INTEGRATION OF POLYMER AND COMPOSITE MATERIALS FOR ENHANCED DESIGN FREEDOM AND COST-EFFICIENCY

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SUMMARY: A novel manufacturing route is currently being investigated for polymer composite structures. The objective is the integration of several materials, including reinforced composite preforms and polymers, in order to offer shorter manufacturing times, greater design freedom and multifunctionality. Combinations of processing techniques for the integration of several material transformation steps have been studied. Two approaches were followed in order to produce parts with hybrid reinforcements and parts with selective reinforcement. Combinations of glass mat thermoplastics with unidirectional prepregs and neat polymers were considered in the first approach. The second approach showed how selective placement of composite tows can open interesting perspectives for tailoring mechanical properties and dimensional stability. Furthermore, the placement of the composite preceded the overinjection of a neat polymer which provided the final part geometry and surface finish. Results reveal how the synergy obtained during the combination of the material transformation steps can reduce the production time. Impregnation, consolidation and placement of the composite preforms, injection of the polymer and in-situ bonding of all materials were achieved during the same shortened processing cycle.

KEYWORDS: Integrated processing, thermoplastic, composite preforms, cost-efficiency, design freedom, consolidation, tow placement.

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INTRODUCTION

Several manufacturing routes exist for the transformation of thermoplastic polymer composites [1]. They include impregnation of the reinforcement fibres by the polymer matrix, heating and melting of the polymer, consolidation and finally cooling of the composite parts. Preimpregnated and preconsolidated preforms such as prepregs or preforms based either on powder impregnated yarns or on commingled yarns are generally delivered by material producers to transformers for the manufacturing of the desired products.

The type of material preform used determines the processing techniques and the material transformation phenomena needed for achieving good consolidation of the final part. Autoclaving or press moulding use preimpregnated reinforcement such as prepregs or fabrics which are placed by hand or in a semi-automatic manner into the mould before the heating and cooling cycle is performed. They are considered as post lay-up consolidation techniques and have been used extensively. Tape placement and pultrusion are more recent techniques for <u>on-line consolidation</u> in which placement, heating and consolidation are performed simultaneously and locally during the automatic lay-up of the preimpregnated tapes or yarns. In comparison to other techniques, automatic tape placement shortens the lay-up time, the heating and cooling times and subsequently offers potential for more cost-effective production. Nevertheless, the process induces fast and highly non-isothermal consolidation phenomena in the material. The transformation of materials under such transient processing conditions is currently studied by several groups of scientists. This example illustrates the need in the field of composites to develop cost-effective techniques which offer a higher degree of material and process integration.

With the above mentioned techniques, continuous fibres are mainly used to obtain high performance laminates but with some limitation in terms of part geometry. Tape placement shows some potential for producing parts with different out-of-plane curvatures. Nevertheless, the continuous preconsolidated tapes limit the feasible curvature and hinder curved in-plane placement. The placement of preimpregnated but un-preconsolidated yarns such as commingled yarns can offer higher potential for a more precise placement of reinforcement. The high deformability of the yarns allows for small radii of curvature and the continuous yarns can be placed only in the locations and directions where stiffness and strength are required, the remaining volume of the part being filled with neat or short-fibres reinforced polymers. This selective and tailored placement of reinforcement will be of interest only when good consolidation of the integrated materials can be ensured.

The combination on the same equipment of preform impregnation, automatic placement of reinforcements, composite consolidation and the addition of polymers has been investigated [2-5]. The combination of materials with different rheological, thermal and mechanical properties requires the study of phenomena related to multi-material structures, such as the optimisation of <u>in-situ consolidation</u> and the control of interfaces between integrated materials. The combination of material transformation steps can promote interesting synergy effects.

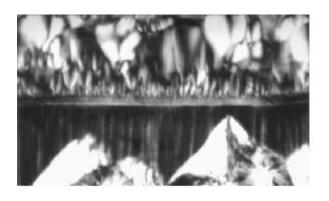


Fig. 1: Interfacial crystallisation during in-situ nonisothermal bonding: transcrystallinity across the initial interface.

It has been shown, for example, that under specific non-isothermal conditions, bonds between two integrated polymers can be processed in shorter times than under isothermal conditions because specific crystallisation across the interface is promoted (Fig. 1). Consequently, by an optimum control of bonding parameters, the cycle times during integrated processing are reduced and costefficiency is improved. Hybrid composites combining thermoplastics glass mat unidirectional prepregs and neat polymers were investigated in this first approach [3].

