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AN EXPERIMENTAL STUDY ON PREFORM EVOLUTION DURING THERMOPLASTIC COMPOSITE OVERMOULDING

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ABSTRACT

Thermoplastic composite overmoulding is an integrated process to manufacture components with combined continuous and short fibre reinforcements. During the manufacturing cycle, such materials undergo characteristic deformations that are highly dependent on temperature, pressure and time. Two experimental setups are used to investigate the compaction behaviour of continuous carbon fibre - Polyphenylene Sulphide (CF-PPS) composites: (1) isothermal compaction tests on small-scale specimens and (2) full scale component overmoulding of a ribbed plate component. In both cases, the results show a clear correlation between the thickness evolution of the material and the temperature. In the component overmoulding experiments, an additional type of deformation is observed at the overmoulded interface, between the continuous fibre and short fibre materials, where the compaction forces the continuous fibres into the rib structure, leaving a matrix-rich zone underneath the rib and significantly distorting the orientation of the reinforcing fibres of the preform.

1 INTRODUCTION

Whilst established manufacturing processes for continuous fibre thermoplastic composites offer excellent mechanical properties, they are limited to relatively simple designs. Overmoulding of short fibre compounds onto continuous fibre preforms, via an injection moulding stage in addition to a forming stage, has enabled the rapid manufacture of thermoplastic composite structures that benefit from high intrinsic mechanical properties, geometric complexity and low production cycle times. One of the key steps in the manufacturing process is the consolidation at the overmoulded interface, where the continuous fibre and short fibre materials interact under high temperature and pressure conditions in a very short time. Such high temperatures and pressures are required to eliminate voids and ensure interply bonding in overmoulding processes. Nonetheless, these conditions can lead to squeeze flow, an inherently transient and inhomogeneous deformation mechanism, resulting in redistribution of the matrix and fibres [1]. Excessive flow can affect the dimensional stability and potentially the mechanical performance, hence an understanding of this behaviour is important when considering the processing of thermoplastic composites.

Thermoplastic composite materials in their melt state can be regarded as consisting of inextensible fibres surrounded by an incompressible viscous fluid [2]. In forming operations, the fibres constrain the matrix flow but are themselves transported with the matrix. In this study, a methodology for the characterisation of a CF-PPS material during a typical overmoulding cycle is proposed, by conducting

simple isothermal compaction tests in addition to full-scale component overmoulding experiments, where the deformation mechanisms are investigated. An understanding of the material behaviour under isothermal compaction will assist in determining the final dimensions of processed preforms in addition to the main deformation mechanisms during overmoulding. Nonetheless, mapping of data from simple experiments to predict the response of highly automated processes such as overmoulding and thermoforming (stamp-forming) for continuous fibre-reinforced thermoplastic preforms is generally complicated by extreme temperature/pressure profiles due to the highly non-isothermal nature of the process.

2 METHODOLOGY

2.1 COMPACTION EXPERIMENTS

The preform material used in this study was *TC1100 Cetex PPS*, with a nominal ply thickness of 0.21 mm and 59 % fibre volume fraction. Two different specimen configurations (Fig. 1a & 1b) were used: a consolidated configuration (autoclaved at 320 °C for 30 mins and then waterjet cut to required dimensions), replicating the state of the material during the overmoulding process and a stack of assupplied tapes, laid up in a cruciform configuration, as described in [3], such that contact is ensured during the lateral spreading of the individual plies. In both specimen designs, the layup sequence is comprised of a [90, 0]_{3, s} 12-ply stack (cross-ply layup), with the effective pressurised area being 15 mm x 15 mm. Kapton tape was applied to the cruciform specimen edges to minimise individual in-plane ply movement, thus ensuring alignment when loading them onto the compaction rig.



Fig. 1. Compaction test setup (a), consolidated (b) and crucifix (c) specimens.

An Instron universal test machine was set up with custom-built temperature-controlled compression platens as illustrated in Fig. 1c, such that controlled load and temperature could be transferred to the specimens. Each platen was mounted with a 325 W thick film conduction heater, supplied by *Watlow*®, enabling temperatures of up to 400 °C to be attained. The loading program consisted of isothermal ramp-dwell steps, where the fast application of load is followed by a longer creep interval, as previously employed by Nixon-Pearson et al. for thermo-set pre-preg [4]. The program includes five 240 second steps with an incremental load of 0.5 kN, starting at 1.0 kN, such that the effective pressure at the end

of the test is 13.3 MPa. FREKOTE 770-NC is applied to the platen surfaces in order to minimise specimen-tool friction and ensure good release properties upon test completion. A very small pre-load is applied to the specimen prior to ramp-dwell program to ensure good thermal contact. Upon completion of the ramp-dwell stage, the heaters are turned off and the specimen is cooled to just below its glass transition temperature. The cooling rate (and thus test time) is improved by directing a cold air stream, generated by a *Vortex Tube* (supplied by *Meech Static Eliminators Ltd.*) towards the centre of the specimen. The specimen is held at the final pressure of 13.3 MPa for the duration of the cooling stage, thus preventing de-consolidation and enabling micrograph analysis of the specimen in its "locked-in" state. Correction for machine compliance and thermal expansion at different temperatures was accounted for by running dummy tests without loading a specimen.

