EFFECT OF PROCESS VARIABLES ON THE PERFORMANCE OF GLASS FIBRE REINFORCED COMPOSITES MADE BY HIGH PRESSURE RESIN TRANSFER MOULDING

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Abstract

The CAFE regulations will require average fuel consumption of cars and light duty trucks to be reduced from 27 mpg to 54.5 mpg by 2025. In order to reach this requirement, automakers have to improve both the vehicle powertrain efficiency and the vehicle weight. Polymer composite materials are a preferred alternative to achieve the needed weight reduction by combining a higher strength to weight ratio than steel and aluminum, superior fatigue and corrosion resistance than metal and very good crashworthiness characteristics. However, high throughput and cost effective composite manufacturing processes are essential for high performance fibre reinforced polymer composites to penetrate the automotive market to their full potential. High Pressure Resin Transfer Moulding (HP-RTM) process is a new process based on the Resin Transfer Moulding (RTM) technology that enables the processing of very reactive resins in very short cycle times (< 5-10 min i.e. >25,000 parts per year) with various types of reinforcement (glass, carbon, natural fibre). With HP-RTM’s unique self-cleaning impingement mixhead, resins that react in less than 1 minute (fast-curing epoxy or polyurethane systems) can still be processed. This process, combining the high mechanical performance of RTM parts with short cure cycle, thus presents a great interest for automotive applications. In this study, the effect of the process parameters, such as the injection flow rate, the vacuum assistance sequence and mould gap control, on the mechanical performance and quality of HP-RTM composite plates was determined and HP-RTM process mapping was established from the obtained mechanical results.

Introduction

According to the new CAFE (Corporate Average Fuel Economy) regulation, fuel consumption of all cars and light duty trucks should increase from 27 miles per gallon (mpg) to 54.5 mpg by 2025. To reach this requirement, vehicle powertrain efficiency and vehicle weight have to be improved. Polymer composite appear to be one of the best alternative to achieve the needed weight reduction by combining a higher strength to weight ratio than steel and aluminum (50-70% weight reduction as a replacement of steel with carbon fibre and 25-35% with glass fibre), superior fatigue and corrosion resistance than metal and very good crashworthiness characteristics, as well as possibility for complex shapes structures with fewer sub-components, thus reducing assembly time and cost.
In the past decades, liquid injection moulding processes have been widely used to manufacture high performance composite structures in marine and automotive applications. Among them, the interest in resin transfer moulding (RTM) process has especially increased for automotive applications. This closed mould system allows the production of large and complex composite structures with high consistency, tight geometrical tolerances and mechanical properties comparable to autoclaved processed parts. Good surface finish up to Class A can be obtained on all the sides of the component. However, the current injection and resin curing times remain very long (30 – 120 min i.e., i.e. 30,000 – 50,000 parts per year), reducing the application of RTM process in domains where high volume production is required. High throughput and cost effective composite manufacturing processes are essential for high performance fibre reinforced polymer composites to penetrate the automotive market to their full potential.

Recently, a related new technology, High-Pressure Resin Transfer Moulding (HP-RTM) has become very attractive for high volume production application, as it enables the processing of very reactive resins in very short cycle times (< 5-10 min) with various types of reinforcement (glass, carbon, natural fibre). Using a unique self-cleaning impingement mixhead, resins that react in less than 1 minute (fast-curing epoxy and polyurethane systems) can still be processed by mixing and injecting the resin and the hardener at a pressure up to 150 bars. The high injection speed allows filling the mould before the resin starts to cure while the use of highly reactive resin significantly reduces the cure time and therefore the cycle time. In order to get a shorter filling time, the injection can be done in a partially opened mould allowing a faster in-plane flow of the resin. The mould is then closed to enable the resin to flow through the preform thickness and fully impregnate it. This process variant is also known as compression Resin Transfer Moulding versus injection Resin Transfer Moulding. Vacuum can as well be applied to the system to further improve the filling time and part quality. This process, combining the high mechanical performance of RTM parts with short cure cycle, presents a great interest for the major automotive manufacturers and automotive part suppliers.

Various studies investigated experimentally and numerically the effect of process parameters on the performance and quality of the manufactured composite parts made by RTM and compression RTM process. Every aspect of the RTM process has been extensively studied in the last two decades from the preforming stage to the mould filling and curing stage [1-3], showing the importance of controlling the thermal and pressure histories as well as the resin properties to obtain high quality parts. Similarly, many RTM numerical studies have been carried out to predict the physical and chemical phenomena involved during the process (i.e. heat transfer, flow and compaction, cure and residual stresses) via simulation of the mould filling stage and simulation of the process cycle [4]. With the intent to increase the volume production, compression RTM was then examined by many studies. The effects of mold gap and gap closure speed, injection pressure or flow rate and compression pressure, temperature and reinforcement properties on the mould filling time, part mechanical properties and void content were investigated [5-13].