The second approach presented hereafter is related to placement of selective reinforcement.

Some results will be presented on the material consolidation phenomena which occur during the different integrated processes: impregnation, placement of composites preforms and polymer overinjection.

EXPERIMENTAL

Integrated processing

A processing machine has been developed which combines several units for impregnation, placement and final consolidation of composite preforms [2-5]. Injection and extrusion units allow polymers to be added to the composite elements. The different processes integrated in this study were, in order, powder impregnation of carbon fibres to form tows, preconsolidation of the impregnated yarns, automatic placement of the yarns into a mould and overinjection by an additional polymer (Fig. 2).

Integrated parts based on powder impregnated tows

The powder impregnation line comprises a fluidised bed containing the polymer powder through which carbon fibres are driven. The preimpregnated tow is then guided into an oven in which the polymer powder is melted and stabilised onto the carbon filaments. The tow is driven through parallel pins to control its preconsolidation level. The powder impregnation mechanisms were studied and the optimum conditions established for the selected material [6]. The tow is then guided through a nozzle on a robot arm, at the outlet of which the tow is positioned into a mould. The nozzle is equipped with heating elements to soften the preconsolidated tow during the placement and to control its temperature and thus its degree of consolidation. The pressure at which the nozzle is applied can be varied in order to study the effect of the consolidation pressure during the placement. For some experiments, a roller is used to apply additional pressure. After tow placement, the mould is closed and additional polymer is injected over the tows.

For this study the powder impregnation line was set to deliver tow with a constant polymer weight fraction of 50 %. The main parameter investigated was the tow placement speed, which was varied from 1 to 150 mm/s. For each sample, four tows of 24K carbon fibres were placed. The nozzle temperature was kept constant at 260°C and the polymer was overinjected at 260 °C.

Integrated parts based on commingled yarns

The same method was used with commingled yarns of polymer and carbon fibres. A yarn was directly introduced into the robot nozzle and was therefore directly heated from room temperature to the desired placement temperature. Sixteen yarns of 6K were placed into each sample to give the same number of carbon fibres as for the powder impregnated tows. The PA12 polymer was then overinjected. The influence of the placement speed on the yarn consolidation was studied.

Materials

Powder impregnated tows were based on carbon fibres which were impregnated with polyamide 12 (PA12) powder. Impregnation parameters were selected to obtain a tow with a volume fraction of carbon fibres of 36 %. Commingled stretch-broken yarns were produced from carbon fibres and PA12 fibres by Schappe Techniques. The volume fraction of carbon fibres was 56 %. The polymer used for the injection process was PA12. The PA12 powder, fibres and granules were supplied by EMS-Chemie and had a melting temperature of 178 °C.

Tests

Samples were cut from the parts produced by the integrated processing and the mechanical characteristic of the tows were measured using a four point bending test (ASTM D790). For the overinjected samples, the additional polymer was removed before mechanical testing. Void content was determined using ASTM D792 and image analysis, and was used to characterise the evolution of the consolidation of the impregnated tows and commingled yarns.

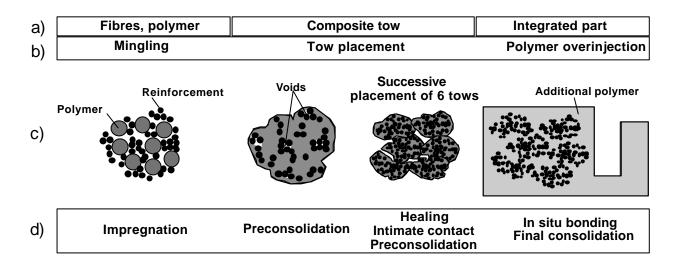


Fig. 2: (a) Materials, (b) processes and (d) material transformation steps integrated to produce samples with selective reinforcement. (c) Evolution of cross-sections from initial tow to the final integrated part.

RESULTS AND DISCUSSION

Demonstration parts were successfully produced with the integrated processing equipment. These parts include selective reinforcement, precisely oriented and covered by an injected neat polymer (Fig. 3). Thus fine tailoring of mechanical properties is achieved in composite parts exhibiting a complex geometry

and good surface finish. In this paper some of primary results related to the evolution of the part consolidation during the different steps of the integrated processing (Fig. 2) will be shown.

