Effect of Material and Process Variables on the Performance of Resin-Transfer-Molded Epoxy Fabric Composites

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ABSTRACT: The effects of material and process variables on glass fabric-reinforced epoxy composites by the resin-transfer molding (RTM) process were studied. It was found that the molded aged resin with 55% fiber exhibited twice the mold-filling time and caused a 7–15% deterioration in the interlaminar shear strength (ILSS) and in the flexural strength of the composites as compared to those of the composites molded with fresh resin. At a 55% fiber volume fraction, composites molded with aged resin resulted in a 35% longer filling time and a 4–12% decreased ILSS and flexural strength as compared to those of the composites at a 44% fiber volume fraction. Moldings with a perimeter inlet exhibited a 65% shorter mold-filling time, 28% reduced void content, and 6% improved flexural strength as compared to those of the composites molded with the center inlet. © 2000 John Wiley & Sons, Inc. J Appl Polym Sci 77: 2149–2155, 2000

Key words: epoxy; composites; material and process variables; resin-transfer molding (RTM); mechanical properties

INTRODUCTION

The resin-transfer molding (RTM) process for the production of fiber-reinforced composites has attracted much interest from aerospace and automotive to power machinery industries because of the potentially high rate of production and the high quality of the finished parts.[1–4] Large polymer composite parts with complex shapes can be efficiently fabricated by the RTM process as compared to the traditional prepreg layup/autoclave cure process.[2,4] The RTM process is a closed-mold processing technique, which involves loading the fiber preform into a mold cavity, followed by an injection of a low-viscosity thermoset polymer resin under pressure and at elevated temperature and then curing the resin *in situ* to form the final parts.[4] The critical step in RTM is to impregnate the preform as quickly as possible while minimizing undesirable features such as void content or nonuniform wetting of the preform.[5]

A large number of variables are related to the RTM processes: the resin characteristics, fiber preform, resin preheated temperature, mold temperature, injection pressure, vacuum assistance, gating method, and mold geometry.[5–12] The design and the control of the critical variables of the RTM process are needed in the process-development stage to maintain optimal conditions.

The Darcy law has been used to describe the resin flow in the filling stage of RTM processes. The viscosity of the resin and the permeability of the fabric preform are the two important parameters in the Darcy law that relates to the proper

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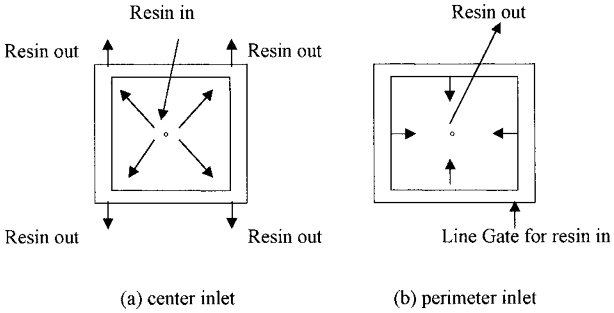
fiber wet-out and the impregnation rate of the fabric preform.[13,14] The viscosity of a thermoset resin is a function of temperature and the degree of cure (chemical reaction).[15] Resin aging arising from an unexpected prolonged storage under ambient conditions may result in the curing of the resin and an increase in its viscosity. Hayward and Harris[6] found that a high-viscosity resin will not properly penetrate the fiber and wet-out the fibers in the RTM molding with 55% fiber as compared to that with 40% fiber in the case of a polyester resin. Akay[16] and Berglund and Kenny[17] studied the aging effect on carbon/epoxy prepregs for an autoclave process. It was found that aging increased the activation energy of the resin but caused no deterioration in the mechanical strength of the resulting composites. The fiber volume fraction of the composites has the most significant impact on the overall strength of the composites.[8] However, an increase in the fiber fraction provides resistance to resin flow and affects the cycle time of the RTM process.[8,9] Hansen[10] and Abraham and Mcllhagger[11] found that the mold filling and the part quality by the perimeter inlet could be controlled much better than that by the center inlet in the RTM process. Cai[12] demonstrated that the mold-filling time by the perimeter inlet is reduced substantially as compared to that by the center inlet in a simplified RTM process analysis.

Although various studies have attempted to characterize the RTM process, most of the previous studies on RTM have been concentrated on flow modeling.[5,13,18,19,20] Studies concerning the effects of material and processing variables on high volume fraction fiber composites by RTM are lacking. The effects of process variables on the physical and the mechanical behavior of an one-part high-performance epoxy composite produced by the RTM process were investigated.[21] Variables studied include injection pressure, injection temperature, and fabric structure.