The compaction experimental investigation is not necessarily focused on reproducing specific processing conditions related to overmoulding, but rather aimed at exploring the response of fibre-reinforced thermoplastic preforms across a range of pressures and temperatures. The generated data is to be used for simple extraction of geometric material parameters in a robust procedure for a 3D hyper-viscoelastic model by Belnoue et al. [5] in future work. As seen from Table 1, typical parameters for consolidating CF-PPS continuous fibre laminates vary between different processes, with thermoforming requiring the highest tool pressures. Further, overmoulding technologies generally require even higher pressures in order to counteract the force imparted on the tool by the injected compound material. For this reason the maximum applied loads on the specimens during the compaction ramp-dwell stage are scaled to match typical overmoulding tool pressures.

	Press Lamination	Autoclave Consolidation	Thermoforming
Target temperature (°C)	320 °C	320 °C	320 °C
Pressure (MPa)	0.7 - 2.1	0.7 - 1.0	1.0 - 4.0
Time at temperature and pressure	15 – 30 mins	20 – 30 mins	\leq 5.0 seconds (depending on tooling and process settings)

Table 1: Typical Processing Parameters for TenCate Cetex© PPS TC1100 [6]

2.2 OVERMOULDED RIBBED PLATE MANUFACTURE

The overmoulded ribbed plate component, which are described in [7], consists of a cross-ply (continuous carbon fibre PPS, 2.5 mm thick) preform and four 120 mm-long short carbon fibre-PPS ribs. The materials used for the preform and ribs were TC1100 Cetex© PPS and Luvocom 1301-0824 (supplied by Lehmann & Voss) respectively. An Arburg 270C Allrounder injection moulding machine was used to overmould the short fibre compound onto the continuous fibre preform. Pre-heating temperatures of 260 °C, 290 °C, 320 °C and 350 °C were used for this study. The heating stage was conducted in an external two-sided horizontal oven comprising of a total of eight KRELUS G14-25-2.5 MINI 7.5 IR-heater banks. K-type thermocouples (Fig. 2b) were used to acquire the midplane and surface temperature profiles of the preforms during the processing cycle. For the midplane temperature readings, a 10.0 mm deep, 1.35 mm diameter hole was drilled into the edge of the preform, as seen in Figure 3, thus providing a sliding fit for the thermocouple. A high temperature adhesive was used to ensure good thermal contact between the thermocouple and the preform. The surface thermocouple was removed prior to the clamping operation as the exposed metallic wire would damage the tool surface. The transfer of the heated preform was carried out via a robot (KUKA KR6) with a custom-built gripper system, ensuring adequate alignment of the preform when it is transported to the injection moulding machine (Fig. 2a.) Once the preform is fixed between the mould tool halves of the injection machine, the robot exits the working space of the machine and the clamping tool presses the preform with a 400

kN force (forming stage.) After this, the short fibre compound material is injected through the mould cavity and a holding pressure of profile of [65, 55, 40] MPa for [6.0, 3.5, 2.0] seconds is applied to compensate for material shrinkage. After a 30 second cooling stage to mould temperature, the preform is demoulded from the tool and left to cool at ambient conditions. The preform thickness is then measured using Vernier callipers.



Fig. 2: Overmoulding setup for ribbed plate component (a), showing positioning of thermocouples for in-situ temperature measurements.

3 RESULTS AND DISCUSSION

3.1 Compaction Experiments

In general, the response of the thermoplastic composite material shows a recurring trend across all tested compaction specimens. The displacement-time plots shown in Fig 3 imply that through-thickness compaction is limited to small deformations when the test temperature is below the melting point of the matrix (285 °C for PPS [8].)



Fig. 3: Transverse widening of cruciform specimen at different times (a) and typical displacementtime curves of specimens under different test temperatures (b).

At these temperatures the material is thermally stable and deforming almost elastically. However, at higher temperatures the material is clearly seen to deform in a viscoelastic manner, illustrated by the characteristic thickness evolution. Under these conditions the viscous response of the molten matrix is expressed as a relaxation of the material under constant load, as can be observed by the decaying displacement at each consecutive loading step. It is interesting to note that the consolidated specimens were able to compact further than the cruciform specimens. This is most likely due to the loss of material underneath the pressurised area as a result of transverse spreading of the plies, whereas the material in the cruciform specimens is retained between adjacent plies as it spreads laterally. For consolidated specimens tested at 300 °C, most of the compaction occurs within the first seconds of the initial loading step, however, the cruciform specimens showed a gradual compaction with increasing load and time, reaching the compaction limit at a much later stage.