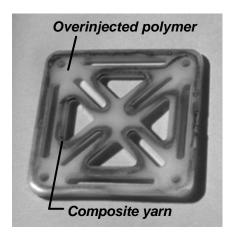


Fig. 3: Example of an integrated demonstration part with selective reinforcement. The composite yarn of carbon fibres and PA12 matrix was precisely placed before polymer overinjection.

<u>Commingled yarns</u> were placed into the mould before the overinjection of the additional surrounding polymer. The yarns could be placed precisely in different directions and the placement parameters were determined to control the preconsolidation level of the yarn passing through the heated placement nozzle. At the outlet of the nozzle the molten yarn is rapidly cooled by the surrounding air and solidifies. The local placement temperature and pressure determine the degree of preconsolidation of the yarn. With a range of placement conditions the void content of the placed yarns ranged between 10 and 15 %.

The consolidation of commingled yarns was modelled in a parallel study [7]. The model is based on the impregnation of reinforcement bundles by the molten polymer matrix (see the preconsolidation step in Fig. 2). Several bundles of fibres form one yarn. A population of bundles with different sizes and numbers of carbon fibres were considered. The model is based on Darcy's law, takes into account the counter pressure created by the entrapped air and can be applied to non-isothermal conditions. It can be applied to the yarn placement step of integrated processing [8]. Figure 4 shows the variation of yarn void content with the placement speed. The agreement between the experimental and predicted values is good. The placement speed influences the consolidation degree via the yarn temperature and the time during which the pressure is applied.

Final consolidation is achieved during the overinjection step. The injection process time was of the order of seconds and a pressure of up to 900 bars was used. Several coupled phenomena occur during this process; compaction of the yarn under the high pressure of the injected polymer, further consolidation of the yarns, additional impregnation of the yarns by the injected polymer and displacement of non-preconsolidated yarns. Experiments showed that yarn compaction is a primary phenomenon when the polymer is injected onto a yarn maintained at room temperature. In this case no significant impregnation by the additional polymer occurred and the above mentioned model can be used since no additional mass transfer needs to be considered. For a yarn with an initial void content of 13 % after placement,

the predicted void content after compaction was 10 %, a value in good agreement with the 9 ± 2 % obtained experimentally.

This demonstrates that the commingled yarns can be placed with a good control of their consolidation level, an important requirement for maintaining their position during the overinjection process. The final polymer injection improves the tow consolidation. If needed, a better control of the yarn temperature during the injection process or the use of a commingled yarn with a higher matrix content would further improve the final consolidation.

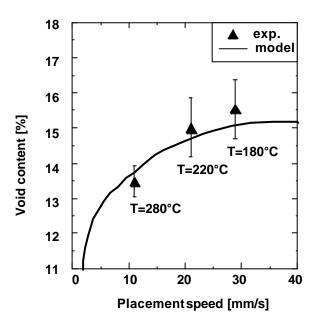


Fig. 4: Variation of predicted and measured void contents with the placement speed of commingled varns, before final overinjection.

Using <u>powder impregnated tows</u>, an additional process was integrated, namely the impregnation of initially dry fibres by a polymer powder (Fig. 2). Thus all the steps for the material transformation, from the neat fibres and matrix to the final production of a composite part, are performed on the same integrated equipment.

Tows with constant matrix and void content were produced on the powder impregnation line. The variation of void content and flexural modulus of the tows placed at different speeds by the robot are represented in Fig. 5. The curve a) represents the results obtained on the tows placed without further polymer injection.

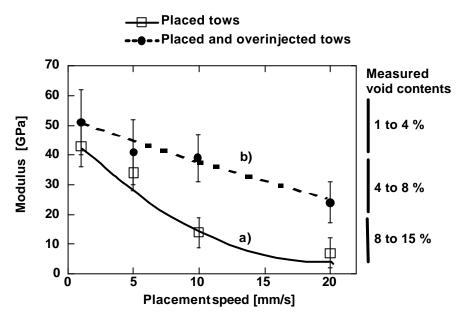


Fig. 5: Evolution of the flexural modulus and void content of powder impregnated tows after placement performed at different speeds: a) before and b) after polymer overinjection.