However, most of these studies examined processes with low injection pressure (1 - 7 bars) and slow-curing resins. The use of high injection pressure and highly reactive resins with HP-RTM process brings a lot of new processing challenges, such as fibre wash-out, filling defect, high porosity level and decrease in mechanical performance. Only few studies examined those issues. Barraza et al. demonstrated that an increase in flow rate and resin viscosity reduced mechanical properties due to higher air entrapment [14]. Chaudhari et al. examined the influence of the gap size and the stacking sequence for compression HP-RTM and found no effect of the gap size on the tensile, flexural and ILSS properties of the laminates [15]. Deléglise
et al. recently developed a numerical approach to model isotherm mould filling with highly reactive resin, where filling and cure occurred simultaneously [16].

In this study, glass fibre reinforced composites were made by HP-RTM using a fast-curing epoxy resin system. The effect of the process parameters, such as the injection flow rate, the vacuum assistance sequence, mould gap control and binder concentration, on the mechanical performance and quality of HP-RTM composite plates was determined and HP-RTM process mapping was established from the obtained mechanical results.

**Experimental**

**Materials**

VORAFORCE™ fast-cure epoxy resin system from Dow was used as thermoset matrix. This resin system was especially developed for fast injection process. Prior to processing experiments, the chemical and rheological behaviours of the resin system were characterized using a Differential Scanning Calorimeter (DSC, Q2000 system from TA Instruments) and a parallel plate geometry rheometer (ARES system from Rheometric Scientific), respectively. Figure 1 shows the resin reactivity at 100 °C which correspond to the typical curing temperature. The resin reaches its gel time in 50 to 60 seconds and fully cures in less than 250 seconds (approx. 4 minutes). Also it can be noticed that the resin reaches a viscosity superior to 1 Pa.s after 40 seconds. From a manufacturing point of view, this implies that the mould filling and impregnation of the reinforcement should be done in less than 40 seconds, in the resin processability region.

![Figure 1: Variation of the degree-of-cure and the viscosity of the VORAFORCE™ epoxy at 100 °C](image)

Dry quadraxial [0°/-45°/90°/45°] glass non-crimp fabric with an areal weight of 1205 g/m² was used as reinforcement. In order to limit fiber wash-out and fibre displacement, an epoxy-based binder was applied to the glass fiber fabric. Different binder concentrations (0%, 4%, 5% and 6%) were applied on one or both surfaces of the fabric.

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HP-RTM process and process variables

The HP-RTM unit used in this project was a Krauss Maffei RTM 8/4-2K metering unit coupled with a 2500 ton Dieffenbacher press. The resin was pre-heated at 80 °C and the hardener at 40 °C. The mixing pressure was set at 90 bars and 1100 g of resin/hardener mixture at a ratio 100:26 per weight was injected per shot. A steel plaque mould was mounted in the press and preheated at 100 °C. The injection gate and the vent were located at each side of the mould. From the resin characterization data, the cycle time was set to 5 minutes. One ply of glass fibre reinforcement (0.762 m x 1.016 m) was used for each manufactured plaque and a compression pressure of 500 tons was applied to the mould.

Table 1 presents the various process variables investigated in this study. Their effects on the mechanical performance and the quality of the composite plate were examined.

<table>
<thead>
<tr>
<th>Process parameters</th>
<th>Variables</th>
</tr>
</thead>
<tbody>
<tr>
<td>Injection speed (g/s)</td>
<td>35, 45, 65 and 85</td>
</tr>
<tr>
<td>Compression gap (mm)</td>
<td>0, 1 and 2</td>
</tr>
<tr>
<td>Vacuum sequences</td>
<td>No vacuum</td>
</tr>
<tr>
<td>Binder concentration and location</td>
<td>Vacuum applied during the filling and impregnation steps</td>
</tr>
<tr>
<td></td>
<td>0%</td>
</tr>
<tr>
<td></td>
<td>2% on each side</td>
</tr>
<tr>
<td></td>
<td>2.5% on each side</td>
</tr>
<tr>
<td></td>
<td>3% on each side</td>
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</tbody>
</table>

Mechanical tests

Tension tests (ASTM standard D3039 [17]) and 3-point bending tests (ASTM standard D790 [18]) were performed to assess the influence of the processing conditions on the mechanical properties of the composite panels. No aluminum tabs were used for the tensile specimen due to the multi-axial orientation of the glass fibre. Because of the large size of the panel and the possibility of property gradient within the panel, samples were taken at the top (injection side), middle and bottom (event side) of the panel.