In this study, the effect of the material and process variables on the performance of glass fabric-reinforced composites based on a two-part epoxy resin were investigated. The composites were produced by the RTM process under constant pressure and isothermal filling conditions. Variables studied include resin aging, fiber volume fraction, and gating arrangement. The mold-filling time, the void content, and the matrix-dominated properties, such as the interlaminar shear strength (ILSS), the flexural strength, and the dynamic mechanical properties of RTM composites are presented.



(a) center inlet
(b) perimeter inlet

Figure 1 Different gating arrangements in the RTM process.

EXPERIMENTAL

Materials

The resin used was a two-part epoxy/amine resin LY564/HY2954 supplied by Ciba–Geigy (Hawthorne, NY). The mixing ratio of the base resin LY564 to the hardener HY2954 is 100:35 by weight. Detailed information pertaining to this resin can be found elsewhere.[22] Both the fresh and the aged resin were used for the processing of RTM moldings. The aged resin is designated as the matrix resin and the hardener, which has been stored in a separate container under ambient conditions of 70% relative humidity and 25°C temperature up to 18 months. The fabric reinforcement used was a highly permeable glass fabric from Brochier (Decines Cedex, France), Injectex EF420-E01-100, with a surface density of 420 g/m^2.

Preparation of RTM Composites

A flat rectangular aluminum mold with the dimensions of $0.325 \times 0.325 \times 0.003$ m was used for the preparation of the RTM composites. It comprised a top and a bottom half as well as a picture frame with a center gate or a perimeter line gate at the corner on the top half, clamped together with 12 evenly distributed bolts around the mold edge. Both the center inlet and the perimeter inlet, as shown in Figure 1, were used in this study. A rubber gasket was placed around the perimeter of the mold halves to provide a proper seal. Heat blankets were used for the heating of the mold and the resin pot. Ten and eight layers of glass fabrics of EF420 were used to produce

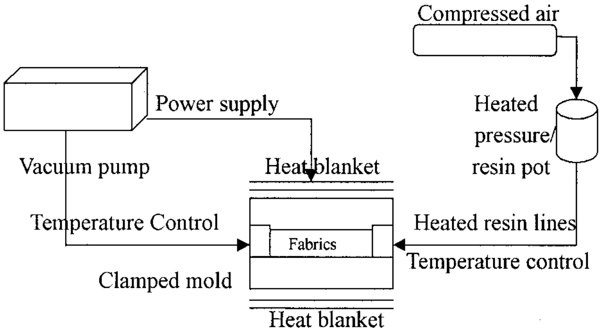


Figure 2 Experimental setup for the RTM process.

Table I Standard Settings of Process Variables for EF420/LY564 RTM Composites

Variables	Settings
Transfer pressure	147 kPa
Mold temperature	70°C
Resin temperature	35°C
Cure condition	140°C, 4 h
Fabric temperature	70°C
Resin viscosity at 25°C	617 mPa s
Pack pressure	196 kPa
Fiber volume fraction	55 ± 2%
Nominal thickness of panels	3.0 mm
Gating arrangement	Center inlet

3-mm-thick plates of of a 55 and 44% fiber volume fraction, respectively. The resin was preheated and degassed at 35°C for 20 min. Then, the resin was injected into the mold maintained at 70°C under 147 kPa. The injection pressure was monitored with a Flutterless® gauge by adjusting the air pressure in the resin pot. The mold was vented to the atmosphere once the resin injection started. Two J-type thermocouples neighboring to the center inlet and the perimeter inlet were used to monitor the temperature throughout the whole process. After the resin flowed out from each outlet, the outlets were opened and closed alternately several times to eliminate any entrapped air in the mold cavity. Upon completion of the mold filling, the mold temperature was increased to 140°C for 4 h to cure the plates. The cured plates were then cooled, demolded to the traditional, followed by examining for quality by ultrasonic C-scanning. The average thickness variation from the center to the edge of the molded samples was 0.14 mm, which indicates that the injection pressure exerted on the mold caused the mold to open slightly. Figure 2 displays the experimental setup for the RTM process. Except when a specific variable was studied, the RTM composites were produced under the standard settings of the process variables as given in Table I.