In addition to the thickness change, optical micrographs of the compacted specimens show a clear difference between the in-plane deformations at different test temperatures. Individual ply deformations are negligible in the consolidated specimen tested at 260 °C and as the viscosity is extremely high below the melt temperature, the presence of regular cracks (as seen in Fig. 4a) suggest that the thermoplastic matrix is deforming elastically under the high stress conditions.



Fig. 4: Optical micrographs (a-c) and corresponding post-compaction images (d-f) of consolidated specimens at the edges.

At 280 °C, evidence of the squeezing phenomenon can be seen in Fig. 4b, where the plies closer to the midplane spread further than the plies located on the specimen surfaces. Images of the post-compaction specimen (Fig. 4e) also show significant spreading of the central plies only. This could be attributed to the lower friction between the individual layers of the same orientation as well as the increased transverse velocity components for plies located near the centre of the laminate, as predicted by squeeze flow models with non-perfect slip [9], indicating the importance of this interaction during processing. Maximum in-plane spreading, and through-thickness compaction is seen in specimens tested above 280 °C and is characteristic of fibre-reinforced thermoplastics at processing temperatures. Fig. 4c clearly shows the high extent to which the plies are squeezing. Fig. 5a & 5b show the micrographs of the cruciform specimens tested at 280 °C and 300 °C respectively. Similarly, very little spreading is observed at lower temperatures and the plies can remain relatively undeformed. There is a clear

distinction of the individual plies due to the interply bond-lines, potentially introducing significant friction locally at that interface. Unlike the consolidated specimens, images of the cruciform specimens (Fig. 5d) show that the spreading of the plies is more constrained by the specimen geometry, evidenced by the lack of matrix material at the inner corners in its post-compacted state, where this is clearly visible in the consolidated specimens. This further implies that the ability of the specimens to compact under a given load is dependent on the geometry.



Fig. 5: Optical micrographs (a-b) and corresponding post-compaction images (c-d) of cruciform specimens at the edges.

3.2 Overmoulded Ribbed Plate Manufacture

Under the different temperatures conditions, two types of deformations are observed in the manufactured overmoulded ribbed plates: (i) a global thickness variation and (ii) local deformation of the overmoulded interface. Both deformations are a consequence of the squeeze-flow behaviour of thermoplastics, deforming predominantly in-plane, where this effect is noticeable at the edges of the ribbed plate (Fig. 6), but also out-of-plane near the overmoulded interface (Fig. 7b.)



Fig. 6: Ribbed plates manufactured at pre-heating temperatures of (a) 260 °C and (b) 320 °C, showing significant squeezing at higher temperatures.

Such material behaviour is likely due to the lower viscosity of the PPS matrix above its melting point. The compaction results support this observation as there is no significant change seen in the specimen thickness until the onset of melting is reached, implying that the molten matrix is free to flow within the preform, observed by the large quantities of deformed material in Fig. 7b. At elevated temperatures, this behaviour also causes significant ply re-orientation near the vicinity of the overmoulded interface, between the continuous and short fibre materials, initiated by build-up of matrix in this region. This is initiated upon closing of the mould tool halves, as previously reported in [7]. It is further observed that the degree to which the preform material protrudes into the rib increases with temperature, given that more material can be squeezed in when the viscosity is lower. Locally it is expected that there will be matrix-rich zones as well as some reduction in load carrying capability due to the distorted orientation of the continuous fibre plies. Nonetheless, at these temperatures, the lack of a visible bondline is indicative of good welding characteristics between the continuous fibre materials.



Fig. 7: Measured preform thickness after overmoulding at different pre-heating temperatures. Error bars indicate standard deviations (a). Optical micrographs of localised preform deformations at overmoulded interface from ribbed plate pre-heated to 290 °C (b) and 320 °C (c).

Increasing the pre-heating temperature of the preform is seen to reduce the final thickness, illustrated by the negative correlation in Fig. 7a, nonetheless, the compaction of the ribbed plates at each pre-heating temperature is considerably less than the compaction of the cruciform specimens tested at the same temperature. This is attributed to the size of the sample in question as the percentage of squeezed material in the ribbed plates is much less compared to the smaller compacted samples, thus limiting the compaction. Further, the difference in the loading and thermal history, as can be illustrated by the temperature profile of the ribbed plate preform during the manufacturing cycle in Fig. 8b, directly affects the compaction behaviour. The midplane thermocouple shows a steep temperature drop at the instant the "colder" mould tool (145 °C) presses against the pre-heated preform and forms it. This causes the viscosity of the matrix material in the preform to rise very quickly and thus inhibits further squeezing for a given load. Furthermore, a significant increase in the lateral spreading of the midplane plies was observed in the ribbed plates pre-heated above the matrix melting temperature.

