap between the individual layers, which confirms good interlayer bond quality in the laminates.

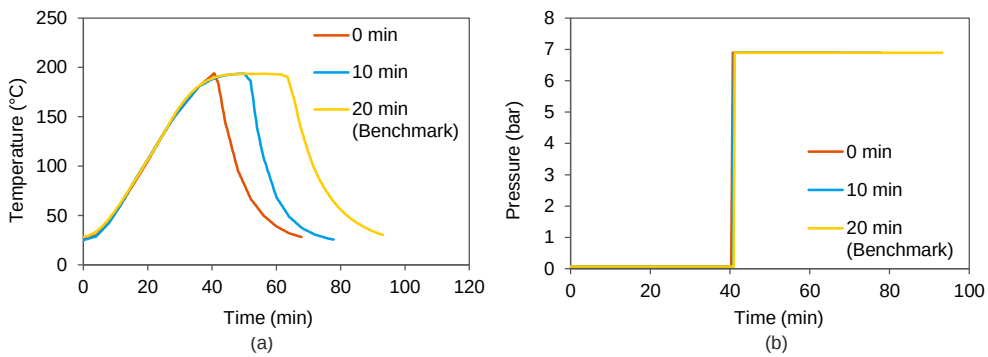


Figure 7. Temperature (a) and pressure (b) profiles of hot press lamination with different dwell times of 0, 10 and 20 min.

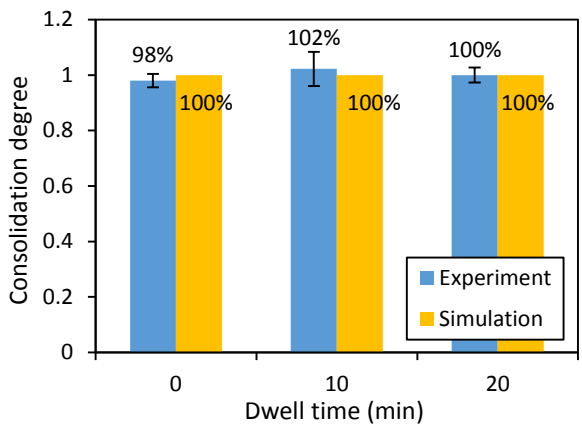


Figure 8. Comparison of simulated consolidation degrees with experimental results for different dwell times (the same consolidation temperature and pressures as the benchmark samples were used: $T_{con} = 194\ ℃, P_{con} = 6.9\ bar$).

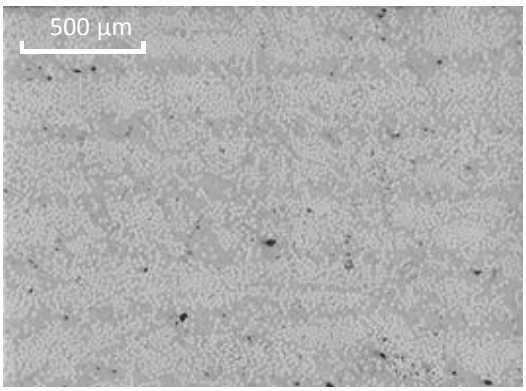


Figure 9. Microscopy image of benchmark laminate, consolidated at 194 °C with a pressure of 6.9 bar and a dwell time of 20 min.

Lamination with different consolidation temperatures was performed to determine the minimum required temperature to get full consolidation. Figure 10 shows a comparison between the model predictions and experimental results, and an overall good match was observed. Insufficient bond quality was obtained in the laminate that was consolidated at a lower temperature of 163 °C. This is clearly indicated by the visible gaps between the plies in Figure 11. For the studied system, full consolidation can be achieved when the processing temperature is above 172 °C.