Measurements of Viscosity and Dynamic Mechanical Properties

The resin viscosity and the dynamic mechanical properties, including storage modulus (G'), loss modulus (G''), and loss tangent (tan δ), of the RTM composites were measured with a Rheometrics mechanical spectrometer Model RMS-605 in accordance with the specification of ASTM D 4473 and ASTM D 4065, respectively. Viscosity measurements were performed with disposable parallel plates of 25 mm in diameter, which were subjected to the forced oscillations with a gap of 0.5 mm. A time sweep was conducted to obtain the viscosity–time profile at 70°C. The composite specimens were subjected to the forced oscillations at a frequency of 1 Hz and 0.08% strain over the temperature range of 25–250°C. The maximum value of G''/G', that is, tan δ, was taken as the glass transition temperatures of the composites.

Testing of Physical and Mechanical Properties

Ultrasonic C-scan inspection and optical image analysis were used to investigate the void content in the composites. The composite plates were first inspected using the ultrasonic C-scan test. The C-scan analysis provided basic information of the

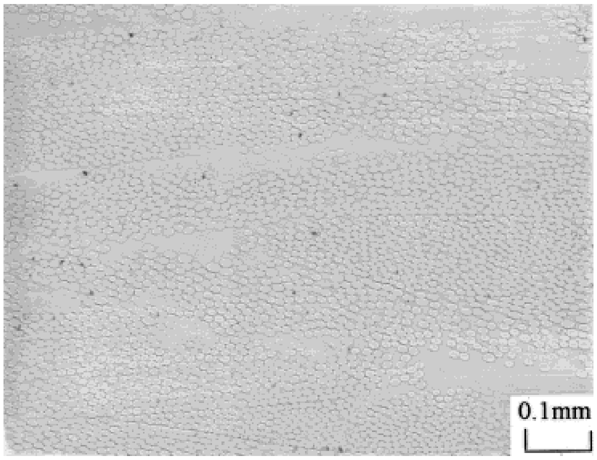


Figure 3 Microphotograph of the cross-sectional view of EF420/LY564 RTM laminates.

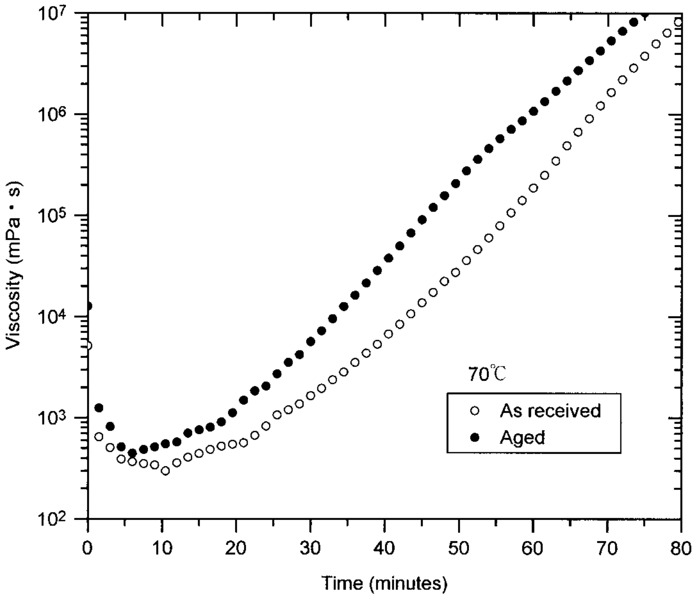


Figure 4 Viscosity profiles as a function of time for as-received and aged LY564/HY2954 resin at 70°C.

void distribution in the composites. After the C-scan analysis, the void content was determined from polished cross sections of the plate with a computerized image analyzer coupled with a ZEISS optical microscope. The detailed procedures of the ultrasonic C-scan and image analysis are described elsewhere.[21] A typical microphotograph of the cross-sectional view of EF420/LY564 RTM laminates is shown in Figure 3. The voids were identified as the black dots in the figure. The ILSS was tested in accordance with ASTM D 2344 using a three-point loading fixture with the span-to-depth ratio of 5. The three-point flexural tests were run in accordance with ASTM D790 at a span-to-depth ratio of 32. Seven specimens were measured for each ILSS and each flexural test.