The localised deformation at the overmoulded interface can further lead to a variety of problems in terms of the manufacturability of such components, as the effective reduction in rib cross-sectional area reduces the pressurisation capability of the compound material to fill the injection cavity, potentially leading to short shots as seen in Fig. 8a. Recommended measures for preventing such defects include increasing the injection pressure and/or flow rate of the compound material, however, this tends to lead

to higher running costs. Alternatively, reducing the pre-heating temperature will lead to less matrix flowing to these regions. In this case, the bonding between the preform and the ribs is expected to be lower due to the reduced ability for the molecular chains to diffuse across the material boundary [10]. A clear trade-off is presented when designing overmoulded components, thus attention must be devoted to the understanding of the material behaviour under these highly non-isothermal processes.



Fig. 8: Incomplete filling of the ribbed plate mould cavity observed at a pre-heating temperature of 350 °C (a). Midplane thermocouple temperature profile for ribbed plate overmoulded at a pre-heating temperature of 350 °C (b).

4 CONCLUSIONS

In this study, the evolution of continuous CF-PPS composite material was investigated under two different processing setups. The first setup consisted of isothermal ramp-dwell, load-controlled tests conducted on two specimen configurations: consolidated and crucifix. The results showed that a compaction limit is reached quicker at lower temperatures, especially when tested below the melting point of the matrix. Optical micrographs and visual inspection further show that the squeezing behaviour of the material is more profound in the plies with fibres oriented transverse to the deformation direction as well as in plies located near the midplane of the material. The second setup consisted of overmoulding flat preforms, composed of the same material system and layup of the compaction specimens. The results show that the final preform thickness is also seen to decrease at higher temperatures, indicating reduced dimensional stability as well as potentially reduced mechanical performance due to the local deformation and re-orientation of the continuous fibre plies at the vicinity of the overmoulded interface. Similar to the compaction specimens, significantly more squeezing of the individual plies is observed at the preform midplane, where the temperature stays hotter for a longer period. These observations contribute to the development of overmoulding-specific design guidelines and manufacturing practices through simple experimental material characterisation, indicating that a well-defined process window is essential to ensure quality components.

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REFERENCES

- [1] Engmann, J., Servais, C., & Burbidge, A. S. (2005). Squeeze flow theory and applications to rheometry: A review. *Journal of Non-Newtonian Fluid Mechanics*, *132*(1–3), 1–27.
- Balasubramanyam, R., Jones, R. S., & Wheeler, A. B. (1989). Modelling transverse flows of reinforced thermoplastic materials. *Composites*, 20(1), 33–37. Shuler, S. F., & Advani, S. G.
 "Transverse squeeze flow of concentrated aligned fibers in viscous fluids." *Journal of Non-Newtonian Fluid Mechanics*, 65(1), 47–74, 1996.
- [3] Ivanov, D., Li, Y., Ward, C., & Potter, K. (2013). Transitional behaviour of prepregs in automated fibre deposition processes. *Proceedings of International Conference on Composite Materials*, 19.
- [4] Nixon-Pearson, O. J., Belnoue, J. P. H., Ivanov, D. S., Potter, K. D., & Hallett, S. R. An experimental investigation of the consolidation behaviour of uncured prepregs under processing conditions. *Journal of Composite Materials*, 51(13), 1911–1924, 2017.
- [5] Belnoue, J. P. H., Nixon-Pearson, O. J., Ivanov, D., & Hallett, S. R. (2016). A novel hyperviscoelastic model for consolidation of toughened prepregs under processing conditions. *Mechanics of Materials*, 97, 118–134.
- [6] TenCate Cetex[©] TC1100 PPS Resin System, Product Datasheet, *TenCate Advanced Composites*.
- [7] Valverde, M. A., Kupfer, R., Kawashita, L. F., Gude, M., & Hallett, S. R. "Effect of processing parameters on quality and strength in thermoplastic composite injection overmoulded components", *ECCM18 18th European Conference on Composite Materials*, 2018.
- [8] Nohara, L. B., Nohara, E. L., Moura, A., Costa, M. L., & Rezende, M. C. (2006). Study of Crystallization Behavior of Poly(Phenylene Sulfide). *Polímeros: Ciência E Tecnologia*, 16(2), 104–110.
- [9] Rogers, T. G. (1989). Squeezing flow of fibre-reinforced viscous fluids. *Journal of Engineering Mathematics*, 23(1), 81–89.
- [10] Yang, F., & Pitchumani, R. (2002). Healing of thermoplastic polymers at an interface under nonisothermal conditions. *Macromolecules*, 35(8), 3213–3224.