An Instron 5582 testing machine was used with a load cell of 100 kN with the traction fixture for the tensile tests. The cross head speed was fixed to 2 mm/min. The composite flexural properties were measured using the three-point bending fixture with a 25 kN load cell on an Instron 55R11213 testing machine. The loading nose and the radial support had a diameter of 6 mm and 3 mm respectively and the cross head speed was set to 0.85 mm/min. The span between the supports was set to 32 mm. Five to six samples were tested for each mechanical test.
Fibre volume fraction and porosity

Following ASTM standard D2584 [19], sample pyrolysis was carried out to determine the sample fibre volume fraction and the void content. The sample was maintained in the oven for 2 hours at 600 °C. Up to nine samples per plaque from different location were burned.

Results

For all the tested conditions, no gradient in properties was observed between the samples taken from the top, middle or bottom of the panel. Therefore, in the following, only the results from the samples located in the middle of the panel will be presented.

Injection HP-RTM

Fibre alignment

Prior testing, an observation of the tensile sample showed fibre misalignment in certain cases. The fibre misalignment was characterized by the ratio of deviation length over the sample width. The fibre deviation was then plotted versus the resin injection rate, as shown in Figure 2. A large fibre misalignment was observed for fabric with no binder, especially with high flow rate. Binder help to reduce the fibre displacement, however, misalignment up to 45% were still observed for high flow rate (85 g/s). The fibre misalignment seems independent of the binder concentration and location, but flow rate dependant.

![Figure 2: Tensile sample fibre deviation with resin injection rate for injection HP-RTM process](image)

Tensile tests

Figure 3 shows the tensile modulus and tensile strength of glass/epoxy composite panels manufactured by injection HP-RTM under various conditions. The varied process parameters were the resin injection rate from 35 g/s to 85 g/s, the binder content and the vacuum sequence (no vacuum and vacuum applied during the resin injection and reinforcement impregnation). Figure 3-a and Figure 3-b present the tensile properties for the panels made without vacuum
assistance while Figure 3-c and Figure 3-d show the tensile properties of the panel made by applying vacuum during the process until the fibre impregnation.

From Figure 3-a, it can be noticed that the tensile modulus decreases with an increase in binder content. Up to 4% binder concentration on one face, the modulus averages is 25 GPa whereas it decreases to 20 GPa above 4% binder content. A slight increase in modulus can be also observed from 0% to 2% binder. On the other hand, the composite tensile strength remains constant around 400 MPa with an increase in binder concentration. Moreover, the resin injection rate does not affect the tensile properties. Similar trends were observed for the panel made with vacuum assistance (Figure 3-c and Figure 3-d). The use of vacuum assistance does not improve the composite tensile properties.

Flexural tests

Figure 4 shows the flexural modulus and flexural strength of glass/epoxy composite panels manufactured by injection HP-RTM under various conditions. Figure 4-a and Figure 4-b present the flexural properties of composite panels made without vacuum assistance while the
As for the tensile properties, the binder concentration is the parameter that affects the mechanical properties the most. A decrease of 20% and 30% in flexural modulus is observed from 4% binder concentration for panels made without vacuum and with vacuum assistance, respectively. A significant decrease in flexural strength, up to 50%, is also observed above 4% binder concentration for the panel made with vacuum. The resin injection rate does not affect the flexural properties, except at 0% binder. In that case, a lower injection rate induces higher flexural modulus and flexural strength.

Figure 4: Glass/epoxy composite flexural modulus and flexural strength made by injection HP-RTM under various process conditions.
Compression HP-RTM

Fibre alignment

Fibre misalignment was also observed for the composite plaques made by compression HP-RTM as shown in Figure 5. Similar trend as for the injection HP-RTM were observed with higher fibre deviation with fast flow rate (up to 60% fibre deviation for a resin injection at 85 g/s).

![Graph showing fibre deviation vs flow rate for compression HP-RTM process]

Figure 5: Tensile sample fibre deviation with resin injection rate for compression HP-RTM process

Tensile tests

The tensile properties of glass/epoxy composite panels manufactured by compression HP-RTM are presented in Figure 6. The process parameters were varied similarly as for the injection HP-RTM process (flow rate and vacuum sequence) with the addition of the influence of the gap size (1 and 2 mm). Contrary to injection HP-RTM process, no influence of the binder concentration was observed. The tensile modulus and the tensile strength appear to remain constant for all the condition tested. Also, the gap size had no effect on the properties.