RESULTS AND DISCUSSION

Effect of Aging

The resin flow in the RTM process depends strongly on its viscosity, which is a function of temperature and the degree of cure.[15] The evolution of viscosity during mold filling at 70°C for the aged and the as-received (fresh) LY564/HY2954 resin is shown in Figure 4. In Figure 4, the aged resin exhibits a higher viscosity than that of the fresh resin. Hence, the time period allowed for the mold filling, at which the viscosity of the aged resin is below 1000 mPa s, was reduced. The increase in the viscosity of the aged resin is probably due to the formation of a polymer network arising from its prolonged storage. In RTM, mold filling can be treated as a pseudo-steady flow of Newtonian fluids through porous media obeying the Darcy law. Hence, the average resin velocity is proportional to the average pressure gradient, but is inversely proportional to the resin viscosity. An increase in the resin viscosity leads to a decrease in the resin flow rate and an increase in the mold-filling time under the same injection pressure. Aged resin exhibited almost twice as much mold-filling time and void content as that in the case of the as-received resin, as given in Table II. The influence of resin aging on the mechanical properties of the EF420 glass fabric composites are also given in Table II.

In Table II, the interlaminar shear strength (ILSS) and the flexural strength of the plates molded from aged resin were 7–15% inferior to that of the plates molded from the fresh resin. The decrease in ILSS and flexural strength was caused by an increase in the void content in the plates of the aged resin, which led to a reduction of the cross-sectional area and an initiation of failure from large discrete voids.[23] Voids appeared because the penetration of the fabrics and the wet-out of the fibers were hindered by the higher viscosity of the aged resin as discussed by

Table II Effect of Aged Resin on the Mechanical Properties and the Filling Time of EF420/LY564 Composites

Resin Conditions	ILSS (MPa)	Flexural Strength (MPa)	Flexural Modulus (GPa)	Void Content (%)	T_g (°C)	Filling Time[a] (s)
As-received	50.3 (4.8)	393.7 (12.4)	22.8 (1.4)	0.83 (0.49)	153	193
Aged	42.8 (1.4)	365.4 (15.8)	22.1 (0.7)	1.60 (1.10)	139	405

( ) denotes standard deviation.

[a] Filling time obtained as the slowest resin flow-out.

Chen et al.[24] The ILSS often decreases with increasing void content.[25] Flexural strength usually serves as an indicator of the reinforcing efficiency of the plate.[6] The difference in the flexural modulus of the composites molded from the aged resin and the fresh resin was very small because the modulus is largely controlled by the fibers. The aged resin also resulted in the deterioration in the storage modulus (*G′*), the early peak of loss modulus (*G″*), and the lower glass transition temperature (T_g), as shown in Figure 5 and given in Table II, due to the poor wetting and the poor quality in the resulting composites.

Effect of Fiber Volume Fraction

The fiber volume fraction is one of the most important factors affecting the mechanical strength of high-performance composites. However, more fibers can retard the flow of resin in the RTM process. The aged resin was used to produce RTM composites of different fiber volume fractions for comparison. The mold-filling time increased by 35% when the fiber volume fraction increased from 44 to 55% as given in Table III. This can be attributed to more resistance to the resin flow in the fabric reinforcement owing to the decreased permeability of the fabric preform. The leading of either the macroflow fronts around the fiber bundles or the microflow fronts within a fiber bundle is likely to form the voids during the filling stage.[18–20] Thus, voids in the 44% fiber fraction composite were 66% less than that of the 55% fiber fraction composite as given in Table III. The difference of the impregnation rates between the

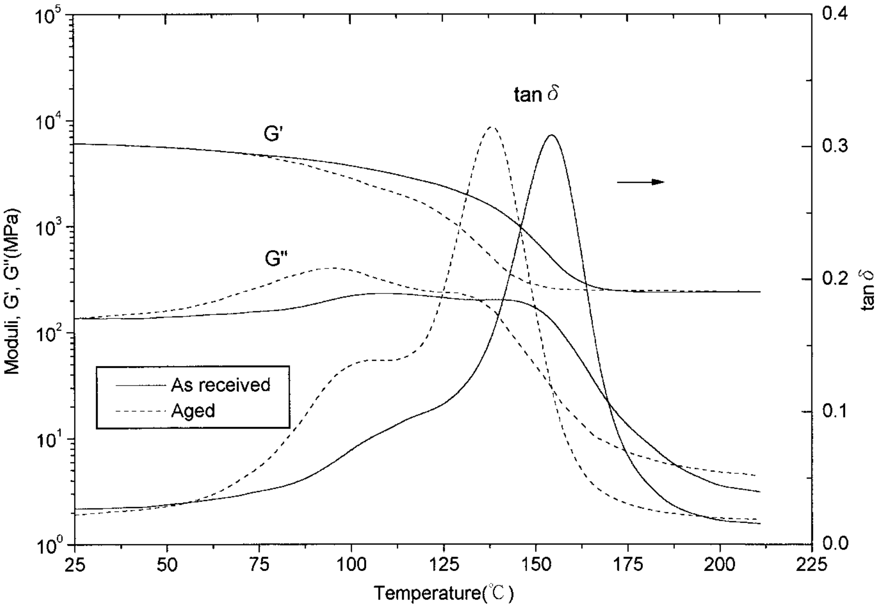


Figure 5 Storage modulus (*G′*), loss modulus (*G″*), and loss tangent (tan δ) as a function of temperature for EF420/LY564 composites produced with both as-received and aged resin.