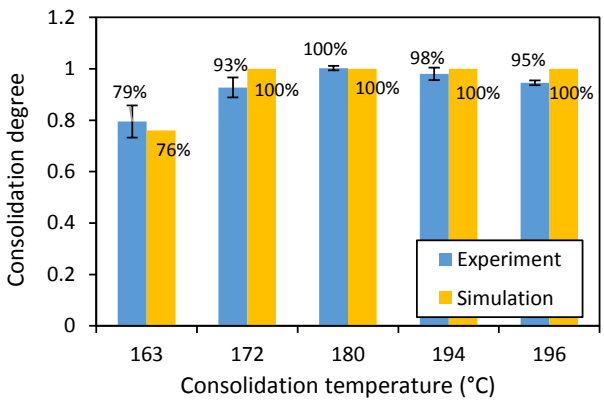


Figure 10. Comparison of simulated consolidation degrees with experimental results for different processing temperatures ($P_{con} = 6.9$ bar, $t_{dwell} = 0$ min).

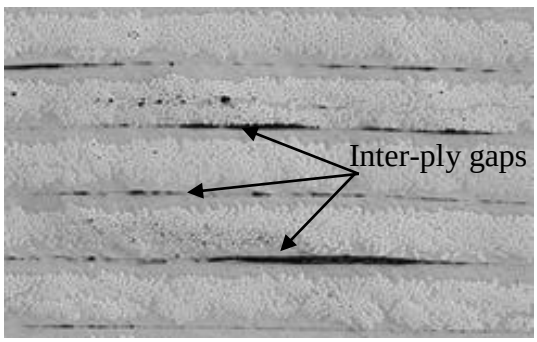


Figure 11. Microscopy image of laminate consolidated at 163 °C with a pressure of 6.9 bar and a dwell time of 0 min.

To study the influence of consolidation pressure on the quality of consolidation, a lower consolidation pressure of 0.07 bar was also used. As shown in Figure 12, both simulation and experiment indicate full consolidation for both studied laminates. It has to be noted that the minimum required consolidation pressure could be lower than 0.07 bar, while a pressure lower than 0.07 bar is not

achievable for the machine used in this study.

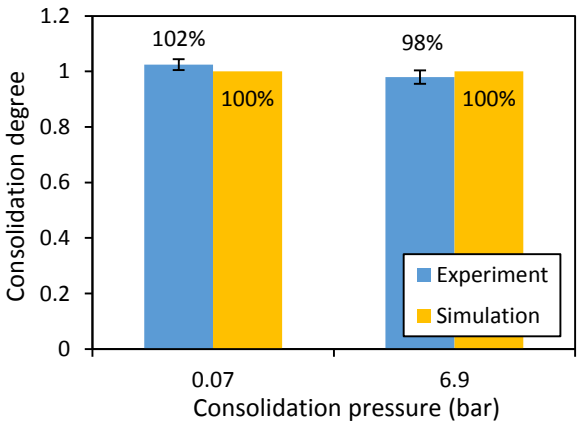


Figure 12. Comparison of simulated consolidation degrees with experimental results for different processing pressures ($T_{con} = 194$ °C, $t_{dwell} = 0\ min$).

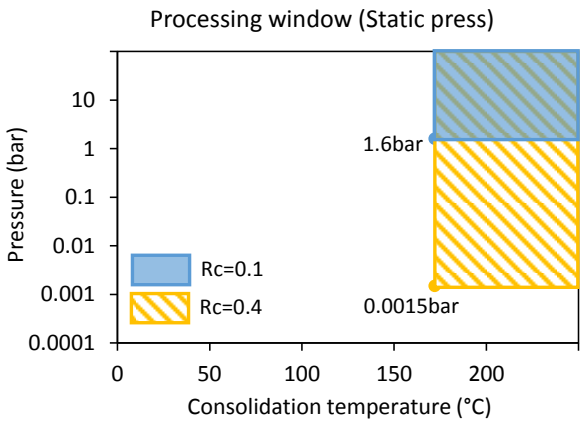


Figure 13. Model predicted processing window for lamination of GF/PP for static press consolidation.