The tow stiffness is significantly influenced by the placement speed. The void content ranged from 4 to 15 % for placement speeds of 1 to 20 mm/s respectively. The presence of voids clearly influences the mechanical behaviour of the placed tows. At high placement speeds the consolidation parameters are no longer optimal and higher void contents were observed. Placement at higher temperatures might improve the consolidation, provided that the degradation of the polymer could be avoided. Furthermore the degree of consolidation can still be improved by increasing the placement pressure or by decreasing the tow deconsolidation with additional cooling (Fig. 6). In fact, during integrated processing, the final degree of consolidation of the composite preforms is determined by the polymer over-injection. Curve b) in Figure 5 shows the variation of the stiffness for tows placed and overinjected. The polymer injection significantly improves the tow consolidation for all the investigated speeds. The improvement is more efficient when the initial void content is high, that is when the placement speed is high. This tendency was confirmed when a tow, better consolidated during placement, by the application of additional pressure, was then overinjected. The modulus increase was smaller than with the standard placement (Fig. 6). Final consolidation could still be improved by tailoring the tow temperature just before the injection phase in order to promote better compaction and further impregnation.

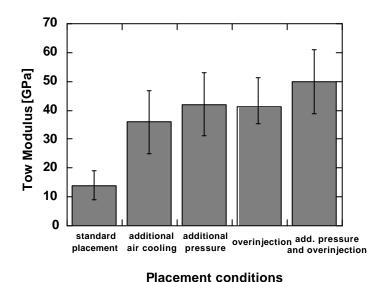


Fig. 6: Influence of the placement conditions on the tow properties. The tow placement speed was 10 mm/s.

This approach demonstrated that commingled yarns and powder impregnated tows can be processed efficiently for the selective reinforcement of integrated parts. The maximum placement speed is related to the material preconsolidation level required for keeping the yarn in place during the following processes. Furthermore, the effect of the polymer injection on the consolidation of the composite is crucial for the control of the final part quality. Figure 7 summarises the variation of the tow consolidation with the different integrated processes. Placement at low speed with additional pressure and cooling systems can provide tows with reasonably low void content and good mechanical properties (curve b) in Fig. 7). These consolidated tows can be used directly for the selective reinforcement of composite parts based on GMT or textile fabrics.

When polymer overinjection is used, impregnation and placement can be performed at higher speeds because the injection process determines the final consolidation of the composite reinforcement (curve a) in Fig. 7). The integration of two fast processes such as high speed placement and injection offers short cycle times. Furthermore the polymer provides added value such as a complex geometry and good surface finish.

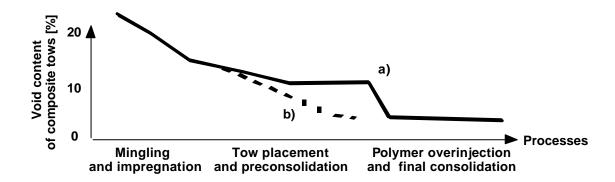


Fig. 7: Evolution of tow consolidation degree during the different integrated processes. a) placement at high speed and overinjection, b) placement at low speed.

CONCLUSION

The production of parts of complex geometry and tailored local mechanical properties is still a challenge for the composite industry. The development of an integrated processing concept and of a multi-process device has opened interesting perspectives for design freedom and cost-effective production of complex shaped parts. Neat polymers and composites are combined in a single operation to reduce the manufacturing cost by the suppression of intermediate processing and assembly steps. Each processing sequence combines different processes and material transformation steps (impregnation, placement and consolidation of composite reinforcement, polymer injection, etc.).

In-situ combination of glass mat thermoplastics, thermoplastic prepregs and neat polymer has been investigated in a first approach. The integration of selective composite reinforcement is currently being evaluated by local placement of commingled yarns or powder impregnated tows into an injected polymer part. The evolution of the composite void content during the different processes was determined and can be tailored to optimise the total processing time. It was shown, for example, that impregnation and placement speeds can be higher when tow consolidation is achieved during the final polymer overinjection.

The two approaches demonstrate that the integrated processing concept introduces new perspectives in the design of composite parts. The local placement of composite can tailor load distribution and dimensional stability within a polymer part of complex geometry. Furthermore, the integration of materials and processes promotes synergy effects: the integration of several consolidation steps and control of non-isothermal in-situ bonding processes reduce production times. Subsequent manufacturing flexibility and cost-efficiency open new potential for the application of composite materials and the integration of more advanced system functions.

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