Table III Effect of Fiber Volume Fraction on the Mechanical Properties and the Filling Time of EF420/LY564A Composites

Fiber Volume (%)	ILSS (MPa)	Flexural Strength (MPa)	Flexural Modulus (GPa)	Void Content (%)	T_g (°C)	Filling Time[a] (s)
44	48.3 (2.8)	382.0 (13.8)	19.3 (0.7)	0.54 (0.38)	134	300
55	42.7 (1.4)	365.4 (15.8)	22.1 (0.7)	1.60 (1.10)	139	405

LY564A: aged resin. ( ) denotes standard deviation.
[a] Filling time obtained as the slowest resin flow-out.

macropores of fiber bundles and the micropores among individual fiber filaments was diminished by having a lower fiber fraction. The 44% fiber fraction composite had a higher flexural strength and a higher ILSS than that of the 55% fiber fraction composite as a result of a reduction of voids in the resulting composites, as listed in Table III. However, the decreased fiber fraction leads to a lower flexural modulus and a lower T_g, as given in Table III, due to the increased resin content and the significant loss of stiffness in the composites. In general, glass reinforcement imposed the additional constraint/stiffness on the resin matrix in a similar way as does increased crosslinking.[26] Crosslinking increases the glass transition of a polymer by introducing restrictions on the molecular motions of a chain.[27]

Effect of Gating Arrangement

The center inlet and the perimeter inlet for the moldings were used to assess the effect of different gatings on the performance of the RTM process and the resulting composites. The mold-filling time was dramatically reduced for the moldings with the perimeter inlet as compared to that with the center inlet, as given in Table IV. The reduction in the mold-filling time in the perimeter inlet case can be explained by the fact that the much higher flow rate in the perimeter line gate and a relatively low resin viscosity at the initial stage of mold filling resulted in such a large reduction of the mold-filling time. For the molding with the center point inlet under the same pressure, the inlet flowrate is relatively small and decreases with time in an outward direction.[11,12] Therefore, the mold-filling rate slows down and the resin viscosity increases with time, and voids are, therefore, formed. It is evident that the difference in the impregnation rates between the macropores and micropores facilitated the formation of voids in the moldings with the center inlet. As shown in Table IV, moldings in the center inlet case exhibited a much higher void content than that in the perimeter inlet case. Additionally, decreases in the ILSS and the flexural strength were observed for the moldings in the center inlet case as compared to those in the perimeter inlet case. As expected, the two moldings exhibited about the same glass transition temperatures (T_g). Evidently, the perimeter inlet is a better gating arrangement for mold filling in the RTM process as compared to the center inlet.

CONCLUSIONS

Resin-transfer molded glass fabric composites based on a two-part epoxy were produced under

Table IV Effect of Different Gating on the Mechanical Behavior and Filling Time of EF420/LY564 Composites

Gating Arrangement	ILSS (MPa)	Flexural Strength (MPa)	Void Content (%)	T_g (°C)	Filling Time (s)
Center inlet	50.3 (4.8)	393.7 (12.4)	0.83 (0.49)	153	193[a]
Perimeter inlet	51.7 (3.7)	419.2 (15.8)	0.60 (0.14)	152	68

( ) denotes standard deviation.
[a] Filling time obtained as the slowest resin flow-out.

various material and processing conditions. Variables investigated included resin aging, the fiber volume fraction, and the gating arrangement. Aged resin resulted in a twofold mold-filling time, 7–15% reduced mechanical properties, and a lower glass transition temperature in the composites as compared to those of the composites molded from fresh resin. The increase in the glass fiber content from 44 to 55% led to a 35% longer filling time and a 4–12% decrease in the mechanical properties because of the increased void content. Molding in the perimeter inlet case exhibited a 65% less mold-filling time, a 28% lower void content, and a 6% better flexural strength as compared to the molding in the center inlet case.

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