As shown in Figure 13, a processing window, showing the limit values of processing parameters to ensure full consolidation, was also constructed using the consolidation model. A larger processing window was found for the material system with a smoother surface, represented by a larger R_C value. The minimum required consolidation temperature was found to be the same, while the minimum required consolidation pressure was found smaller for the smoother tape. It may be assumed that there is no maximum pressure for consolidation, however there will be a maximum temperature above which unwanted degradation of the polymer will take place.

5 CONCLUSIONS & OUTLOOK

A consolidation model has been implemented and applied for the lamination process of thermoplastic tape based composites. The influence of tape surface roughness and processing parameters on the consolidation quality have been investigated, and the model has been validated for hot press lamination of GF/PP composites. A good agreement was achieved between the model predictions and the experimental results.

Temperature and pressure were found to be the most important parameters that influence the quality of lamination for the considered material system. Furthermore it was found that the surface roughness plays an important role as it might influence the process settings needed to obtain full consolidation. Due to the short reptation time of PP, additional dwell time was found not necessary for static press lamination of GF/PP composites.

With the aid of this consolidation model, process window were constructed for the studied material system and the lamination process. The developed model framework will enable process optimization for different material systems and lamination processes,

ACKNOWLEDGEMENTS

The authors would like to acknowledge the technical support from the University of Delaware, Center for Composite Materials (UD-CCM), on material characterization.

REFERENCES

[1] SABIC, https://frt.sabic.com/products/udmax/index.html, 2017.
[2] ASTM, (2013).
[3] J.A. Hinkley, J.M. Marchello, B.C. Messier, NASA Langley Research Center, Hampton, Virginia, 1996.
[4] C.A. Butler, R.L. Mccullough, R. Pitchumani, J.W. Gillespie, Journal of Thermoplastic Composite Materials, 11 (1998) 338-363.
[5] C. Ageorges, L. Ye, M. Hou, Composites Part A: Applied Science and Manufacturing, 32 (2001) 839-857.
[6] F. Yang, R. Pitchumani, Polymer Composites, 24 (2003) 263-278.
[7] H. Shi, I.F. Villegas, H.E.N. Bersee, 18th International Conference on Composites Materials, Jeju, South Korea, 2011.
[8] S.C. Mantell, G.S. Springer, Journal of Composite Materials, 26 (1992) 2348-2377.
[9] P.G. de Gennes, The Journal of Chemical Physics, 55 (1971) 572-579.
[10] R.P. Wool, K.M. OConnor, Journal of Applied Physics, 52 (1981) 5953 - 5963
[11] Y.H. Kim, R.P. Wool, Macromolecules, 16 (1983) 1115-1120.
[12] V. Agarwal, Universit of Delaware, Newark, DE, USA, 1991, pp. 237.
[13] L.J. Bastien, J.W. Gillespie, Polymer Engineering & Science, 31 (1991) 1720-1730.
[14] R. Pitchumani, S. Ranganathan, R.C. Don, J.W. Gillespie Jr, M.A. Lamontia, International Journal of Heat and Mass Transfer, 39 (1996) 1883-1897.
[15] C. Ageorges, L. Ye, Y.-W. Mai, M. Hou, Composites Part A: Applied Science and Manufacturing, 29 (1998) 911-919.
[16] P.G. De Gennes, Macromolecules, 9 (1976) 587-593.
[17] P.G. De Gennes, Macromolecules, 9 (1976) 594-598.
[18] M. Doi, S.F. Edwards, Journal of the Chemical Society, Faraday Transactions 2: Molecular and Chemical Physics, 74 (1978) 1789-1801.
[19] M. Doi, S.F. Edwards, Journal of the Chemical Society, Faraday Transactions 2: Molecular and Chemical Physics, 74 (1978) 1802-1817.
[20] M. Doi, S.F. Edwards, Journal of the Chemical Society, Faraday Transactions 2: Molecular and Chemical Physics, 74 (1978) 1818-1832.
[21] M. Doi, S.F. Edwards, Journal of the Chemical Society, Faraday Transactions 2: Molecular and Chemical Physics, 75 (1979) 38-54.