In compression HP-RTM, the resin flows first in the empty space between the mould and the reinforcement and then impregnates the reinforcement through the thickness at the compression. In that case, the binder does not restrict the in-plane flow of the resin in the fabric, most probably leading to a better impregnation at high binder concentration. The use of vacuum reduces the results discrepancy and improves the tensile strength by approximately 10%.
Figure 6: Glass/epoxy composite tensile modulus and tensile strength made by compression HP-RTM under various process conditions

Flexural tests

The flexural properties of glass/epoxy composite panels manufactured by compression HP-RTM are presented in Figure 7. Similarly to the tensile properties, no influence of the binder, the resin injection rate or the gap size was noted for the panel manufactured with compression HP-RTM. However, the use of vacuum assistance reduces the scatter in property for a given condition and overall increases the flexural modulus and flexural strength by 15%.
Fibre volume fraction and porosity

Fibre volume fraction and porosity measurements were carried out on composite plaques with 6% binder made by injection HP-RTM with a resin flow rate of 45 g/s. An average fibre volume fraction of 52 ± 5 % was measured. Overall, less than 4% void content was measured in the composite plaques. From Figure 8, it can be noticed that the use of vacuum assistance until the fibre impregnation significantly helped to reduce the void content. The vacuum improves the void migration out of the preform toward the vent.
Discussion

Good quality large glass fibre reinforced epoxy composite panels were obtained with a highly reactive resin. This shows that the 50 second resin process window is sufficient to well impregnate the reinforcement. As the fabric impregnation does not seem to be an issue, the process cycle could be further reduced from 5 minutes to 3 - 4 minutes, still allowing the resin to cure at more than 95%. From the different HP-RTM process parameters tested, the binder concentration is the parameter affecting the most the mechanical performance of the manufactured composite plaques. The effect of binders and tackifiers on the mechanical properties of composite was investigated by other researchers and a reduction of the flexural properties, interlaminar strength and fracture toughness was often reported [20-24]. In particular, Shih and Lee [22] determined that the binder location in the fabric had a strong influence on the fabric permeability and thus the resulting mechanical properties. They found that binder located outside the tow can block the resin flow and decreasing the size of the empty channel between the tows and therefore reduces the permeability. On the other hand, binder located inside the tow helps the fabric permeability but also lead to an increase in void content which is detrimental for the mechanical properties. From the results obtained in this study, it can be assumed that for injection HP-RTM, low binder concentration does not affect the permeability of the quadraxial fabric and a good fabric impregnation is obtained. At higher binder concentration, it can be assumed that the binder starts to affect the fabric permeability and the fabric impregnation, creating void content and lower mechanical properties. In the case of the compression HP-RTM, the binder does not affect the resin flow, as the resin mainly flows through the thickness of the fabric. From the results of this study, low binder concentration on both sides of the fabric, vacuum assistance and medium resin flow rate appeared to be the most favorable parameters to make glass fabric reinforced epoxy composites by injection or compression HP-RTM. Compression HP-RTM seems to be a more versatile process as the composite performances were not affected by any tested process parameters. This might be suitable for the manufacturing of complex tri-dimensional geometry where high binder concentration is needed.
Conclusion

In this experimental study, the effect of HP-RTM processing conditions (resin flow rate, vacuum sequence, compression gap size, and binder concentration and location) on the mechanical properties of glass fibre reinforced composite plaques was investigated. Two process variants were examined: injection HP-RTM and compression HP-RTM. Both processes allowed the manufacturing of large composite plaques in less than 5 minutes. The cycle time could be further reduced based on the resin reactivity. In both case, the resin flow rate did not affect the mechanical properties of the composite but induced a fibre displacement during the resin injection. However this fibre displacement appeared to be controllable with the use of binder particularly below 65 g/s. The use of vacuum assistance slightly increased the mechanical properties and reduced the void content in the composite part. Binder concentration was determined to be the parameter affecting the most the composite mechanical properties, leading to decrease in mechanical performance from 4% binder concentration on one side of the fabric.

From this study, it is suggested that low binder concentration on both sides of the fabric, vacuum assistance and medium resin flow rate are the most favorable parameters to make glass fabric reinforced epoxy composites by injection or compression HP-RTM.

This study demonstrates the great potential of HP-RTM processes to manufacture high performance composite for high volume production such as in automotive industry.

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